MATERIALE PLASTICE
https://revmaterialeplastice.ro
https://doi.org/10.37358/Mat.Plast.1964

Optimization of Pan Coating Process for Increased Efficiency of Controlled-Release Urea Fertilizer

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Abstract: *The goal of the research is to develop an experimental mathematical model of pan coating process effect on the biodegradable polymer and to determine the optimal process parameters. The polymer solution was conducted with phosphated di-starch phosphate, polyvinyl alcohol, and polyacrylic acid and performed as material coating for the controlled-release urea fertilizer. The image analysis method has been used to determine the particle size distribution, Sauter mean diameter of the particle and layer thickness that is novel. The central composite rotatable design has been selected to determine the regression models of the process, which described the relationship between two objective variables as layer thickness, release time with angle of pan, spray flow, and coating time. The statistical analysis results indicate the fitness of model.*

Keywords*: biodegradable polymer, controlled-release fertilizer, image analysis, optimization, pan coating*

1.Introduction

New fertilizers with almost higher quality usage than traditional fertilizers were enhanced efficiency fertilizers (EEFs) [1]. These fertilizers incorporate a mechanism for regulating the release of nutrients, supplying the essential nutrients at the right time, right rate, right to meet the needs of plants. They were also referred to as smart fertilizers (SFs) [2]. Recently, EEFs are becoming increasingly common in agricultural production and are used as a strategy for sustainable agriculture [3]. They save energy, reduce climate change, using water, CO_2 and N_2O emissions during production and application [4–6], and improve the crop productivity [7, 8], costs, and benefits. Based on the mechanism of nutrient release and the structure of fertilizers, they are classified into 3 groups as slow-release fertilizers (SRFs), stabilised fertilisers (SFs), and the controlled-release fertilizers (CRFs) [2] in which, CRFs were almost used in modern farming [9]. The CRFs have a coating layer (encapsulation) to prevent the direct interaction of the nutrients with the environment to regulate the release of nutrients. The coating layer has been typically made of the substance beeing water-insoluble, semi-permeable, or impermeable [1].

For more than 50 years, the sulphur has been used as the first coating material to manufacture sulphur-coated fertilizers (SCFs) [10]. Due to the presence of micropores, cracks and insufficient sulphur coverage, the consistency of the sulphur coating was poor [6]. The polymers are used as the coating materials and more advanced than the sulphur. They are often classified into synthetic and natural polymers. Synthetic polymers as polyacetate, polyacrylamide, polyacrylic acid, polydopamine, polylactide, polyolefines, polystyrene, polysulphonate, polyvinyl chloride, and poly-urethane have been performed as coating materials. They were costly, harmful, polluting and mostly non-degradable [11]. The natural polymers almost indicate the benefits over synthetic polymers due to non-toxic, low cost, easily accessible, biodegradable and environmentally sustainable.

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Chitosan, sodium alginate, starch and its derivatives, cellulose, lignin, agricultural residues, biochar, and polydopamine could be used [2, 11–14].

CRFs were produced by the coating particle process to create the surrounding layer of coating materials, which avoids changes in existing production technology, reducing production, and investment costs. The particle coating processes can be classified as wet coating, dry coating, gas phase melt coating, and liquid phase encapsulation. The several coating technologies are used on an industrial scale. They are divided into two categories: mechanical agitation system, and pneumatic solid mixing system. The first type, the mixing of the solid has been achieved by the movement of the apparatus itself or using an agitator, which includes drum, pan, and disc coaters. Others have used as the fluidized-bed, spouted-bed or Wurster apparatus coaters [15]. The tablet velocity in pan coaters is predicted by using blue tracer tablets for video imaging. The velocity has been defined as a regression model using design of experiment (DoE) of the three variables, such as pan speed, angle and pan diameter [16]. Several studies have described the influence of the operational conditions (rotation speed, fill level, number of nozzles, and spray rate) on the coating uniformity of table in rotating drums [17]. Recently, the discrete element method (DEM) has been used to model and simulate the coating process in pan coater [18] and rotating drum [19, 20]. Few studies are mostly applied in pharmaceutical products to make the coating tablets. the coating process has been often prepared the coating fertilizers to determine the characteristics of the coating and release time of the product in fertilizer [21]. There are significant without few studies on mathematical modeling and optimization of the coating process. Recently, Da and Rocha [22] have evaluated the influence of the inlet air temperature, the flow rate, and the atomizing air pressure on the coating efficiency for coating urea in a spouted bed using a central composite rotatable design (CCRD) of experiments. The study has not described and determines the optimal parameters of the coating process.

In this study, CRUF was performed using a polymer coating process in a pan coater. This polymer was synthesized by poly-reaction of phosphated di-starch phosphate (PDSP), polyvinyl alcohol (PVA), and polyacrylic acid (PAA) [23]. The regression model clarified the effect of the coating process variables on the thickness of the layer and the release time, by DoE. The optimal results of the model were calculated using multi-objective optimization in MATLAB [24].

2.Materials and methods

2.1. Materials

Phosphated di-starch phosphate (PDSP, E1412, Nam Bao Tin Co., Vietnam), polyvinyl alcohol (PVA, PCT1316, 99%, HiMedia Ltd., India), sodium tetraborate ($Na_2B_4O_7.10H_2O$, 99.5%, Xillong Co., China), glycerol ($C_3H_8O_3$, 99%, Guangdong Co., China), polyacrylic acid (pure emulsion, PAA, 2030, Nuplex Resins Co., Vietnam) were purchased. Commercial granular urea (Ca Mau Fertilizer Co., Vietnam) was used to yield CRUF. The chemicals were used for determination of urea such as pure urea (analytical grade, Guangdong Co., China), *p*-dimethyl aminobenzaldehyde (*p* – DMAB, GRM1425, 99%, HiMedia Ltd., India). All chemical reagents were used as received without further purification.

2.2. Experimental procedure

30 g of gelatinized PDSP in 1000 mL of distilled water at 75°C was stirred at 350 rpm for 30 min, added 0.6 g $Na_2B_4O_7.10H_2O$, and then, 30 g of PVA was slowly added and stirred for 30 min. This solution was mixed with PAA ratio of solution PAA was 3:7 (v:v) to create a mixing-polymer solution for the coating material [23]. The viscosity of the forming solution was measured by a technical viscometer (Prona RV-2, Taiwan) and adjusted with distilled water to achieve an appropriate viscosity for spray process. The diagram of the pan coating system one was set up and showed in Figure 1. The coating solution was placed into the pressure tank (Prona RC-2E, Taiwan) and sprayed by an automatic spray gun (Prona RA-100RC-08R, Taiwan) with a diameter of 0.8 mm. The spray process was operated at 0.18 MPa pressure with spray flow of 1.5–2.5 mL/min.

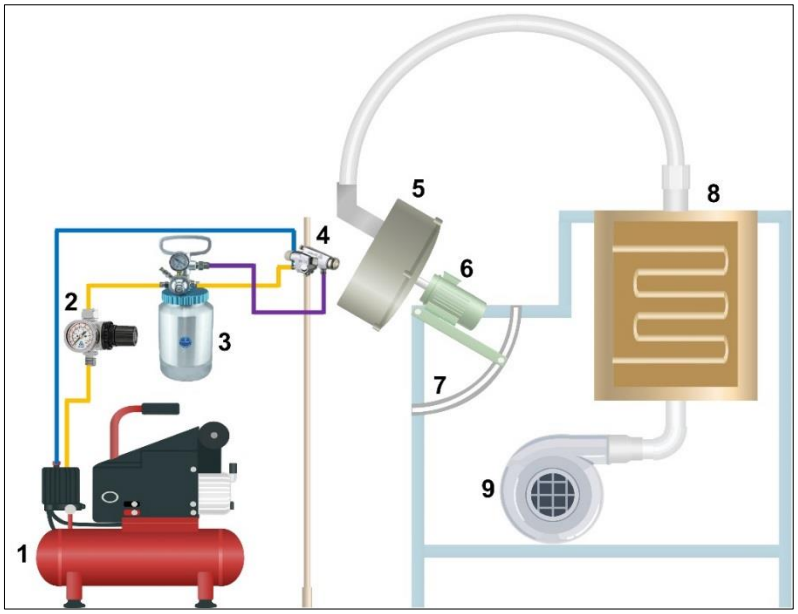

Figure 1. The diagram of pan coating system: (1) air compressor, (2) air regulator, (3) pressure tank, (4) automatic spray gun, (5) pan, (6) motor, (7) inclinometer, (8) air heater, and (9) fan

A pan (diameter of 0.5 m) was dried at 70°C, for 10 min. Then, 100 g of granular urea which was classified by a sieve shaker (RETSCH, Germany) with mesh sizes from 2.8 mm to 4.75 mm and used as the core CRUF. The rotational speed, inclination angle of pan, drying temperature, flow rate and coating time were controlled according to the experimental conditions. Finally, the product was dried at 70°C for 15 min. The particle size distribution of uncoating and coating urea was determined with use of a digital camera (Sony RX100 Mark V, Japan) and the image processing toolbox were conducted by MATLAB software [25]. *Sauter* mean diameter (SMD) was also calculated as Equation (1) [26].

$$SMD = \frac{\sum(n_i.d_i^3)}{\sum(n_i.d_i^2)} = \frac{1}{\sum\left(\frac{x_i}{d_i}\right)} \quad (1)$$

The n_i, d_i, and x_i were the number of particles, the particle size, and the mass fraction, respectively. The concentration of urea was measured according to the spectrophotometric method [27] on a spectrophotometer (GENESYS 20, Thermo Scientific, USA). 1.00 g of uncoating or coating urea was put into net bags. The bags were placed in sealed flask containing 200 mL of distilled water at temperature 25°C. After a certain time, the samples took out 2 mL and measured the urea concentration. The ratio of urea release was determined by Equation (2).

$$\%\ urea\ release = \frac{C_i}{C_*} \quad (2)$$

where the C_i was urea concentration of samples at a certain time, the C_* was total urea concentration, which was determined after taking 1.00 g of fertilizers, cutting, putting into a beaker containing 200 mL of distilled water, stirring at 200 rpm for 30 min to be totally released.

2.3. Design of experiment

The coating process has been a complex process and affected by several factors. The coating factors have been fully classified into the following categories: coating formulation, core, pan coater, spraying system [28], and thermodynamic factors [29]. Based on conducting influence analysis and the experimental conditions, the independent factors have affected the coating layer thickness. They were

showed in Table 1. The effect of the factors was calculated by screening experiment with Plackett-Burman design to determine significant factors [30].

Table 1. Factors and levels for the design of experiments

Factor	Variable		Level	
	Coded	Uncoded	–	+
Inclination angle of pan (°)	X_1	x_1	40	60
Spray flow (mL/min)	X_2	x_2	1.5	2.5
Coating time (min)	X_3	x_3	30	60
Pan loading (kg)	X_4	x_4	0.04	0.06
Rotational speed of pan (rpm)	X_5	x_5	50	70
Drying temperature (°C)	X_6	x_6	60	80
Drying flow rate (m/s)	X_7	x_7	6.1	9.5

To optimize the coating process for two predicted responses such as layer thickness and release time, three significant independent variables were selected from a screening experiment as the critical variables and designated as Z_1, Z_2 and Z_3. The regression model was the second-order equations, as shown in Equation (3).

$$y = \beta_0 + \sum_{i=1}^{3} \beta_i . Z_i + \sum_{i=1}^{3} \beta_{ii} . Z_i^2 + \sum_{i=1}^{3} \sum_{j \neq i=2}^{3} \beta_{ij} . Z_i . Z_j + \varepsilon \quad (3)$$

The $\beta_0, \beta_i, \beta_{ii}$ and β_{ij} were constant, linear coefficients, quadratic coefficients, and interaction coefficients, respectively and ε was the statistical error. The regression coefficients were determined from experimental data by using CCRD and the linear regression model [30]. The effects of significant factors and their interactions were also analyzed by analysis of variance and the response surface plots. Finally, the optimal results were calculated by multi-objective optimization for minimal layer thickness and maximum release time [31].

3.Results and discussions

3.1. CRUF characterization

A CRUF was produced by the pan coating process system as shown in Figure 2. As shown in Figure 2a, the colour of the particles is the same and the range of size is not wide. This proved that the coating layer was created from the coating, which was uniform. The SEM image of layer surface as shown in Figure 2b. It revealed that the surface structure was smooth, thicker, and completely covered core. A cross-sectional of SEM image of a CRUF as shown in Figure 2c, the layer structure was well tight, conjunct, without the presence of micropores, cracks and other defects. So, it reduced the possibility of swelling, crack of layer, and surface evaporation and increased storage capacity and release time.

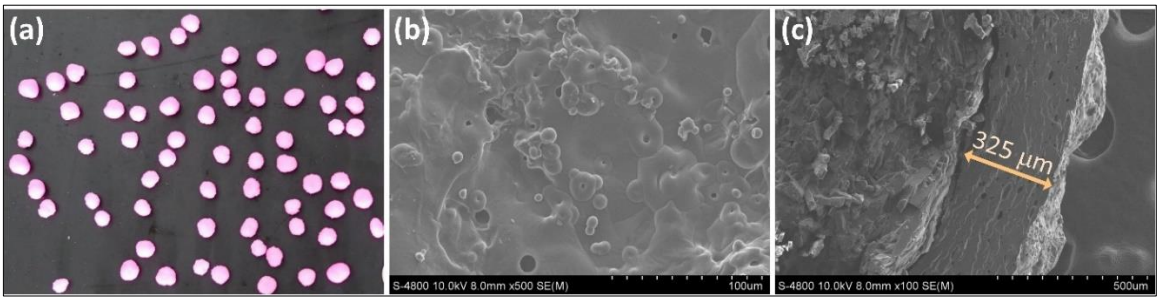

Figure. 2 The image of CRUF: (a) image of the particles, (b) surface SEM and (c) cross-sectional SEM image

The thickness of the coating layer is also determined based on Figure 2c. This value is equivalent to a thickness of 0.321 mm as it is calculated in Equation (4). The method of image analysis is therefore reliable and suitable for the determination of SMD and layer thickness.

$$\bar{y}_1 = \frac{(SMD)_{CRUF} - (SMD)_{core}}{2} \text{ (mm)} \quad (4)$$

The $(SMD)_{CRUF}$ is SMD of CRUF and $(SMD)_{core}$ is SMD of core (uncoated urea).

3.2. Screening experiment to select significant factors

The design matrix in the Placket-Burman model [32] with response values was presented in Table 2. The effects of selected factors are analyzed and presented in Figure 3. A positive value of the effect indicates an increase in the layer thickness if the variable increases. Conversely, a negative value exposes that the one is better at low levels of the variables. This bar chart demonstrates that the higher effects are derived from the factors such as inclination angle of pan, spray flow, and coating time. The variables of these factors are going to be used to determine the mathematical models and the optimal values.

Table 2. Plackett-Burman design matrix and response values

No.	Factors							Response
	X_1	X_2	X_3	X_4	X_5	X_6	X_7	$\bar{y}_1$
1	+	–	–	+	–	+	+	0.1137
2	+	+	–	–	+	–	+	0.1220
3	+	+	+	–	–	+	–	0.1841
4	–	+	+	+	–	–	+	0.3009
5	+	–	+	+	+	–	–	0.1423
6	–	+	–	+	+	+	–	0.1690
7	–	–	+	–	+	+	+	0.1958
8	–	–	–	–	–	–	–	0.1621

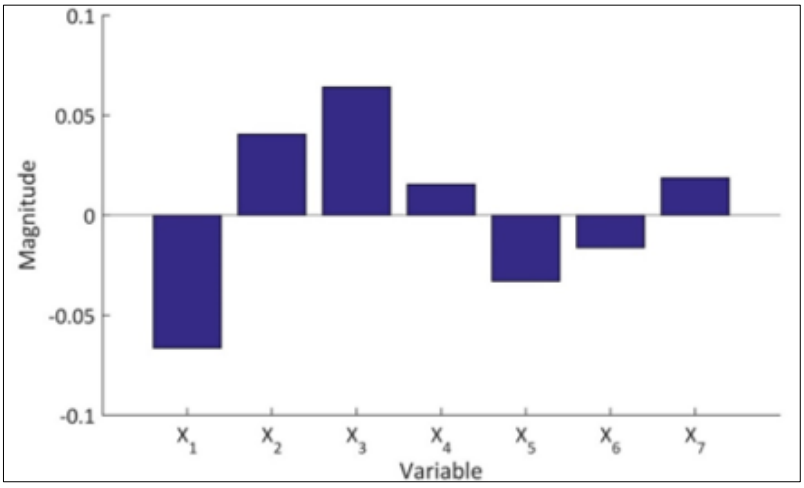

Figure 3. The magnitudes of the factors

3.3. Optimization of the experimental conditions

Three independent variables like the angle of the pan (Z_1), the spray flow (Z_2) and the coating time (Z_3) were selected as the critical variables and designated at three distinct levels as the lower (–), zero (0), and upper levels (+). Design matrix of CCRD for three variables with two predicted values of layer thickness ($\bar{y}_1$) and eighty percent release time (y_2) are showed in Table 3.

Table 3. Matrix of central composite rotatable design

No.	Z_1	Z_2	Z_3	$Z_1.Z_2$	$Z_1.Z_3$	$Z_2.Z_3$	Z_1^2	Z_2^2	Z_3^2	$\bar{y}_1$	y_2
1	–	–	–	+	+	+	+	+	+	0.3009	4.1016
2	+	–	–	–	–	+	+	+	+	0.0225	4.3594
3	–	+	–	–	+	–	+	+	+	0.4004	4.6289
4	+	+	–	+	–	–	+	+	+	0.0523	4.1836
5	–	–	+	+	–	–	+	+	+	0.0659	4.9453
6	+	–	+	–	+	–	+	+	+	0.3458	4.7344
7	–	+	+	–	–	+	+	+	+	0.2397	4.6875
8	+	+	+	+	+	+	+	+	+	0.3090	4.2188
9	-1.682	0	0	0	0	0	2.829	0	0	0.3221	4.4648
10	1.682	0	0	0	0	0	2.829	0	0	0.1432	3.7969
11	0	-1.682	0	0	0	0	0	2.829	0	0.1031	4.6875
12	0	1.682	0	0	0	0	0	2.829	0	0.2472	4.6992
13	0	0	-1.682	0	0	0	0	0	2.829	0.1375	4.5469
14	0	0	1.682	0	0	0	0	0	2.829	0.4381	5.1094
15	0	0	0	0	0	0	0	0	0	0.3563	4.3125
16	0	0	0	0	0	0	0	0	0	0.3340	4.3828
17	0	0	0	0	0	0	0	0	0	0.3431	4.5352
18	0	0	0	0	0	0	0	0	0	0.3186	4.6758
19	0	0	0	0	0	0	0	0	0	0.3511	4.4648
20	0	0	0	0	0	0	0	0	0	0.3035	4.4180

The regression coefficients were calculated, and their statistical significances were checked by the linear regression model of MATLAB. The outcomes of experiments such as $\bar{y}_1$ and y_2 were indicated in Table 4 and Table 5. The effect of the regression coefficients in the models could not be significant if the P-value is greater than 0.05 and is removed from the regression equation.

Table 4. The regression coefficients with the response value as the layer thickness ($\bar{y}_1$).

Coefficients	Estimate	Standard error	t-Stat	P-value
β_0	0.33471	0.018589	18.006	< 0.0001
β_1	- 0.04233	0.012333	- 3.432	0.0064
β_2	0.03724	0.012333	3.020	0.0129
β_3	0.05051	0.012333	4.096	0.0022
β_{12}	- 0.03504	0.016114	- 2.174	0.0548
β_{13}	0.12196	0.016114	7.569	< 0.0001
β_{23}	0.00096	0.016114	0.060	0.9536
β_{11}	- 0.03779	0.012004	- 3.148	0.0104
β_{22}	- 0.05812	0.012004	- 4.841	0.0007
β_{33}	- 0.01830	0.012004	- 1.524	0.1584

Table 5. The regression coefficients with the response value as eighty percent release time ($\boldsymbol{y_2}$).

Coefficients	Estimate	Standard error	t-Stat	P-value
β_0	4.46721	0.051766	86.297	< 0.0001
β_1	- 0.14574	0.034343	- 4.244	0.0017
β_2	- 0.02945	0.034343	- 0.858	0.4113
β_3	0.16537	0.034343	4.815	0.0007
β_{12}	- 0.12011	0.044874	- 2.677	0.0232
β_{13}	- 0.06151	0.044874	- 1.371	0.2004
β_{23}	- 0.14061	0.044874	- 3.134	0.0106
β_{11}	- 0.13357	0.033428	- 3.996	0.0025
β_{22}	0.06525	0.033428	1.952	0.0795
β_{33}	0.11290	0.033428	3.378	0.0070

A check of fit of these models pointed out that the value of the determination coefficients (R^2) for each model was 0.929 and 0.905. The values of probability with the Fisher's statistic were also very low, 0.000124 and 0.000496. They indicated that the goodness of the fit models can be large enough to use

optimizing coating process. The second-order polynomial equations in real variables (z_1, z_2, z_3) were indicated as Equation (5) and Equation (6).

$$\bar{y}_1 = 0.20061 - 0.00303.z_1 + 1.00436.z_2 - 0.03728.z_3 + 0.00081.z_1.z_3 - 0.00038.z_1^2 - 0.23247.z_2^2 \quad (5)$$

$$y_2 = 1.71291 + 0.16704.z_1 + 2.04480.z_2 + 0.00336.z_3 - 0.02402.z_1.z_2 - 0.01875.z_2.z_3 - 0.00134.z_1^2 + 0.00050.z_3^2 \quad (6)$$

The relationship between multiple input variables and one output variable are indicated by a response surface plot as shown in Figure 4 for layer thickness ($\bar{y}_1$) and Figure 5 for eighty percent release time (y_2).

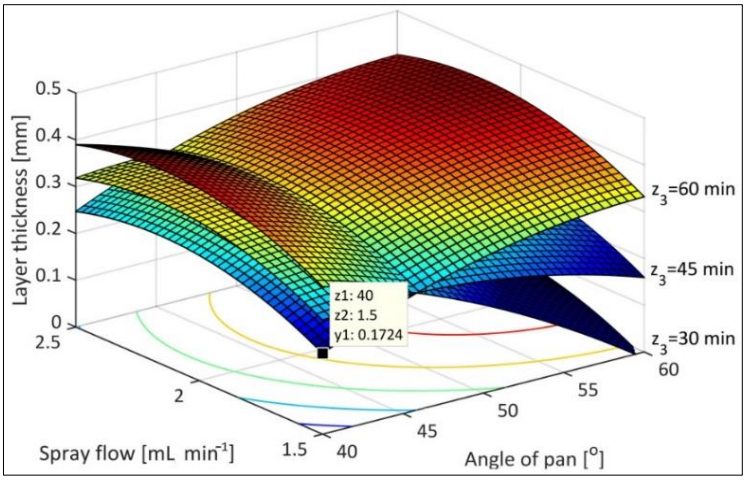

Figure 4. Response surface of layer thickness with spray flow and angle of pan at a coating time of 30, 45, and 60 min

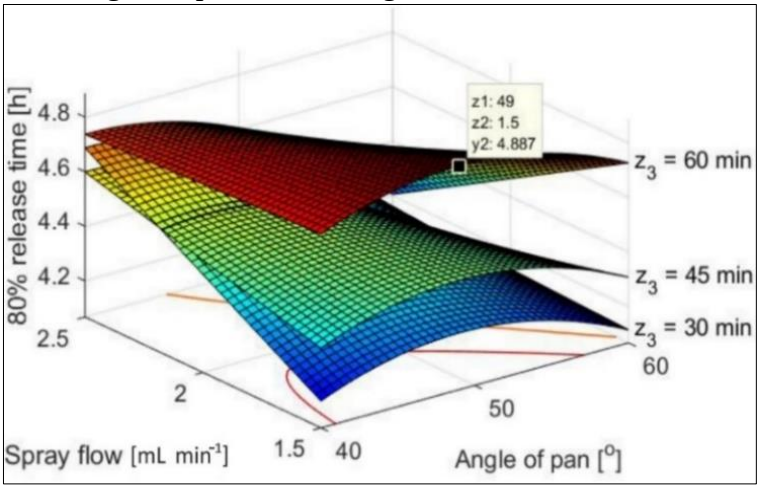

Figure 5. Response surface of eighty percent release time with spray flow and angle of pan at a coating time of 30, 45, and 60 min

The impacts of the main variables (z_1, z_2, z_3) and interaction between them ($z_1.z_2$, $z_2.z_3$, $z_1.z_3$) on response values are presents in Figure 4 and 5. The coating time greatly affects layer thickness and release time. When the coating time increases, the slope of the response surface and their value increases. The response value reaches the minimum at the boundary value of the experimental region.

As shown in Figure 4, the relationship between layer thickness and spray flow is a quadratic curve and the maximum value obtains at spray flow of 2.16 mL/min. At the high spray flow, the impact force of the spray flow affects the particle surface and droplet size is smaller, thereby reducing the interaction

between the surface particles and the coating material, and being easily exhausted into the environment. Therefore, the layer thickness of coating material decreases. In addition, the release time increases with increasing spray flow when the value of coating time is low, but vice versa is at high coating time values. The change of the release time is insignificant as shown in Figure 5. The properties of the layer are greatly affected at the first stage which is the contact stage of the coating material with the core to form the layer. At the second stage its properties do not change when the layer is stably formed.

The angle of the pan also has a great influence on layer thickness. The larger the angle value, the layer thickness reaches the maximum value at 60^o in Figure 4. As shown in Figure 5, the release time decreases when the angle is greater than 49^o. Due to the increasing angle, the particle is spread over the surface of pan so that surface area in contact with the coating solution increases where the centrifugal force, the particle friction force and the turning force also decrease. The layer thickness increases but its properties such as uniformity, tightness and compression reduces and the release time is less.

The selection of the target variable is decisive in determining the optimal value of the pan coating process. This study has two-objective fitness function as Equation (5) and Equation (6). The results of Pareto optimization to find a minimum layer thickness are satisfy the maximum of percent eighty release time as shown in Figure 6. The release time is considered to be the most important parameter of CRUF. The optimal value is the angle (z_1) of 48.051^o, spray flow (z_2) of 1.502 mL/min, coating time (z_3) of 59.965 min with maximum percent eighty release time of 4.884 h, and layer thickness of 0.273 mm. This result is a key factor that to help calculate, design of the coating process and its application in the production of CRUF by pan coater.

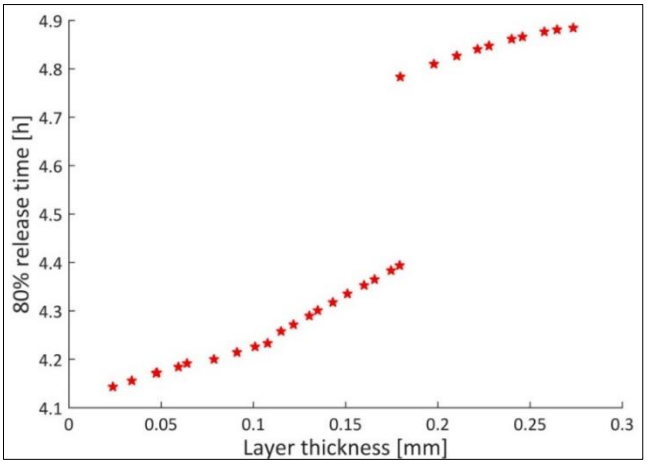

Figure 6. Pareto front of the optimization results

4.Conclusions

The study developed a suitable pan coating system to use biodegradable polymer synthesized from PDSP, PVA and PAA as coating material to produce CRUF. The particle size distribution and Sauter mean diameter and layer thickness were determined by novel image analysis method. The factors of the process were identified, analyzed and evaluated to determine the level of effects and significant factors by the Plackett-Burman model. The regression equations were established from the experimental results using the linear regression model and were analyzed statistically. Finally, the optimal value with a maximum percent eighty release time and a minimum layer thickness was calculated by multi-objective optimization using genetic algorithm. The result was an angle of 48.05^o, spray flow of 1.502 mL/min and coating time of 59.965 min. The optimal value is at the boundary of the experimental region so that the next project determines the best optimum value for this coating process.

Acknowledgments. This research is funded by Ho Chi Minh City University of Technology, VNU-HCM, under grant number BK–SDH–2020–1581011.

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Manuscript received: 3.06.2020