# MODELING OF GRANULATION IN HIGH SHEAR MIXERS

**E. Masanja**
Chemical & Process Engineering, University of Dar Es Salaam,
P.O. Box 35131 Dar es Salaam, Tanzania.

## ABSTRACT

*Granulation experiments were carried out in a high shear mixer/granulator, type fm 50. The influence of varying the binder liquid viscosity was investigated. Population balance modeling was made out using Hounslow's sectional method.*

*The result of the experiments are in conformity with theory and indicate that binder liquid viscosity has a pronounced effect on agglomeration behaviour. At high viscosities, nucleation was the only growth mechanism observed; this occurred concurrently with breakage. In contrast, at lower viscosities other mechanisms, like coalescence and crushing, and layering, were noted. Breakage was also noted, the causes of this phenomenon is still unknown, most likely it is caused by the ploughshares, however no experimental proof was found. More research is needed to clarify this mechanism.*

*There is still scope for improving the modeling, e.g. to include optimisation procedures which can save considerable computer time and also to develop and/or test other kernel forms.*

## INTRODUCTION

Granulation is an indispensable technique for improving powder properties. The addition of a dry or wet binder to a powder mixture and the continuous colliding of the particles will cause the particles to sick together and thus grow. Simple as it might seem to be, granulation is still not a well understood.

There are several advantages in using granules instead of powders, e.g. homogeneous enzyme activity of the washing powder can be obtained, when the granules are of comparable size and density to the other ingredients of the washing powder. The flowability of granules is much

better than that of powders, granules have a higher density than powders, which makes them more compact and easy to transport. Also granules are considerably less dusting. There is a range of equipment suitable for making granules: rotating drums, fluidized beds, high-speed granulator and high-shear mixers.

## THEORY

### Granulation Mechanisms

Granulation takes place in several regimes, all of which consist of one or more mechanisms. The mechanisms and regimes which are most important during granulation in the high shear mixer are nucleation, compaction/coalescence, crushing and layering, and breakage stages.

### Nucleation stage

This stage is called non-inertial regime by Ennis [1]. In this stage every collision is successful, when two particles collide, they stick together instead of rebounding. As pointed out by Ennis [1], the most important parameter is the liquid viscosity and not surface tension, which was originally thought to be the most important parameter. During collisions, the liquid sublayer between sub-particles is deformed, turning them into dynamic bonds, making liquid viscosity an important parameter. Two particles will stick together after a collision depending on the magnitude of the kinetic energy of the particle dissipated. This is predominantly determined by the binder liquid viscosity and not by the surface tension. The surface tension does play a role in stabilising the formed granules. Nucleation stage lasts as long as the viscous Stokes' number is smaller than the so called critical viscous Stokes' number, i.e.:

$$St_v < St_v^*$$

or

$$\frac{8\rho_g \bar{r} u_o}{9\eta} < \left[1+\frac{1}{e}\right]\ln\left[\frac{h}{h_a}\right] \qquad (1)$$

equation (1) (and also later on equation (2) are of a more qualitative value.

After every successful collision, liquid creeps into the newly formed interstitial voids, due to the surface tension. This causes the thickness of the liquid layer to diminish and thus lowering $St_v^*$. Also the particles become larger during nucleation, thus raising $St_v$. The adding up of these two effects causes $St_v$ to become equal to $St_v^*$, and nucleation ends.

### Compaction stage

This stage is frequently called the equilibrium stage, because no growth is observed. The reason for this is that the internal structure of the granules keeps changing. During this stage, liquid is squeezed out onto the surface again until the condition for successful collisions as stated in equation (1) is met. When particles are struck, the smaller sub-particles creep in the interstitial spaces and press the liquid out onto the surface. This effect can also be attributed to the structure and operation of the high shear mixers, e.g. a chopper in the mixers might be responsible for the rearrangement of the granules. It is obvious that the duration of the compaction stage will depend upon parameters such as binder liquid amount and viscosity, and initial particle size distribution. For instance, when using a more viscous binder liquid, the time for compaction will increase.

### Coalescence stage

This is the so-called inertial stage. In this stage not all the collisions are successful. Growth is preferential towards the larger particles, because of their higher deformability. This is in complete contrast with the nucleation stage, where growth is presumed to be random. After each successful collision the granule is compacted again until enough liquid has been squeezed out, so that further coalescence becomes possible. During this stage equation (1) is still valid, however, throughout this stage the viscous Stokesí number is approximately equal to the critical Stokes' number, which explains why after every collision compaction is

Stokes' number, which explains why after every collision compaction is necessary.

### Crushing and layering

During the coalescence stage, the governing mechanism shifts from coalescence to crushing and layering, which has a much lower growth rate. During crushing and layering the smaller particles are crushed, while the larger particles pick up the fragments from the crushed smaller particles. This growth phenomena is preferential towards the larger particles.

### Breakage

When conditions are favourable, particles can reach considerable sizes. Once this happens, breakage, which is highly undesirable, can occur although does not occur in a rotating drum like equipment. As yet, not much research has been carried out in this field.

### Data analysis and Presentation

In order to get more insight on the mechanisms and processes occurring during granulation, one can use the graphs of the size distributions of the different samples. These give more information than just the mean particles size.

In size distribution plots, it is of utmost importance that the right width is chosen. A third type of graph also used to study particle growth is the reduced particle size distribution (Fig.1). This graph is comparable to a normal cumulative undersize distribution but instead of using the absolute particle size, a dimensionless size is used. In this case the particle size is divided by the mass mean diameter. This is based on the fact that when particles grow by one breath mechanisms, the reduced size distributions fall together. The three graph types mentioned in this section will be used throughout this paper in order to present the experimental results.

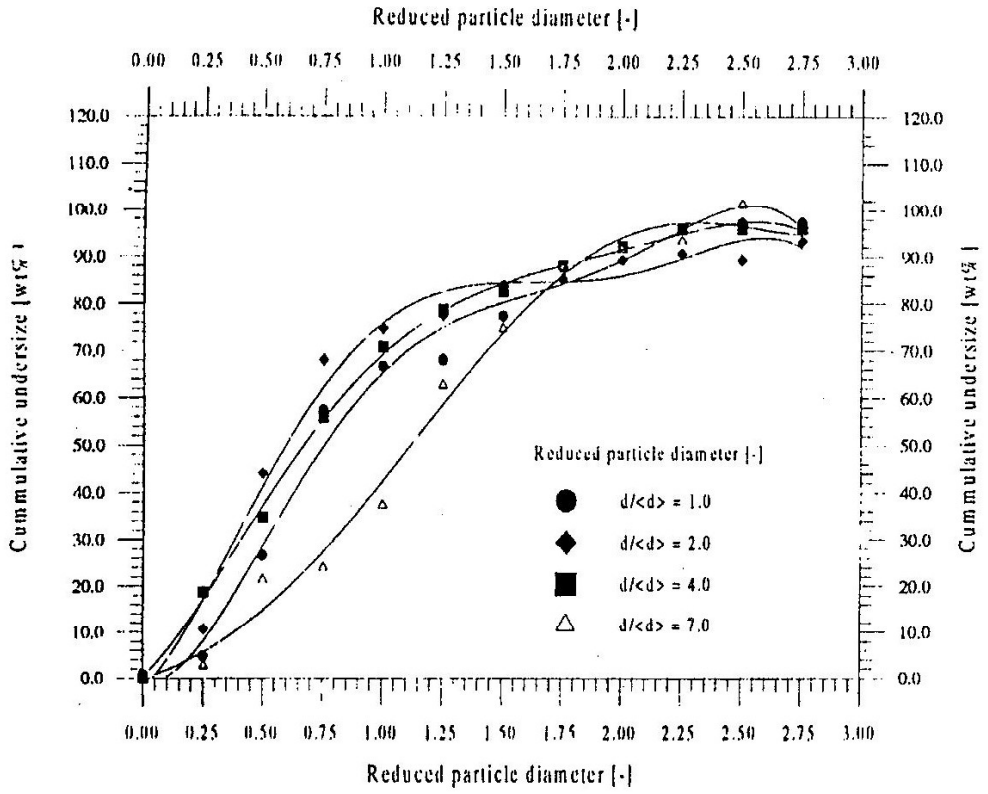


**Fig. 1: An example of a reduced particle size distribution, experimental result for initial binder viscosity = 16.4 mPas**

## Modeling

A common technique in modeling particle growth is population balance modeling. Here Hounslow's sectional method [2,3] was taken as a basis. This method relies on discretisation of the particle size distribution using particle volume as the internal coordinate. The smallest particles in a class i have a volume of $2^i$ and the largest particles have a volume of $2^{i+1}$. In other words: $v_i = 2v_{i-1}$. This method has the convenience of describing a wide particle distribution with substantially less classes, which shortens computation time considerably.

For every class i four mechanisms play a role in the population balance:

- Birth of a particle, due to a successful collision between two particles from class i-l (this will always lead to particles belonging to class i).
- Birth of a particle, due to a successful collision between a particle from class i-l and a particle from a lower class (this will not always lead to particles belonging to class i).
- Death of particle, due to a successful collision between a particle from class i and a particle from class i and a particle from class i or a higher class (this will always lead to the formation of a particle with a volume larger than 2i+l).
- Death of a particle, due to a successful collision between a particle from class i and a particle from a lower class (this will not always lead to particles with a volume larger than 2i+l).

Following Hounslowís procedure, the overall equation is given [3]:

$$\frac{dN_i}{dt} = \frac{1}{N_T}\left[ N_{i-1}\sum_{j=1}^{i=2} 2^{(j-i-1)}\beta_{i-1,j}N_j + \frac{1}{2}\beta_{i-1,j-1}N_{i-1}^2 - N_i\sum_{j=1}^{i=1} 2^{(j-i)}\beta_{i,j}N_j - N_i\sum_{j=1}^{\infty}\beta_{i,j}N_j \right] \qquad (2)$$

where Ni is the number of particles in class i at a certain moment and i,j is the so-called coalescence kernel (which represents the fraction of all possible collisions between particles from classes i and j that lead to agglomeration, per unit time).

Although the coalescence kernel is a very important parameter, it can not be derived theoretically, because it is impossible to determine all the forces which play a role during granulation. Therefore, a semi-empirical coalescence kernel will be used. Usually the kernel consist of two parts; i.e. a constant and a volume-dependent function

$$\beta_{i,j} = kf(v_i, v_j) \qquad (3)$$

In this modeling, two types of kernels were used: the random kernel, which is a constant volume function and therefore shows no preference towards any of the growth classes, and the sum kernel, this shows

$$\beta_{i,j} = kf(v_i + v_j) \tag{4}$$

## EXPERIMENTS

### Granulation Experiments

All experiments were carried out in a batch wise operated high shear mixer, type fm 50, which had a total volume of 50 litres, and was equipped with three ploughshare shovels attached to a rotating horizontal shaft and a side-mounted stationary spinning chopper, as shown in Fig. 2. The rotational speed of the ploughshares was variable, the clearance distance between the wall and ploughshares was set at 3100 μm. In order to prevent excessive heating of the granulating mass, the high shear mixer was equipped with a cooling jacket.

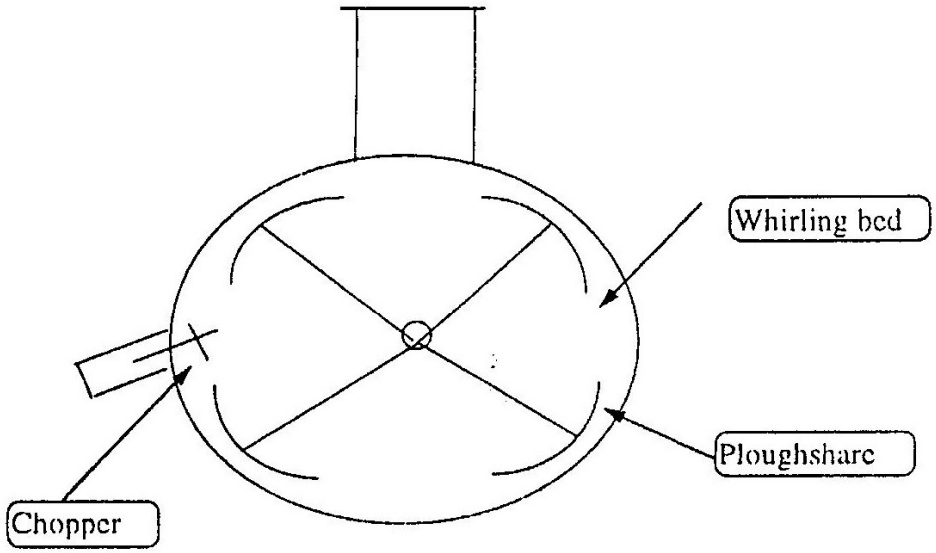


**Fig. 2: A schematic representation of the shear mixer.**

The experiments were carried out by adding 3.3. kg of water (which served as a binder liquid) to a homogenised powder mixture. At regular time intervals the temperature was recorded and also sample were taken

time intervals the temperature was recorded and also sample were taken for analysis.

## RESULTS AND DISCUSSION

A FORTRAN program which was not yet optimised was used to fit experimental data to the different theoretical kernel forms [3].

### Experimental Results

After the addition of the binder liquid to the high shear mixer, the viscosity changed drastically due to the increased temperature and the dissolution of the filler ($Na_2SO_4$ in these experiments). Viewed in the light of equation (1), it is easy to see that due to a higher viscosity the duration of the nucleation stage was longer and thus the particles grew larger. From equation (1) it is clear that the higher the viscosity the longer will be the compaction stage. This is due to the fact that it takes more energy to squeeze out a more viscous liquid.

Growth in the coalescence regime was faster. Again the increased dissipation caused more collisions to be successful. The fact that the increased viscosity also caused slower compaction seemed of less importance. Finally, growth in the crushing and layering regime was not as fast with the more viscous binder liquid. The particles became less sensitive to breakage due to the increased energy dissipation, and because of that there was less smaller particles at hand to provide for the layers on the large particles.

It was expected that the lower the initial binder liquid viscosity the shorter will be both the nucleation stage as well as the equilibrium stage. These stages might not even be measurable, meaning they might last for a shorter time than the sampling time interval. If the theory is extended, it can be envisaged that growth in the nucleation stage is slower (due to the lower viscosity dissipation due to considerably low energy dissipation) while crushing and layering will be faster (the particles are broken more easily).

This was done for all experiments, and the results are in Figs. 3 and 4.

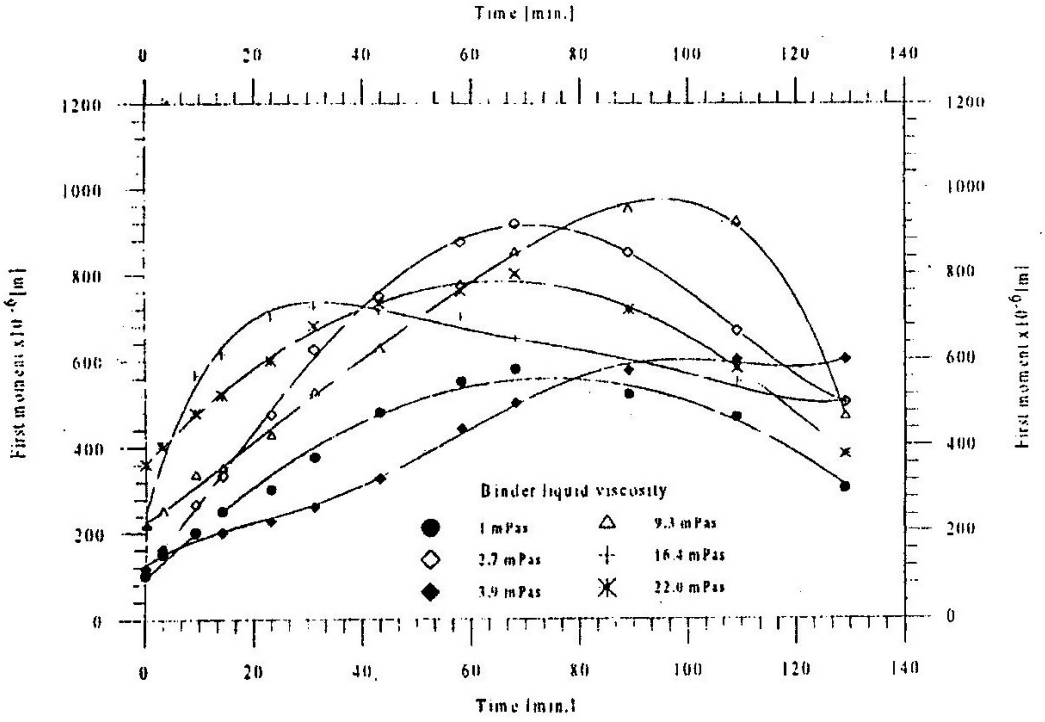


**Fig. 3: The history of mass mean diameter for various binder liquid viscosities.**

From Figs 3 and 4 it can be seen that the lower the binder viscosity, the shorter the nucleation stage. It is generally excepted that the nucleation stage will end well before the first sampling time (after 1 minute) One can also conclude from Fig. 3 that equation (1) is indeed valid, because the extent of nucleation, which is defined by the relative growth of the particles in the nucleation stage, was higher with higher viscosity.

It was also observed that both the nucleation and the compaction stages are terminated before the first sample was taken. From t = 7 minutes growth took place solely by one mechanism, being coalescence.

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The particle size distributions prove that crushing and layering took place, since the amount of smaller particles remained unaltered. The medium particles diminished and the larger particles grew. This indicates that the medium particles were crushed and the large particles took up the small fragments, thus explaining why the maximum in the particle size distribution shifted to the right and the amount of small particles stayed about the same.

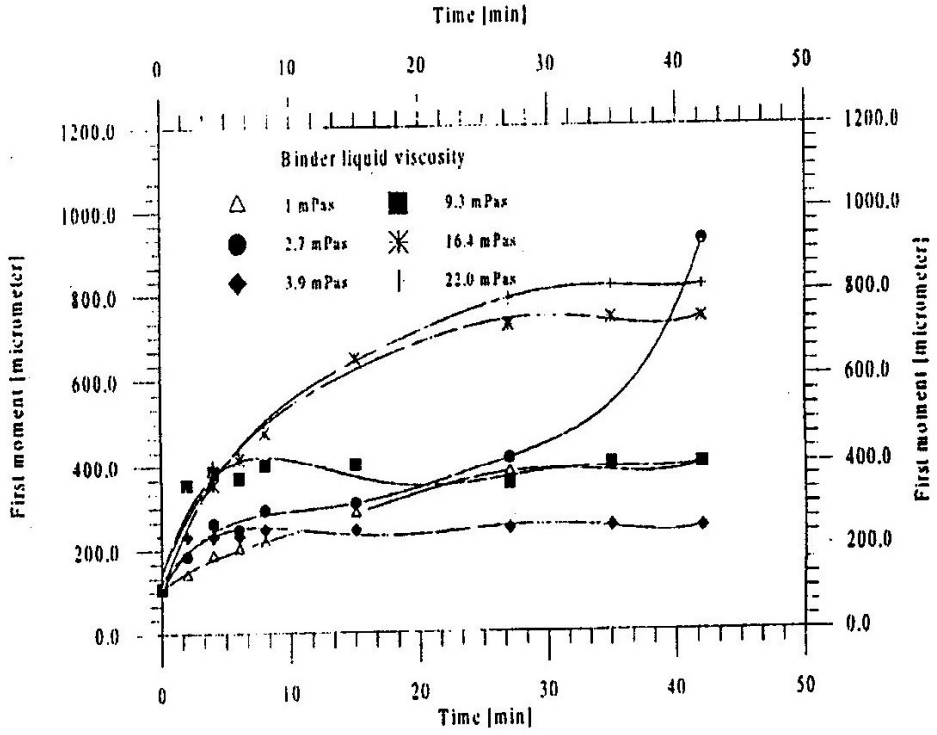


**Fig. 4: The course of the mass mean diameter in time, first stages**

Obviously, the governing growth mechanism was shifting. It is postulated that the governing growth mechanism is of `hindered nucleation'. This trend can be divided into a stage of fast and slow growth. During the first seven minutes the fast growth took place, where the number of small particles ($d < 200\ \mu m$) diminished, the large ($d > 512.5\ \mu m$) and medium particles ($200\mu m < d < 512.5\ \mu m$) grew. From t = 7 minutes till t = 30 minutes growth was slower. Here, the

yet known what triggered this breakage.

It is expected that both the nucleation stage as well as the equilibrium stage will last shorter with lower initial binder liquid viscosity. They might not even be measurable, meaning they will last shorter than the time in between two samples. When the theory is extended, it can be envisaged that growth in the nucleation stage will be slower (due to the lower viscosity dissipation will be considerably less) while crushing and layering will be faster (the particles are broken more easily).

An explanation for the agglomeration behaviour for these experiments is that both nucleation and breakage occur concurrently throughout the experiment and that breakage gradually becomes more important. This would explain the hindered nucleation during the first minutes of the experiment. However, as stated before, not much is known about breakage, so no definite conclusion can be drawn from these experiments as well.

One unexpected result is the high growth rate during coalescence. Obviously the low viscosity, which makes it easier for the binder liquid to be squeezed out of the interstitial space during rearranging after a successful collision. Again, the extent of nucleation, the compaction time and the rate of coalescence will be expected to be higher. However, when looking at Fig. 2, it becomes clear, that the results do not differ substantially from the experiment with binder liquid viscosity 16.4 mPas. From these experiments the thought arises that at high viscosities 16.4 mPas changes in viscosity have little or no effect.

## Modeling Results

A number of experimental results were fitted to the model. The nucleation stage and the first few minutes of the coalescence stage (if this stage was entered) were modeled.

When studying both the random and the sum kernel for the nucleation stage, one can see that when more binder liquid is present the kernels are larger. These kernels represent the average rate of growth during the first minute. Although the rate of nucleation is equal for all cases, the extent

stage, one can see that when more binder liquid is present the kernels are larger. These kernels represent the average rate of growth during the first minute. Although the rate of nucleation is equal for all cases, the extent of nucleation varies. Since it is known that nucleation is terminated within 1 minute, the average growth in the first minute, is thus proportional to the difference in values for the growth kernels in the first minute. If the kernels are plotted against the solution phase ratio 1 a linear fit can be obtained (Fig. 4).

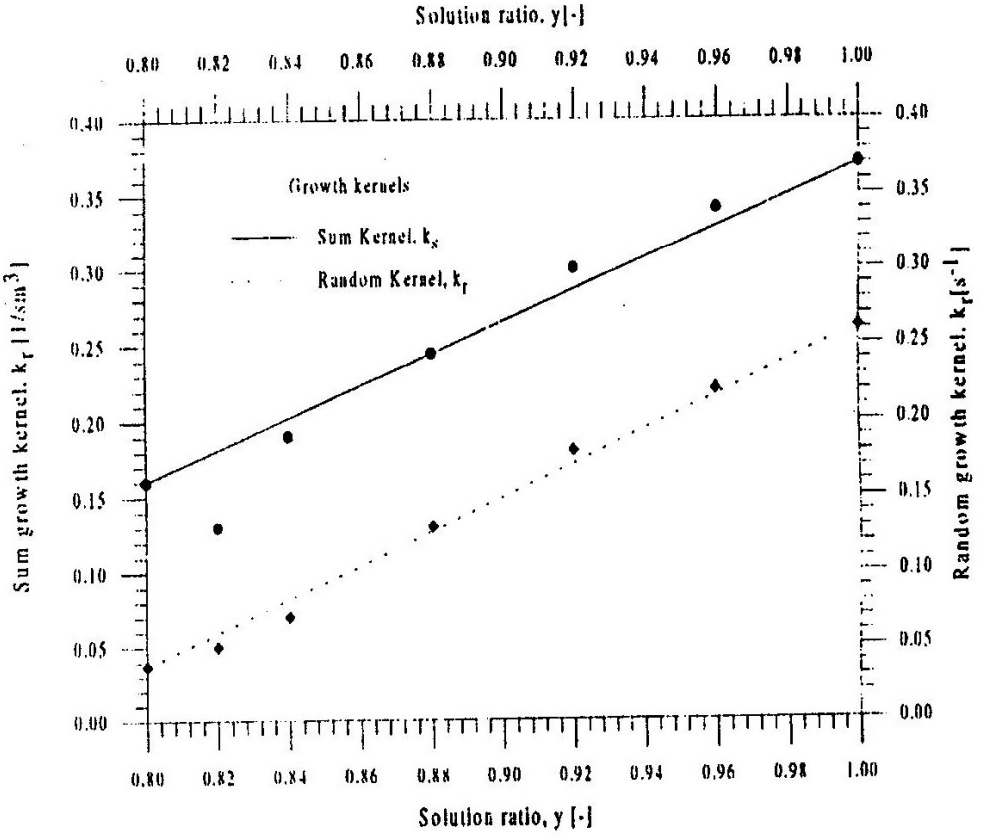


**Fig. 5: Dependence of random and sum growth kernel on solution phase ration y during nucleation stage**

The results yielded for the sum kernel: $k_{1,s} = 1.05y-0.68$ for the random kernel: $k_{1,r} = 1.12x10^{-11}-8.59yx10^{-12}$. This is a very important result, which will make predictions concerning granulation in the nucleation

kernel became larger. This enhances the idea that growth in the coalescence stage is preferential towards the larger particles, which is in accordance with earlier findings [1,2].

When looking at the least favourable result, where the experimental points lie exactly between the sum kernel fit and the random kernel fit, an alternative, n exponential kernel might be used

$$\beta_{i,j} = k(v_i + v_j)^{\alpha} \tag{5}$$

where (in this case) $\beta_{i,j}$ would lie between 0 (the random kernel) and 1 ( the sum kernel). This has been suggested before [2,3] but it could not be incorporated in the computer program. One misleading result was that for some samples, the random kernel gave the best fit. This did not imply that at some time during an experiment growth was not preferential towards the larger particles, because when looking at the comparisons between the random and sum kernel fits, it can be seen that a sum kernel fit is a bit flat and a random kernel fit is too steep. So, naturally, when the first stage is fitted with a sum kernel, the fit will be a bit too flat compared to the experimental curve. When fitting the second stage with a random curve this can be compensated. However, at later times the same kernel has again given the best results, so it is safe to say that using the same kernel yields the best result.

## CONCLUSIONS & RECOMMENDATIONS

It was found that binder liquid viscosity has a pronounced effect on agglomeration behaviour. The experimetal results can be explained with the theory. However, at high viscosities higher than 16.4 mPas, the only growth mechanism observed is nucleation, which occurs concurrently with breakage. This is in contrast with experimental results at lower viscosities, where other mechanisms like coalescence and crushing and layering occur. It is advised that future research be carried out to find the viscosity at which this change in granulation behaviour occurs and also to find a reason for this.

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carried out to find the viscosity at which this change in granulation behaviour occurs and also to find a reason for this.

Another interesting feature observed in the experiments was the breakage. More research will have to be carried out to clarify this mechanism. At this moment, not much is known about it, not even what caused this phenomenon. It is most likely that it is caused by the ploughshares but as yet no experimental proof exists

Modeling is still at an early stage and so there is still a lot to be done. For instance, an optimisation procedure should be added to the computer program, which would save considerable amount of time, which will come in handy if the remaining experimental results are to be fitted. Secondly, the model can be expanded by using other kernel forms.

## NOMENCLATURE

| Symbols | Description |
|---|---|
| d | particle diameter, m |
| h | thickness of binder liquid, m |
| $h_a$ | characteristic length of surface, m |
| $k_{i,r}$ | random growth kernel, $s^{-1}$ |
| $k_{i,s}$ | sum growth kernel, $s^{-1}m^{-3}$ |
| $\bar{r}$ | mean particle radius, m |
| t | time, s |
| $u_0$ | initial granule veleocity, m/s |

### Greek Symbols

| | |
|---|---|
| $\beta_{i,j}$ | coalescence kernel |
| $\eta$ | viscosity of binder liquid, mPas |
| $\rho$ | granule density, $kg/m^3$ |

**Dimensionless numbers**

| | |
|---|---|
| a | exponent for exponential kernel |
| e | coefficient of restitution |
| $N_i$ | number of particles of class i |
| $N_T$ | total number of particles |
| $St_v$ | viscous Stokesí number |
| $St_v^*$ | critical viscous Stokesí number |
| y | solution phase ratio |

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