Studies on Effect of Process Parameters on Particle Growth in a Fluidized Bed Granulator

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By

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CERTIFICATE

This is to certify that the project report entitled, "**Studies on Effect of Process Parameters on Particle Growth in a Fluidized Bed Granulato**r" submitted by **Lisa Sahoo** in partial fulfilments of the requirements for the award of Master of Technology Degree in Chemical Engineering at National Institute of Technology, Rourkela is prepared by her under my supervision and guidance. The candidate has fulfilled all prescribed requirements and the thesis, which is based on candidate's own work, has not been submitted elsewhere.

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ABSTRACT

In present work effect of various process parameters affecting the particle growth in a FBG is observed by performing experiments. The effect of the process parameters such as amount of liquid to be sprayed (V), fluidizing gas temperature (T), fluidizing gas flow rate(F) and amount of bed materials (W) on the growth rate of particles fluidized in a fluidized bed granulator is studied accordingly. The correlation for particle growth is also derived using dimensional analysis method. The dimensional analysis method adopted in this work is exponential in nature unlike the conventional method. The experimental value of particle growth and the calculated values of the particle growth obtained through dimensional analysis are compared. The results found from the comparison shows good agreement with each other. The optimization of the process parameter has been carried out to improve the performance of the fluidized bed granulator. MATLAB coding is developed for the optimization of developed correlation for particle growth using Interval halving method.

Keywords: Granulation, particle growth, Fluidized bed drying and granulation, Interval halving method, MATLAB coding

CHAPTER 1

INTRODUCTION

Particle products are drawing attention for its high surface to volume ratio availability in the reaction, solubility, coating and dispersion. Therefore they have becoming the goal in many industrial systems. Particle science and technology are also applied in industries and also taught in academics. These include reaction engineering, crystallization, precipitation, coating, granulation, drying, solids transport, solid/liquid and solid/gas separation etc. Granulation,as the name suggest is a size enlargement process by which fine powders are agglomerated into large agglomerates. Granulation improves flowability and appearance which finds great application in pharmaceutical industries. Dissolution rate, bulk density, reduction in caking formation and strength of granule is also improved. It improves handling of powder which are difficult to handle because of their cohesiveness and low flowability (Litster and Ennis,2004).Among the various granulation techniques, fluid bed granulation (FBG) is one of the most widely used.

Both dry and wet processes can achieve granulation. In wet granulation usually called binder granulation, a liquid binder such as a solution or a melt is pumped, poured or atomized into an agitated bed of powders contained in a granulator whose main role is to provide shearing forces to the powder mass. Binders are used for generating cohesiveness to agglomerating powders. Consolidation by shear forces and drying result in solvent evaporation from the binder or melt thickening as a result of which inter-particle bridges strengthen, powder particles stick together, and larger granules of the original powder are formed.

The control of fluidized bed granulation process is very much difficult as it involves wetting, drying and mixing of particles simultaneously. If the liquid is excessive most of the region of the bed may defluidize and particles stick to each other forming large wet lumps called wet quenching. While, if excessive particle growth occurs, the minimum fluidization velocity will exceed the operating velocity thereby causing defluidization which is called dry quenching. Therefore it is necessary to understand the mechanism of particle growth and to study the effect of various parameters on the growth rate of particles in a fluidized bed. There are various methods for optimization such as single variable and multi-variable optimization. One of the first numerical methods developed to find the root of a nonlinear equation is Interval Halving method. In the present study Interval Halving method is used for its several advantages over the other methods.

1.1 ADVANTAGES

Fluidized bed granulation (FBG) has many advantages over conventional methods. The conventional granulation methods require separate equipment. But FBG is performed in one unit which saves labour cost, transfer losses and time. Another advantage is that once the conditions affecting the granulation have been optimized the process can be automated. Granulation technology has a wide spectrum of applications ranging from pharmaceutical to food, fertilizer, and detergent to mineral, ceramic, waste processing, and advanced materials.

One of the simplest methods of finding the solution of a nonlinear equation is Interval Halving method. The major advantage of Interval halving is that the error in the solution is bounded. The absolute error is halved at each step so the method converges linearly, although it is slow.

1.2 OBJECTIVES

The objective of present work is

- ✓ To study the effect of various parameters such as amount of liquid sprayed, fluid temperature, superficial velocity, bed material on the growth rate in a fluidized bed granulator.
- ✓ To optimize the process parameter to improve the performance of the fluidized bed granulator.

1.3 PLAN OF THE THESIS

Present work has been planned to be reported in five chapters.

Chapter-1 deals with the introduction regarding to the granulation process and fluidized bed granulator including its key advantages and applications.

Chapter-2 deals with the literature surveys on the previous works related to the fluidized bed granulator and the granulation process.

Chapter-3 deals with the experimentation including the experimental set-up and the materials with the methods adopted for granulation.

Chapter-4 deals with results and discussion where the experimental observations are reported. These observations have been correlated against the different system parameters. The experimental and calculated values on particle growth are thus compared with each other. Optimization of the developed correlation is carried out using Interval Halving method. MATLAB coding is also used for the same.

Chapter-5 deals with the conclusion implying its use over a wide range of system parameters efficiently in a cost effective manner.

CHAPTER 2

LITERATURE REVIEW

Fluidized bed granulation is a process for the conversionof liquid products, e.g. suspensions, solutions ormelts, into granular solids. It is a fairly complex process because granulation and drying occur simultaneously. Agglomeration will occur if a wet particle collides with another particle and is bound by a liquid bridge which becomes solidified during the subsequent drying period.

2.1 GRANULATION

Granulation is a size enlargement process in which small particles are agglomerated together by spraying a liquid binder on to a dry powder bed (Roy, 2009). Agglomerates are formed by the aggregation of particulate solids that are held together by short-range physical or chemical forces acting among particles, by chemical or physical modification of the particles triggered by specific process conditions or by substances that act as binders by adhering physically or chemically to form material bridges among particles (Pietsch, 2003).

2.2 DRYING

Once a particle is wetted its drying begins immediately. Since agglomeration takes place primarily in the fluidized bed, particles will have time to dry during their residence time in the fluidizing column.In the drying stage, the agglomerated droplets are brought into contact with heated gas for the evaporation to take place equally from the surface of all droplets.

2.3 REASONS FOR GRANULATION

1.Many powders are cohesive and having poor flow properties for their small size, irregular shape or surface characteristics. This often results in a wide variation of mass in the final product resulting variable fill of tablet dies etc. Granules formed by granulation process will therefore contribute to improved flow properties.

2. Some powders are difficult to compact but granules easily get compacted and produce stronger tablets.

3. The hazards associated with the toxic materials will reduce that may arise during handling of powders. Proper precautions must be taken during the granulation process.

4. Hygroscopic materials may adhere and form a cake when stored as a powder. Granulation reduces this hazard as the granules absorb some moisture and retain their flowability.

5. As granules are denser than the parent powder material, they occupy less volume per unit weight. Therefore they are more convenient for storage or transportation.

2.4 TYPES OF GRANULATION

Granulation methods can be divided into two types: wet methods, which use a liquid in the process and dry methods in which no liquid is used.

1) Wet granulation

Wet granulation involves the massing of dry primary powder particles using a granulating fluid known as binder. The binder must be volatile so that it can be removed by drying, and be non-toxic. Typical binders include water, ethanol and isopropanol or combination of them. Water is commonly used from economic and ecological point of view. Its disadvantages are it may adversely affect drug stability and it needs longer drying time as compared to organic solvents. The main advantage of water is that it is non-flammable which reduces the safety risks.When water-sensitive drugs are formulated organic solvents are used or when a rapid drying time is required.

In the traditional wet granulation method the wet mass of powder is forced through a sieve to produce wet granules and subsequent screening breaks agglomerates and removes the fine material, which can then be recycled. The processes or techniques used for wet granulation are wet massing, fluidized bed granulation, spray drying, pan granulation and extrusion and palletizing.

2) Dry granulation

In the dry granulation method the primary powder particles are aggregated under high pressure. It can be of two types. Either a large tablet known as 'slug' is produced in a heavy-duty tabletting press (a process known as 'slugging') or the powder is squeezed between two rollers to produce a sheet of material ('roller compaction'). In both the cases a proper milling technique is adopted to produce granular materials which are then sieved to separate the desired size fraction. Dry methods are used for the drugs that do not compress well after wet granulation or those which are moisture sensitive. This granulation method includes Roller compaction and Slugging (Aulton, 2000).

2.5 FLUIDIZED BED GRANULATOR

Granulation in fluidized bed involves suspension of particulates in a stream of air and spraying a liquid from the top of the fluidized bed. Particles get wet when comes in contact with the sprayed liquid. The wet particles then collide with other particles and adhere to form granule.

In fluidized bed granulation particles are produced in a single piece of equipment by spraying a binder as solution, suspension, or melt on the fluidized powder bed. A fluidized bed granulator generally has high volumetric intensity because of its low voidage, and the size of equipment is much smaller compared to other types of granulators. This process involves two or more steps e.g., mixing and granulation, drying, decomposition and agglomeration etc. (Roy, 2009). Granules formed in Fluidized bed granulator are homogeneous. All the particles in the powder are sprayed evenly with liquid or binder. By adjusting various parameters the type of granule formed by this technique can be influenced over a wide range.

Mechanism

The rate processes in FBG is a combination of aggregation, binder solidification and breakage as illustrated in Fig 2.1 (Tan et al, 2004).

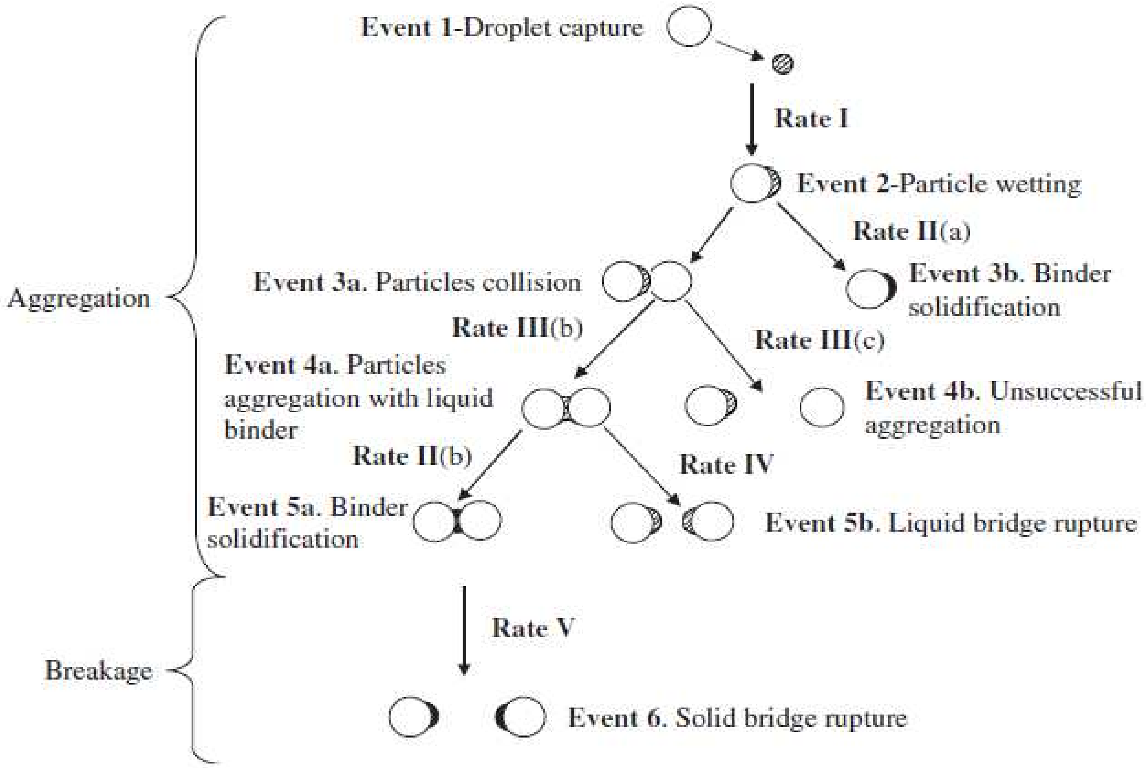


Fig 2.1 Sequence of rate processes in FBG (Tan et al, 2004)

In a top sprayed granulation, the binder droplets first collide with particles (Event 1) and consequently wet the particles in the spray zone (Event 2). The rate of droplet capture (Rate I) in this region is determined by the binder addition rate and the droplet size. There is a total separation in time scales of the various rate processes. The particle collision occurs on a µs time scale while binder solidification time is of the order of ms. Hence the probability of binder solidifying even before colliding with other particles (Event 3b) is very low. However, a more probable scenario is that the wetted particles (Event 2) will collide with other particles

(Event 3a) to form aggregates (Event 4a). The collided particles may also rebound (Event 4b), if the binder is not capable of causing successful aggregation upon collision. Depending on the binder solidification rate, the liquid bridge binding the particles will either solidify to form a granule (Event 5a) or rupture (Event 5b). However as the process proceeds, the solidified binder bridges may also rupture if the bonds are not strong enough to withstand the agitation of the fluidized bed (Event 6), creating a breakage event.

2.6 VARIABLES INFLUENCING PARTICLE GROWTH IN FBG

In order to get successful operation and desired end product control of the FBG process is essential. Many parameters have significant influence on the process.The main material and process variables affecting the quality of final product in FBG are listed in Table 2.1. Careful and accurate control of these parameters in FBG is necessary. Hence many studies have been carried out for betterunderstanding of these parameters and their effect on the process.

Table-2.1. Classification of fluid bed granulation parameters, modified from Schafer and Worts(1977) and Aulton and Banks (1981)

Apparatus parameters	Process parameters	Product parameters
Air distribution plate	Bed load	Type of binder
Shape of granulator body	Fluidizing air flow rate	Quantity of binder
Nozzle height	Fluidizing air temperature	Binder solvent
Positive or negative pressure operation	Fluidizing air humidity	Concentration of granulating solution
Scale-up	Atomization Nozzle type Spray angle Spraying regime Liquid flow rate Atomizing air flow rate Atomizing air pressure Droplet size	Temperature of granulation solution Starting Materials Fluidization Powder hydrophobicity

To troubleshoot any process, understanding the product, the equipment being used and the process is very important. The pharmaceutical industry and the suppliers of fluidized beds have implemented improvements in related hardware and process controls to minimize granulation problems. Factors affecting the fluid bed granulation can be divided into three broad categories.

2.6.1 Processes

There are several process parameters that can be adjusted in FBG. Although many studies have been carried out for single parameters in FBG it is important to recognize that the parameters do have inter-relationships and they may influence each other. The bed moisture content and the droplet size are two important elements in FBG. To prevent the over wetting of the powder mass, there should be equilibrium between the moisture intake and evaporation during the FBG (Kristensen and Schafer, 1987). Increase in liquid flow rate and inlet air humidity result to larger droplet size and higher bed moisture and hence larger granules are usually obtained (Davies and Gloor, 1971; Schaefer and Worts, 1978a; Schaafsma et al., 2000). However, if the air-to-liquid mass ratio is kept at a constant level, the increased liquid flow rate decreases the droplet size (Schafer and Worts, 1977b). Increase in atomising air decreases the droplet size and therefore smaller granules are obtained (Merkku et al., 1993; Juslin et al., 1995a-b; Yu et al., 1999; Hemati et al., 2003; Bouffard et al., 2005). Increased inlet air temperature and excess gas velocity enhance evaporation and hence decrease the granule size (Lipps and Sakr, 1994, Wan et al., 1999). According to pilot scale studies by Cryer and Scherer (2003), binder spray rate and binder droplet size explained 65% and 10% of the granule size variance, respectively. Rambali et al. (2001b) found that granule size can be optimized by using 3 fundamental variables: the powder moisture content, the droplet size and the air flow rate. Too high air flow rate can however result to attrition of the granules (Parikh, 1991). The granule breakage during the drying process is also dependent on the moisture content of the granules; dry granules are more prone for attrition and the fines fraction increases under stress (Nieuwmeyer et al., 2007a). Different testing methods have also been developed to study the attrition and breakage of the granules (Tardos et al., 1997; Airaksinen et al., 2000; Reynolds et al.,2005).

2.6.2 Materials

The properties of the starting material have an important role in FBG process. Since the wetting and the free water present on the surface of the particles are essential in the formation of granules, the particle size of the starting materials affects the granulation. As the decrease in particle size increases the total surface area of the mass, it also results in a smaller granule size (Schafer and Worts, 1977a; Ormos and Pataki, 1979b; Abberger et al., 2002). Small particle size and needle-like shape of the particles can also lead to problems in fluidization (Kristensen and Schafer, 1987; Juslin and Yliruusi, 1996b). Kristensen and Hansen (2006) used a rotary processor in FBG to compensate the impaired fluidization activity due to the increased cohesivity of the starting material. Absorbing materials, e.g. starch, result in incomplete wetting of the surface and thus the amount of liquid should be higher (Schaefer and Worts, 1977a; Schinzinger and Schmidt, 2005). The solubility of the starting material into the binding solution also influences the granule growth. Ormos and Pataki (1979a) compared 5 different materials that had different solubilities and found that the highest growth rate was obtained with the materials having the highest solubility. Drying is simultaneously occurring during the spraying phase in FBG. When higher binder concentrations are used, the evaporation of the solvent results in more viscous liquid bondings and more stabilized agglomerates. Consequently also the granule size is increased (Kristensen and Schafer, 1987). Relationship between final granule size and binder concentration has been well established (Davies and Gloor, 1972, 1973; Schaefer and Worts, 1978b; Alkan and Yuksel, 1986; Wan and Lim, 1991; Wan et al., 1992; Rohera and Zahir, 1993; Liu et al., 1994; Abberger, 2001b; Bouffard et al., 2005; Rajniak et al., 2007). Also, the type of binder has a role in the agglomeration phenomenon. In general, an increase in viscosity also results in increased agglomerates. Gelatin, however, has been found to form a portion of big granules even at low binder concentrations (Schaefer and Worts, 1978b; Ormos et al, 1979c; Georgakopoulos et al., 1983; Rohera and Zahir, 1993).

2.6.3 Variables for Equipment Design

Equipment variables in FBG have not been found to be as relevant as in high shear mixing (Kristensen and Schaefer, 1987). Proper fluidization can be obtained by different distributor plates as well as by varying FBG container shapes. Davis and Gloor (1971) found that the decrease in the nozzle height increased the average granule size slightly and decreased the friability of the granules. This was explained by the binder's increased ability to wet and penetrate the fluidized solids due to the shorter distance. If the nozzle is located at a too high position, the risk of spray drying and walls wetting also increases (Hemati et al., 2003). In other studies, the height of the atomizing nozzle or the nozzle diameter has had only little or no effect at all on the FBG process (Rambali et al., 2001a; Cryer and Scherer, 2003). On the other hand, too low position of the nozzle may result to clogging of the nozzle. The size of the granulator, however, can have a significant impact on the particle size and therefore the moisture content in the bed is the key parameter to be controlled in scale-up studies (Faure et al., 2001).

2.7 PREVIOUS WORKS ON FLUIDIZED BED GRANULATION

The first pharmaceutical experiments of fluid bed granulation were made by Wurster (1959, 1960) and ever since much effort has been focused on developing, studying and utilizing this method.

The fluid bed granulation process is an influenced by various processing parameters like atomization pressure, binder addition rate, inlet temperature etc. These process variables largely affect the granulation efficiency and the final granule characteristics. The effect of starting materials on granule properties was previously studied.

Liske and Mobus (1968) studied inlet air temperature and humidity, flow rate of air, nozzle position, spray rate and spray pressure in fluidized bed granulator and hence compared fluidized bed granulation with wet granulation process.

Parameters such as binder addition rate, air pressure to the binary nozzle, inlet air temperature in fluidized bed granulator has been studied by Davies and Gloor (1972) to describe effect of process variable on physical property of final granules.

Binder addition rate has received a lot of attention in terms of research. Therefore this parameter has been studied by many authors. Walker (2006), Smith and Nienow (1983),Hemati (2003) showed that an increase in the amount of binder added increased the mean size of the granules. Tan (2006) studied the effect of the binder spray rate on the growth kinetics of the process and found that increase in the spray rate increase the granule growth rate. A higher spray rate of binder will cause a higher rate of collisions between particles and binder droplets and thus speed up the rate of agglomeration. Then it was suggested that the binder spray rate is the rate determining step in the whole process.

The binder viscosity is a key operating parameter which largely affects the overall process in a fluidised bed and hence has been studied extensively.Seo (2002), Walker (2006), Chen (2009)shown that as the average binder concentration, i.e., as the amount of hydroxyl propyl methylcellulose powder dissolved in the solution increasesthe average size of the granules also increases. Because greater the viscosity the more energy is required to break up the liquid droplets and hence larger droplets if the energy is unchanged. These larger droplets cause larger granules to be formed. However, this result is only up to a point as increasing the viscosity too high can then become a hindrance to granule growth rate.

Smith et al., (1983) proposed a mechanism of particle growth in which the strength of inter-particle bridges and the extent of fluid drag and inertial forces on particles are described.

Rawley (1989) studied bag shaking cycle in fluidized bed granulator to improve particle size distribution of granules formed if shake time and corresponding interval between bag shakes are optimized.

Granulation experiments were run by spraying a certain amount of a suspension into the fluidized bed of inert material discontinuously. For the suspension fine CaCO3 primary particles with 2 mm diameter were suspended in water and polyvinylpyrrolidone added as binder. This suspension was sprayed on glass spheres of an average diameter of 700 mm. At the end of one experiment, Becher and Schlunder (1998) concluded that the drying zone is affected by three process parameters, the bed height, the nozzle gas flow rate and the gas temperature in the bed and above the plenum of the bed. In practice, if we want surface layering as the desired particle growth mechanism, shallow fluidized beds with a nozzle spraying upwards into the fluidized bed are recommend. Then, an increase in the outlet gas temperature or in the nozzle gas flow rate reduces the formation of agglomerates.

Controlled spraying and atomization of the binder solution is necessary for an optimal process. It is important to determine dependence of spray properties on the atomization parameters at different scales used during development, as this information can be used as a basis for establishing process parameters for the manufacturing process. Gemci et al., (2002) presented the spray characteristics of hydroxypropyl Cellulose (HPC) solution sprayed through two different size nozzles used for the top spray granulation process in a fluidized bed column.

A series of experiments are carried out batch wise in a fluidized bed granulator with malic acid to understand the growth mechanism and the growth rate of particles with respect to the operating parameters such as the flow rate of the spray solution, temperature and flow rate of the fluidizing air, the concentration of the solution and the particle diameter. The increase in concentration of the spray solution or the increase in flow rate is found to increase the growth rate of particles. Srinivasakannan and Balasubramaniam (2003) concluded that an increase in the flow rate or an increase in the concentration of the spray solution increases the particle growth rate. Neither the increase in the flow rate nor the temperature of the fluidizing air is found to alter the growth rate; however, they facilitate to operate the bed at high liquid flow rates without wet quenching.

Hemati et al., (2003) studied the coating and granulation of solid particles by aqueous solutions of polymers or inorganic salts, aims to understand the effect of process-related variables such as the excess gas velocity, atomizer location, liquid flow rate and concentration, and atomizing air flow rate and also physicochemical-related variables such as the viscosity of solutions, wettability of the granulating liquid on solid particle surfaces, initial particle mean size, and porosity of the particles on the agglomeration kinetics of solid particles in a fluidized bed.

Goldschmidt et al., (2003)modeled top-spray fluidized bed granulation by DEM coupled with a detailed hydrodynamic model of the fluid phase. A two-dimensional situation was considered for this purpose. Binder droplets and solid particles were explicitly accounted for in the DEM simulation, and criteria for the occurrence of coalescence as function of the fractional liquid coverage of the colliding particles have been applied. The solidification of binder has also been modeled. The simulations gave the size and composition distributions of the resulting granules. The influence of several parameters on granulation kinetics has been investigated which includes binder spray rate, droplet size, fluidization velocity.

Walker et al., (2006) investigated co-melt granulation of lactose and PEG in a fluidized bed granulator. The process parameters such as binder amount and binder viscosity were correlated to granulation time and particle size distribution.

In many applications agglomerate stability is an important criterion for production. Since agglomerates do not conform to specific standards of size and shape, they may create problems in operations such as tableting and thus reduces the efficiency of the entire process and lowersthe product quality.Weber et al., (2006) studied liquid, amount of agglomerate liquid, particle shape, particle size, particle size distribution, and fluidizing gas velocity to determine the parameters affecting the stability of wet agglomerates in a fluidized bed by taking glass beads and water in the fluidizing column.

Grunewald et al., (2010)determine the influence of process parameters on internal nucleation rates by performingsteady-state experiments with a bottom-spray configuration considering binder concentration, granule hold up.

Patnaik and Sriharsha (2010) studied the mechanism of granule growth in a fluidized bed. They performed experiment in a main glass fluidization column having an ID of 10.16cm and heightof 24cm. This glass column is placed over a mild steel column of the same diameter and height 98cm.The binder solution was fed through a 2mm stainless steel tube and the compressed air from a 6.35mm tube at the tip of the atomizer. The binder solution was atomized due to the impingement of the air jet. They investigated the concept of granule formation by individual drops and to quantify the relationship between drop size and granule size by direct measurement.

Patel et al., (2010) investigated the influence of fluid bed process parameters such as binder addition rate, atomizing pressure, and binder concentration on the granule growth process and the final physical characteristics of the granules. They performed the experiment using PVP K-30, starch, HPMC 15cps and carbopol 934p as binders and diclofenac sodium has taken as the bed material. They found that the granulation process is almost controlled by the amount of binder added and atomization air pressure also affects the granule breakage and flow properties of granules.

Singh et al., (2011) performed experiment on the aqueous solution of 5% PVP K-30 as a binder and sprayed it on lactose monohydrate bed. A full factorial design was applied to optimized the granulation process variables like inlet air temperature, spray rate and batch size and other granule properties, namely the Carr's index, Hausner index, the angle of repose and the moisture content, were evaluated at the optimal operation conditions.

2.8 OPTIMIZATION

With the advent of computers optimization has become a part of computer aided design activities. Optimization is a mathematical discipline that concerns the finding of minima and maxima of functions, subject to so-called constraints. They are extensively used in engineering design problems where the emphasis is on maximizing or minimizing a certain goal.It is essential to formulate an optimisation problem before selecting any optimisation method for optimising the process/parameters/equipments.

2.8.1 Steps in formulisation of an optimization problem

- Need identification
- Choose design variables
- Formulation of constraints
- Formulate the objective function
- Restrict the search space
- Choose the optimisation algorithm
- Obtain solution(s)

Various classical optimization techniques are there based on what is to be optimised (Deb, 2003). Based on process parameter optimisation, optimization method is classified into two principal categories. Again these two types are classified in several sub-categories as described below.

2.8.2 Classification of Optimisation methods:

I. SINGLE-VARIABLE OPTIMIZATION

- Bracketing methods
  1. Exhaustive search method
  2. Bounding phase method
- Region-Elimination methods
  1. Interval halving method
  2. Fibonacci search method
  3. Golden section search method
- Gradient-based methods
  1. Newton-Raphson method
  2. Bisection method
  3. Secant method

II. MULTI-VARIABLE OPTIMIZATION

1. Direct search method
2. Gradient based method

Because of complex nature of fluidized bed granulation and complexity of multivariable effects, single variable optimisation technique has been used in the present work for parameter optimization. In this work Interval Halving method has been adopted for optimisation of process parameter.

2.8.3 SINGLE VARIABLE OPTIMISATION

2.8.3.1 Bracketing methods

The minimum of a function is found in two phases. First, a crude technique is used to find a lower and an upper bound of the minimum. Thereafter, a more sophisticated method is used to search within these limits and find the optimal solution with the desired accuracy.

1. Exhaustive search method

This method is the simplest of all other methods. In the exhaustive search method, the optimum of a function is bracketed by calculating the function values at a number of equally spaced points. Usually, the search begins from a lower bound on the variable and three consecutive function values are compared at a time based on the assumption of unimodality of the function. Based on the outcome of comparison, the search is either terminated or continued by replacing one of the three points by a new point. The search continues until the minimum is bracketed.

2. Bounding phase method

Bounding phase method is used to bracket the minimum of a function. This method guarantees to bracket the minimum of a unimodal function. The algorithm begins with an initial guess and thereby finds a search direction based on two more function evaluations in the vicinity of the initial guess. Thereafter, an exponential search strategy is adopted to reach the optimum.

2.8.3.2 Region-Elimination methods

Once the minimum point is bracketed, a more sophisticated algorithm needs to be used to improve the accuracy of the solution. In this section, three algorithms that primarily work with the principle of region elimination are described. Depending on the function values evaluated at two points and assuming that the function is unimodal in the chosen search space, it can be concluded that the desired minimum cannot lie in some portion of the search space. Internal Halving is a sub type of this method.

1. Interval halving method

In this method, function values at three different points are considered. Three points divide the search space into four regions. Fundamental region elimination rule is used to eliminate a portion of the search space based on function values at the three chosen points. Three points chosen in the interval (a,b) are all equidistant from each other and equidistant from the boundaries by the same amount. Two of the function values are compared at a time and some region is eliminated. Figure 2.1 shows these three points in the interval.

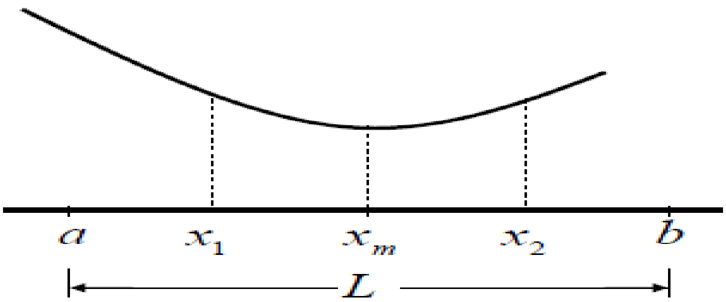


Fig. 2.2 Three points x_1, x_m and x_2 used in the interval halving method

2. Fibonacci search method

In this method, the search interval is reduced according to Fibonacci numbers. The property of the Fibonacci numbers is that, given two consecutive numbers F_{n-2} and F_{n-1}, the third number is calculated as follows:

$$F_n = F_{n-1} + F_{n-2} \qquad (2.1)$$

Where n = 2, 3, 4…..

One difficulty of the Fibonacci search method is that Fibonacci numbers have to be calculated and stored. Another problem is that at every iteration the proportion of the eliminated region is not the same.

3. Golden section search method

In order to overcome the two problems encountered in Fibonacci search method, the Golden section search method is used. In this algorithm, the search space (a,b) is first linearly mapped to a unit interval search space (0,1). Thereafter, two points at'‘t’ from either end of the search space are chosen so that at every iteration the eliminated region is (1-t) to that in the previous iteration (Fig. 2.2). This yields the golden number t=0.618.

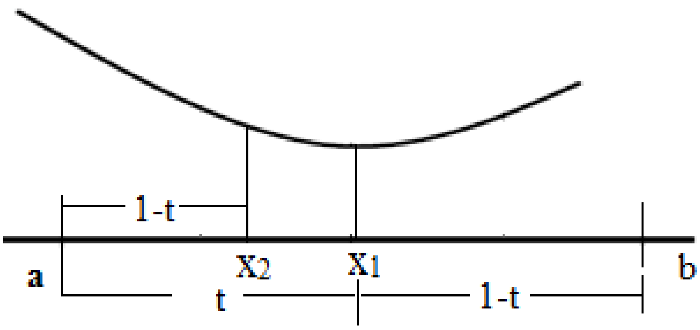


Fig. 2.3The points (x_1 andx_2) used in the golden section search method

2.8.3.3 Gradient-based methods

In many real-world problems, it is difficult to obtain the information about derivatives, either due to the nature of the problem or due to the computations involved in calculating the derivatives. Despite these difficulties, gradient-based are popular and effective. The optimality property at a local or a global optimum the gradient is zero cam be used to terminate the search process.

1. Newton-Raphson method

In the Newton-Raphson method, a linear approximation to the first derivative of the function is made at a point using the Taylor's series expansion. That expression is equated to zero to find the next guess. If the current point at iteration at t is $x^{(t)}$,the point in the next iteration is governed by the following simple equation obtained by considering up to the linear term in Taylor's series expansion.

$$x^{(t+1)} = x^{(t)} - \frac{f'(x^{(t)})}{f''(x^{(t)})} \tag{2.2}$$

2. Bisection method

The Newton-Raphson method involves computation of the second derivatives, a numerical computation of which requires three function evaluations. In the Bisection method, the computation of the second derivative is avoided; instead, only the first derivative is used. Both the function values and the sign of the first derivative at two points is used to eliminate a certain portion of the search space.

3. Secant method

In the secant method, both the magnitude and sign of derivatives are used to create a new point. The derivative of the function is assumed to vary linearly between the two chosen boundary points. Since boundary points have derivatives with opposite signs and the derivatives vary linearly between the boundary points, there exists a point between these two points with zero derivative. If at two points x_1 and x_2, the quantity $f'(x_1)f'(x_2) \leq 0$, the linear approximation of the derivative x_1 and $x_2$will have a zero derivative at the point z given by

$$z = x_2 - \frac{f'(x_2)}{\left(f'(x_2) - f'(x_1)\right) / \left(x_2 - x_1\right)} \tag{2.3}$$

2.8.4. MULTI-VARIABLE OPTIMIZATION

1. Direct search method

This method requires many function evaluations to converge to the minimum point. Computationally this method may be expensive.

2. Gradient based method

These methods are usually faster search methods. For problems where the objective function is discrete or discontinuous or the variables are discrete the gradient based methods cannot be applied. On the contrary, in problems where the derivative information is easily available, gradient based methods are very efficient.

2.9 INTERVAL HALVING METHOD

In the interval halving method the interval is always divided in half. The function value at the midpoint is evaluated if the function changes sign over a given interval. Where the sign change occur the location of the root is determined lying within the sub-interval. Then the sub-interval becomes the interval for the next iteration and the process is repeated until the root obeys the required precision.

Due to the following advantages of interval halving method this technique has been used in present study.

a) Since the method brackets the root, the interval halving method is always convergent.

b) The interval gets halved as iterations are conducted, so the error can be guaranteed in the solution of the equation.

Algorithm (Interval Halving Method)

Step 1: Choose a lower bound “a” and an upper bound “b”. Choose a small number ε. Let x_m= (a+b)/2, L=b-a. Compute f (x_m).

Step 2: Set x_1=a +L/4, x_2=b-L/4.Compute f(x_1) and f(x_2).

Step 3: If f(x_1) <f (x_m) set b=x_m; x_m= $x_1$go to step 5; Else go to step 4.

Step 4: If f(x_2) <f (x_m) set a = x_m; x_m= x_2; go to step 5, Else a=x_1; b=x_2; go to step 5.

Step 5: Calculate L= b-a, if L<ε Terminate; else go to step 2.

CHAPTER 3

EXPERIMENTATION

The experimental set-up for granulation experiment is shown in fig 3.1.

Fig.3.1. Photograph of experimental set-up

3.1 PARTS OF EXPERIMENTAL SET-UP

The experimental set-up is consisting of:-

i) The fluidizing column:

A transparent Perspex glass column of 5cm ID, 80cm Height and 2.5mm wall thickness is used.

ii) Temperature sensor:

A digital thermal sensor is attached to the main apparatus that shows the temperature of the inlet fluidizing air.

iii) Heater:

A heater is provided on the way of fluidize air, which heats the air to desired temperature of operation.

iv) Variac:

A variac is also provided that controls the heater.

v) Rotameter:

A Rotameter is used in the line for measuring the flow rate of the air i.e. used as fluidizing medium.

vi) Air Compressor:

It is a multistage air compressor of sufficient capacity 25 kgf/cm^{2}which supplies air to the apparatus, which is controlled with the help of a valve.

vii) Air Accumulator/Receiver:

It is a horizontal cylinder used for storing the compressed air from compressor. There isone G.I. pipe inlet to the accumulator and one by-pass line from one end of the cylinder. The exit line is also a G.I. pipe taken from the central part of the cylinder. The purpose of using the air accumulator in the line is to dampen the pressure fluctuations. The operating pressure in the cylinder is kept at 20 psig.

viii) Pressure Gauge:

A pressure gauge in the required range (1-50 psig) is fitted in the line for measuring theworking pressure. The pressure gauge is fitted with an air accumulator/receiver.

ix) Valves:

A globe valve of ½ inch ID is provided in the by-pass line for sudden release of the linepressure. A gate valve of 1/2 inch ID is provided in the line just before rotameter to control theflow rate of air to the fluidizing bed.

x) Air Calming Section:

This is an important component of the experimental set-up. It consists of a cylindrical portion (4.5 cm id. and 7.5 cm length) followed by a conical bottom. The cone angle is about 35^0- 40^0. The larger side is of 45 mm id. and the smaller of 12 mm id., the height of the cone being 6.5 cm. The cone is brazed with G.I. flange of 11.4 cm O.D. The central bore of the flange is also of 45 mm dia. The cone is made of ordinary G.I. sheet

xi) Hot Air Generator:

A hot air generator is provided which heats the air to be supplied to the fluidizing column through the rotameter.

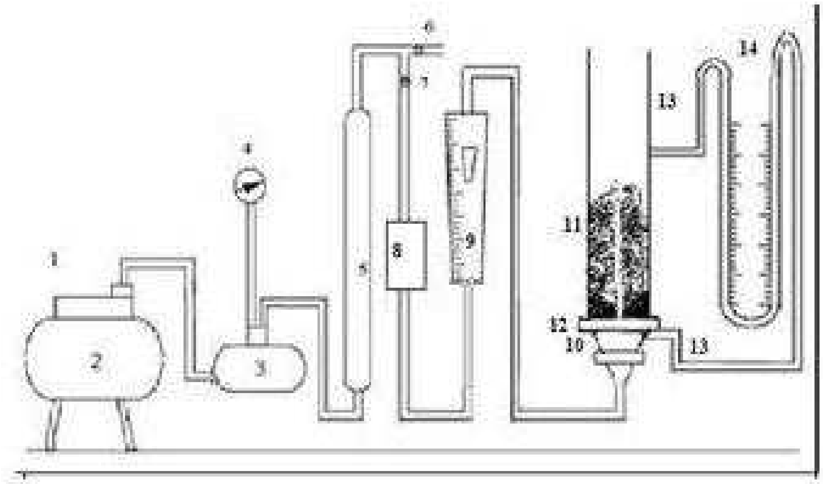
xii) Canvas Cloth:

A thin canvas cloth is used in place of air distributor to fluidize powder in the column for granulating experiment.

xiii) Manometer Panel Board:

A U tube manometer is used to measure the bed pressure drop.

The schematic diagram of the experimental set-up is given in fig.3.2.



1. Compressor; 2. Receiver; 3. Constant pressure tank; 4. Pressure gauge; 5. Silica gel tower; 6. By pass valve; 7. Line valve; 8. Hot air generator; 9. Rotameter; 10. Calming section; 11. Fluidizing column; 12. Distributer; 13. Pressure tapping 14. U tube manometer

Fig.3.2. Schematic diagram of the experimental set-up

3.2. MATERIALS AND METHODS

For the granulation experiment, the sample was taken in the column and was measuredand weighed. Then the heater was switch on and air was supplied into the column from bottom passing through the heater. The desired temperature of the fluidized air is maintained with thehelp of the sensor. The air is supplied by the compressor and the flow is controlled by the valveand rotameter. First of all the air is passed till the minimum fluidized condition of the bed isattained and then it is used to heat the particles. The liquid solution or binder is then passed from the top of the column onto the hot particles. The flow rate of the fluidizing air is then increased gradually. The adhesive powder under fluidize condition binds with each other in the presence of binder.For the experiment sucrose solution is taken as the binder and in the bed titanium dioxide powder of average size 25 micron and density 0.77gm/cc powder and Calcium carbonate powder of average size 15 micron and density 0.7gm/cc is taken. Various parameters like the volume of binder used, the amount of powder taken in the bed, temperature of inlet air and flow rate of the fluidizing air has been varied and accordingly the particle growth has been studied. The observations are noted down by varying one process parameter and at the same time other parameters are kept constant. After the completion of the process the final product is taken out and they are separated into desired size fraction by sieving the product material. Then the bigger sized granules are separated for the study.

The experiments were carried out by varying the different system parameters are discussed asscope of the experiment in Table – 3.1.

Table -3.1 (A): Scope of Experiment for TiO_2

Sl. No.	Material	W, gm	F, Nm^3/h	T, ^oC	V, ml
1	TiO_2	25	32	50	2
2	TiO_2	40	32	50	2
3	TiO_2	60	32	50	2
4	TiO_2	80	32	50	2
5	TiO_2	25	32	50	2
6	TiO_2	25	60	50	2
7	TiO_2	25	70	50	2
8	TiO_2	25	85	50	2
9	TiO_2	25	32	50	2
10	TiO_2	25	32	60	2
11	TiO_2	25	32	70	2
12	TiO_2	25	32	80	2
13	TiO_2	25	32	50	2
14	TiO_2	25	32	50	5
15	TiO_2	25	32	50	10
16	TiO_2	25	32	50	15

Table -3.1 (B): Scope of Experiment for $CaCO_3$

Sl. No.	Material	W, gm	F, Nm^3/h	T, ^oC	V, ml
1	$CaCO_3$	50	26	50	3
2	$CaCO_3$	50	26	50	5
3	$CaCO_3$	50	26	50	7
4	$CaCO_3$	50	26	50	10
5	$CaCO_3$	30	26	50	5
6	$CaCO_3$	40	26	50	5
7	$CaCO_3$	45	26	50	5
8	$CaCO_3$	50	26	50	5
9	$CaCO_3$	40	26	50	5
10	$CaCO_3$	40	24	50	5
11	$CaCO_3$	40	22	50	5
12	$CaCO_3$	40	20	50	5
13	$CaCO_3$	40	26	50	5
14	$CaCO_3$	40	26	55	5
15	$CaCO_3$	40	26	60	5
16	$CaCO_3$	40	26	65	5

CHAPTER 4

RESULT AND DISCUSSION

4.1 DEVLOPMENT OF CORRELATION FOR PARTICLE GROWTH

Graph between process parameters such as weight of the material, temperature and flow rate of the fluidizing air and volume of the binder is plotted against particle growth of TiO_2in fig.-4.1, 4.2, 4.3 and 4.4 using the data from Table- 3.1 (A).

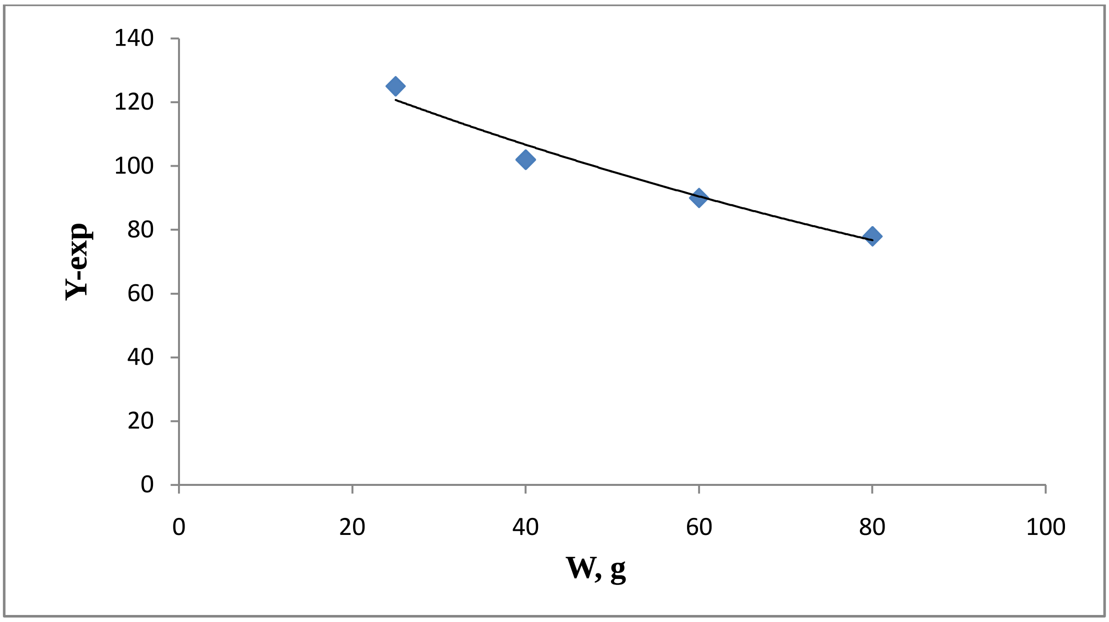


Fig. 4.1 Variation of weight of particle in the bed on the growth rate of TiO_2

It is observed that Y-exp i.e. experimental particle growth decreases as weight of particle (W) increases.When the particle load was increased, more particles had to be agglomerated with the same amount of binder. The ratio between the binder volume and the number of particles decreased leading to a lower growth rate and smaller agglomerates.

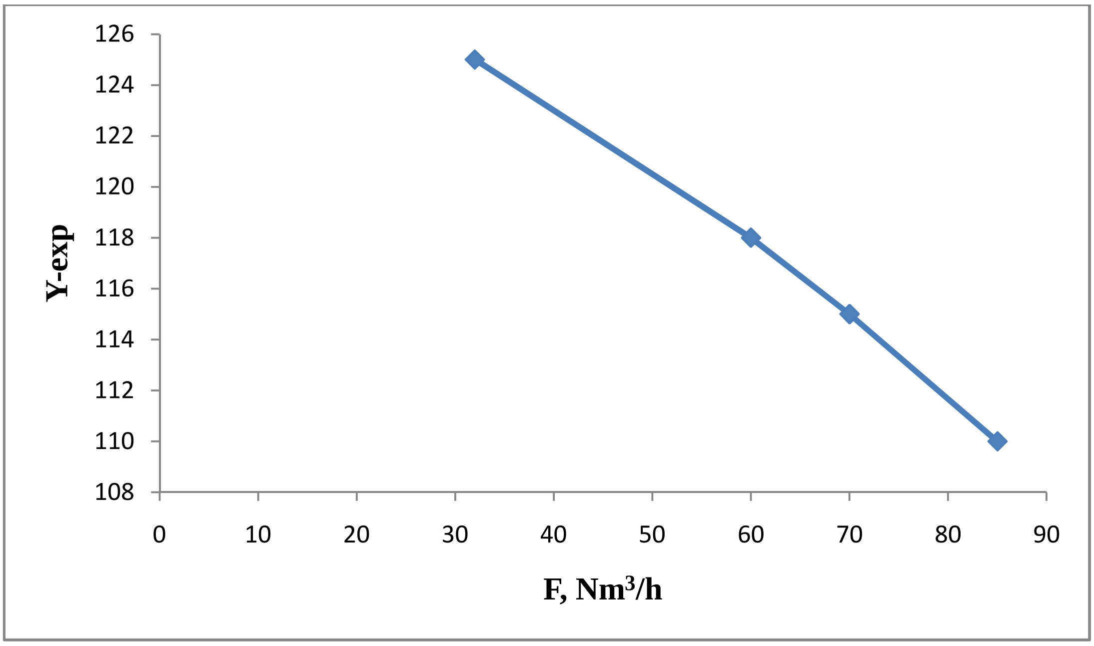


Fig. 4.2 Variation of Fluidizing air flow rate on the growth rate of TiO_2

It can be observed from fig. 4.2 that smaller size granules are formed on increasing the air flow rate. This happens because larger fluid velocity makes the granules to collide with each other and with the wall vigorously reducing large agglomerates to small size granules.

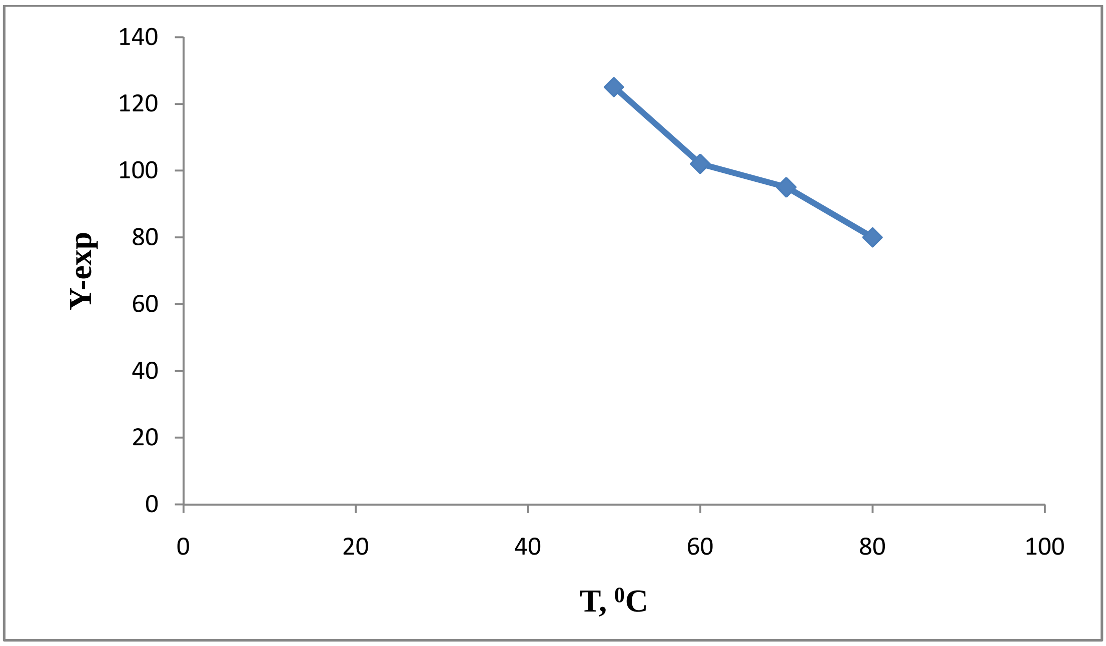


Fig. 4.3 Variation of Temperature of fluidizing air on the growth rate of Tio_2

Increase in inlet air temperature seems to decrease the granule size as can be seen from fig. 4.3. This can happen because at higher temperature the sprayed liquid may evaporated at a faster rate leading to small sized granules.

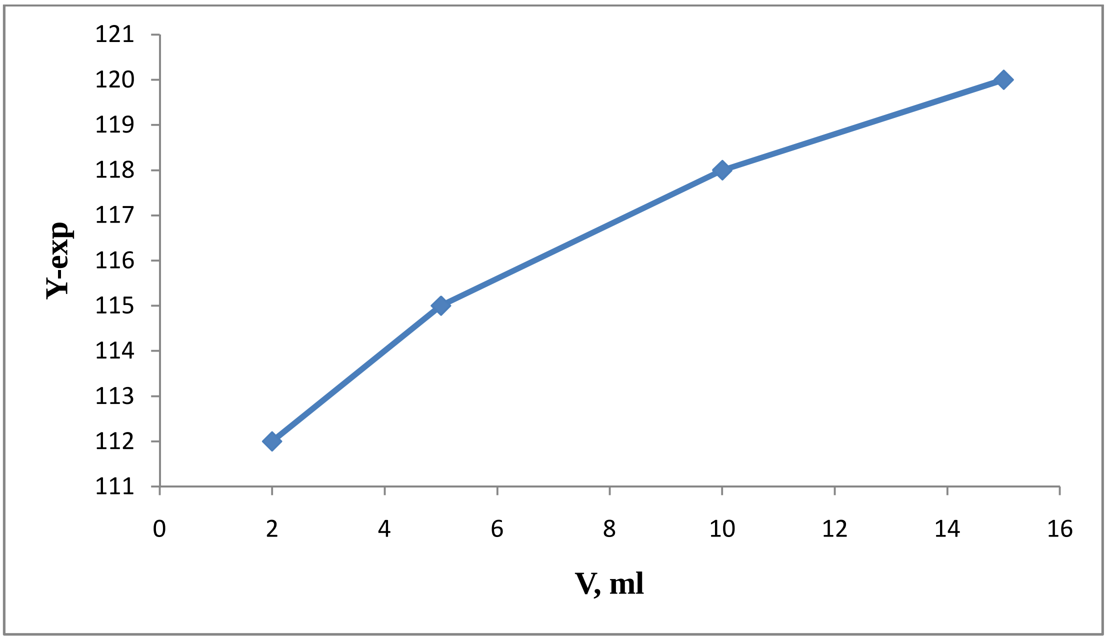


Fig. 4.4 variation of Volume of binder used on the growth rate of TiO_2

It was observed that more the amount of binder used, more is the fraction of particle growth. It issuch because on adding more binder, the powder agglomerates are formed thus the granule size is increasing. The granules obtained for 5 ml binder volume and 15 ml binder volume are shown in fig 4.5. It can be observed that at 15 ml volume of binder bigger size granules are formed as compared to volume of 5ml.

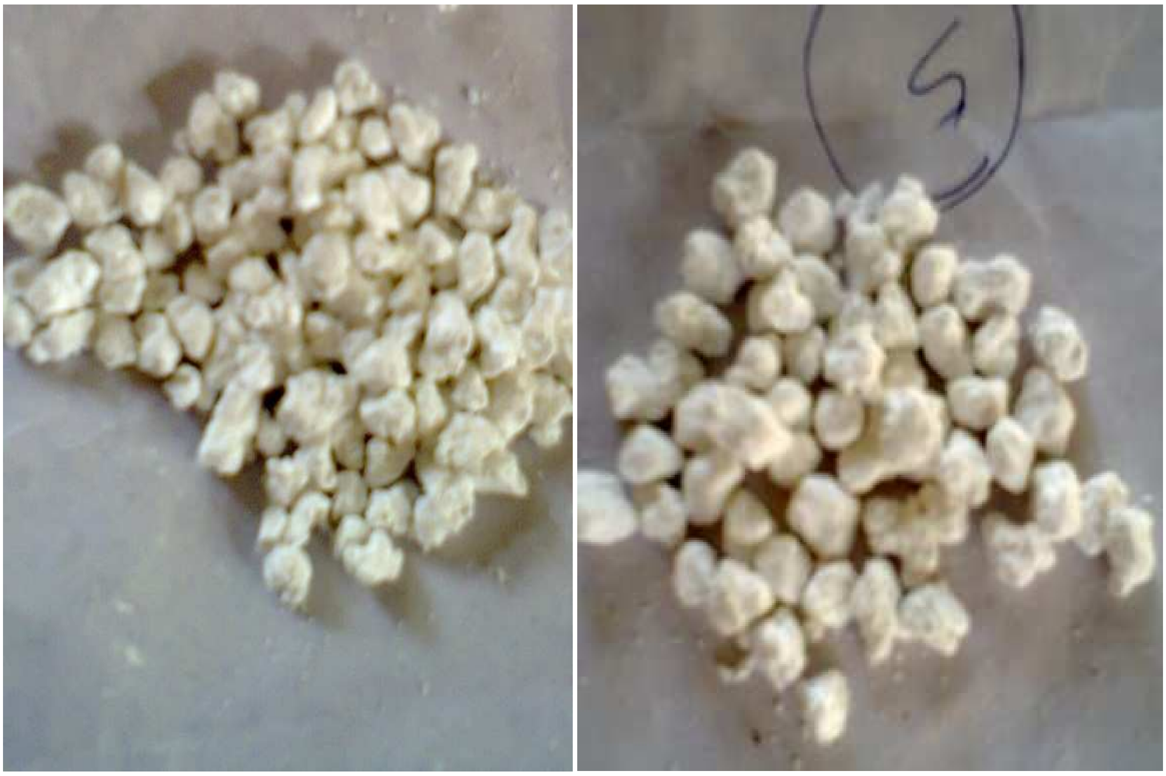


Fig. 4.5 Experimental result of granulation of Titanium dioxide at binder volume of 5 ml and 15 ml respectively

The correlation for growth of the particle has been developed on the basis of dimensional analysis by using data on various parameters and experimentally observed particle growth size. The dimensional analysis method adopted is exponential in nature unlike the conventional method. The correlation plot is shown in Fig- 4.6. The calculated values of particle growth obtained through dimensional analysis have also been compared against the experimental values and the percent deviation for the same is shown in Table-4.1 (A).

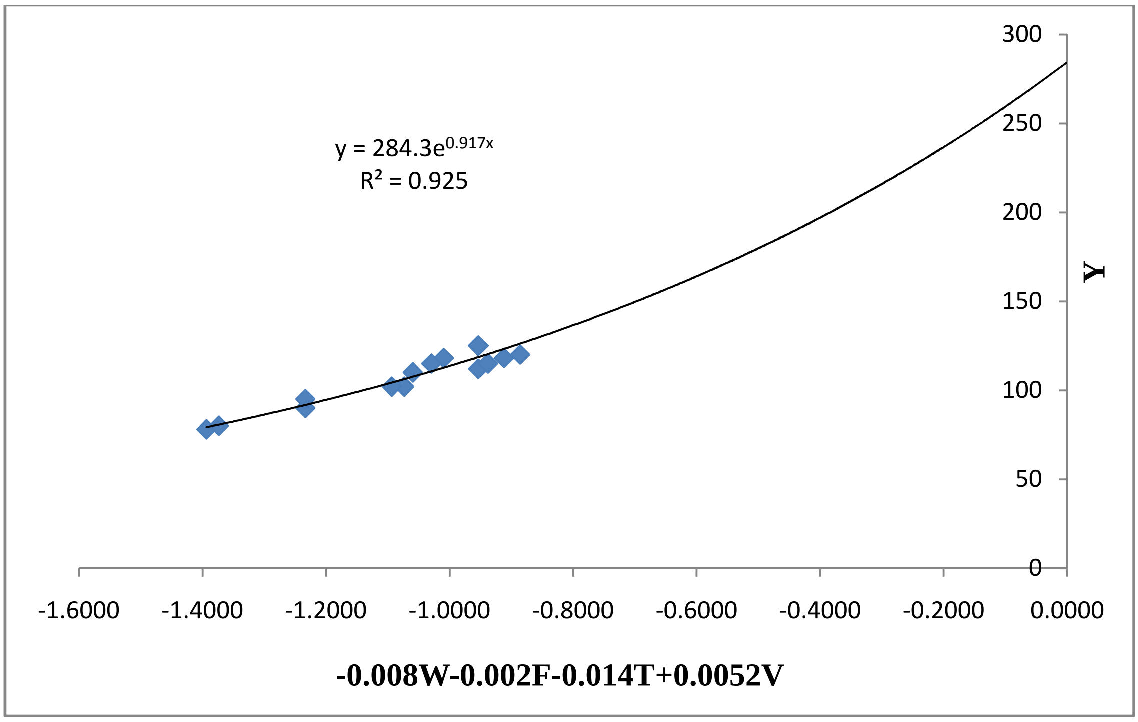


Fig. 4.6 Plot of particle growth rate against the system parameters

The correlation developed based on Dimensional analysis is as follows:

$$Y = 284.3 \times e^{0.917 \times [-0.008W - 0.002F - 0.014T + 0.0052V]} \tag{4.1}$$

Table-4.1 (A): Calculated and experimental values for TiO_2

Sl. No.	Material	W, gm	F,Nm^3/h	T, ^oC	V, ml	Y-exp	Y-cal	%dev
1	TiO_2	25	32	50	2	125	118.583	5.13
2	TiO_2	40	32	50	2	102	106.227	-4.14
3	TiO_2	60	32	50	2	90	91.7304	-1.92
4	TiO_2	80	32	50	2	78	79.2125	-1.55
5	TiO_2	25	32	50	2	125	118.583	5.13
6	TiO_2	25	60	50	2	118	112.647	4.54
7	TiO_2	25	70	50	2	115	110.6	3.83
8	TiO_2	25	85	50	2	110	107.599	2.18
9	TiO_2	25	32	50	2	125	118.583	5.13
10	TiO_2	25	32	60	2	102	104.296	-2.25
11	TiO_2	25	32	70	2	95	91.7304	3.44
12	TiO_2	25	32	80	2	80	80.6787	-0.85
13	TiO_2	25	32	50	2	112	118.583	-5.88
14	TiO_2	25	32	50	5	115	120.292	-4.60
15	TiO_2	25	32	50	10	118	123.194	-4.40
16	TiO_2	25	32	50	15	120	109.65	-5.14
17	TiO_2	40	22	50	5	124	110.66	11.57
18	TiO_2	40	22	50	7	129	112.19	14.08
19	TiO_2	40	22	50	10	132	114.80	15.00
20	TiO_2	40	22	50	15	138	112.19	16.57
21	TiO_2	40	22	50	10	132	112.61	15.00
22	TiO_2	40	20	50	10	136	113.02	17.20
23	TiO_2	40	18	50	10	131	113.44	13.86
24	TiO_2	40	16	50	10	124	113.02	8.52
25	TiO_2	40	18	50	10	131	108.95	13.86
26	TiO_2	45	18	50	10	127	105.03	14.35
27	TiO_2	50	18	50	10	125	101.24	15.84
28	TiO_2	55	18	50	10	122	113.02	17.01
29	TiO_2	40	18	50	10	131	105.99	13.86
30	TiO_2	40	18	55	10	128	99.40	17.19
31	TiO_2	40	18	60	10	120	93.22	17.16
32	TiO_2	40	18	65	10	113	93.22	17.36

To find the validation of the correlation obtained in equation 4.1,the calculated particle growth Y-cal for other experimental data (i.e. from Sl. No. 17 – 32 in Table-4.1 (A)) is obtained using the above correlation (4.1).

Table-4.1 (B): Calculated and experimental values for $CaCO_3$

Sl.No.	Material	W, gm	F,Nm^3/h	T, ^oC	V,ml	Y-exp	Y-cal	%dev
1	$CaCO_3$	50	26	50	3	187	100.227	46.31
2	$CaCO_3$	50	26	50	5	189	101.15	46.58
3	$CaCO_3$	50	26	50	7	192	102.082	46.83
4	$CaCO_3$	50	26	50	10	200	103.496	48.25
5	$CaCO_3$	30	26	50	5	213	117.135	45.09
6	$CaCO_3$	40	26	50	5	200	108.849	45.58
7	$CaCO_3$	45	26	50	5	191	104.929	45.16
8	$CaCO_3$	50	26	50	5	189	101.15	46.58
9	$CaCO_3$	40	26	50	5	200	108.849	45.58
10	$CaCO_3$	40	24	50	5	213	109.249	48.79
11	$CaCO_3$	40	22	50	5	223	109.651	50.76
12	$CaCO_3$	40	20	50	5	227	110.054	51.45
13	$CaCO_3$	40	26	50	5	200	108.849	45.58
14	$CaCO_3$	40	26	55	5	197	102.082	48.27
15	$CaCO_3$	40	26	60	5	193	95.7352	50.48
16	$CaCO_3$	40	26	65	5	180	89.783	50.12

The standard deviation and mean deviation for CaCO3 and TiO_2 sample are listed in Table-4.2.

Table-4.2: Mean deviation and standard deviation using Dimensional analysis

Sample	Standard deviation	Mean deviation
Tio_2	2.4	4.17
$Caco_3$	2.17	1.88

4.2 OPTIMIZATION OF THE DEVELOPED CORRELATION FOR PARTICLE GROWTH

The correlation obtained in equation (4.1) has to be optimized to get maximum particle growth. Therefore varying one variable at a time and keeping other variables constantoptimisation of process variables was carried out one by one.

First of all volume of the binder parameter was selected for optimization. Substituting the values of other variables, the equation-(4.1) can be written as follows.

$$Y = 284.3 \times e^{0.917 \times [(25)\times(-0.008)+(32)\times(-.002)+(50)\times(-0.014)+(V)\times(0.0052)]} \tag{4.2}$$

$$\Rightarrow Y = 284.3 \times e^{0.917 \times (-0.964 + 0.0052 \times V)}$$

$$\Rightarrow Y = 284.3 \times e^{-0.884 + (4.77 \times 10^{-3})V} \tag{4.3}$$

Similarly following equations are obtained for inlet air temperature (T), fluidizing air flow rate (F) and amount of bed material (W),

$$Y = 284.3 \times e^{-0.23 + (-0.012)T} \tag{4.4}$$

$$Y = 284.3 \times e^{-0.82 + (-1.834 \times 10^{-3})F} \tag{4.5}$$

$$Y = 284.3 \times e^{-0.75 + (-7.34 \times 10^{-3})W} \tag{4.6}$$

Above process has been followed to convert the multivariable equation into a single variable equation to apply the single variable optimization technique. MATLAB code has been developed for this purpose using Interval Halving method of optimization which is given as follows. The defining or objective function only changes for different equations such as eq. (4.4), (4.5) and (4.6).

MATLAB PROGRAMMING

Defining function (1st m-file)

function y = inte(x) ;

$$y = -284.3 \times e^{-0.884 + (4.77 \times 10^{-3}) \times x}$$

Main programme (2nd m-file)

a = input ('enter the initial bound');

b= input ('enter the final bound');

c = input (‘enter the value of the smallest interval');

l= b - a ;

```
while ((b-a) > c )

xm = ( a+ b) /2 ;

x1 = a + ( l / 4) ;

x2 = b - ( l/4) ;

y1 = inte (x1 );

y2 = inte (x2);

ym = inte (x m);

if ( y1 > y m&& y2 > y m)

        a = x1 ;

        b = x2 ;

else if ( y1> y m&& y2< y m )

        a = x m ;

else if ( y1< y m&& y m< y2 )

        b = x m;

end

    l = b - a;

end

x = (a+ b) /2;

y = inte(x);
```

```
disp('the maximum value of the function is ');

disp(y);

disp('the maxima is ');

disp(x);
```

CHAPTER 5

CONCLUSION

Granulation in fluidized bed is a complex process since a number of variables affect the process thereby controlling the particle growth. Therefore selection of process parameters becomes very important.

From above discussion following conclusions can be drawn.

1. The process parameters have been studied to observe the influence of these parameters on granule growth.

2. With low binder content, fine granules are produced, which turns to bigger agglomerates on increasing binder amount. But the inlet air temperature, bed material and air flow rate have counter effect on the final granule size.

3. For TiO_2 material the experimental observed values of particle growth were compared with dimensional analysis. The percentage of deviation observed is approximately between -6 to +6 for dimensional analysis approach.

4. Again the developed correlation using dimensional analysis is used for the other extra experimental data which seems to be in good agreement as the percentage standard deviation ranges between 8 to 17 for Tio_2 material but it is approximately between 45 to 50 percent for $CaCO_3$material which is not in good agreement with each other.

5. As the percent deviation is not too much for TiO_2 material, so the correlation developed can be applied for a wide range of parameters.

NOMENCLATURE

W	Weight of particle in the bed (g)
F	Fluidizing air flow rate (Nm^3/h)
T	Temperature of fluidizing air (^oC)
V	Volume of binder used (ml)
Y-exp	Experimental Fractional growth
Y-cal	Calculated Fractional growth
%dev	Percentage deviation
Y	Fractional growth

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