



**Faculty of Manufacturing Engineering**

**OPTIMIZATION OF AMMONIA RELEASE MODEL IN A  
FLUIDIZED BED GRANULATION SYSTEM USING RSM AND PSO**

**Norhidayah binti Mohamad**

**Doctor of Philosophy**

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**OPTIMIZATION OF AMMONIA RELEASE MODEL IN A FLUIDIZED BED  
GRANULATION SYSTEM USING RSM AND PSO**

**NORHIDAYAH BINTI MOHAMAD**

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in fulfillment of the requirements for the degree of  
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**2020**

## DECLARATION

I declare that this thesis entitled “Optimization of Ammonia Release Model in a Fluidized Bed Granulation System Using RSM and PSO” is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

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Date : .....

## APPROVAL

I hereby declare that I have read this thesis and in my opinion this thesis is adequate in terms of scope and quality for the award of Doctor of Philosophy.

Signature : .....

Supervisor Name : Associate Professor Dr. Mohd Rizal Salleh

Date : .....

## **DEDICATION**

This thesis is dedicated to my dearly missed late parents Haji Mohamad Bin Lamin and Hajah Temah Bte Seman. Thank you both for believed in me, gave faith and strength to pursuing my higher Degree. Ya Allah, I beg for Your mercy on their soul and grant them a place in Jannah. Ameen.

My beloved husband, Khavier Ismail, my precious kids, Khawlah Azwar and Khaer Hamzah as well as my family members for their everlasting love and prayers throughout my research journey.

## ABSTRACT

Granulation is an important class of production processes in food, chemical and pharmaceutical industries. The optimization of granulation in combining the particles together through the creation of bonding between the powder particles are critically important. In the urea fertilizer industries, the granulation processes are critically severe due to its ammonia ( $\text{NH}_3$ ) emission from these processes.  $\text{NH}_3$  gas can be highly poisonous and varying impact to a variety of live being. It is estimated that global  $\text{NH}_3$  emissions from urea fertilizer processes are approximately at 10 to 12 Tg N/year which representing 23% of overall  $\text{NH}_3$  released globally. Therefore, this research conducts the study on the experimental works, optimization modelling process and validate the output in order to minimize the  $\text{NH}_3$  gas emission in the fluidized bed granulation process for the urea fertilizer manufacturing. Correspondingly, the manipulated variables in this research are pressure (Mpa), binder feed rate (rpm) and inlet temperature ( $^{\circ}\text{C}$ ). Whereas, the type of powder to be used is urea with the usage quantity of powder for every run of experiment is 150 g. While, the other parameters such as binder volume (ml), mixture concentration (%) and running times (min) will be kept constant throughout the experimental process. Using Response Surface Method (RSM), the amount of  $\text{NH}_3$  release detected during granulation process is used as the objective function. A mathematical model for the  $\text{NH}_3$  release as a function of the fluidized bed granulation system had been empirically proposed. A source code was developed with the MATLAB for computation of algorithm. Particle Swarm Optimization (PSO) soft computing method was used to optimize the process parameters and the coefficient of equation parameters. The results showed that the optimum processing parameter to minimize  $\text{NH}_3$  gas release happened at the temperature range of  $50^{\circ}\text{C}$  to  $80^{\circ}\text{C}$  with the value of pressure at 0.4 Mpa and the binder feed rate was 4 rpm. The experiment has been designed using Design of Experiment (DOE) particularly RSM via Central Composite Design (CCD) to compare with the simulation results. The measured experimental responses average values obtained are 0.01%. The average percentage of errors between the PSO models and experimental validated models are approximately 0.79% and it was found that the error value falls within the acceptable range. This research will be useful for optimizing the urea granulation production process and reducing the  $\text{NH}_3$  gas release to the environment.

## **PENGOPTIMUMAN MODEL PELEPASAN AMMONIA DALAM SISTEM GRANULASI LAPISAN TERBENDALIR MENGGUNAKAN RSM DAN PSO**

### **ABSTRAK**

*Granulasi adalah merupakan kelas utama proses pengeluaran dalam industri makanan, kimia dan farmaseutikal. Pengoptimuman proses granulasi dalam menggabungkan partikel-partikel amat penting melalui penghasilan ikatan antara partikel serbuk tersebut. Di dalam industri baja urea, proses granulasi merupakan satu proses yang amat kritikal disebabkan berlakunya pelepasan gas ammonia ( $\text{NH}_3$ ) ketika ianya berlangsung. Gas  $\text{NH}_3$  adalah merupakan gas yang sangat beracun dan boleh memberi pelbagai impak yang amat besar kepada pelbagai hidupan. Dianggarkan pelepasan gas  $\text{NH}_3$  dunia dari industri baja urea adalah sekitar 10 hingga 12 Tg N/tahun mewakili kira-kira 23% daripada keseluruhan gas  $\text{NH}_3$  dunia. Kerja penyelidikan ini melibatkan kerja-kerja eksperimen dan permodelan proses pengoptimuman untuk mengawal pelepasan gas  $\text{NH}_3$  dalam proses granulasi lapisan terbendalir bagi penghasilan baja urea. Seterusnya, pemboleh ubah yang dimanipulasi dalam penyelidikan adalah tekanan (Mpa), kadar suapan pengikat (rpm) dan suhu masukan ( $^{\circ}\text{C}$ ). Manakala jenis serbuk yang digunakan adalah urea dengan jumlah kuantiti 150 g untuk setiap eksperimen yang dijalankan. Selain daripada itu, lain-lain parameter antaranya isipadu pengikat (ml), kepekatan campuran (%) dan jangka masa (min) adalah dalam keadaan tetap. Dengan menggunakan Response Surface Method (RSM), jumlah pelepasan  $\text{NH}_3$  yang dikesan semasa proses granulasi digunakan sebagai fungsi objektif. Model matematik bagi pelepasan  $\text{NH}_3$  sebagai fungsi sistem granulasi lapisan terbendalir dicadangkan secara empirikal. Satu kod sumber telah dibangunkan menggunakan MATLAB bagi pengiraan algoritma. Kaedah pengkomputeran lembut Particle Swarm Optimization (PSO) digunakan untuk mengoptimumkan parameter proses dan pekali parameter persamaan. Keputusan menunjukkan bahawa parameter yang optimum bagi kadar pelepasan gas  $\text{NH}_3$  yang minimum adalah pada suhu antara  $50^{\circ}\text{C}$  hingga  $80^{\circ}\text{C}$  dengan nilai tekanan 0.4 Mpa dan kadar suapan pengikat pada 4 rpm. Eksperimen ini telah direka dengan menggunakan Design of Experiment (DOE) khususnya RSM melalui Composite Central Design (CCD). Kiraan nilai eksperimen secara purata respon yang didapati ialah 0.01%. Peratusan ralat purata antara model Particle Swarm Optimization dan model yang disahkan ujikaji dianggarkan 0.79% dan dalam julat yang ditentukan. Didapati bahawa perbezaan nilai antara simulasi dan eksperimen berada dalam julat yang dapat diterima. Kajian ini amat berguna untuk mengoptimumkan proses pengeluaran granulasi urea dan mengurangkan pelepasan  $\text{NH}_3$  ke alam sekitar.*

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## LIST OF SYMBOLS AND ABBREVIATIONS

wt%	- Weight percentage
°C	- Degree celcius
µm	- Micrometer
g	- gram
%	- percentage
ANOVA	- Analysis of Variance
CCD	- Central Composite Design
DOE	- Design of Experiment
FBG	- Fluidized Bed Granulation
NH <sub>3</sub>	- Ammonia
N	- Nitrogen
ppm	- Part per molecule
PSO	- Particle Swarm Optimization
RSM	- Response Surface Method
rpm	- Round per minute
UG	- Urea Granule

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## LIST OF PUBLICATIONS

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1. N.Mohamad, D. Shafeeq, A.Shaaban, M.A.Huda, 2017. Modeling and Simulation Affect of Binder Addition Rate On Ammonia Emission During Granulation Process, *Journal of Advanced Manufacturing Technology*, 11 (1), pp. 69-76.
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# CHAPTER 1

## INTRODUCTION

### 1.1 Granulation system

Granulation is an important class of the production processes in the food, chemical and pharmaceutical industries. It is used to produce the granules from the liquid products such as solutions or suspensions. Granulation is the combination and formation process of the particles together through the creation of bonding between those powder particles. The bonding of powder particles can be happened through the application of binding agent.

The granulation process is used to combine one or more types of powder particles and transforms it into a granule that will allow the tableting process and produce the quality of tablets at the required tablet pressing speed range (Fries et al., 2014 and Veliz et al., 2015).

The granulation mechanisms will include the process of nucleation and formation, agglomeration, consolidation, coalescent and breakage. The initial step of this process is wetting of the feed powders. The formation of granules will be happened with the assistance of binding fluid which is strongly influenced by the spraying and/or fluid distribution as well as the feed formulation properties, in comparison with the mechanical mixing (Da Silva et al., 2014).

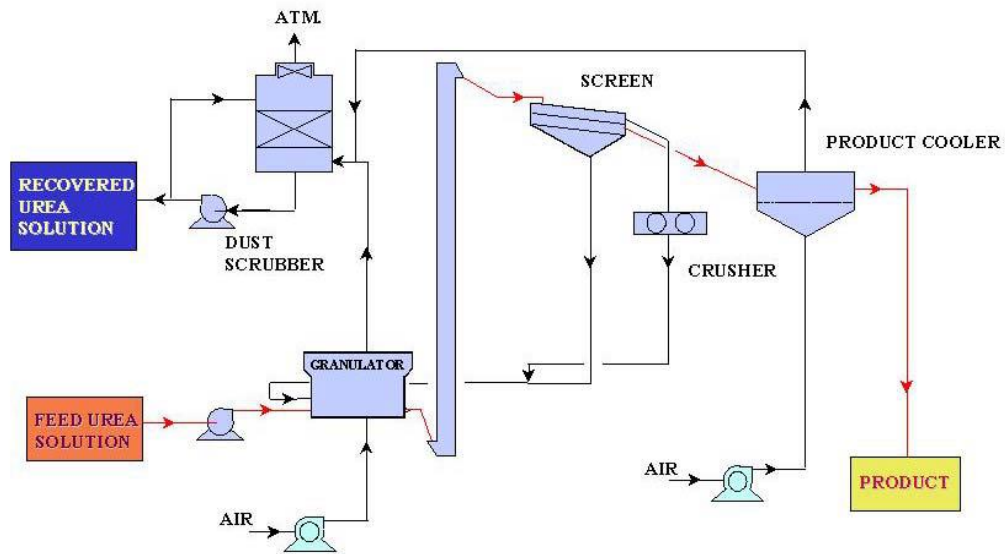


Figure 1.1: Heavy industry urea granulation process flow diagram (Fageria et al., 2011)

The most challenging issues in controlling the urea granulation process is maintaining the entire fluidized bed system while producing within the range of required quality of urea granules. Figure 1.1 shows the diagram of urea granules process flow of the heavy urea fertilizer industries. It is essential to control the equipment operating procedure in order to obtain the optimum operation with the desired quality of the end product. Here, the main parameters of the fluidized bed granulation system should be thoroughly monitored for the better operating performance. There are several identified parameters that have the significant impacts on the process performance especially the process variables which will affect the quality of the final product of fluidized bed granulation process. For this research, there are few selected parameters had been identified that will give the significant impacts on the ammonia gas released during the granulation process and those parameters are binder feed rate, temperature, and atomization pressure.

Furthermore, the challenges in controlling the process of granulation including the importance of regulating the granule density, the granule size formation, sensor for the critical product attributes such as the bulk density and particle size distribution that are available with the large measurement suspensions (Patel et al., 2010). Additionally, the shortage of manipulated variables which were the binder spray and the rate of mixing or tumbling also have been highlighted.

Da Silva et al. (2014) did the compilation of techniques used on observing and controlling the fluidization procedures, particle sizes and moisture contents during the coating and granulation processes in the fluidized bed system. The development technique of operating and scheming systems for coating and granulation of particles is highly required, not only to allow the operation in a stable bubbling fluidization regime, which intensifies the heat and mass transfer, but also to ensure strict quality specifications for products, such as, uniform particle sizes distribution, low moisture contents and good flow ability. The author, Da Silva et al. (2014) also focused on the discussion of techniques used and results obtained in studies on operating and procedure of granulation and coating process in the fluidized bed system that was reported for the last few decades.

Bucalá et al. (2017) were comprehensively discussed on the fluidized bed granulation systems. These researchers were focused on developing the techniques and methodologies for monitoring and controlling the coating and granulation processes in fluidized bed systems. They also covered some of the methodologies applied to monitor the fluidization regime and identified the fluid dynamic instabilities in the particle granulation processes and highlighting the time series analysis of pressure fluctuation. Some difficulties were determined and the future prospects on the development of the strategies to monitor and control granulation processes also had been identified (Bucalá et al., 2017).

In a fluidized bed system, the granules can be obtained from the combination of solid particles or powders. Then, a liquid is passing or spraying through the solid material. The important properties of the fluidized bed system are the fluid like behaviour, an enlarged active surface caused by the increased of bed porosity and good particle mixing. A laboratory scale of pilot plant of fluidized bed granulation system as shown in Figure 1.2 had been installed for this study at the workshop of Fakulti Kejuruteraan Pembuatan (FKP), Universiti Teknikal Melaka Malaysia.

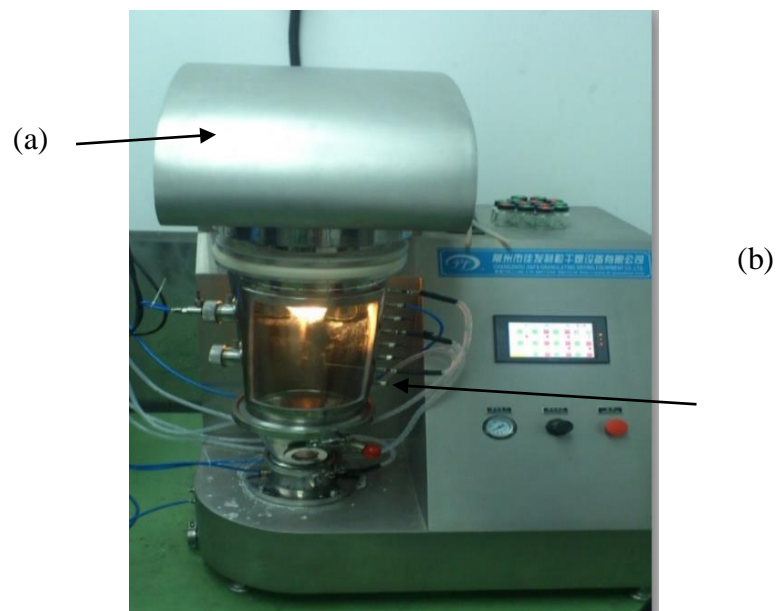


Figure 1.2: Fluidized bed system (a) fluidized bed spray granulator and (b) process chamber

The urea powders are fluidized by the air stream at the predetermined pressure and temperature. Then, a binder liquid will be injected or sprayed and it will be dropped onto the particles. Due to the low humidity and the increased of temperature, the liquid fraction, such as the solvent or the external phase is evaporated. The remaining solid forms of a new layer on the particle surface as shown in Figure 1.3.

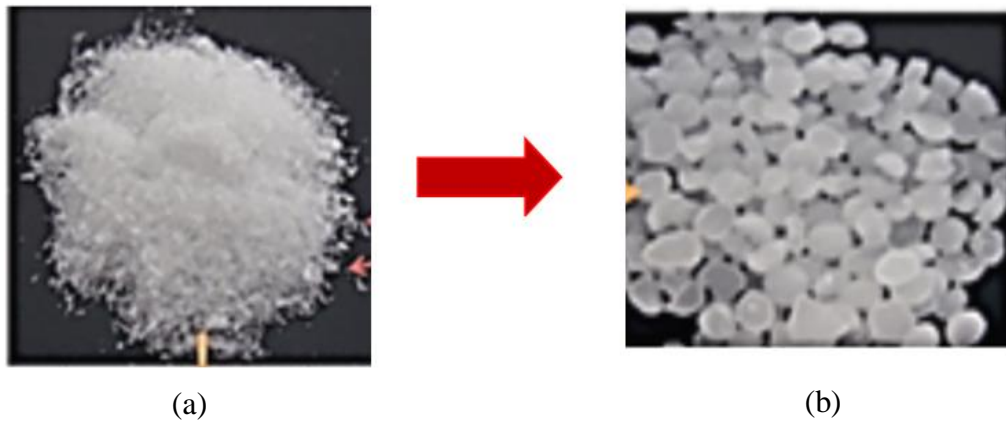


Figure 1.3: Transformation of (a) fine urea powders to (b) uniform urea granule seed

Urea granule is used as fertilizers to supply additional nitrogen for the paddy plant growth and also sustaining the soil fertility. The utilization of urea fertilizers on the paddy fields has been well accepted for producing the high rice grain yields and improved the nutrient availability. The urea fertilizer is also cost effective and it is having high nitrogen contents comparing to the other resources.

The urea fertilizer has been widely commercialized and it can be purchased as a prills or granulated material. In the previous years, the urea fertilizer was usually produced by dropping the liquid urea from a prilling tower. The height of the tower was corresponding to the principle of the product drying process. The different between urea granule and urea prills are, it was formed a smaller and softer substance than other materials commonly used in fertilizer blends. After going through the innovations processes, the urea is then manufactured as granules. Urea granules are larger, harder, and more resistant to moisture. Today, due to the demand from agriculture industry, the granulated urea has become more suitable material for fertilizer blends. The fluidized bed granulation process is selected as the most high-end method to produce the urea granules.

Now, the fluidized bed granulation is more dominant for urea solidification rather than prilling method. In this process, granules are usually formed by the successively spraying and drying of concentrated urea solution onto recycled granules (Jannat et al., 2016).

## **1.2 Problem statement**

The elevated number of ammonia emissions in this world are mainly came from the agriculture activities such as manure storage, slurry spreading and the use of inorganic nitrogen fertilizers. The ammonia gas can contribute to eutrophication, an oversupply of nitrogen and acidification to the ecosystems. It also forms the particulate matter in the atmosphere which has adverse effects on the human health. The plant operators are facing with the demands from the environmental authorities to reduce the emissions of ammonia to the environment. Figure 1.4 shows the graph of gases pollution that were retrieved from the industrial activities. It shows that the agricultural activity had contributed to the highest ammonia released compared to the other activities. It also includes the activities such as the process of producing the urea fertilizer and farming. According to Danielou (2012), annually 1.2% of total world energy had been used for fertilizer industry and approximately 90% of it (199 million tone) was used for ammonia production. The ammonia gas that was released from the mineral fertilizer will depend on many factors, including, type of fertilizer applied, soil properties and method of the production.

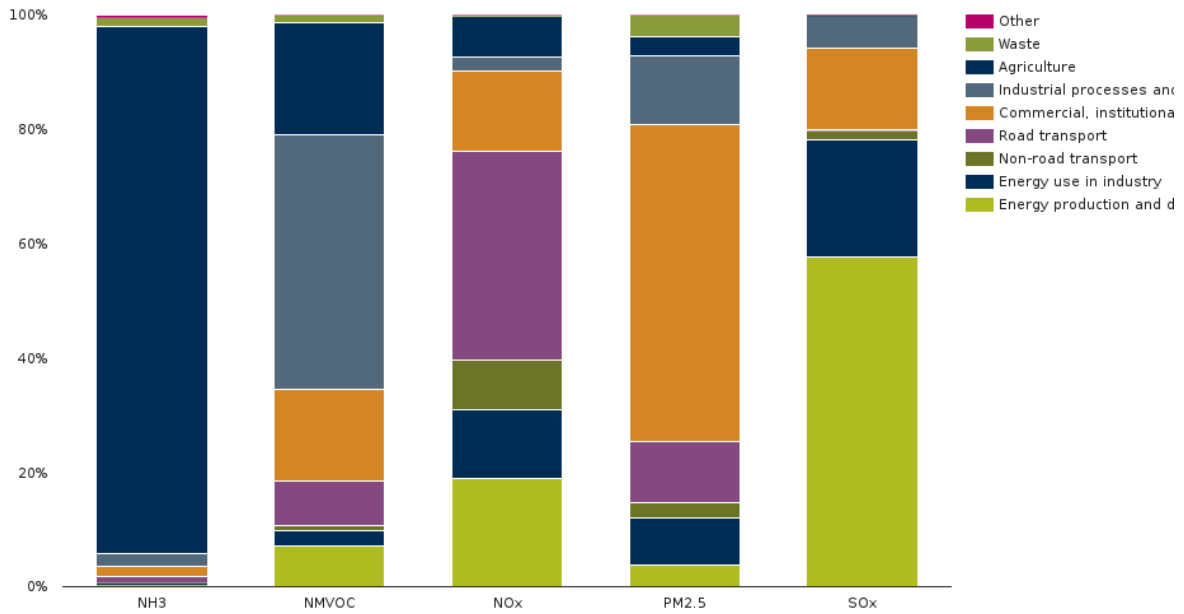


Figure 1.4: Type of gases pollution's collection data source from industry activities (Beusen et al., 2008)

Global ammonia emissions from nitrogen fertilizer are estimated approximately at 10 to 12 Tg N/yr which is tera gram of nitrogen released per year (Beusen et al., 2008). These ammonia gas released raised the concern from economic, environmental and national policy perspectives. The European Union (EU) members have committed to encounter the ammonia released issues under the National Emissions Ceiling Directive (European Commission, 2015). Figure 1.5 represents the percentage of ammonia released according to the management categories.