——技術資料——

On the Operation of a Continuous Granulation Plant-Manufacturing of Fertilizer Granules

Chang Dae Han*

1. Objective

The objective of this review is to dicuss some of the problems associated with the operation of industrial granulators of fertilizer manufacturing, from the point of view of process control. A smooth operation of any chemical plants requires a better understanding of the basic principles involved in various processes and in unit operations. And a better control scheme for a given process requires both the steady-state and dynamic behaviors of various pieces of equipments involved in the process. This is ths hope of this literature review to give some insight of this aspect of the problems to the readers, with an example of granulation process of fertilizer manufacturing.

2. Introduction

Granulation—the act of forming or crystallizing into grains, granules or small masses-occurs in many processing in dustries. It is becoming more and more important in the pharmaceutical, chemical, ceramic, powdered metal, non-ferrous metal and iron and steel industries.

Although granulation has been employed for the past few decades as an industrial manufacturing porcess, the control of such a process (based on the modern concepts of automatic control theory, rather than on the operator's experience or intuition) does not seem to have been touched upon. At a first glance, this kind of process may appear unattractive for a more sophisticated control approach because it looks "awkward." The movement and handling of solids is obviously not as simple and readily definable as the piping of liquids and gases. However, recent developments in the chemical engineering technology of this and related chemical and physical processes have given us a better understanding of the main features of the granulation process.

Consider a typical granulation process as sketched in Figure 1. We are considering here only granulators of the rotating cylinder type, rotating about the axis, which is slightly inclined to the horizontal. The granulator exit stream goes to the screen, through which particles are classified into three streams:

i) undersized particles which pass both the upper and lower sieve.

ii) on-sized particles which pass the upper sieve but are retained on the lower.

iii) oversized particles which are retained on the upper sieve.

The undersized particles are recycled to the inlet of the granulator. Oversized particles are crushed in the mill and the crushed particles recycled. Depending on the particular process, a portion of the on-sized particles may also be recycled (sometimes after crushing, although this is not shown in Figure 1).

Continuous granulation processes have a certain similarity to the continuous crystallization process in the sense that both nucleation and particle growth occur in the process loop, but they differ in that the granule size is increased by continuously adding feed material to a tumbling bed of granules. Therfore, it should

* The author is Associate Professor of Chemical Engineering, Department of Chemical Engineering, Polytechnic Institute of Brooklyn, Brooklyn, New York, 11201, U.S.A.

be pointed out that expressions for nucleation and growth rate must be included in the differential material balance equations.

Under steady-state conditions there must be a definite and constant number of granules in the granulator at all times. These granules will have a definite size distribution and a steady-state is reached if the rates of formation and growth do not change with time. Since a constant number of granules will be formed per unit time, the rate of formation of granules must be equal to the rate at which granules are withdrawn from the granulation plant as product. Also the rate of growth must adjust itself in such a manner that granules formed will grow to the desired size in the time available for their growth.

A literature review(Section 3) indicates that continuous operation of industrial granulators suffers from, for one thing, sudden increases of recycle rate which lead very frequently to plant shutdown. By "recycle rate" is meant the mass flow rate of granules entering the granulator. Control of the recycle is related to control of the solid-liquid ratio in the granulator and seems to be related to the grinding operation. The former determines granule growth and the latter determines the formation of new particles (nucleation).

For the purposes of this review, we will concentrate on one specific plant class, typified by that for the production of the fertilizer diammonium phosphate (DAP). In DAP plants, it is customary to react dilute phosphoric acid (in the region of 55% concentration) with ammonia vapor to produce a fine suspension of mixed monammonium and diammonium phosphates in water (hereinafter called "slurry", although the particle size is in the sub-micron range). The slurry is then piped into an inclined rotating drum so as to spray upon a tumbling bed of DAP granules. Ammonia vapor is also piped into the granulator, but the distributor is located in the bottom of the drum, so that it is always submerged in the granule bed. It will be assumed (based on the literature) that granules grow by being wetted with slurry, which then reacts with ammonia to complete the reaction to DAP, a large part of the moisture being driven off by the heat of reaction. A succesison of such wettings, reactions and dryings will deposit a sufficient number of layers to build up granules to the required size. This growth mechanism is referred to in the literature as the "Onion-skin" mechanism.

Any further drying of the granules will be done external to the granulator. The granules must be cooled before going to storage, and cooler can be installed to handle either the total flow from the dryer or only that portion which is withdrawn as product.

3. Literature Survey

3.1. Theory and Practice of Granulation

The term granultion is applied generally to many processes in the fields of fertilizers, pharmaceuticals, ceramics and others. These processes differ markedly in the types and forms of feed streams, and in the manner in which individual particles increase their size. However, they all have in common the fact that the granulator product is a stream of prticles having a comparatively wide size range, that this stream is screened to remove particles whose sizes fall in a range acceptable for packaging and sale, and those particles rejected are recycled to the granulator inlet after a size reduction stage if necessary.

With such a wide disparity of processes to review, we restrict ourselves to a class of fertilizer manufacturing granulation processes. Even here, one is faced with a choice because of the variety of products marketed in the granular form. Examination of the literature (see references 1 through 11) discloses that most granular fertilizers are of the mixed type. In the preparation of mixed fertilizers(references 1 through 7), two or more materials, such as superphosphate, potassium chloride, ammonium sulfate, etc., are to be made up into granules containing all components. The raw materials are prepared as powders and fed at controlled rates into the rotating granulator where the powder grains are made to adhere together by spraying with some fluid (water, acid, steam, etc.). Obviously, the granules build up by agglomeration of grain to grain and particle to particle, the so-called "Snowball" mechanism. If we seek a somewhat simpler process, with, say, only one solid feed stream and one fluid stream, we find that the process for the manufacture of diammonium phosphate (references 8 through 11) fits the bill. At first glance (see Fig. 2, it appears that we have two fluid streams, one a dilute slurry of sub-micron particles of mono- and diammonium phosphates in water, and the other ammonia vpoar. However, these two streams stand in a fixed relation to each other and can be regarded as one. The diammonium phosphate granulation process is the one we have chosen to discuss.

In general, the literature shows very little attention paid to mathematical analysis. A great deal of thought has been devoted to the chemitry of the systems, with pragmatic determinations of such parameters as the mole ratio of ammonia to phosphoric acid in the slurry (8, 9, 10). Further attention has been devoted to the manual operation of units, and the fluctuations and shutdowns they are prone to. The first real attempt at analysis is Han's (11), who proposes a model which assumes uniform residence time in the granulator and uniform particle size in the crusher output.

If the overall process is considered, analogies can be drawn between granulation and crystallization. In both processes, nucleation takes place and particles grow by deposition of "solute" on nuclei either from solution (crystallization) or by wetting with slurry followed by contacting with ammonia (granulation). If the crystallizer has facilities for screening of product and recycling of off-size, the analogy is closer still. In the light of this resemblance, the approach used by Han in crystallizer studies (12, 13, 14) is worth consideration.

3.2. Particle Growth Mechanism

Two basic mechanisms can be postulated for particle growth in environments in which particles tumble in a liquid medium. In one case wetted particles collide and adhere to each other, possibly due to the surface tension of the interstitial liquid, i,e. the snowball effect. In the other case, wetted particles absorb solute from the liquid, with excess liquid driven off by some means, thus depositing a layer. Successive wettings and dryings will then build the particle size up to the required value. Particles grown under these conditions show a typical pattern of concentric rings when sectioned, and the mechanism is therefore called the "Onionskin" mechanism.

In the manufacture of mixed fertilizers, where no chemical reaction occurs between the components, the only mechanism for growth is the snowball one. However, in DAP units, where material is deposites as the result of a chemical reaction, particles can grow by both the snowball and the onionskin method. The factor which determines which indeed takes place is the ratio of solid to liquid feed, in "wetter" systems, particles tend to grow by agglomeration, in "dryer" ones by deposition (15, 16, 17, 18, 19). Since the greater part of granular fertilizer production is devoted to mixed fertilizers of more than one component, most published material is devoted to particle agglomeration. Some insight into onionskin growth can be obtained from work done in other processes (20, 21, 22, 23).

Katz and some of his associates (24, 25) have given mathematical formulations for determining particle size distributions in systems where particles grow or shrink according to some prescribed differential law. They make use particularly of the various moments of the particle size distribution functions in order to determine the dynamic behavior of the particle parameters of size, surface and mass.

3. 3. Particle Size Analysis

The mathematical treatment of particle size distributions is a branch of probability theory and has been covered in several useful books. Cadle (26) and, particularly Herdan (27), present clear and thorough expositions of the subject. The basic mathematics is, of course, inherent in Katz's work (24, 25) mentioned above. In order to simplify experimental calculations, Kaye and Treasure (28) have developed a graphical method for determining third moments for given particle size distributions. This is, of course, necessary in the transformation of a distribution by number to one by weight. Capes (29) has contributed some useful information on the development of particle size distributions as granulation proceeds.

3. 4. Comminution and Classification

We are not in this review primarily concerned with the relation between power input to size reduction machinery and the characteristics of the product. Most of the older literature in this field is more or less restricted to this consideration and is conveniently summarized in Perry (31). One's main concerns are to ascertain the functional forms of the particle size distributions encountered in "real life" particulate solids, the relation between the particle size characteristics of mill feed material and mill product, and so on. Beke (30) appears to be one of the few to study such matters in comminution theory. He has found that for dynamic impulse machinery (which categorizes the hammer mills most often used in granulation plants) the product size distribution has the log normal form regardless of the feed size distribution, a result of the product classifier usually present in such machines. Beke further establishes a qualitative relationship between the product size distribution and the machine rotational speed, i. e. the faster the rotation, the finer the product, the smaller its mean size and standard deviation and hence the larger the number of particles.

Mining companies in Australia appear to have devoted a great deal of effort to the analysis of the grinding part of mining operations (32, 33, 34, 35). Their aim has been to ultimately control their operations automatically and they claim fair success. A basic tool they have used is the matrix representation of particle assemblies.

Arbiter and Harris (36) have studied batch milling and have developed a three-dimensional representation to show how particle size distribution varies with the number of mill revolutions. If one relates the speed of a continuous mill to the number of revolutions of a batch mill, their results exactly confirm Beke's observations.

An analytical basis for particle size classification still appears to be far away. Most published work pertains to fine powders, 50μ and smaller, whereas we are dealing with size ranges of 1—8 mm. Perry (31) has summarized the older literature, and none of it is really predictive, that is no method is made available to predict screen efficiency given a particle size distribution, flow rate and screen size. Rendell and Mullin(38) have devised a simple single-aperture model to investigate the flow of particles through screens, but their results so far are inconclusive. Beke (30) quotes an analytical proof that any screen is 100% efficient only when a given particle is in contact with it for an infinite time.

3.5. Residence Time Distribution in Granulator

Little work has been published on residence time in granulators per se. However, various workers have studied the problem in similar rotary equipment such as kilns(39), dryers(41, 44) and coolers (45). However, while the authors are aware of the fact that residence time is not uniform in such machinery, they apply their efforts to seeking theoretical or empirical expressions for average residence time.

Danckwerts(40) has studied the problem in generalized continuous flow systems, using a probabilistic approach. Naor and Shinner(42) have developed the method further on the same generalized basis, while others(43) have applied it to experimental studies on fluidized and moving beds with continuous flow.

4. Areas of investigation for process control

The state of the art of controlling granulation processes seems to be quite primitive as yet. Most industrial granulation plants are being operated by skilled operators, using the experience they have gained in similar processing units. Needless to say, the successful operation of a process based on a sophisticated control technique depends on a successful modelling of the process. Therefore, the successful simulation model is essential to the control study.

Han's paper(11) on a simplified steady-state model indicates clearly how overall plant performance is bound up with the recycle ratio. The total recycle consists of three components, the undersize, part of the on-size and the crushed oversize. This immediately suggests the areas to be investigated in the control study:

a) Relation between recycle and screen specifications. Given the particle size distribution of the granulator discharge, what fractions are rejected as oversize and undersize? A minor consideration (at this point) might be the influence of screen loading on screen efficiency.

b) Relation between recycle and granulator operation. What effect does particle growth rate have on the particle size distribution and hence on the fractions of off-size rejected? This problem is closely tied into the solid to liquid ratio. For stable operation two conditions are necessary. First, the average particle size should not be a too sensitive function of the ratio of liquid to solid feed. Second, the average particle size should be near the value corresponding to minimum recycle, otherwise the recycle ratio will vary too strongly. It is obvious that one cannot operate the process continuously unless the recycle ratio remains fairly close to the design level because of the design limitations of the auxiliary equipment. Therefore, a knowledge of the exact form of the relation between the average product size and the recycle ratio as a function of liquid to solid feed ratio would be very important for a quantitative investigation of useful control schemes.

c) Relation between recycle and grinding. Any increase in particle surface area caused by reducing the crushed particle size will not only change the particle balance in the process loop but also decrease the average particle size of the granulator exit stream.

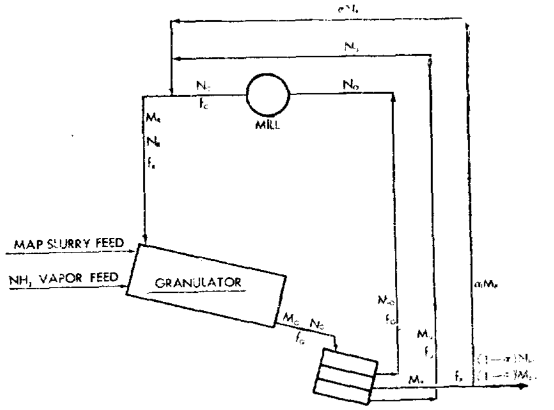

Figure 1. Schematic of DAP Granulation Process.

This in turn will increase the recycle ratio because of the increase in the fraction of under size passing the lower sieve. Consequently, an increased recycle rate decreases the liquid content of the granulating mixture, thereby decreasing particle size.

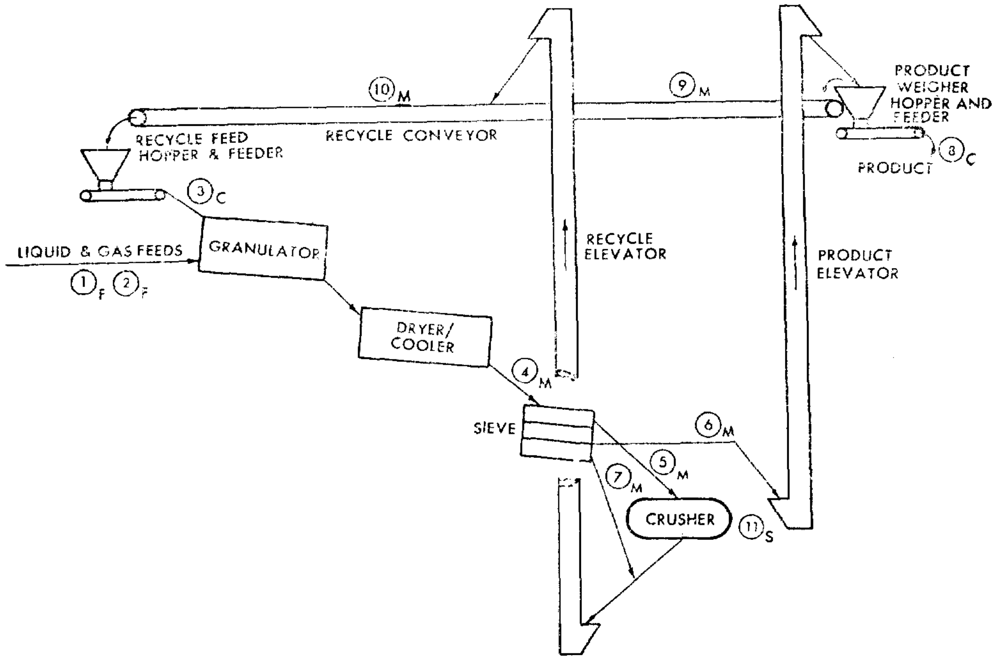

Figure 2. Simplified Flow Diagram of Granulator Plant.

References

1) Hein, L. B., Hicks, G. C., Silverberg, J. and Seatz L. F., *Granulation of High Analysis Fertilizers*, Agricultural and Food Chemistry, Vol. 4, No. 4, p. 318, April 1956.

2) Hignett, T. P., *The Changing Technology of Granulation and Ammoniation*, Agricultural Chemicals, Jan. 1967.

3) Brook, A. T., *Developments in Granulation Te-*

chniques, Proceedings No. 47, The Fertilizer Society (London, U. K.), Nov. 1957.

4) Phillips, A. B., Hicks, G. C., Jordan, J. E. and Hignett, T. P., *Effect of Particle Size of Raw Materials on Granulation of Fertilizers*, Agricultural and Food Chemistry, Vol. 6, No. 6, p. 449, June 1958.

5) Sauchelli, V., *Chemistry and Technology of Fertilizers*, Reinhold, New York, 1960.

6) TVA, Wilson Dam, Ala., *Report on Pilot Plant Demonstrations of the Production of Granular Fertilizers, June* 9 & 10, 1959.

7) Hignett, T. P., *Granulation of Fertilizers*, Farm Chemicals, Jan., Feb., March & April, 1963.

8) Young, R. D., Hicks, G. C., Davis, C. H., *TVA Process for Production of Granular Diammonium Phosphate*, Agricultural and Food Chemistry, Vol. 10, No. 6, p. 442, Nov. –Dec. 1962.

9) Horn, W. R. and Fouser, N. D., *Production of* 18-46-0 *Fertilizer-A Comparison of Processes*, Chemical Engineering Progress, Vol. 62, p. 109, July 1966.

10) Davis, C. H., Meline, R. S., Graham, H. G., *TVA Mixed Acid Nitric Phosphate Process*, Chemical Engineering Progress, Vol. 64, No. 5, p. 75, May 1968.

11) Han, C. D., *Steady-State Behavior of Continuous Granulators An Elementary Mathematical Analysis*, Submitted to I. & E. C. Fundamentals.

12) Han, C. D., *Determination of Crystal Growth Rate by Analog Computer Simulation*, Chemical Engineering Science, Vol. 22, p. 611, 1967.

13) Han, C. D., *Evaluation of Some of the Kinetic Parameters in Crystallization*, Chemical Engineering Science, Vol. 23, p. 321, 1968.

14) Han, C. D. and Shinnar, Reuel, *The Steady State Behavior of Crystallizers with Classified Product Removal*, A. I. Ch. E. Journal. Vol. 14, No. 4, p. 612, July 1968.

15) Newitt, D. M., Conway-Jones, J. M., *A Contridution to the Theory and Practice of Granulation*, Trans, Instr. Chem. Engrs., Vol. 36, p. 423, 1958.

16) Farnand, J. R., Smith, H. M. and Ruddington, I. E., *Spherical Agglomeration of Solids in Liquid Suspension*, The Canadian Journal of Chemical Engineering," p. 94, April 1961.

17) Sutherland, J. P., *The Agglomeration of Aqueous Suspensions of Graphite*, The Canadian Journal of Chemical Engineering, p. 268, Dec. 1962.

18) Capes, C. E. and Danckwerts, P. V., *Granule Formation by the Agglomeration of Damp Powders; Part I: The Mechanism of Granule Growth; Part II: The Distribution of Granule Sizes*, Trans. Instr. Chem. Engrs., Vol. 43, p. T 116, 1965.

19) Capes, C. E., *Mechanism of Pellet Growth in Wet Pelletization*, I. & E. C. Process Design and Development, Vol. 6, No. 3, p. 390, July 1967.

20) Dunlop, D. D., Griffin, L. I., Moser, J. F., *Particle Size Control in Fluid Coking*. Chemical Engineering Progress, Vol. 54, No. 8, p. 39, August 1958.

21) Lee, B. S., Chu, J. C., Jonke, A. A. and Lawroski, *Kinetics of Particle Growth in a Fluidized Calciner*, A. I. Ch. E. Journal, Vol. 8, No. 1, p. 53, March 1962.

22) Grimmett, E. S., *Kinetics of Particle Growth in the Fluidized Bed Calcination Process*, A. I Ch. E. Journal, Vol. 10, No. 5, p. 717, Sept. 1964.

23) Grimmett, E. S., *Particle Growth and Size Distribution in Fluidized Bed Processes A Mathematical Model with Computer Solutions*, Presented at 58th Annual Meeting of A. I. Ch. E., Dec. 1965.

24) Behnken, D. W., Horowitz, J. and Katz, S., *Particle Growth Processes*, I. & E. C. Fundamentals, Vol. 8, No. 8, p. 212, August 1963.

25) Hulbert, H. M. and Katz, S., *Some Problems in Particle Technology; A Statistical Mechanical Formulation*, American Cyanamid Company report, July, 1963.

26) Cadle, R. D., *Particle Size: Theory and Industrial Applications*, Reinhold, New York, 1965.

27) Herdan, G., *Small Particle Statistics*, Academic Press, 2nd edition, 1960.

28) Kaye, B. H. and Treasure, C. R. G., *Data Handling Techniques for Particle Size Analysis*, British Chemical Engineering, Vol. 11, No. 10, p. 1220, Oct. 1966.

29) Capes, C. E., *A Note on Size Distribution in Granulation, Balling and Wet Pelletization*, Trans. Instr. Chem. Engrs., Vol. 45, No. 3, p. CE 78, 1967.

30) Beke, Béla, *Principles of Comminution*, Akadémiai Kiadó, Budapest, 1964.

31) *Perry's Chemical Engineers Handbook*, McGraw-Hill, 4th edition, 1963, Section 8, *Size Reduction and Size Enlargement* Section 21, Subsection on Screening Performance.

32) Lynch, A. J., *A Mathematical Model of a Multi-Stage Comminution System*, 58th Annual Meeting, A. I. Ch. E., Dec. 1965.

33) Calcott, T. G., *The Production of Constant Size Distributions*, 58th Annual Meeting, A. I. Ch. E., Dec. 1965.

34) Broadbent, S. R. and Calcott, T. G., *A Matrix Analysis of Processes Involving Particle Assemblies*, Phil Trans. Roy. Soc., Part A., Vol. 249, p. 99, 1956.

35) Reid, K. J., C. S. I. R. O., *A Solution to the Batch Grinding Equation*, Chemical Engineering Science, Vol. 20, p. 953, 1965.

36) Arbiter, N. and Harris, C. C., *Particle Size Distribution-Time Relationships in Comminution*, British Chemical Engineering, Vol. 10, No. 4, p. 240, April 1965.

37) Suzuki, Akira and Tanaka, Tatsuo, *Crushing Efficiency in Relation to Some Operational Variables and Material Constants*, I. & E. C. Process Design and Development, Vol. 7, No. 2, p. 161, April 1968.

38) Rendell, M. and Mullin, J. W. *The Flow of Particles Through a Screen Aperture.*

39) Saeman, W. C., *Passage of Solids Through Rotary Kilns-Factors Affecting Time of Passage*, C. E. P. 47, pp. 508-514, Oct. 1951.

40) Danckwerts, P. V., *Continuous Flow Systems, Distribution of Residence Times*, Chemical Engineering Science, Vol. 2, No. 1, p. 1, Feb 1953.

41) Miskell, F. and Marshall, W. R., *A Study of Retention Time in a Rotary Dryer*, Chemical Engineering Progress, Vol. 52, No. 1, p. 35-J, Jan. 1956.

42) Naor, P. and Shinnar R., *Representation and Evaluation of Residence Time Distributions*, I. & E. C. Fundamentals, Vol. 2, p. 278, Nov. 1963.

43) Morris, D. R., Gubbins, K. E., Watkins, S. B., *Residence Time Studies in Fluidized and Moving Beds with Continuous Solids Flow*, Trans. Instr. Chem. Engrs., Vol. 42, p. T 323, 1964.

44) Schofield, F. R. and Glikin, *Rotary Dryers and Coolers for Granular Fertilizers*, Trans. Instr. Chem. Engrs., Vol. 42, p. 183, 1962.

45) Turner, G. A., *The Thermal History of a Granule in a Rotary Cooler*, The Canadian Journal of Chemical Engineering, p. 13, Feb. 1966.