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# Granulation of Ammonium Sulphate Fertilizer in a Spouted Bed

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## Abstract

Ammonium sulphate was successfully granulated in a spouted bed by atomizing a solution of the salt into a bed of seed particles spouted with hot air. The granules thus produced had a uniform layered structure. The particle growth rate was correlated with the solution feed rate, and an estimate was made of the rate of size reduction due to attrition which occurred concurrently.

It was found that under a given set of operating conditions, the net growth rate increased in approximate proportion to solution feed rate raised to the power of 0.8, but remained independent of the seed particle diameter.

Considerable amount of fine dust was produced. It was caused by incomplete deposition of sprayed solution and the attrition of the particles in the spout. The rate of attrition under granulation conditions was independent of the particle diameter, and was strongly affected by the moisture content of the bed solids.

## 1. Introduction

Granulation is a process of size enlargement, and is commonly employed in the fertilizer industry to produce particles of 1–4 mm. size range. The process is often carried out in a rotary drum, and a slurry or solution of the material to be granulated is sprayed onto the rolling bed of particles in the drum.

Growth can occur by two mechanisms. The first mechanism is agglomeration<sup>1)</sup>, or sticking together of smaller particles with the liquid providing the binding force. The second mechanism is growth by layering process which comes about by deposition of solids after the evaporation of water<sup>2,3,4)</sup>. Successive layers are thereby formed on the particles.

The similar process can be carried out more effectively in a spouted bed than in a rotary drum, since the spouted bed not only provides very systematic agitation of the bed solids but also allows intimate gas-solids contact which facilitates evaporation of water<sup>5)</sup>. Granulation can also be carried out in a fluidized bed, but the use of a spouted bed enables granules of a much larger size to be produced. Apart from this, the existence of a high voidage zone at the bottom of the spout provides an ideal location for injecting the liquid spray. Rapid evaporation of water in this region causes the temperature of the hot gas to fall sharply before it comes in contact with the bulk of the bed solids. Higher inlet gas temperatures can therefore be used without causing thermal damage to the bed

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particles than would be permissible in a fluidized, with consequent improvement in evaporation rates.

The most important advantage, however, arises from the systematic cyclic movement of particles in a spouted bed, as against the more random motion in a fluidized bed, since with the former, agglomeration is avoided so that the granules produced are uniform in size, and possess a homogeneous layered structure. The granules produced are of improved appearance, have better storage properties and require shorter drying times<sup>6)</sup>.

The variables that are likely to affect the growth rate of the particles in a spouted bed are as follows:

- (1) Solution spray rate.
- (2) Seed particle diameter.
- (3) Attrition: the surface abrasion of particles to create fine dust.

Therefore, the effect on the net growth rate of the operating variables (seed sizes, feed rate of solution, temperature variations across the bed and attrition rate) were investigated.

The object of this research was to correlate the above mentioned variables with the net growth rate, thereby to acquire a better understanding of the nature of the granulation process.

## 2. Apparatus and Experimental Procedures

### 2.1 Apparatus

The equipment used in this investigation is schematically shown in Figure 1. The main granulation column consisted of a 6" dia.  $\times$  1000 mm. high stainless steel column with a 60° conical base. The approach pipe for the spouting air consisted of a 2" I. D. stainless steel pipe, which was constricted by a 3/4" dia. orifice at the point of entry into the bed. The atomizer is also shown in Figure 1. It was installed in the centre of the orifice and the strong shearing action at the 3 mm. nozzle broke up the solution into a fine spray.

A series of iron-constantin thermocouples were used to measure the temperature of inlet and exit air streams and at various levels along the height of the bed.

The ammonium sulphate solution was stored in a 6" dia.  $\times$  1000 mm. high Pyrex

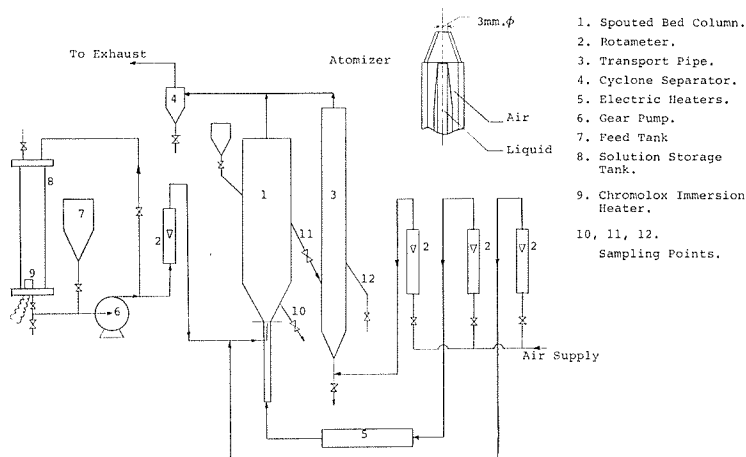


Fig. 1 A Schematic Diagram of the Spouted Bed Granulator

glass column, which was graduated to enable measurement of solution feed rate. A Chromolox nickel-plated immersion heater was placed at the bottom of the column, and the temperature of the solution was regulated by means of a thermostat. The solution was pumped to an atomizer which sprayed it as a fine mist into the base of the spouted bed.

## 2.2 Experimental Procedures

a. Growth runs A solution about 40% by weight of ammonium sulphate was made, and was stored in the solution reservoir. The solution was heated to 70°C by means of the immersion heater.

A bed of seed particles of ammonium sulphate of known weight (6.2–7.1 kg) and of a particular size range (1.17–3.36 mm) was put into the spouting column. The particles were used as nuclei upon which the atomized ammonium sulphate solution was sprayed during the experiment.

The particles were spouted with hot air at approximately 200°C. The supply of solution to the atomizer was started after allowing ten to fifteen minutes for preheating of the bed to its expected temperature of about 70°C during the experiment.

Solids were withdrawn periodically from the bed through the outlet in the cone, so that the average bed height was maintained constant throughout a run.

The experiments were carried out for five to ten hours. The withdrawn particles were screened to determine the size distribution.

b. Attrition runs Attrition runs were carried out with the objective of determining the extent of attrition occurring in a spouted bed. The particles were spouted with air at 200°C, but without injection of ammonium sulphate. In some runs water was injected into the bed as feed to simulate the conditions during growth runs, while the other runs were carried out with no water injection at all.

The entire bed was removed periodically for screen analysis and then returned to the column to continue the run.

c. Granule structure Some large granulated particles from the product of the granulation experiment were selected. The particles were set in a mould of organic polymeric material, and the surface of the hardened mould was sandpapered until the cross-section of the particle close to its equator could be seen clearly. The sectioned particles were observed under a microscope, and were also photographed.

## 3. Theory

The spouted bed in the granulator can be divided into two zones; one is a "spray zone (the spout)" in which the particles grow by the spray droplets, and the other is a "drying zone (the annulus)" in which the moisture is removed from wet particles. The particles grow gradually by repeating this cycle.

Since it was confirmed by observing the granulated particles that agglomeration did not occur in the spouted bed granulator, the mass rate of growth of a particle, on passing through a spray zone, is assumed to be proportional to its surface area<sup>7,8)</sup>, then

$$\frac{dm}{dt_s} = k_1 S_p \quad (1)$$

The surface area and mass of a particle in equation (1) are expressed in terms

of its diameter as follows,

$$S_p = \frac{\pi D_p^2}{\phi_s} \quad (2)$$

$$dm = \frac{\pi \rho_s D_p^3}{2\phi_s} dD_p \quad (3)$$

Substituting equations (2) and (3) into equation (1) then gives;

$$\frac{dD_p}{dt_s} = \frac{2k_1}{\rho_s} \quad (4)$$

However, the time, a particle spends in the spray zone ( $t_s$ ), is not an easily measurable quantity. Therefore, assuming that the time, a particle spends in the spray zone, is directly proportional to the residence time of a particle in the granulator ( $t$ ), we may write

$$\frac{dt_s}{dt} = k_2 \quad (5)$$

where  $k_2$  is designated as the residence time coefficient and depends on flow characteristics of the spouted beds.

Combining equations (4) and (5), we get

$$\frac{dD_p}{dt} = \frac{2k_1 k_2}{\rho_s} \quad (6)$$

The rate of size reduction of a particle due to attrition and fragmentation in the spouted bed would also be expected to be proportional to the particle surface area as described in section 4. 3.

Hence

$$\frac{dm}{dt} = -k_3 S_p \quad (7a)$$

and

$$\frac{dD_p}{dt} = -\frac{2k_3}{\rho_s} = R \quad (7b)$$

Combining equations (6) and (7), one obtains the net growth rate equation

$$\frac{dD_p}{dt} = \frac{2}{\rho_s} (k_1 k_2 - k_3) \quad (8)$$

Upon integration of equation (8) under the initial condition  $D_p = D_{p_0}$  at  $t = 0$ , the following equation is obtained:

$$D_p = D_{p_0} + \frac{2}{\rho_s} (k_1 k_2 - k_3) t = D_{p_0} + Kt \quad (9)$$

where  $K [2(k_1 k_2 - k_3)/\rho_s]$  is the net growth rate.

The validity of equation (9) will be tested against experimental data, and an attempt will be made to relate the net growth rate ( $K$ ) with those operating variables which are expected to have an effect on particle growth rate.

#### 4. Experimental Results

The operating conditions and the experimental results are summarized in Table 1.

The first step in data analysis was to find a suitable mean diameter of the particles to characterize the size distribution as determined by screen analysis. Several types of mean diameters (length mean, surface mean, weight mean, sauter mean or reciprocal mean) were calculated from the screen analysis data. The most consistent trends of mean diameter versus time plot were obtained with reciprocal mean diameter. The reciprocal mean diameter were therefore selected

**Table 1** The Operating Variables and the Experimental Results of the Growth and Attrition Runs

Run No.	Seed size (mm.)	Bed height (cm.)	Air velocity (cm./sec)	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> feed rate* (gm./min)	Net growth rate (mm./hr)	% of feed lost as fine dust***
1	1.17-1.68	46.0	60.6-70.7	13.0	0.0863	17.0
2	1.17-1.68	46.0	57.6-77.8	16.2	0.1083	11.5
3	1.17-1.68	46.0	58.6-80.8	17.5	0.1111	18.9
4	1.17-1.68	46.0	55.6-80.8	20.3	0.1024	30.4
5	1.68-2.00	46.0	72.7-85.9	20.0	0.1048	22.2
6	2.00-2.38	46.0	80.8-121	19.3	0.1123	11.9
7	2.38-2.83	46.0	101-143	35.2	0.2044	30.2
8	2.83-3.36	46.0	101-143	38.4	0.1493	45.0
9	0.99-3.36	46.0	63.6-85.9	22.6	0.0957	25.9
10	0.70-3.36	46.0	62.6-96.0	17.7	0.0713	9.6
11	2.38-4.70	26.0-36.5	60.6-126	40.0**	-0.1600	-
12	2.38-4.70	30.5-36.5	98.5-126	0	-0.0400	-
13	2.38-2.83	20.0-26.0	56.6-66.7	0	-0.0264	-
14	2.38-2.83	19.0-26.0	56.6-66.7	11.0**	-0.0953	-
15	2.38-2.83	18.0-26.0	52.5-68.7	20.5**	-0.1320	-
16	1.68-2.00	18.0-20.0	48.5-52.5	0	-0.0323	-
17	2.00-2.38	18.0-20.0	52.5-64.6	0	-0.0130	-

\* Expressed as 100% (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>.

\*\* Water was used instead of ammonium sulphate solution.

\*\*\* Estimated from material balance.

as a basis for further analysis of the data. This choice is also justified on physical grounds for it gives an indication of surface area available per unit volume<sup>9</sup>).

#### 4.1 Temperature Profiles in Beds

A typical temperature profile in the bed is shown in Figure 2. The bed temperature in both the spout and the annulus dropped sharply from 200°C to 75°C in the bottom 10 cm. of the bed and remained constant above the  $l/Z=0.5$  level. Therefore, it was concluded that all of the heat transfer from the air to the particles took place in this region by means of the high transfer rate in the spouted bed.

The air temperature differences between the bottom and the top of the bed steadily increased with the increase of solution feed rate as would be expected from heat balance considerations.

#### 4.2 Net Growth Rate (K)

Typical data for a growth run

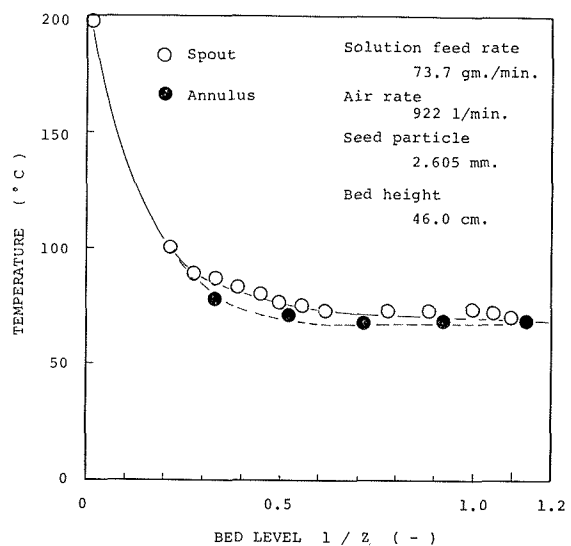


Fig. 2 A Typical Temperature Profile in Bed

are presented graphically in Figure 3, which shows the size distribution of the growing bed particles at different time intervals.

When the reciprocal mean diameters of the granulated particles were plotted against times, straight lines were obtained. Two such plots are shown in Figure 4 and Figure 5 which represent the minimum and the maximum scatter of data respectively. The slope of the line gives the net growth rate of the granules for the particular run in accordance with equation (9). The net growth rate for the various runs are shown in Table 1. In all cases, the slopes remained positive, indicating that normal growth due to solid deposition predominated over size reduction due to attrition.

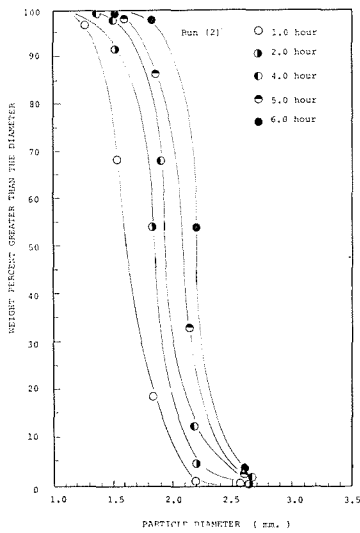


Fig. 3 Particle Size Distribution During Granulation Experiment.

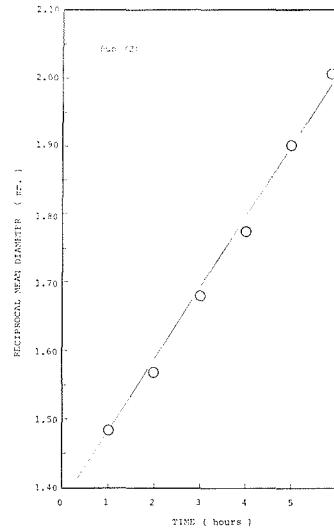


Fig. 4 The net Growth Rate of  $(\text{NH}_4)_2\text{SO}_4$  Particles (1).

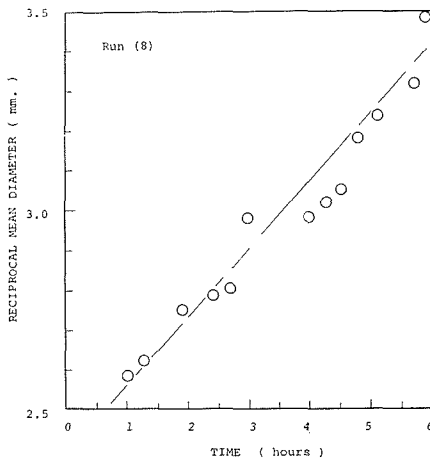


Fig. 5 The Net Growth Rate of  $(\text{NH}_4)_2\text{SO}_4$  Particles (2).

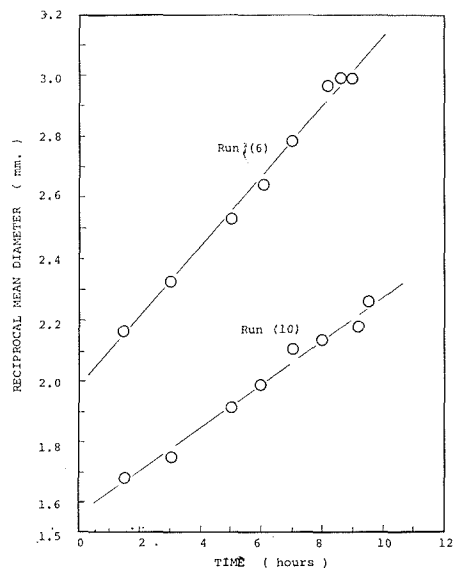


Fig. 6 The Net Growth Rate of  $(\text{NH}_4)_2\text{SO}_4$  Particles (3).

Most experiments were carried out over five to six hour periods except for two runs which were extended to 9.0 and 9.5 hours respectively. The results for these longer runs, shown in Figure 6. indicated that the mean size of the particles was still increasing.

The net growth rate would be expected to vary linearly with the feed rate of ammonium sulphate solution<sup>7)</sup> since with the bed weight hold constant, the amount of solution available for coating per unit weight of the bed particles increased. When the net growth rate values were plotted against the corresponding solution feed rates on a log-log scale, a straight line was obtained as shown in Figure 7. The slope of the line was 0.8. This indicated the dependence of the net growth rate ( $K$ ) on solution feed rate was weaker than expected. It could be related to the significant amount of dust being produced during the experiment, as any fine particles elutriated were equivalent to the loss of material that could have contributed to the growth of the particles in the bed.

The effect of the seed size on the net growth rate is shown in Figure 8. It was found that the net growth rate was independent of the seed size under these operating conditions.

#### 4.3 Attrition Rate (R)

Typical data for the attrition runs are presented graphically in Figure 9 and Figure 10. Figure 9 shows the size distribution of the ground particles at different time intervals and Figure 10 shows the size reduction rate of the particles.

Particle breakdown in a spouted bed would be expected to occur by two mechanisms as suggested in a jet-suction pulverizer<sup>10)</sup>. The first mechanism is surface grinding, which comes about by the surface abrasion of particles. The second mechanism is bulk crushing, which occurs by breakage of particles into smaller fragments. If bulk crushing is predominant in the spouted bed, particles are initially crushed into the intermediate size particles and the fines appear thereafter. On the other hand, when surface grinding is predominant, the fine

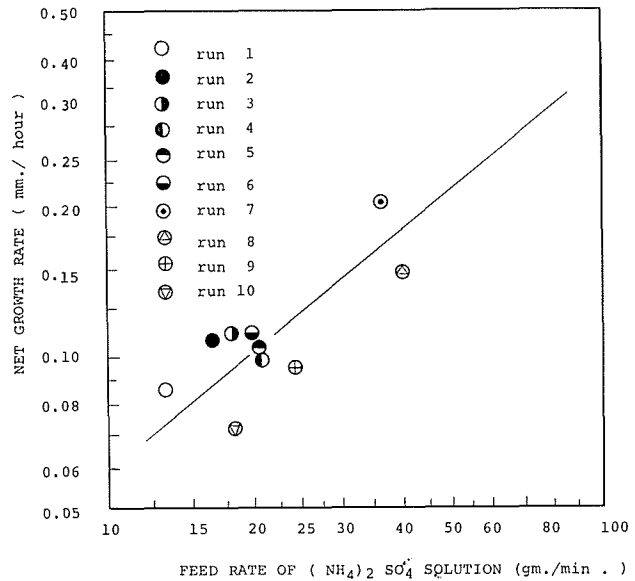


Fig. 7 The Variation of Net Growth Rate with Solution Feed Rate

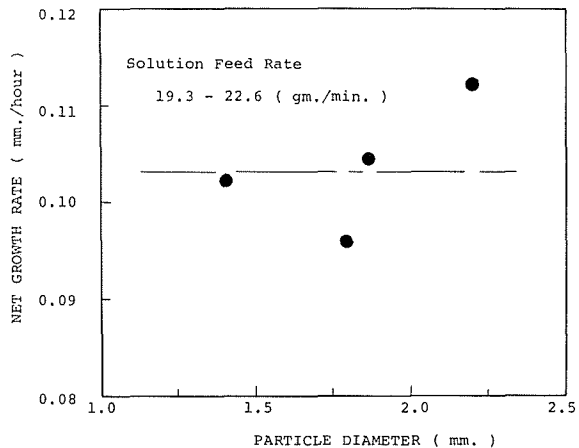


Fig. 8 Relation of Particle Diameter and Net Growth Rate

particles are produced at the beginning of grinding and the fraction of them increases gradually during the run. Therefore, the size reduction rate would be expected to be proportional to the particle surface area when surface grinding predominates over bulk crushing.

It was found that the size reduction of particles in the spouted bed occurred by the surface abrasion as shown in Figure 9. Furthermore, the linearity of the graph in Figure 10 proved that it was a reasonable assumption in equation (7a) that the size reduction rate was proportional to the particle surface area, and the validity of equation (7a) was confirmed.

The attrition rate for the various runs are shown in Table 1, and it is found that the attrition rate is independent of the seed particle size under these operating conditions. The effect of water feed rate on the attrition rate is shown in Figure 11, and it indicates the attrition rate is dependent on moisture content in the particles. When water was sprayed into the bed, the particles would have a higher moisture content than without injection. The moist particles were more friable and hence were abraded faster.

The data in Figure 11 are insufficient for giving any quantitative correlation between the attrition rate and the particle moisture content, but the fact that the attrition rate increase with moisture content is qualitatively established.

The results also indicated the size reduction rate during a granulation run were not negligible relative to the net growth rate.

#### 4.4 Residence Time Coefficient and Normal Growth Rate

Deposition of solids would be expected to occur mostly in the spout region of the bed, since the feed solution was injected into the centre of the bed at the conical base of the granulator. This indicates the seed particles grow only while passing through the spout. Hence, the time a particle growing with the spray droplets can be estimated from the residence time of a particle in the spout which

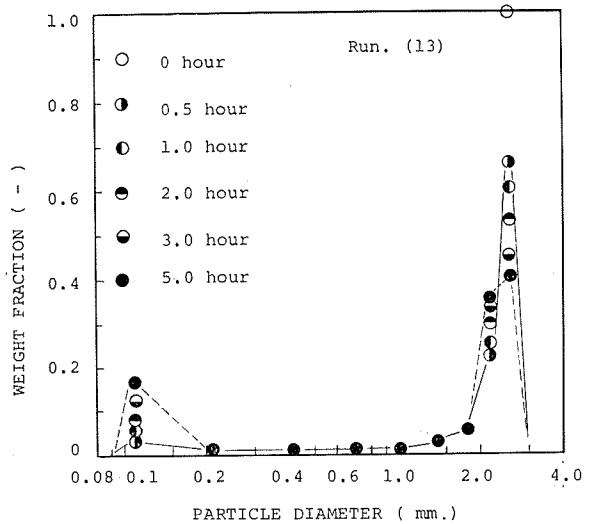


Fig. 9 A Typical Size Distribution Pattern of Grinding in a Spouted Bed

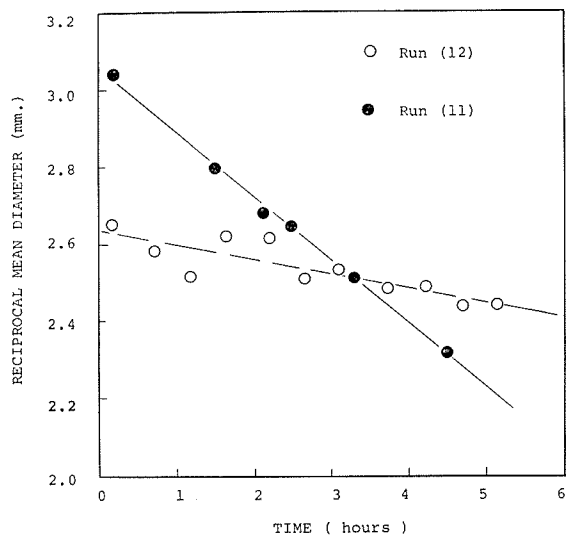


Fig. 10 The Attrition Rate of  $(\text{NH}_4)_2\text{SO}_4$  Particles

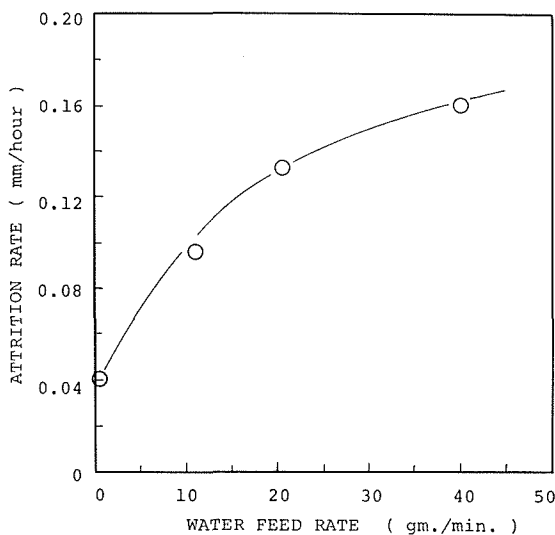


Fig. 11 Relation of Water Feed Rate and Attrition Rate

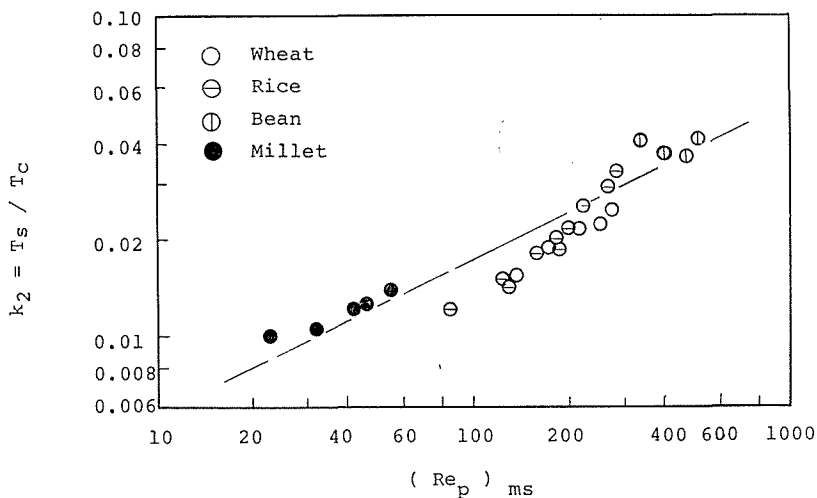


Fig. 12 Relation of  $(Re_p)_{ms}$  and Residence Time Coefficient

is given by the hydrodynamic features of the spouted bed.

An attempt to relate the residence time of particles in the spout with the variables of the system has been made by Yokogawa<sup>11)</sup>. He solved numerically the equation of motion and obtained some diagrams which provided the relationship of the mean particle residence time in the spout and  $Z/D_c$  as well as  $U/U_{ms}$  on the basis of his experimental data

The proportion of time spent by the particles passing through the spout to the mean one-cycle time of the particles in the spouted bed ( $T_s/T_c$ ), calculated by the authors from his data, is shown in Figure 12, and these data could be correlated by the following equation ;

$$k_2 = \frac{T_s}{T_c} = 0.0017 \sqrt{(Re_p)_{ms}} \quad (10)$$

where

$$(Re_p)_{ms} = \frac{D_p U_{ms} \rho_f}{\mu_f} \quad (11a)$$

$$U_{ms} = \left(\frac{D_p}{D_c}\right) \left(\frac{D_i}{D_c}\right)^{1/3} \sqrt{\frac{2gZ(\rho_s - \rho_f)}{\rho_f}} \quad (11b)$$

In the above equation the effects of solids properties, bed height and orifice diameter were all reflected in  $(Re_p)_{ms}$ . Figure 12 shows that  $k_2$  increases with increasing particle diameter and bed height.

The residence time coefficients estimated from equation (10) were 0.015–0.02 for the present experimental date. Thus, the normal growth rate ( $2k_1/\rho_s$ ) in equation (9) could be deduced from the net growth rate ( $K$ ), the attrition rate ( $R$ ) and the above mentioned residence time coefficient ( $k_2$ ). The values of the normal growth rate calculated by equation (9) were 11.2–21.4 mm./hr.

#### 4.5 Granule Structure

The sectioned particle is shown in Figure 13. The picture shows distinctly the boundary separating the nuclei and the surrounding shell deposited during the granulation experiment. The interface appeared to be the weakest region of the particle.

The sectioned particles were observed under a high power microscope (100X). It showed that the particle growth was due solely to the layering process, since the structure of the deposited material was uniform and without any agglomerates.

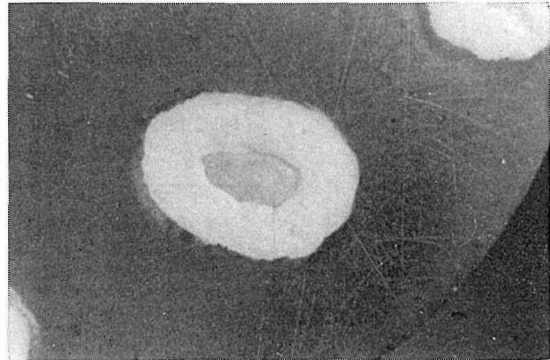


Fig. 13 Granule Structure

### 5. Conclusions

A spouted bed technique was applied to the granulation of fertilizer and the effects on the growth rate of operating variables were investigated.

(1) The growth mechanism in a spouted bed granulator was solely by layering process, and the net growth rate, the attrition rate and the residence time coefficient, given by equation (9), were determined by the experimental data.

(2) The bed temperature both in the spout and the annulus fell sharply in the bottom of the bed and remained constant above the half-level.

(3) The net growth rate of the ammonium sulphate particles in the spouted bed granulator was independent of the seed particle diameter. With increasing solution feed rate, the net growth rate increased in approximate proportion to feed rate raised to the power of 0.8.

(4) Size reduction rate of ammonium sulphate particles in the spouted bed due to attrition of surface grinding was dependent on moisture content in the particles.

(5) Considerable amount of fine dust as shown in Table 1 was produced and was elutriated out of the bed during granulation. It was caused by incomplete deposition of sprayed solution and the attrition of the particles in the spout.

### Nomenclature

- $D_c$  = column diameter, cm  
 $D_i$  = diameter of gas inlet orifice, cm  
 $D_p$  = particle diameter, cm  
 $K$  = net growth rate, cm/sec or mm/hr  
 $k_1$  = constant defined in equation (1), gm/sec·cm<sup>2</sup>  
 $k_2$  = constant defined in equation (5), dimensionless  
 $k_3$  = constant defined in equation (7a), gm/cm<sup>2</sup>·sec  
 $l$  = height above gas inlet, cm  
 $m$  = mass of particle, gm  
 $R$  = size reduction rate, cm/sec or mm/hr  
 $S_p$  = surface area of particle, cm<sup>2</sup>  
 $t$  = residence time of particle in bed, sec or hr  
 $T_c$  = particle cycle time in bed, sec or hr  
 $t_s$  = residence time of particle in the spout, sec or hr  
 $U$  = superficial gas velocity, cm/sec  
 $U_{ms}$  = minimum superficial gas velocity for spouting, cm/sec  
 $(Re_p)_{ms}$  = Reynolds number at minimum spouting, dimensionless  
 $\mu_f$  = fluid viscosity, gm/cm·sec  
 $\rho_f$  = fluid density, gm/cm<sup>3</sup>  
 $\phi_s$  = sphericity of particle, dimensionless

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