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## Alternative method for producing organic fertiliser from anaerobic digestion liquor and limestone powder

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## Accepted Manuscript

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**Abstract**

Generally, the solid and liquid fractions (digestate) from Anaerobic Digestion (AD) energy production are considered as waste. This has a negative impact on the sustainability of AD processes because of the financial outlay required to treat digestate before being discharged into municipal water treatment plants or natural water bodies. The main aim of research was to investigate feasibility of producing an organic fertiliser using anaerobic digestate and limestone powders as the raw materials employing a high shear granulation process. Two- level factorial experimental design was used to determine the influence of granulation process variables on, the strength, resistance to attrition and yield of the granules. It was concluded from the study that it is technically feasible to produce organic fertiliser granules of acceptable strength and product yield. Increasing the liquid-to-solid ratio during granulation leads to increased granule strength and better product yield. Although the strength of the granules produced was lower than typical strength of commercial synthetic fertiliser granules (about 5 to 7 MPa), this could be improved by mixing the digestate with a polymeric binder or coating the particles post granulation.

**Keywords**

Granulation; digestate; granule strength; anaerobic digestion; attrition; experimental design

## 1. Introduction

The increase in the price of natural gas over several years has impacted on the production cost of synthetic fertilisers [1]. High global phosphate and natural gas prices represent a significant cost addition to production of synthetic fertiliser. The world demand of fertiliser is expected to increase due to increase in the population and demand for food [2]. The decision by governments to increase the proportion of renewable energy sources, such as bio-fuels, will also add to the demand for synthetic fertiliser. Therefore, there is an opportunity to develop alternative methods to produce fertiliser, which are less energy intensive.

Generally the digestate from the Anaerobic Digestion (AD) energy production is considered as waste which has a negative impact on the sustainability of AD processes because of the financial outlay required to treat the waste before discharge to municipal water treatment plants or natural water ways. The digestate is known to contain nutrients that are beneficial to plant growth, namely nitrogen, phosphorus and potassium [3]. In theory this makes the digestate a suitable raw material for the production of NPK fertiliser. In the UK, it is common practice for the digestate to be applied to fields as liquid fertiliser under strict government regulations. However, there are problems associated with the direct application of digestate to land as a liquid, for instance nutrient leaching, which can lead to eutrophication of natural water bodies often leading to changes in animal and plant populations and degradation of water and habitat quality. It is also difficult to store and transport the digestate once it has been produced by the AD biogas plant, this usually leaves the biogas plant operator with no option except treatment of the digestate in convectional municipal sewage water treatment plants.

Converting the liquid digestate into a solid product would offer several advantages such as ease of storage and transportation. Also, the properties of the solid product can be tailored to give desired release profiles, which would alleviate the problem of run-off and leaching of nutrients, which is associated with direct application of liquid fertiliser.

Granulation, defined as a particle enlargement or agglomeration process, is accomplished by the formation of inter-particle bonds between primary particles to form new entities called granules [4]. Granulation processes are used extensively within pharmaceutical, food and agricultural industries to impart improved flowability, strength, product appearance, shape and structural form, on powdered materials. Granulation can be used to convert digestate into a solid product with improved physical properties (i.e., ease of storage, improved appearance and more controlled nutrient release). The granulation process requires a solid material and limestone was identified as a suitable material because it is common usage for pH adjustment on agriculture. In addition to improving the handling properties over conventional powder application, a granulated limestone containing digestate would introduce additional nutrients to the product. In this study a high shear granulation process was used to produce limestone-digestate fertiliser granules.

Essentially, this research investigated the production of organic fertiliser through nutrient recycling. Product properties such as granule size and strength were determined. It is important that a high percentage of the product granules be in the size range 2 to 4mm which is the typical size range of commercial granular fertiliser. Strength of the granules is also important as the granules should survive mechanical stresses involved during handling, storage and transportation.

The compressive strength of the granules is also critical quality attribute; the granules have to be strong enough to survive the compressive load during storage when bags of the granules are stacked onto piles. It has been reported in literature that typical granule strength should be about 3 to 4.8 MPa to reduce caking during storage [5]. The granules also need to have high resistance to attrition. In this project a Design of Experiment (DoE) approach has been applied to granulation to study influence of different process variables on granule yield, compressive strength and attrition strength [6]. The influence of process variables on the mechanical properties of organic fertiliser produced from granulation of limestone and digestate was studied using a  $2^K$  factorial design.

## 2. Materials and Methods

### 2.1 Materials

Limestone powder supplied by Killwaughter Chemicals Ltd, UK, was used as one of the raw materials. The particle size distribution of the limestone was measured using laser diffraction technique with Malvern Master Size. The powder has a mean diameter of 50 microns, the size distribution is shown in Figure 1. For X-Ray Diffraction (XRD) a Philips X'pert PRO diffractometer was used to determine the mineral content within the samples at each stage. The XRD trace was recorded in the range of  $5^\circ \leq \varphi \leq 100^\circ$  and quantified using Philips X'Pert High Score software. The XRD analysis of the powder showed that the limestone used in the granulation experiments was mainly composed of calcite and quartz minerals.

### 2.2 Digestate

Digestate supplied by Agriculture and Food Biosciences Institute (AFBI) Hillsborough, Northern Ireland, UK, was used as a granulating liquid. It was used as

received (i.e., no filtration or dilution). The AD plant at AFBI processes only accepts on-site cattle waste as a feedstock. Composition of the wastes in terms of NPK nutrient was analysed and results shown that the concentrations of K and P were 1797 and 459 mg/kg respectively. Ammonia composition of the filtrate from the digestate was found to be 883 mg/L. Full details of the analysis are given elsewhere [7].

### 2.3 Experimental Design

A  $2^k$  - factorial experimental design was used to choose the experiments required to investigate the effect of the process variables on the yield and mechanical properties. Details of the  $2^k$  factorial described in greater detail in literature [8]. Each of the three variables were varied at two different levels; low (-1) and high (+1). The granulation time was varied at two levels; 3 min (low level) and 6 min (high); for liquid to solid ratio the two levels were 0.175 and 0.2 respectively. Two levels of impeller speed were used i.e., 103 rpm and 160 rpm (corresponding to settings of 2 and 4 respectively). The list of experiments undertaken is shown in Table 1. Please note that batches listed in Table 1 were done in duplicate, with the average value reported.

### 2.4 Production of granules

The granules were produced in a high shear granulator Kenwood-KM070 (Kenwood, UK). The granulator has a stainless steel mixing bowl with a capacity of 3 litres and is also equipped with two blade impeller which also undergoes “planetary mixing” motion around the mixer during granulation. The measured mass of limestone powder was added to the mixer and the predetermined amount of digestate added. For all batches produced, binder addition time was 30 seconds and this binder



was added whilst the impeller was in motion at the pre-determined speed for the experiment. All experiments were performed at room temperature. The rotational speed of the impeller can be varied between 100 and 213 rpm.

## 2.5 Drying

The moisture content of the granules after granulation ranged was between 17 and 20 % (w/w). After processing the granules were dried in an oven at 110 °C for 24 hours prior to analysis. The moisture content of the granules after drying was less than 4% (w/w).

## 2.6 Determination of product yield

For the purpose of this work, granules in the size range 2 to 4 mm were required. The yield of the process  $\eta$ , is defined as the percentage of the product which meets the size requirement i.e.

$$\eta = \left( \frac{m_p}{m_{tot}} \right) \times 100\% \quad (1)$$

where:  $m_p$  and  $m_{tot}$  are the mass of granules in the specified size range and total mass of granules produced respectively.

## 2.7 Determination of attrition strength

There are various standard methods that can be used to assess that attrition strength of granular materials [9, 10]. In this work a typical method for assessing the attrition strength similar to one previously used by our group [11] was adapted. The reason for the choice was that this method does not require use of specialised equipment. Though the use of this method might result in an over-estimation of attrition resistance of the granules (due to the absence of the steel balls used in [9]) it still provides a qualitative way of comparing attrition resistance of two different

samples. Granules in the target size were first sieved through the 1mm sieve to remove any fines or broken particles. For each of the batches a sample of granules initial mass,  $m_o$ , was measured and transferred into the 1mm sieve. The stack consisting of pan (for collection of fines, 1 mm sieve and lid), was placed on the sample shaker (Stuart Orbital Shaker, Cole-Parmer UK). The speed of shaker was set to 180 rpm. The samples were subjected to attrition by agitating them in 1 mm sieve and recording the mass of granules retained on the 1mm sieve after every 5 minutes for 45 minutes. The attrition loss which is defined as the percentage mass of the granules due to attrition was calculated using;

$$A_r = \left( \frac{(m_o - m_t)}{m_o} \right) \times 100\% \quad (2)$$

The attrition resistance constant,  $\tau$ , was determined by fitting the  $A_r$  versus time plots to equation;

$$A_r = 100(1 - \exp(-t/\tau)) \quad (3)$$

In equation (3), the attrition resistance coefficient,  $\tau$ , can be interpreted as the time it takes to loss 67.7% by mass due to attrition. A higher attrition resistance coefficient indicates better resistance of the material to attrition. The difference in the resistance of the granules from the different batches to attrition is evaluated using this coefficient.

## 2.8 Determination of granule strength

The granule strength was determined from diametric compression of the single granules using method described previously [12-14]. The granule strength was determined from the failure load,  $F_f$ , and granule diameter,  $D$ , which is taken as the

distance between the platen when first contact is made between the granule and the movable platen, [15];

$$\sigma = 2.8 \times \left( \frac{F_f}{\pi D^2} \right) \quad (4)$$

### 3. Results and Discussion

The results obtained from the experiments are summarised in Table 1. The first column of this table shows the experiment ID. The next three columns show the experimental input variables namely granulation time, impeller speed and liquid to solid ratio respectively. The last three columns show the response variables.

#### 3.1 Effect of process variable on product yield

In this work the product yield has been defined as the mass fraction of granules in the size range 2 to 4mm expressed as a percentage. Detailed ANOVA analysis results of the effect of process variables on the product yield are given in Table 2. The results of the yield from 8 batches of granulation are shown in Fig. 2, which shows that the highest product yield of approximately 46% was obtained for batch EX\_7. This corresponds to the case where maximum amount of digestate; impeller speed was low whilst the granulation time was high. The minimum yield was obtained for batch EX\_4. For the low liquid to solid ratio, maximum yield was obtained when granulation time and impeller speed were both at high levels.

During the granulation process competing mechanisms may occur at any given time; these include granule breakage, consolidation and growth [16]. The rate at which these processes occur depends on a number of process variables. The balance between the competing constructive forces and destructive forces will determine the

overall population balance, i.e., if an increase or decrease in the mean granule size is observed with respect to time.

The effect of the process variables on the product yield is summarised by the empirical model, equation (5);

$$\eta = -86.02 + 4.32 t_g + 498.99 L_s - 0.0118 t_g \Omega L_s \quad (5)$$

The combined effect of the process variables on the fertiliser product yield is summarised by the surface response plots presented in Fig. 3 (a) and (b).

### 3.1.1 Effect of impeller speed on product yield

In line with equation (5) if the granulation time and liquid-to-solid ratio are kept constant, increasing the impeller speed would result in a reduction in the product yield. There could be two possible explanations for this observation. Increasing the impeller speed may result in an increase in breakage of the granules; however it may also promote granule growth. It has been reported in literature that increasing the impeller speed results in an increase in the mean granules size of the granules [17, 18]. On the contrary, there are also some other reports in literature where a reduction in the mean granules size was observed when the impeller speed was increased [12, 19, 20]. Earlier studies on different formulations revealed that increasing the impeller speed would result in a reduction of the product yield [21]. Whether the product yield would increase or decrease if the impeller speed is increased whilst keeping the granulation time constant depends on the balance between the two rate processes of growth and breakage. The product yield will increase if the conditions prevailing in the granulator are that the amount for granules leaving the product fraction is less than the amount of granule entering the product fraction. If the amount of granules leaving

the product fraction is more than the amount of granules entering the product fraction, the product yield will decrease. Fig. 3 (a) shows that when the granulation time is 3 minutes, increasing the impeller speed would result in a reduction in the product yield. This may be explained by domination of the breakage mechanism when the speed is increased. However same figure shows that at longer granulation time increasing the impeller speed would result in an increase in the product yield, which is contrary to what is observed at shorter granulation times. This anomaly may be explained by the fact that in the later case that broken granules have sufficient time to grow again since the granulation time is longer hence the increase in the yield. The trends are reversed when more binder is available in the system (see Fig. 3 (b)). The decrease in the product yield observed at higher liquid to solid ratio and longer granulation times in Fig 3(b) can be explained by overgrowth of the granules into coarse granules due to improved availability of binder.

### 3.1.2 Effect of granulation time on product yield

Consider the case where after a granulation time of  $t_i$ , the mass of granules meeting the product specification is  $m_p$ , mass of granules greater than the product (coarse granules) is  $m_c$  and  $m_f$  is mass of granules smaller than the product (fine granules). On extending the granulation time, the balance between these three fractions the batch will change depending on balance between the destructive forces and the constructive forces. Some granules leave the product size fraction due to breakage and some enter into this fraction due to growth.

Let us focus on what happens to granules within the product fraction when the granulation time is extended. If the granulation time is extended to  $t_{i+1}$ , let  $\phi_{p,c}$  be mass fraction of granules in the in product specification at  $t_i$  that do not change size.

Also let  $\phi_{p,b}$  be the mass fraction of product granules at  $t_i$  that break into fines and  $\phi_{p,g}$  is the mass fraction of product granules that grow into coarse granules. The following expression can be written for these mass fractions, assuming conservation of mass.

$$\phi_{p,b} + \phi_{p,c} + \phi_{p,g} = 1 \quad (6)$$

Similar expressions can be written for the two other fractions i.e., the fines and coarse granules;

$$\phi_{f,b} + \phi_{f,c} + \phi_{f,g} = 1 \quad (7)$$

For the coarse granules we have;

$$\phi_{c,b} + \phi_{c,c} + \phi_{c,g} = 1 \quad (8)$$

In simple terms, the mass of the granules in the product fraction at time,  $t_{i+1}$ , is given by the sum of granules in the product fraction at time  $t_i$  and the change in the mass of in the product fraction during this time interval. Thus;

$$m_p(t_{i+1}) = m_p(t_i) + \frac{dm_p}{dt} \Delta t \quad (9)$$

The 2<sup>nd</sup> term in equation (9) represents the change in the mass of the granules in the product fraction. This can be written as;

$$\frac{dm_p}{dt} = \frac{m_{p,E} - m_{p,L}}{\Delta t} \quad (10)$$

Where:  $m_{p,E}$ , and  $m_{p,L}$  are mass of granules entering and leaving the product fraction respectively during the time interval.

The granules enter the product fraction by either breakage of the granules in the coarse fraction or by growth of the granule in the fines fraction. This can be written as;

$$m_{p,E} = \phi_{f,g} m_f(t_i) + \phi_{c,b} m_c(t_i) \quad (11)$$

The granules leave the product fraction is due to either breakage of the granules in the product fraction or by growth of the granule in the product fraction to coarse granules.

This can be written as;

$$m_{p,L} = \phi_{p,g} m_p(t_i) + \phi_{p,b} m_p(t_i) \quad (12)$$

Now equation (10) can be re-written to obtain;

$$\frac{dm_p}{dt} = \frac{\phi_{f,g} m_f(t_i) + \phi_{c,b} m_c(t_i) - m_p(t_i)(\phi_{p,g} + \phi_{p,b})}{\Delta t} \quad (13)$$

Substituting equation (13) into equation (9) gives;

$$m_p(t_{i+1}) = m_p(t_i) + \frac{\phi_{f,g} m_f(t_i) + \phi_{c,b} m_c(t_i) - m_p(t_i)(\phi_{p,g} + \phi_{p,b})}{\Delta t} \Delta t \quad (14)$$

Consistent with equation (14), whether the product yield increases or decreases with an increase in granulation time would depend in the overall sign of second term; if this term is positive then the product yield increases, a negative sign would lead to a reduction in product yield. If this term is zero then there will be no net change in the product yield.

The effect of the granulation time on the product yield is illustrated in Fig. 4, (a) which shows results when the liquid to solid ratio is kept constant at 0.175. In both cases of impeller speed increasing the granulation time increases the product yield. Higher rate of increase in product yield is obtained at higher impeller speed. An opposite trend is observed at higher liquid to solid ratio in Fig. 4 (b); here increasing the granulation time has more pronounced effect on the product yield when the impeller speed is lower.

According to equation (14), increasing the granulation time would result in increased product yield if the second term is positive; implying the amount of granules leaving the product fraction by growth into the coarse fraction and breakage

into the fines is less than the amount of granules entering the product fraction. This effect is observed in Fig. 4 (a) and (b).

### 3.1.3 Effect of liquid to solid ratio on product yield

Increasing the liquid to solid ratio increases the availability of binder for agglomeration. It is therefore expected that increasing this variable should result in an overall increase in the mean size of the granules. Results presented in Fig. 5 show an increase in the liquid to solid ratio with increase in product yield. Increasing the liquid to solid ratio by a factor of 1.15 will result in an increase in the product yield of about 10%. It is worth noting that increasing the amount of binder added would increase both the product yield and the amount of the nutrients in the resultant granules. However there is a limit on the amount of binder that can be added to the systems; excessive amounts of binder would result in paste formation. Specifically, there is a threshold beyond which further increase in liquid binder would result in over-growth of particles hence reduced product yield.

## 3.2 Effect of process variables on granule tensile strength

Experimental data from compression analysis of individual granules are shown in Fig. 6 (a) and detailed ANOVA analysis results are presented in Table 3. The granules tensile strength for the 8 batches ranges from about 0.3 to 1.1 MPa. The highest strength granule was obtained for batch Ex\_2 and the least was obtained for batch Ex\_4. The correlation between the crushing strength of the fertiliser granules and their attrition strength is shown in Fig. 6 (b). This correlation shows a linear relationship between the attrition strength and the compressive strength of the granules, which is in agreement with previous research [22].



The influence of the process variables on the fertiliser granule strength is summarised by the surface plots in Fig. 7(a) and (b). Fig. 7(a) shows surface plot for the case where the liquid to solid ratio is 0.175 and in Fig. 7 (b) results for liquid to solid ratio of 0.2 are presented. The independent effects of the process variables on the granule strength are discussed in the sub-sections that follow. The tensile strength of the fertiliser granules is related to the process variables according to equation (15).

$$\sigma = 0.706 - 0.0997t_g - 0.00394\Omega + 0.0014t_g\Omega + 0.00612\Omega L_s \quad (15)$$

### 3.2.1 Effect of granulation time on granule tensile strength

Results presented in Fig. 7 show the effect of granulation time on the strength of the fertiliser granule. Increasing the granulation time increases the strength of the granules. It has been shown previously that the granule porosity decreases exponentially with granulation time due to consolidation [23]. It would be logical to expect the strength of the granules to increase with granulation, however granulating for too long may also lead to formation of weak granules due to bonding weakening.

### 3.2.2 Effect of impeller speed of granule strength

The results presented in Fig. 7 show that increasing the impeller speed results in an increase in the granule strength. However the extent of the increase in the strength depends on both the amount of binder present in the granulator and the duration of granulation. When less binder is available in the granulator the effect of increasing the impeller speed on granules strength is less significant as can be seen in Fig. 7 (a); increasing the impeller speed from 103 to 160 rpm increased the strength from ~0.3 to 0.4 MPa. When the liquid to solid ratio was 0.2, the strength remained almost constant at 0.175MPa, these results are consistent with previous findings by [12] .

### 3.2.3 Effect of liquid to solid ratio on tensile strength

The results presented in Fig. 7, which is a plot of equation (15); shows the effect of changing the liquid to solid ratio on granule strength. It is clear from this figure the changing the liquid to solid ratio does not have a significant influence on the strength of the granules (less than 5% increase in granule strength is observed). Changing the liquid to solid ratio used for granulation changes the amount of granulating liquid that is used. During granulation the liquid droplets forms bridges between the powder primary particles. The liquid bridges solidify to form solid bridges during the subsequent drying process. The amount of liquid available would affect the number and strength of bonds formed between the primary particles, which in turn affect the overall strength of the agglomerate formed.

### 3.3 Effect of process variables on attrition strength

For all the batches of granules produced and subsequently analysed, the attrition loss was less than 8% percent (see Fig. 8(a) and (b)). Granules that exhibited a higher mass loss during the test are considered to be more prone to attrition. The least attrition loss was obtained for batch Ex\_6, while the maximum loss was obtained for batch Ex\_5. The statistical analysis on the influence of process variables on the attrition strength is summarised in the ANOVA analysis presented in Table 4.

The attrition resistance of the fertiliser granules from different batches can be related to the process variables according to equation (16).

$$\tau = -1900.63 + 331.5 t_g + 14.832 \Omega + 4.894 \Omega L_s \quad (16)$$

The effect of process variables on the attrition strength is summarised by response surface plots shown in Fig. 9. The effect of impeller speed on the attrition strength of

the granules becomes more pronounced when the amount of granulating liquid is increased (see Fig. 6 (b)). The detailed discussion of the effect of the process variables is given in following sections.

### 3.3.1 Effect of granulation time on attrition strength

It can be deduced from equation (16) that increasing the granulation time should result in an increase in the strength of the fertiliser granules. As the granulation time increases the granules are subjected to more collisions which results in further consolidation of the granules [16, 24]. The granules become smoother and rounder as granulation time is increased [25]. The smooth surface of the granules makes them less susceptible to attrition. This concurs with the results presented in Fig. 9 (a) and (b).

### 3.3.2 Effect of impeller speed on attrition strength

The effect of changing the impeller speed on attrition strength is shown in Fig. 9 (a) and (b). Fig. 9 (a) shows the effect of changing both impeller speed and granulation time at the same level of liquid to solid ratio 0.175; the results show that the granulation time has more significant effect on the attrition when compared to impeller speed. Changing the impeller speed from 103 to 160 rpm does not result in significant change on the attrition strength. However when the liquid to solid ratio is 0.20 the attrition coefficient increases from about 300 minutes to about 1100 minutes, when the impeller speed is changed from 103 to 160 rpm. This implies that an increase in the granules resistance to attrition or an increase in the attrition strength as illustrated in Fig 9 (b), which shows the interaction effect of the process variables.

### 3.3.3 Effect of liquid to solid ratio on attrition strength

The liquid to solid ratio does not have a significant effect on the attrition strength of the granules as can be observed from Fig. 10. Increasing the amount of binder used in granulation is expected to result in formation of stronger bonds between the primary particles and also result in formation of more smooth granules [25]. To confirm this, digital images of representative granules from different batches were analysed using the ImageJ software (National Institutes of Health, US) using the surface plot function. Surface plots of granules produced with different liquid to solid ratio presented in Fig. 10 show that smoother granules surface can be obtained when using a higher liquid to solid ratio. This in turn should affect the attrition strength of the granules. The attrition strength is a measure of the resistance to loss of mass by mainly loss of particle at the outer surface of the particles. This result is at odds with data obtained in earlier researchers [26] who observed that more granule breakage occurred at higher moisture contents, which they attributed to attrition. One possible explanation for this discrepancy would be that the range of values of liquid to solid ratio used in this work was too narrow for this effect to be noticed.

## 4 Conclusions

The fertiliser product yield is more sensitive to changes in the granulation time and amount of liquid added during granulation. Impeller speed does not have a significant effect on the product yield, although it had significance on other granule properties such as granule strength. Increasing liquid-to-solid ratio during granulation leads to increased granule strength and better product yield. It was concluded from the study that it is technically feasible to produce organic fertiliser granules of acceptable strength and yield. Future studies will focus on further improving the mechanical

strength of the granules and studying the homogeneity of the granules in terms of nutrient composition.

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## Nomenclature

$A_r$	Attrition ratio	[-]
$d$	Granule diameter	[mm]
$F$	Compression force required to break the granule	[N]
$\phi_{c,b}$	Fraction of coarse granules that break into product in time interval $\Delta t$	[-]
$\phi_{c,c}$	Fraction of coarse granules that do not change size in time interval $\Delta t$	[-]
$\phi_{c,g}$	Fraction of coarse granules that grow in time interval $\Delta t$	[-]
$\phi_{f,c}$	Fraction of fine granules that do not change size in time interval $\Delta t$	[-]
$\phi_{f,b}$	Fraction of fine granules that break in time interval $\Delta t$	[-]

$\phi_{f,g}$	Fraction of fine granules that grow into product in time interval $\Delta t$	[-]
$\phi_{p,c}$	Fraction of product granules that do not change size in time interval $\Delta t$	[-]
$\phi_{p,b}$	Fraction of product granules that break into fines in time interval $\Delta t$	[-]
$\phi_{p,g}$	Fraction of product granules that grow into fines in time interval $\Delta t$	[-]
$\eta$	Product yield (equation 3)	[%]
$L_s$	Liquid-to-solid ratio	[-]
$m_{bin}$	Mass of digestate used	[kg]
$m_c$	Mass of coarse granules produced (larger than 4mm)	[kg]
$m_f$	Mass of fines, (less than 2mm)	[kg]
$m_o$	Initial sample mass	[kg]
$m_{tot}$	Total mass of granules produced per batch	[kg]
$m_{pow}$	Mass of powder used	[kg]
$m_t$	Mass of granules remaining on sieve at time $t$	[kg]
$m_p$	Mass of the granules in the size range 2 to 4mm	[kg]
$m_{p,E}$	Mass of the granules entering the product fraction in time interval $\Delta t$	[kg]
$m_{p,L}$	Mass of the granules leaving the product fraction in time interval $\Delta t$	[kg]
$\tau$	Attrition constant	[-]
$\Delta t$	Time interval under consideration	[s]
$R$	Granule radius	[mm]
$\sigma$	Granule tensile strength defined by equation (4)	[MPa]
$t$	time during attrition testing	[Min]
$t_g$	Granulation time	[Min]
$\Omega$	Granulation speed	[Rpm]

**Table 1: Summary of the data used in the Factorial Design of Experiment.**

Std	Run	Factor 1 A: $l_s$ -	Factor 2 B: $\Omega$ rpm	Factor 3 C: $t$ min	Response 1 $\eta$ %	Response 2 $\sigma$ MPa	Response 3 $\tau$ min
14	1	-1	1	1	31.31	1.08	2841
6	2	-1	1	-1	10.61	0.28	1125
1	3	-1	-1	-1	17.15	0.33	1214
16	4	1	1	1	31.10	0.94	3202
9	5	-1	-1	1	19.07	0.51	2160
10	6	-1	-1	1	19.07	0.51	2160



2	7	-1	-1	-1	17.15	0.33	1214
11	8	1	-1	1	45.54	0.43	1081
3	9	1	-1	-1	20.76	0.28	664
13	10	-1	1	1	31.31	1.08	2864
5	11	-1	1	-1	10.61	0.28	1125
7	12	1	1	-1	30.05	0.54	1658
12	13	1	-1	1	45.54	0.43	1087
4	14	1	-1	-1	20.74	0.28	664
15	15	1	1	1	31.05	0.95	3202
8	16	1	1	-1	30.05	0.54	1658

Table 2: ANOVA Table for Product Yield response variable

Source	Sum of Squares	df	Mean Square	F- Value	P-value
Model	822.22	3	274.07	56.6	0.001
<i>A-t<sub>g</sub></i>	292.58	1	292.58	60.43	0.0015
<i>C-L<sub>s</sub></i>	302.58	1	302.58	62.49	0.0014
<i>ABC</i>	227.06	1	227.06	46.89	0.0024
Residual	19.37	4	4.84		
Cor Total	841.58	7			

$R^2 = 0.9770$

Adj  $R^2 = 0.9597$

Table 3: ANOVA table for strength response variable.

Source	Sum of Squares	df	Mean Square	F Value	p-value
Model	0.24	4	0.061	9.54	0.047
A- $t_g$	0.13	1	0.13	20.03	0.0208
B- $\Omega$	0.08	1	0.08	12.52	0.0384
AB	0.029	1	0.029	4.49	0.1244
BC	$7.18 \times 10^{-3}$	1	$7.18 \times 10^{-3}$	1.13	0.3665
Residual	0.019	3	$6.38 \times 10^{-3}$		
Cor Total	0.26	7			

$R^2 = 0.9271$

Adj  $R^2 = 0.83$

Table 4: ANOVA table for Attrition strength response variable.

Source	Sum of Squares	df	Mean Square	F -Value	p-value
Model	$4.85 \times 10^6$	3	$1.62 \times 10^6$	10.55	0.0227
<i>A-GT</i>	$1.98 \times 10^6$	1	$1.98 \times 10^6$	12.93	0.0229
<i>B-W</i>	$1.61 \times 10^6$	1	$1.61 \times 10^6$	10.53	0.0315
<i>BC</i>	$1.25 \times 10^6$	1	$1.25 \times 10^6$	8.19	0.0459
Residual	$6.12 \times 10^5$	4	$1.53 \times 10^5$		
Cor Total	$5.46 \times 10^6$	7			
$R^2 = 0.8878$					
Adj $R^2 = 0.8036$					

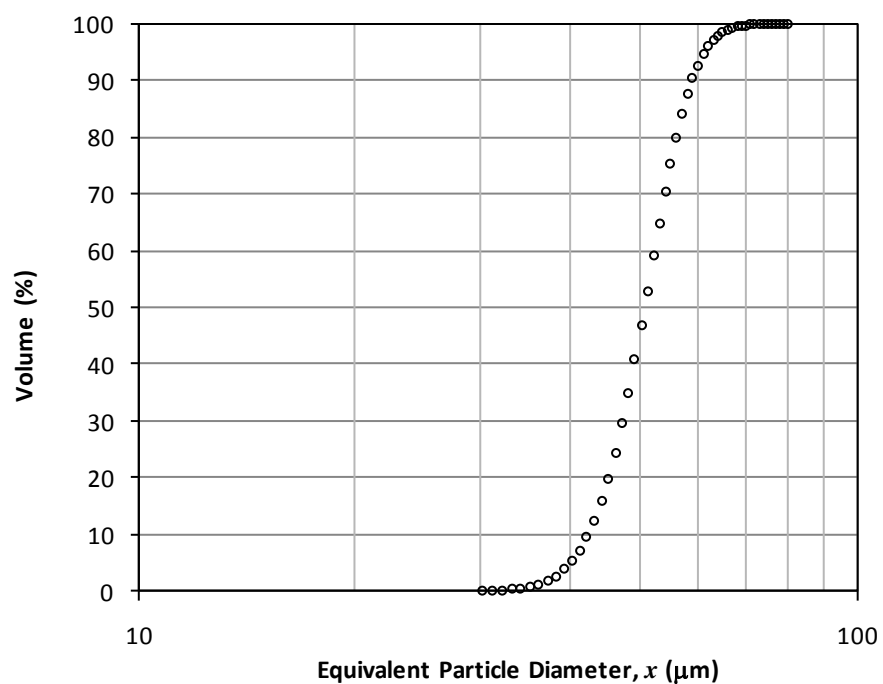


Fig. 1: Particle size distribution of the limestone powder

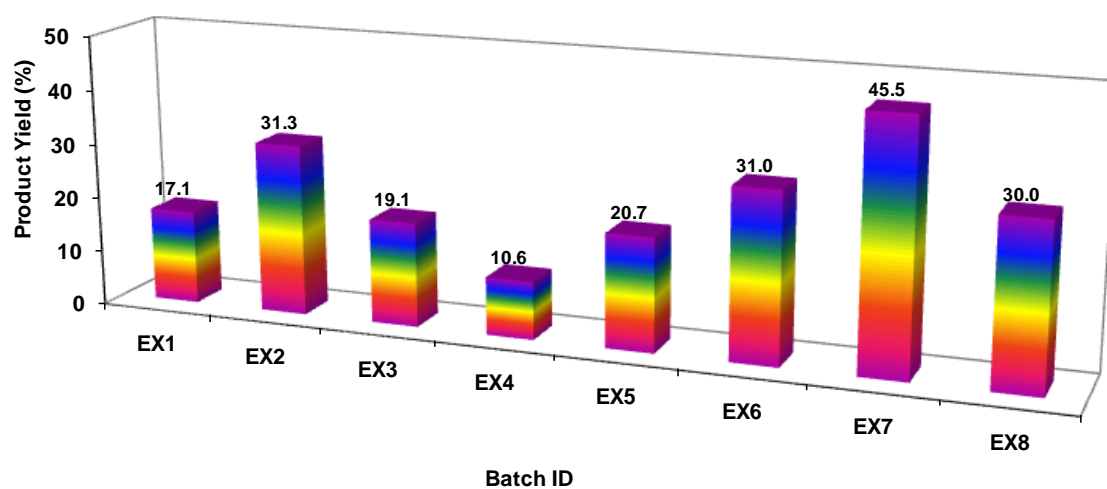
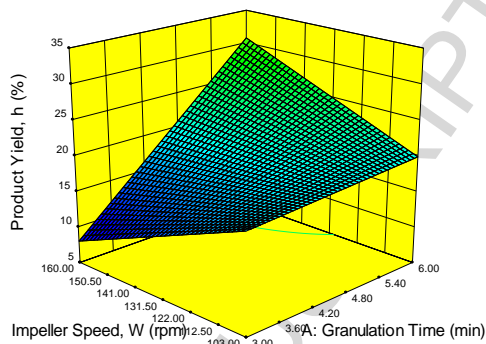


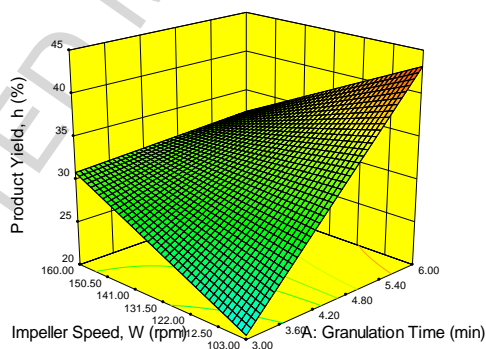
Fig. 2: The product yield for 8 experiments N.B. Values shown in this plot are average of two replicates.

Design-Expert® Software  
Factor Coding: Actual  
h  
45.54  
10.61  
X1 = A: GT  
X2 = B: W  
Actual Factor  
C: Ls = 0.17



(a)

Design-Expert® Software  
Factor Coding: Actual  
h  
45.54  
10.61  
X1 = A: GT  
X2 = B: W  
Actual Factor  
C: Ls = 0.20



(b)

Fig. 3: Response surfaces plots showing effect of process variables on product yield at two different levels of liquid to solid ratio (a) 0.17 and (b) 0.2. The surface plots are obtained from (5).

Design-Expert® Software  
Factor Coding: Actual  
h

X1 = A: GT

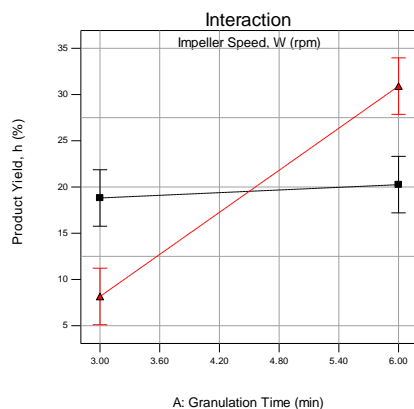
X2 = B: W

Actual Factor

C: Ls = 0.17

■ B- 103.00

▲ B+ 160.00



(a)

Design-Expert® Software  
Factor Coding: Actual  
h

X1 = A: GT

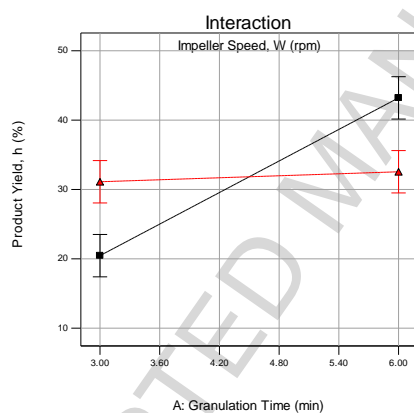
X2 = B: W

Actual Factor

C: Ls = 0.20

■ B- 103.00

▲ B+ 160.00



(b)

Fig. 4: Effect of granulation time on product yield.



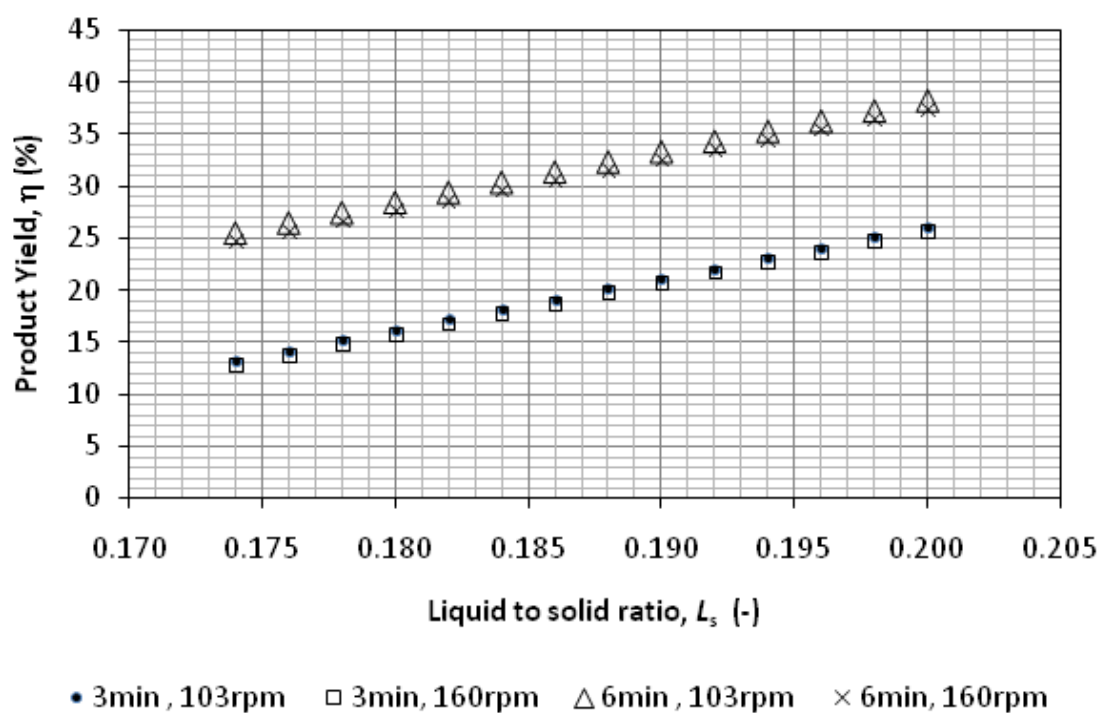
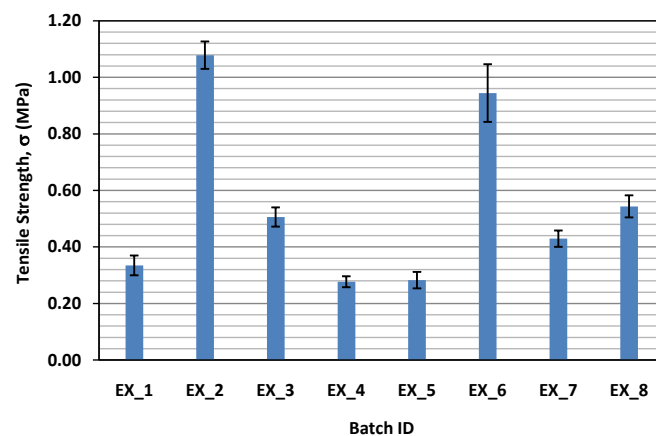
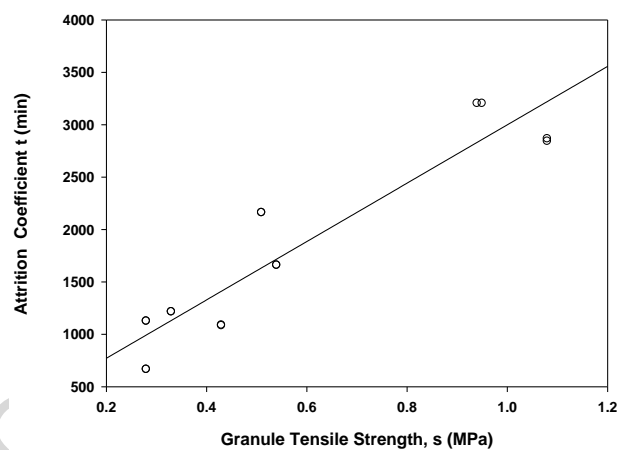


Fig. 5: Effect of liquid to solid ratio on product yield.



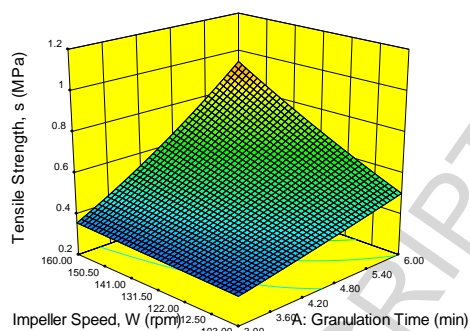
(a)



(b)

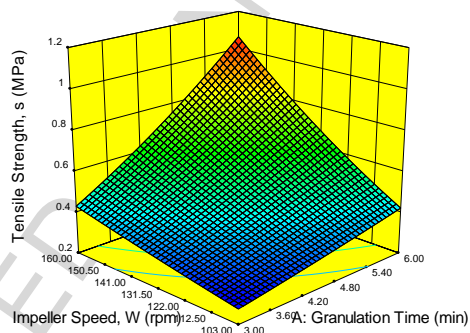
Fig. 6:(a) Granule tensile strength results for the different batches.  $n = 25$  and error bars indicate the standard deviation (b) Correlation between granule tensile strength and attrition strength.

Design-Expert® Software  
Factor Coding: Actual  
Original Scale  
s  
1.08  
0.28  
X1 = A: GT  
X2 = B: W  
Actual Factor  
C: Ls = 0.17



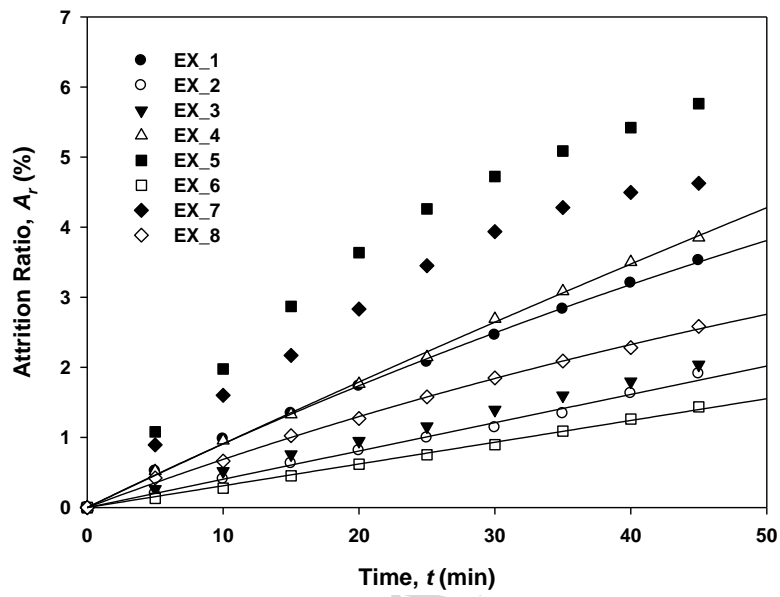
(a)

Design-Expert® Software  
Factor Coding: Actual  
Original Scale  
s  
1.08  
0.28  
X1 = A: GT  
X2 = B: W  
Actual Factor  
C: Ls = 0.20

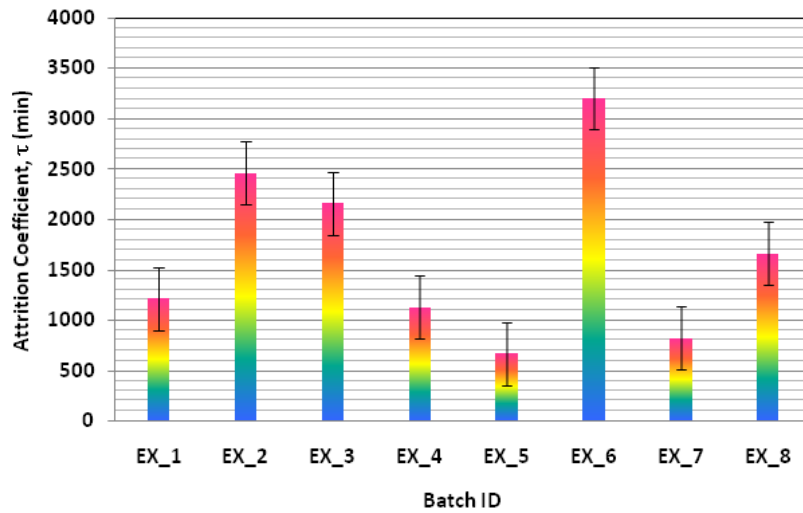


(b)

Fig. 7: Surface plot showing effect of process variables on granule strength.



(a)



(b)

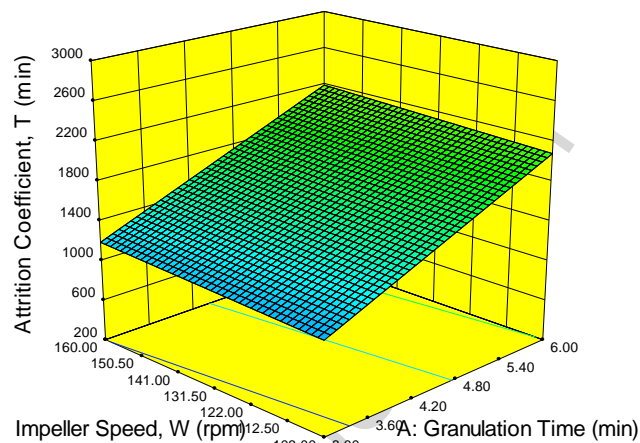
Fig. 8: (a) Attrition ratio as a function of testing time (b) comparison of the attrition coefficients of different batches.

Design-Expert® Software  
Factor Coding: Actual



X1 = A: GT  
X2 = B: W

Actual Factor  
C: Ls = 0.17



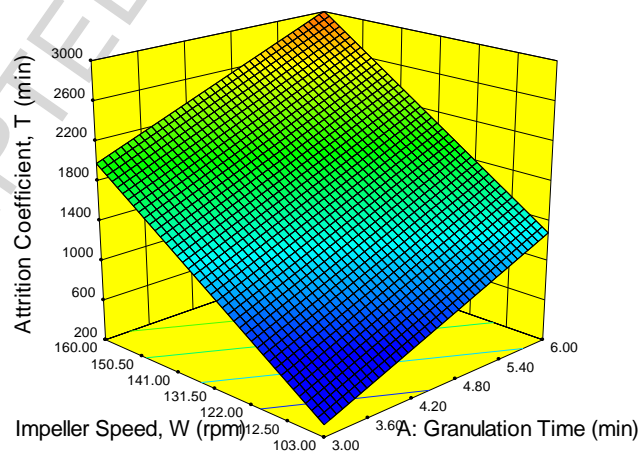
(a)

Design-Expert® Software  
Factor Coding: Actual



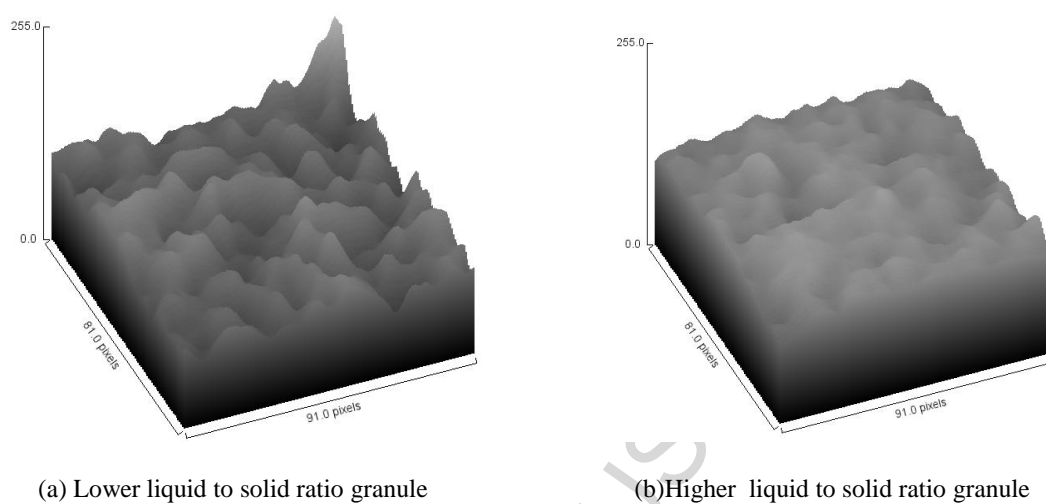
X1 = A: GT  
X2 = B: W

Actual Factor  
C: Ls = 0.20

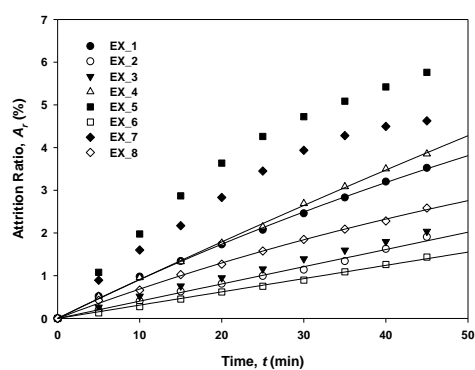


(b)

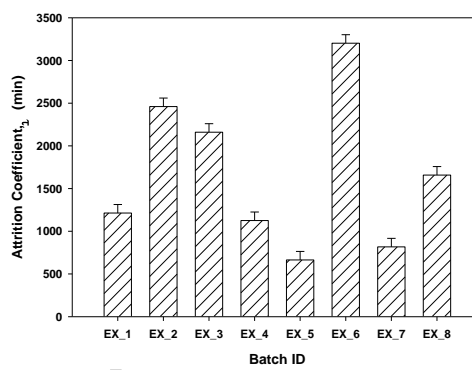
Fig. 9: Effect of process variables on granule tensile strength for different liquid to solid ratios (a)  $I_s = 0.175$  b)  $I_s = 0.20$ .



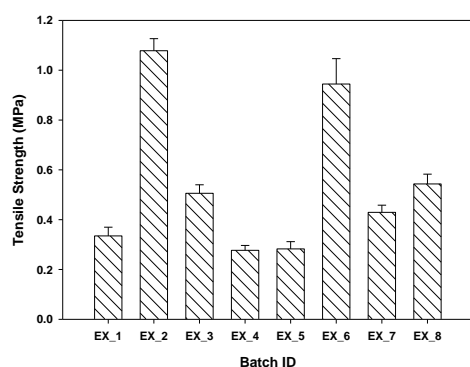
**Fig. 10: comparison of surface roughness of granules with different liquid to solid ratios.**



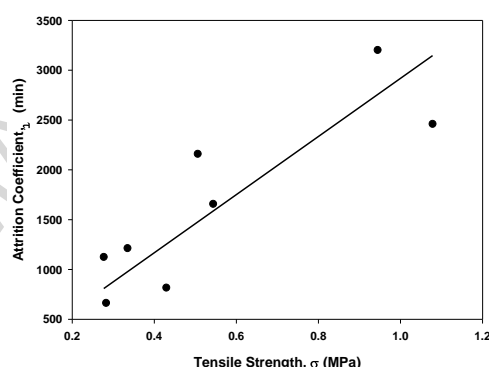
(a)



(b)



(c)



(d)

# Graphical abstract

**Research Highlights**

- It is technically feasible to produce granular organic fertiliser from AD liquor and limestone.
- Effect of granulation process variables on granule yield and strength was investigated.
- Granulation time and amount of digestate available strongly affect the product yield.
- Granule strength is mainly influenced by granulation time and impeller speed.
- Attrition strength of the granules is sensitive to granulation time and impeller speed.