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Optimization of Coating Uniformity Using Modified Bio-Polymer in a Tangential Fluidized Bed Coater

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Abstract: This study presents the coating uniformity using modified bio-polymer in a tangential fluidized bed coater via Taguchi's method. The study focused on the implementation of coating uniformity using modified bio-polymer toward granular urea. In this study, efficiency of mass coating per granular urea was used as indicator for coating uniformity. A series of coating experiments were carried out to determine the effect of inlet air temperature, disc rotation speed and spraying rate to the efficiency of mass coating per granular urea. An orthogonal array, the signal-to-noise ratio and analysis of variance (ANOVA) were used. Besides achieving optimal parameters for the efficiency of mass coating per granular urea, the findings explain parameters that affect the results. The confirmation run had carried out to verify the conclusion from the variance analysis.

Key words: Bio-polymer, Taguchi's method, signal to noise ratio

INTRODUCTION

Urea is the most widespread nitrogen fertilizer that generally used in agriculture. The demand for its usage was expected to grow because of its high content of nitrogen (45%). On the other hand, urea used as fertilizer throughout the world causing severe pollution due to huge lost to the environment. Nitrogen is lost mainly through ammonia evaporation and denitrification. Once ammonia emitted to the environment, especially atmosphere, it plays a central role on environmental issue such as eutrophication, acidification and greenhouse effect (Erisman *et al*., 2007). The need to create controlled-release urea places a new demand on the typical urea fertilizer. The controlled release technology such as coating is one way to increase the effectiveness of urea fertilizer. Many works had done to enhance the coating quality by using sulphur as coating agent (Choi and Meisen, 1997; Singh *et al*., 2013).

Recently, there was an attention in the usage of biodegradable material as coating agent (Suherman and Anggoro, 2011). Starch is well-known as biodegradable polymer that is cheap. Therefore, starch-based material was used in this study. In order to achieve a good coating performance, it is important to select parameters based on experience or by the use of previous studies. This study focusing on three parameters, e.g., inlet air temperature, rotation disc speed and spraying rate. The inlet air temperature was monitored using a thermocouple at the bottom of the tangential fluidized bed and the desired bed temperature was achieved through this. The influence of inlet air temperature was significant to the particle growth (Da Rosa and dos Santos Rocha, 2010), efficiency of coating (Paulo Filho *et al*., 2006), premature droplet evaporation and final moisture content (Palamanit *et al*., 2013), enhance polymer film generation and coating quality and spreading of droplets results in the uniform coating (Chen *et al*., 2009). Rotational speed affects minimum fluidization velocity (Nakamura and Watano, 2007), total pressure drop (Huang *et al*., 2010; Shi *et al*., 2000), moisture content, drying rate (Lim *et al*., 2009) and shearing movement among particles that eliminate break agglomerate (Huang *et al*., 2010). Spraying rate affects particle agglomeration and layering (Ronsse *et al*., 2012; Srivastava and Mishra, 2010), moisture content (Fries *et al*., 2011; Palamanit *et al*., 2013), economics of coating process (Fries *et al*., 2011) and film characteristics (Lan *et al*., 2011).

Since the mid-1960s, the Taguchi's method had effectively applied to companies that allowed them to successfully become world economic competitors (Ross, 1995). A key subject in the Taguchi method is the determination of the combination factors and levels which will provides the experiment with the desired information. Thus, orthogonal arrays are introduced; which is process improvement decisions by using the minimum amount of test data. Orthogonally means that factors can be evaluated independently of each other; the effect of one

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factor does not interfere with the estimation of the influence of another factor (Ross, 1995). In this study, the Taguchi method for design of experiment was used for the optimization of the coating uniformity using modified bio-polymer in a fluidized bed. The previous studies that had been mentioned had demonstrated that inlet air temperature, disc rotation speed and spraying rate were the key process parameters for the coating uniformity characteristics. Due to that, these three parameters, e.g., inlet air temperature, disc rotation speed and spraying rate had been selected, with three different levels (1-3). Then, the results from analysis of variance were computerized to derive the optimal level combinations. Finally, the confirmation experiment was conducted to verify the analytical results.

MATERIALS AND METHODS

Material: Granular ureas were obtained from PETRONAS Chemicals Fertilizer Kedah, Malaysia. The ranges of diameter for granular ureas were 2.5-4 mm with an average tablet weight of 0.048 ± 2 g and an average tablet volume of 143.8 mm^3. It composes more than 97% of urea. The 200 g (4 000 granular) granular ureas was used as the starting material for this study.

Coating solution: The composition for coating solution in 300 mL of water was as stated in Table 1.

Coating solutions were made by compressing a mixture of starch (Tapioca), urea (Assay min 99.5%, Sigma Aldrich), borate (Di-Sodium Tetraborate Decahydrate, Merck) and lignin (Lignin-Alkali, Sigma Aldrich). The composition for the blending ratio and preparation of coating solution was based on Ariyanti *et al*. (2013). Distilled water was mixed with starch at the temperature of 80°C for 30 min for gelatinization process to occur. Next, urea, borate and lignin were added to the gelatinized starch for another 180 min at 80°C. The 60 min prior the mixing time ended, 1.5 g of Bromothymol Blue (BB) was added as standard substance to calculate mass coating solution when using UV visible spectrophotometer.

Experimental setup: The experimental setup used in the present study to carry out the coating experiments was a Fluid Bed Granulator Coater Dryer assembly Model FLP 1.5 developed by Changzhou Jiafa Granulating Drying Equipment Co., LTD. The tangential fluidized bed coater is made with viewing window and light window to view the process. Tangential fluidized bed was shown in Fig. 1.

Table 1: Coating solution composition

Materials	Ratio (%)	Mass (g)
Starch	57.05	15.00
Urea	22.82	6.00
Borate	5.13	1.35
Lignin	15.00	3.95

In these experiments, the coating duration was 50 min and then the coated granular ureas were dried for an additional 10 min in the chamber using the same flow of fluidizing air.

Efficiency of mass coating per granular urea: Ten coated granular ureas were randomly selected from each experiment and each was dissolved in 5 mL of distilled water. The 5 mL of solution containing the coated granular urea was then allowed to dissolve overnight. Later, the concentration of the blue dye was determined by measuring the absorbance at a wavelength of 325 nm using UV visible spectrophotometer (UV-2401PC, Shimadzu). At each experiment, one average was taken. The mass of blue dye per coated granular urea, which is proportional to the total mass of coating per coated granular urea, was then calculated. The efficiency of mass coating per granular urea (Eff.) was calculated as:

$$\text{Eifficiency (\%)} = \frac{Y_a}{Y_e} \times 100 \quad (1)$$

and:

$$Y_e = \frac{1.5\text{g of BB} \times \frac{\text{Volume of solution being sprayed}}{300\text{ mL}}}{4000\text{ granular ureas}} \quad (2)$$

Fig. 1: Tangential fluidized bed coater

Table 2: Experimental layout using L_9 orthogonal array

Exp. No.	Inlet air temperature	Disc rotation speed	Spraying rate error
1	1	1	1
2	1	2	2
3	1	3	3
4	2	1	3
5	2	2	1
6	2	3	2
7	3	1	2
8	3	2	3
9	3	3	1

where, the volume of solution being sprayed depends upon the spraying rate. Y_e denotes expected mass of blue dye per coated granular urea Y_a denotes actual mass of blue dye per coated granular urea received.

Taguchi design of experiment: In this part, the orthogonal array used to reduce the size of experiments for design optimization of the parameters was discussed. The results from the experiments was interpreted using S/N ratio and ANOVA analysis. Afterward, the condition to achieve the optimum efficiency of mass coating per granular urea was obtained.

Design of orthagonal array: In order to get the suitable orthogonal array for the experiments, the number of parameters and levels were referred. Here, three parameters were being studied and each parameter has a three-level design parameters. Therefore, L_9 with design for 4 parameters and three-level design parameters (3^4) was preferred. Principally, L_9 orthogonal array with four columns and nine rows was used. As a result, only nine experiments were essential to study the parameters. The experiments were repeated twice to ensure the consistency of experimental data for a signal-to-noise analysis. The experimental layout for the three parameters using the L_9 orthogonal array was shown in Table 2. Given that the L_9 orthogonal array has four columns, one column of the array was left blank for the error of experiments; orthogonality is not lost by letting one column of the array remains empty (Yang and Tarng, 1998).

Signal-to-noise analysis: In the Taguchi method, a transformation of the replication data to another value created is called signal-to-noise (S/N) ratio. There are several S/N ratios available base on type of characteristic; higher is better (HB), lower is better (LB), or nominal is better (NB) (Ross, 1995). To obtain the optimal efficiency of mass coating per granular urea, the-higher-the-better feature characteristic for each parameter must be engaged. The equation for calculating S/N ratios for HB characteristic is:

$$SN_{HB} = -10\log\left(\frac{1}{r}\sum_{i=1}^{r}\frac{1}{y_i^2}\right) \quad (3)$$

where, r is the number of tests in a trial (number of repetitions) and y_i is the value of efficiency of mass coating per granular urea and the i-th test.

ANOVA analysis: The analysis of variance (ANOVA) was developed by Sir Ronald Fisher in the 1930s as a way to interpret the result (Ross, 1995). It is executed to take variation into account and is statistically based. In this case, three-way ANOVA method was used because three controlled parameters in the experiments. The ANOVA was established based on the sum of the square (SS), the degree of freedom (υ), the variance (V) and the percentage of the contribution to the total variation (P). The five parameters symbols typically used in ANOVA (Ross, 1995) are described below:

- **Sum of Squares (SS):** The total sum of square (SS_T), from S/N ratio can be calculated as:

$$SS_T = \sum_{i=1}^{n}\eta_i^2 - \frac{1}{n}\left[\sum_{i=1}^{n}\eta_i\right]^2 \quad (4)$$

where, n is the total number of experiments and η is the S/N ratio at the i-th test.

The total sum of squares from the parameters (SS_p), can be calculated as:

$$SS_P = \sum_{l=1}^{r}\frac{S_{\eta l}}{r} - \frac{1}{n}\left[\sum_{i=1}^{n}\eta_i\right]^2 \quad (5)$$

where, p represents one of the parameters, l the level number of this specific parameter p, r repetition of each level of the parameter p and $S_{\eta l}$ is the sum of the S/N ratio involving this parameter and level l.

- **Degree of freedom (υ):** A degree of freedom in a statistical sense is associated with each piece of information that is estimated from the data. As being simplified by Ross (1995), the degree of freedom formula was written:

$$\upsilon_p = k_p - 1 \quad (6)$$

where, υ_p denotes degree of freedom for each parameter, p and k_p denotes number of level for each parameter (p).

- **Variance (V):** Variance is defined as the sum of squares of each trial sum result engaged the parameter, divided by the degrees of freedom of the parameter:

$$V_p = \frac{SS_p}{\nu_p} \quad (7)$$

- **Expected sum of squares due to parameter (SS'_p):**

$$SS'_p = SS_p - \upsilon_e V_p \quad (8)$$

where, SS'_p is defined as the sum of squares of parameter minus the error variance times the degree of freedom of each parameter.

- **Percent of the contribution to the total variation (P_p):**

$$P_p\,(\%) = \left[\frac{SS'_p}{SS'_T}\right] \times 100 \quad (9)$$

where, P_p denotes the percentage of the total variance of each individual parameter.

RESULTS AND DISCUSSION

Efficiency of mass coating per granular urea: Table 3 shows the experimental results for efficiency of mass coating per granular urea using Eq. 1 and 2 and the corresponding S/N ratio using Eq. 3. Since the experimental design was orthogonal, it was then likely to split out the result of each parameter at different levels. As per done by Yang and Tarng (1998), for example, the mean S/N ratios for the inlet air temperature at levels 1, 2 and 3 are calculated by averaging the S/N ratios for the experiments 1-3, 4-6 and 7-9, respectively. The mean S/N ratios for each level of the other parameters were computed in the same manner. The mean S/N ratio for each level of the parameters is summarized and called the S/N response table for parameters (Table 4).

Determination of optimal levels: Figure 2-4, show the effect of the three parameters; inlet air temperature, disc rotation speed and spraying rate on the mean S/N ratios, respectively. Figure 2 demonstrates the response of S/N ratio to inlet air temperature. It can be noticed from Fig. 2, as the inlet air temperature increase from 40-80°C, the S/N ratio keeps decrease. It has been showed by Paulo Filho *et al.* (2006), the highest efficiency was achieved at lowest inlet air temperature. They also explained that the major effect of the early drying of the droplet before getting on the particle surface was due to high inlet air temperature. As a consequence of dried, the powders were easily flowed out by the atomizing air. Furthermore, it has been observed that the response of

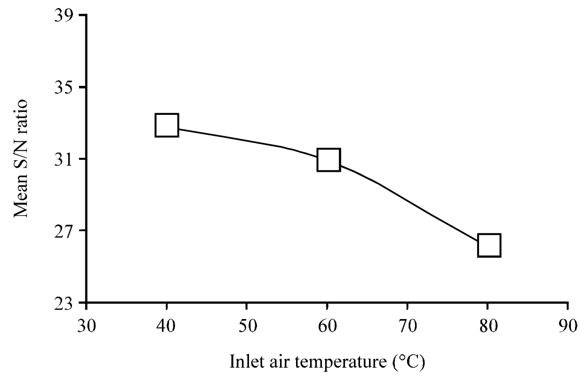

Fig. 2: Effect of inlet air temperature on mean S/N ratio

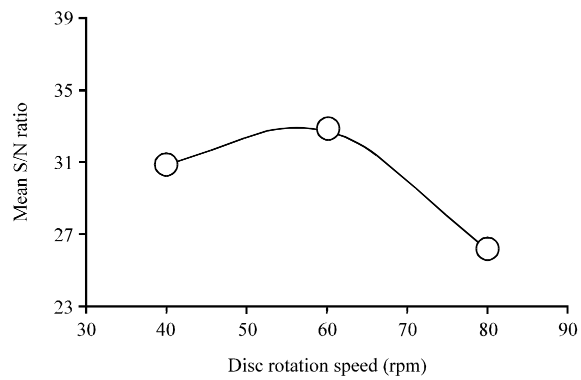

Fig. 3: Effect of disc rotation speed on mean S/N ratio

Table 3: Experimental results for parameters and S/N ratio

				Efficiency of mass coating per granular urea		
Exp. No.	Inlet air temperature (°C)	Disc rotation speed (rpm)	Spraying rate (rpm)	Test 1	Test 2	S/N ratio (dB)
1	40	40	0.5	14.86	15.03	23.49
2	40	60	1.0	62.47	70.22	36.39
3	40	80	2.0	83.88	83.26	38.44
4	60	40	2.0	41.75	29.07	30.56
5	60	60	0.5	87.84	89.30	38.94
6	60	80	1.0	14.02	14.87	23.18
7	80	40	1.0	99.08	76.98	38.69
8	80	60	2.0	14.17	14.12	23.01
9	80	80	0.5	6.83	6.89	16.73

S/N ratio to inlet air temperature was quite big (6.63 dB). This infers that the temperature plays an important role in enhancing efficiency of mass coating per granular urea.

The response of the S/N ratio to disc rotation speed was shown in Fig. 3. The S/N ratio goes up until it reaches a peak at disc rotation speed of 1 rpm. From then on, the S/N ratio decreases as disc rotation speed increases. According to Pisek *et al*. (2000), high rotation speed leads to attrition. In this issue, friction between wall, granular urea and disc surface create the damage and disturbance during the coating process. Thus, it causes the efficiency of mass coating per granular urea to decrease at high speed disc rotation. The fine particles (small mass) pursue the sweeping force and move up with air from the bottom plate (Tracton, 2006). Apart from this, the small granular ureas from attrition process will drag the suspended droplet to follow the sweeping force to upper compartment. The observation on the response of the S/N ratio to disc rotation speed was relatively big (6.67 dB). Figure 4 shows the response of the S/N ratio spraying rate. The mean S/N ratio increases linearly with spraying rate. It has been suggested by Terrazas-Velarde *et al*. (2009), agglomerate growth rate increases as the mass flow rate of solution increases. This may result the efficiency of mass coating per granular urea to be increases with the spraying rate because the suspended droplet of solution was tended to hit the particle surface and agglomerate at high spraying rate. The response of S/N ratio to spraying rate was the biggest value (15.46 dB). This suggests that the spraying rate plays a major role in enhancing efficiency of mass coating per granular urea.

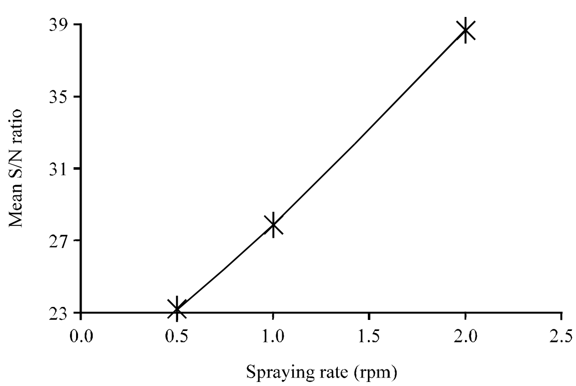

Fig. 4: Effect of spraying rate on mean S/N ratio

Table 4: Signal to noise ratio response table for parameters

	Mean S/N ratio (dB)					
Parameters	Level 1	Level 2	Level 3	Max	Min	Max-Min
Inlet air temperature	32.77	30.90	26.14	32.77	26.14	6.63
Disc rotation speed	30.91	32.78	26.12	32.78	26.12	6.67
Spraying rate	23.23	27.89	38.69	38.69	23.23	15.46

Parameters contribution: The contribution of each parameter to the efficiency of mass coating per granular urea can be determined by performing analysis of variance base on Eq. 4-9. The results of analysis of variance (ANOVA) were displayed in Table 5. The contributions for each parameter, e.g., inlet air temperature, disc rotation speed and spraying rate is 1.184, 1.324 and 54.011%, respectively. The contribution of spraying rate (54.011%) was very significant since the other parameters only contribute not more than 2%. Thus, it shows spraying rate was the main influence on the efficiency of mass coating per granular urea. The ranking of parameters base on their contribution was used as indicator.

Since, inlet air temperature has a minor influence on the efficiency of mass coating per granular urea, it does not matter to use the high inlet air temperature (80°C) or low inlet air temperature (40°C). Thus, in term of energy saving, low inlet air temperature (40°C) was favorable. In the ANOVA analysis, if the percentage error contribution to the total variance is lower than 15%, no important parameter is missing in the experimental design. In contrast, if the percentage contribution of the error exceeds 50%, certain significant parameters have been overlooked and the experiments must be re-designed (Aliofkhazraei *et al*., 2007). As shown in Table 5, the percentage error is 43.481%. This shows that the important parameter in the experimental design was missing but it was not a significant parameter.

Confirmation run: A confirmation experiment is the final step in the design of experiment process (Ross, 1995). It verifies the conclusion from the previous experiments. The confirmation experiment was executed by setting the experimental conditions of the three parameters such as:

Table 5: Results of the ANOVA for efficiency of mass coating per granular urea

Symbol	Parameters	Degree of freedom (υ)	Sum of squares (SS)	Variance (V)	Expected sum of squares (SS')	Contribution (P %)	Rank
A	Inlet air temperature	2	70.123	35.061	6.889	1.184	3
B	Disc rotation speed	2	70.937	35.468	7.703	1.324	2
C	Spraying rate	2	377.439	188.719	314.205	54.011	1
Error		2	63.233	31.616		43.481	
Total		8	581.734			100.000	

Table 6: Prediction results at optimum condition

Parameters	Level description	Level
Temperature (°C)	40.000	1
Disc rotation speed (rpm)	60.000	2
Spraying rate (rpm)	2.000	3
Current grand average of performance (dB)	29.937	
Expected result at optimum condition (dB)	39.550	

40°C for inlet air temperature, 60 rpm for disc rotation speed and 2 rpm for spraying rate. The experimental conditions was performed base on the highest value of mean S/N ratio (dB) obtained. Experiment was conducted with optimum levels of parameters. The observed S/N ratio value was 39.55 dB efficiency of mass coating per granular urea. The optimized process parameters were executed using Qualitek-4 (ANOVA software) and displayed in Table 6; 40°C for inlet air temperature, 60 rpm for disc rotation speed and 2 rpm for spraying rate. It was displayed that under normal condition, the S/N ratio current grand average of performance was 29.937 dB efficiency of mass coating per granular urea. While under optimal condition, confirmation run had showed that S/N ratio efficiency of mass coating per granular urea was found to be 39.55 dB. The improvement of S/N ratio efficiency of mass coating per granular urea from the average of performance to the optimal parameters is 9.613 dB. The results are valid only for the levels of selected parameters.

CONCLUSION

The Taguchi method was used to optimize the efficiency of mass coating per granular urea. It leads to achieve optimization for coating uniformity process parameters. Spraying rate was found to be the main parameters affecting the efficiency of mass coating per granular urea, while inlet air temperature and disc rotation speed had smaller effect on the efficiency of mass coating per granular urea. The optimized process parameters were 40°C for inlet air temperature, 60 rpm for disc rotation speed and 2 rpm for spraying rate.

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