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Optimization of Biodegradable Urea Production Process to Minimize Ammonia Release through Response Surface Method Experimental Design

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Abstract – *This paper investigated the influence of temperature and binder speed rate as a process variable towards amount of ammonia emission (NH_3) in the fluidized bed top spray granulation. Response Surface Method (RSM) was employed in this experiment to optimize the process parameters in a top-spray fluid-bed processor. Three significant variables (binder feed rate, atomize pressure and temperature) were selected for the optimization studies. The statistical model was constructed via central composite design (CCD) using three screened variables. These methods are exclusively used to examine the "surface," or the relationship between the response and the factors affecting the response. The response in this experiment is the quantity of gas release, then the goal would be to find the factor settings that minimize the amount.The inlet temperature was identified as the most critical parameter, followed by the binder addition rate and atomize pressure, respectively.* ***Copyright © 2016 Penerbit Akademia Baru - All rights reserved.***

Keywords: Fluidized Bed, Binder Feed Rate, Gases, Granulation

1.0 INTRODUCTION

In recent years, many environmental regulations have been amended by the regulatory bodies. This has led to a significant reduction in the permissible emission levels for ammonia (NH_3) and urea dust in new urea fluid-bed granulation plants. On the other hands, existing plant operators are also face with the demands from the environmental authorities to reduce the emissions of ammonia to the environment. From the Environmental Hazards Associated with Emissions, the statutory emission limit values (ELVs) into air normally refer to NH_3 emissions and urea dust from specific emission plant point sources. Frequently, ELVs are negotiated between the site operator and the local licensing authority. The ELVs for existing plants may reflect staged values over a defined period to enable the operator to achieve compliance. In Europe, ELVs for urea dust range from 75 to 150mg.Nm-3 and for NH_3, from 100 to 200mg.Nm-3.With the development of new technology, the emission limits recommended by the regulatory authorities will continually reduce.

According to Pothoff [1], NH_3 gas substance which released to the atmosphere is basically not just emitted in the urea granulation plant itself. It is produced with the urea powder and the urea binder solution coming from the evaporation section of the urea synthesis process. Based on their findings, the equilibrium achieved in the evaporation section, is unavoidable. NH_3 will be released as gas during spraying of the urea solution into the fluidized bed. Hence, reducing ammonia emission by optimizing the urea granulation process parameter, the energy consumption will be reduce and lead to greener technology. According to Irfan [2], the vaporization becomes negligible when the melt concentration is reduced to 95%. The spreading of the binder liquid droplets in the powder bed controls most of the agglomeration phenomenon. Here binder speed rate and temperature in the fluidized bed are important parameters that affect the whole process. The urea granule can be dry or wet when it reappears at the spray surface. Dried agglomerates will absorb more binder liquid at their particles surfaces in wet condition compared to wet particles which absorbed less and left more binder liquid available. [3]. By utilizing high binder liquid concentration, the emission of NH_3 could become uncontrollable [4]. This research investigates the correlation of binder speed rate and the temperatures with quantity of NH_3 released while sustain the quality of granules as fine output.

2.0 MATERIALS AND METHODOLOGY

The urea powder whose size is from 400 to 500 μm size was used for granulation in the preliminary experiment. The binder solution is made by adding cassava starch ($(C_6H_{10}O_5)_n$) and urea into deionised water under constant stirring at 325 rpm. For each batch of granulation experiment, 200 g of urea powder and 100 g of the binder solution are used. The lab scale fluidized bed was referred in Fig. 1.

Figure 1: Lab Scale Fluidized Bed Granulation System

A schematic diagram for urea granulation is shown as Figure 2. The fluidized bed granulator consists of a chamber, nozzle for air flow pressure and feeder which is made from silicon tube. The binder solution was transferred to the chamber and granulated using different parameters.

The nozzle was located in the upper port above the bed, facing downward. Spray nozzle distributes air flow at constant pressure. The urea powder and binder solution were mixed continuously inside the chamber at predefined rate.

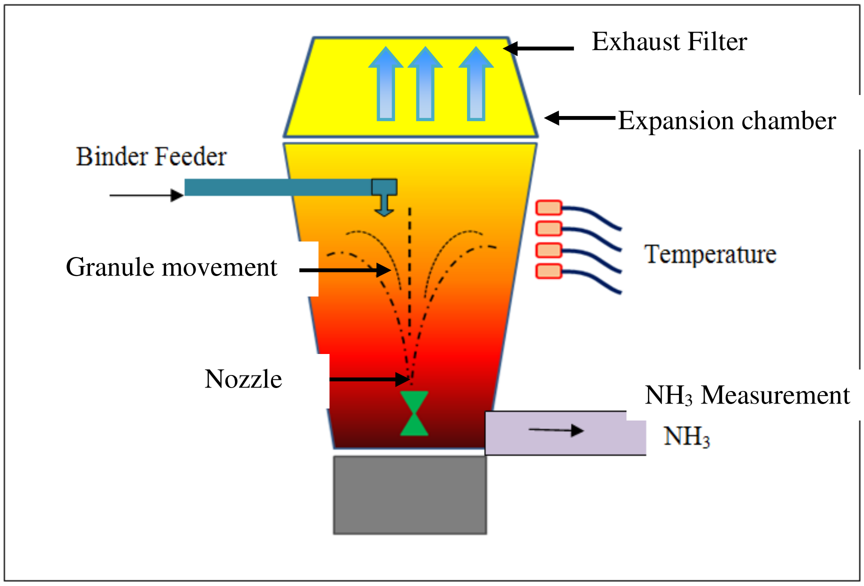

Figure 2: Schematics of the fluidized bed granulation

The binder solution is channeled to the powder bed from top spray through a silicon tube which fed by a peristaltic pump and the binder feed rate is controlled by the pump revolution setting. According to Tejas [6], the total quantity of solution added varied with the binder concentration. Rambali [7] set the feed rate according to the specific run and was continued until all the binder solution was used.

2.1 Gas Sampling Method

The ToxiPro equipment was used to measure quantity of NH_3 concentration in ppm released during granulation. Normal air movements carried the NH_3 to the sensor.

Figure 3: Setting-Up The Toxipro (NH_3 Gas Detector)

The NH_3 gas was recorded at every interval of time for every different set binder feed rate. During remote sampling, the gas was drawn into the sensor compartment through the probe assembly and a length of tubing. The diffused gas entered the sensor port on the front of the instrument. The sensor countered to changes in the concentration of the gas being measured. Figure 3 showed the manual set of Toxipro. The gas detector will turn on by pressing the button for 5 seconds until the red indicator appeared.

2.2 Model Set-up: Response Surface Methodology (RSM)

Response Surface Methodology (RSM) is useful for the modeling and analysis of programs in which a response of interest is influenced by several variables and the objective is to optimize this parameters used. The next step in the formulation of the medium was to determine the optimum levels of significant variables for minimizing NH_3 gas release. For this purpose, the response surface methodology (RSM), a central composite design (CCD) was adopted for this experiment. The significant variables utilized were as follows: Atomize Pressure (Bar), Binder Feed Rate (rpm) and Temperature (^{0}C) each of which was assessed at three coded levels (-1.682, 0, and +1.682). Table 1 represents the design matrix of the variables together with the experimental results of NH_3 emission.

Table 1: Experimental variables at different levels by using Central Composite Design (CCD)

Std	Run	Atomize Pressure (Bar)	Binder Feed Rate (rpm)	Temperature (^{0}C)	Response (ppm)
6	1	0.6	4	40	0.0
2	2	0.2	4	40	0.0
38	3	0.4	3	67	1.8
5	4	0.6	4	40	0.0
34	5	0.0	5	67	1.7
20	6	0.2	4	94	0.8
19	7	0.2	4	94	0.8
16	8	0.6	6	40	0.0
46	9	0.4	5	67	2.3
39	10	0.4	7	67	2.7
27	11	0.2	6	94	2.9
4	12	0.2	4	40	0.0
3	13	0.2	4	40	0.0
26	14	0.2	6	94	2.9
21	15	0.6	4	94	1.5
14	16	0.6	6	40	0.0
36	17	0.8	5	67	2.4
40	18	0.4	7	67	3.0
9	19	0.2	6	40	0.0
45	20	0.4	5	67	2.5
28	21	0.2	6	94	2.9
11	22	0.2	6	40	0.0
15	23	0.6	6	40	0.0
18	24	0.2	4	94	1.1
42	25	0.4	5	13	0.0
17	26	0.2	4	94	1.5
50	27	0.4	5	67	2.5

31	28	0.6	6	94	1.6
23	29	0.6	4	94	1.3
7	30	0.6	4	40	0.0
43	31	0.4	5	121	3.2
29	32	0.6	6	94	2.6
32	33	0.6	6	94	2.7
24	34	0.6	4	94	1.5
47	35	0.4	5	67	2.2
10	36	0.2	6	40	0.0
49	37	0.4	5	67	2.2
13	38	0.6	6	40	0.0
8	39	0.6	4	40	0.0
1	40	0.2	4	40	0.0
48	41	0.4	5	67	2.2
41	42	0.4	5	13	0.0
44	43	0.4	5	121	3.2
12	44	0.2	6	40	0.0
30	45	0.6	6	94	1.5
35	46	0.8	5	67	2.7
37	47	0.4	3	67	1.9
22	48	0.6	4	94	1.5
25	49	0.2	6	94	1.3
33	50	0.0	5	67	0.0
43	51	0.2	58	6	94.0
2	52	0.6	58	4	40.0
41	53	0.2	62	4	94.0
6	54	0.6	58	6	40.0
55	55	0.4	60	5	67.0
44	56	0.6	58	6	94.0
30	57	0.4	60	5	67.0
3	58	0.2	62	4	40.0
11	59	0.2	62	4	94.0
14	60	0.6	58	6	94.0

A total of 60 experiments were conducted. All variables were taken at a central coded value, which was considered as zero. The minimum and maximum ranges of the variables were used, and the full experimental plan with regard to their values in actual and coded form is provided in.

3.0 RESULTS AND DISCUSSION

Central composite design (CCD), a popular second order experimental design was used to design the experiment. The CCD is an effective design that is used for sequential experimentation and provides reasonable amount of information for testing the goodness of fit and does not require unusually large number of design points thereby reducing the overall cost associated with the experiment. CCD has following three set of experimental runs: (1) Fractional factorial runs in which factors are studied at +1, −1 levels. (2) Center points with all factors at their center points that help in understanding the curvature and replication helps to estimate pure error. (3) Axial points which is similar to center point, but one factor takes values above and below the median of two factorial levels typically both outside their range. Axial points make the design rotatable.

3.1 Optimization of granulation process by CCD design

In order to approach the optimum response region of the NH_3 emission during granulation process significant independent variables; binder feed rate and the bed temperature were further explored, each at three levels. Three-dimensional response surface plots were constructed by plotting the response on the Z-axis against any two independent variables, while maintaining other variables at their optimal levels. Figures 4 and 5 showed that slow rate of binder feed and low bed temperature supported reduction of NH_3 emission during granulation process.

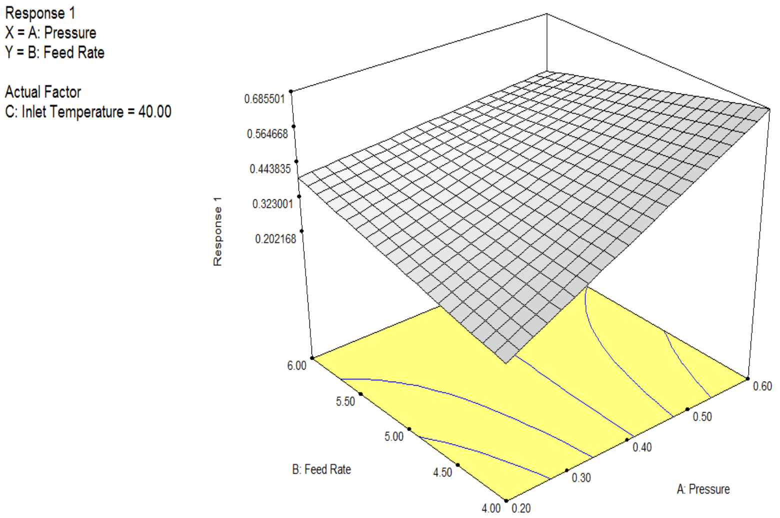

Figure 4: Three-dimensional response surface plot for NH_3 gas emission showing the interactive effects of the binder and atomize pressure.

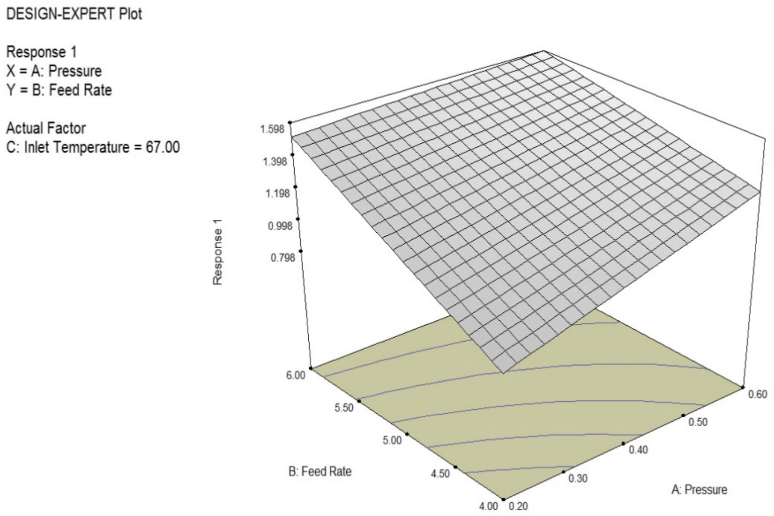

Figure 5: Three-dimensional response surface plot for NH_3 gas emission as response 1 showing the interactive effects of the binder and atomize pressure at constant inlet temperature 67^0C.

The rising of temperature increase the amount of NH_3 emission. The gas starts to release when the temperature reached to 40^0 C. This temperature need to be controlled not exceed more than 85^0 C and the process ended until all the urea powders completely solidified. The results show that NH_3 emission increases with the higher rate of binder feed and the rising of bed temperature. NH_3 emission can be decreased by injecting the binder at certain rate into the granulation chamber.

3.1. Validation of the model

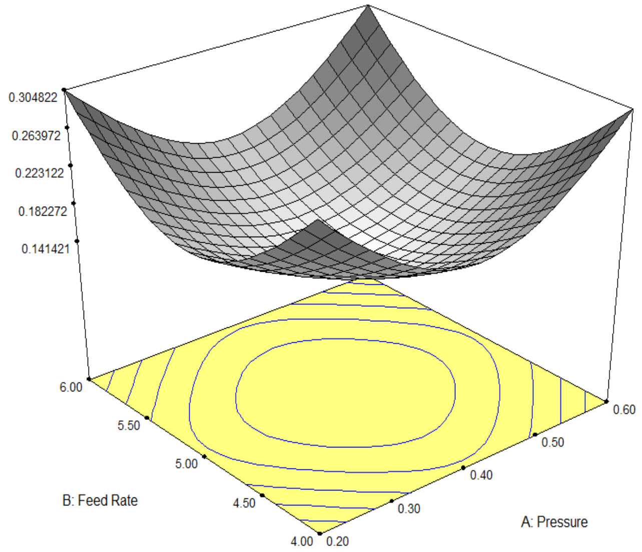

Figure 6: Three-dimensional response surface plot showing the interactive effects of atomize pressure and starch binder feed rate. Hold value: inlet temperature 67^0C.

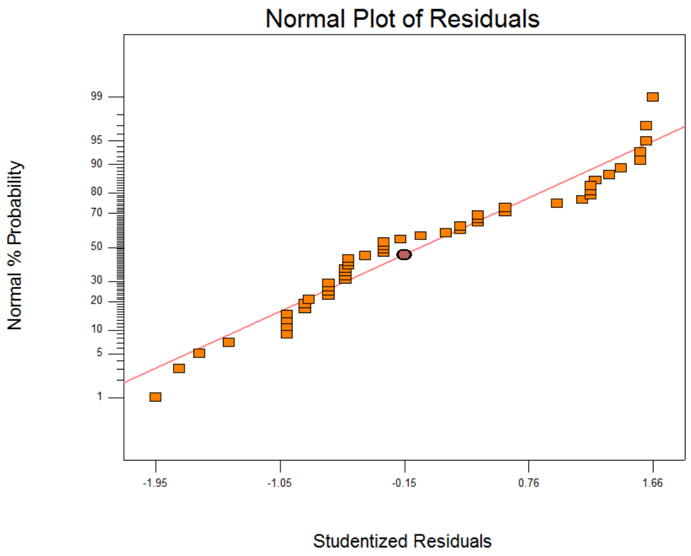

Figure 7: Time courses of NH_3 release production by urea granulation under optimized conditions.

These results confirmed the validity of the model, and the experimental values were determined to be quite close to the predicted values. Time courses of NH_3 release production by urea granulation under optimized condition is shown in Fig. 7.

The RSM applied to the optimization of urea granulation process in this investigation suggested the importance of a variety of factors at different levels. The CCD design plan exploited in the present study enabled us to study and explore the culture conditions that would support a reduction of NH_3 gas release in urea granulation.

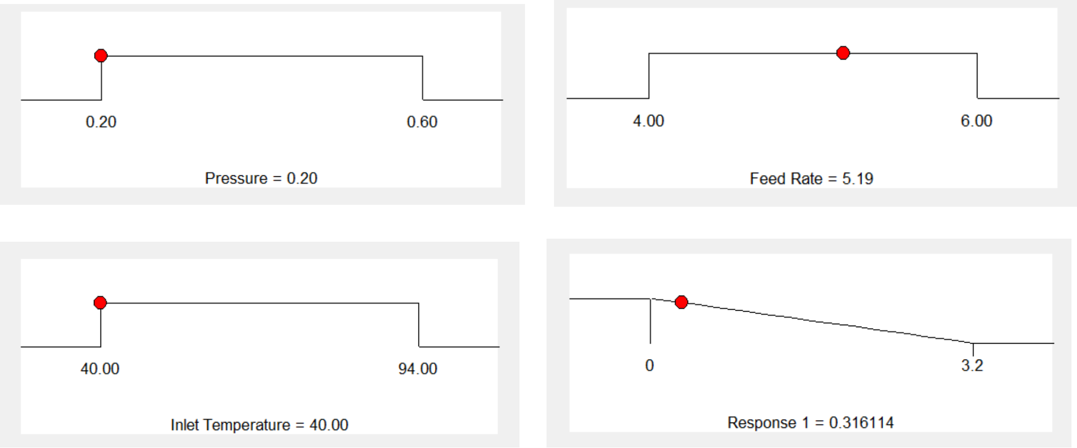

Figure 8: Optimal parameter condition fluidized bed granulation

The NH_3 gas released increased in a linear fashion with binder feed rate, and the response curves shown in Figs. 4 and 5 did not evidence a comparable curvature. Thus, the surface plots suggested that maximum production depends on higher temperature of granulation and the amount of binder inject. The contour plots are not perfectly elliptical and are, rather, horizontal. This indicates that there may be less interaction occurring among the independent variables corresponding to the response surfaces. Refer figure 8, the optimal levels of pressure, feed rate and inlet temperature could be predicted as approximately 0.20 bar, 5.19 rpm and 40.00 ^{0}C , respectively.

3.2 Analysis of variance for (ANOVA) Response Surface 2FI Model

Based on the DOE design, fifty experimental sets were constructed into a CCD experiment. The center point in the experimental design, is the combination of all factors at the center-level, was replicated three times to estimate the experimental accuracy. The corresponding analysis of variance is tabulated in Table 2.

The Model F-value of 12.28 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.0500 indicate model terms are significant. In this case B, C are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve this model. The "Lack of Fit F-value" of 16.47 implies the Lack of Fit is significant. There is only a 0.01% chance that a "Lack of Fit F-value" this large could occur due to noise. Significant lack of fit is bad. The small probability value indicates that the experimental data were fitted well by the regression model. Table 3 showed the determination coefficient of the regression model, *R*-Squared, is 0.6316.

Table 2: Analysis of variance table [Partial sum of squares]

	Sum of		**Mean**	**F**	
Source	**Squares**	**DF**	**Square**	**Value**	**Prob > F**
Model	42.06	6	7.01	12.28	<0.0001 significant
A	0.96	1	0.96	1.69	0.2007
B	3.20	1	3.20	5.61	0.0224
C	35.36	1	35.36	61.98	<0.0001
AB	0.32	1	0.32	0.56	0.4580
AC	0.000	1	0.000	0.000	1.000
BC	2.21	1	2.21	3.86	0.0558
Residual	24.53	43	0.57		
Lack of Fit	19.39	8	2.42	16.47	<0.0001 significant
Pure Error	5.15	35	0.15		
Cor Total	66.59	49			

Table 3: Coefficient of the regression model

Std. Dev.	0.76	R-Squared	0.6316
Mean	1.30	Adj R-Squared	0.5801
C.V.	58.19	Pred R-Squared	0.5269
PRESS	31.50	Adeq Precision	12.148

The regression equation obtained from the ANOVA showed that the "Pred R-Squared" of 0.5269 is in reasonable agreement with the "Adj R-Squared" of 0.5801 (a value >0.76 indicates fitness of the model). The "Adequate Precision" value is an index to measure the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 12.148 indicates an adequate signal. This indicates that the values of higher than 4 are essential prerequisites for a model to be a good fit. This also suggests that the model can be used to navigate the design space. The ‘predicted R-Squared’ is 0.5269, which indicates that the model is-good (for a good statistical model, the R-Squared value should be in the range of 0–1.0, and the nearer to 1.0 the value is, the more fit the model is deemed to be.

4.0 CONCLUSSION

As the temperature increases the rate of NH_3 gas also increases. This approach also increases the rate of drying granules but cannot be used because urea solution is harmed by high temperature. If temperature is high, it leads to higher NH_3 emission. Then if temperature is too low, the quality of granules will be affected.

Binder feed rate has significant positive effects on the NH_3 emission during granulation. The experiment showed that, the range of optimum temperature is 40^0C and binder feed rate is 5 rpm.

In summary, the two most critical process parameters in top spray fluid-bed granulation are processing temperature and the binder addition rate. Atomization air pressure volume has lesser effects on NH_3 emission; however, good control of all these parameters is important to minimize the NH_3 release and maintain the size of quality granules.

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