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Research Article

Optimization of Spraying Process via Response Surface Method for Fabrication of Cellulose Nanofiber (CNF) Film

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Cellulose nanofiber (CNF) is a sustainable bionanomaterial which has fibril's width varying from 5 nm to ~73 nm with an average length of 8 μm. It can be used as a base material for various functional materials such as barrier, flexible electronic substrates, and membrane etc. Though several methods such as solvent casting and vacuum filtration are available for the production of CNF film in laboratory scale, the major constraints are film formation time, shrinkage on the film, and poor uniformity. Spraying CNF suspension is one of the emerging methods which forms the film rapidly. The present investigation deals with the optimization of critical parameters such as CNF suspension concentration, velocity of the conveyor, and spray distance involved in the spraying process via central composite design (CCD) in the response surface methodology (RSM). The influence of these parameters on the basis weight and thickness of the CNF film was evaluated from the linear models. It concludes that the CNF suspension concentration is a strong parameter for controlling the basis weight and the thickness of the CNF film. The developed linear models were validated with experimental data confirming that it was a good fit. Given this correspondence, these models may be used for scaling up the spraying process for the fast production of CNF film.

1. Introduction

Cellulose nanofiber (CNF) is a carbohydrate nanomaterial derived from the cellulose, and it can be used as a base biomaterial for fabrication for films [1], coating [2], gels, membranes [3], and substrates for flexible electronics [4]. CNF has become a potential alternative to replace the synthetic plastics [1], and it is an ecofriendly biomaterial having excellence in their biodegradability in the soil [5]. CNF was produced via delaminating cellulose fibers through mechanical [6], chemical [7], or biological methods [8, 9]. The width of nanofibrils in CNF varied from 5 nm to ~73 nm [4] and acts as a building block for fabrication of CNF films. Cellulose nanofibrils offer a considerable value in aspect ratio and mechanical strength. The reduction of fibers from macroscale to nanoscale improves flexibility and gives large specific surface area and biodegradability [7]. In addition to that, cellulose nanofibers give high mechanical strength with

Table 1: Factor for central composite design.

Factor	Name	Units	Type	Subtype	Minimum	Maximum	Coded low	Coded high	Mean	Std. dev.
A	Suspension concentration	wt. %	Numeric	Continuous	0.5000	2.25	−1 ↔ 0.85	+1 ↔ 1.90	1.38	0.4532
B	Velocity of the conveyor	cm/s	Numeric	Continuous	0.2500	0.6000	−1 ↔ 0.32	+1 ↔ 0.53	0.4250	0.0906
C	Spray distance	cm	Numeric	Continuous	30.00	50.00	−1 ↔ 34.05	+1 ↔ 45.95	40.00	5.18

Table 2: Experimental design.

Std.	Run	Factor 1 *A*: suspension concentration wt. %	Factor 2 *B*: velocity of the conveyor cm/s	Factor 3 *C*: spray distance cm	Response 1 Basis weight g per sq.m	Response 2 Thickness Microns
3	1	0.8547	0.5290	34.0539	48.34	0.07325
6	2	1.8952	0.3209	45.9460	202.05	0.1955
18	3	1.375	0.425	40	100.45	0.11426
15	4	1.375	0.425	40	100.45	0.11426
10	5	2.25	0.425	40	193.01	0.1935
16	6	1.375	0.425	40	100.45	0.11426
14	7	1.375	0.425	50	102.24	0.13939
13	8	1.375	0.425	30	89.95	0.06375
5	9	0.8547	0.3209	45.9460	48.34	0.06575
1	10	0.8547	0.3209	34.0539	53.87	0.05278
7	11	0.8547	0.5290	45.9460	48.34	0.07325
19	12	1.375	0.425	40	100.45	0.11426
4	13	1.8952	0.5290	34.0539	189.95	0.1857
2	14	1.8952	0.3209	34.05396	202.05	0.1955
17	15	1.375	0.425	40	100.45	0.11426
9	16	0.5	0.425	40	30.87	0.02787
8	17	1.8952	0.5290	45.9460	202.05	0.1955
11	18	1.375	0.25	40	90.05	0.12505
12	19	1.375	0.6	40	85.93	0.11426

high stiffness and toughness [10]. These properties could decide the applicability of CNF film for various functional applications such as barrier against water vapor barrier [1], gas barrier and membrane for virus removal [11], and wastewater treatment [3].

Time consumption is a major challenge in the conventional methods for the preparation of CNF films [12]. More than 24 hours is required in the casting process for the formation and drying of film by evaporating the solvent from the CNF suspension. The conventional method for fabrication CNF film is vacuum filtration; however, the draining time increased exponentially with CNF suspension consistency. The time consumed for forming CNF film in the filtration process varied from 10 min to 30 min [12, 13].

Spraying CNF on the fabric surface is a newly reported method for fabrication of CNF film, and the time taken for forming the CNF film is 30 minutes followed by vacuum filtration to remove the water from wet film via applying vacuum and consumed good time for dried sheets [13]. Spraying CNF on the polished metal surface such as smooth stainless steel is a well-cited method for production of CNF film with an operation time of less than 1 min [12, 14]. Spraying CNF on the stainless steel produces the film with two unique surfaces, namely, the rough side which is exposed to air and the smooth side which adhered to the stainless steel surface. Unlike vacuum filtration, the operation time for spraying CNF is independent of CNF suspension consistency. As this process gives promising results, the optimization of the spraying process is required to evaluate the variables mainly controlling the property of CNF film [12].

The developed spray experimental setup was used to fabricate the CNF film. When operating the spray coating experimental setup, CNF suspension consistency and velocity of the conveyor and distance between spray tip and base are effective variables and are experimentally optimized. During the operation of experimental setup, the CNF film can be produced by either fixing the suspension consistency and changing the velocity of the conveyor or changing the CNF suspension consistency at constant velocity. Earlier, the reported variables through experimentation are CNF suspension consistency and velocity of the conveyor in the experimental setup [12, 14].

The identified variables are optimized via a statistical modelling for process scale-up and design and also the interaction between these variables evaluated via response surface method (RSM) which is both a mathematical and statistical

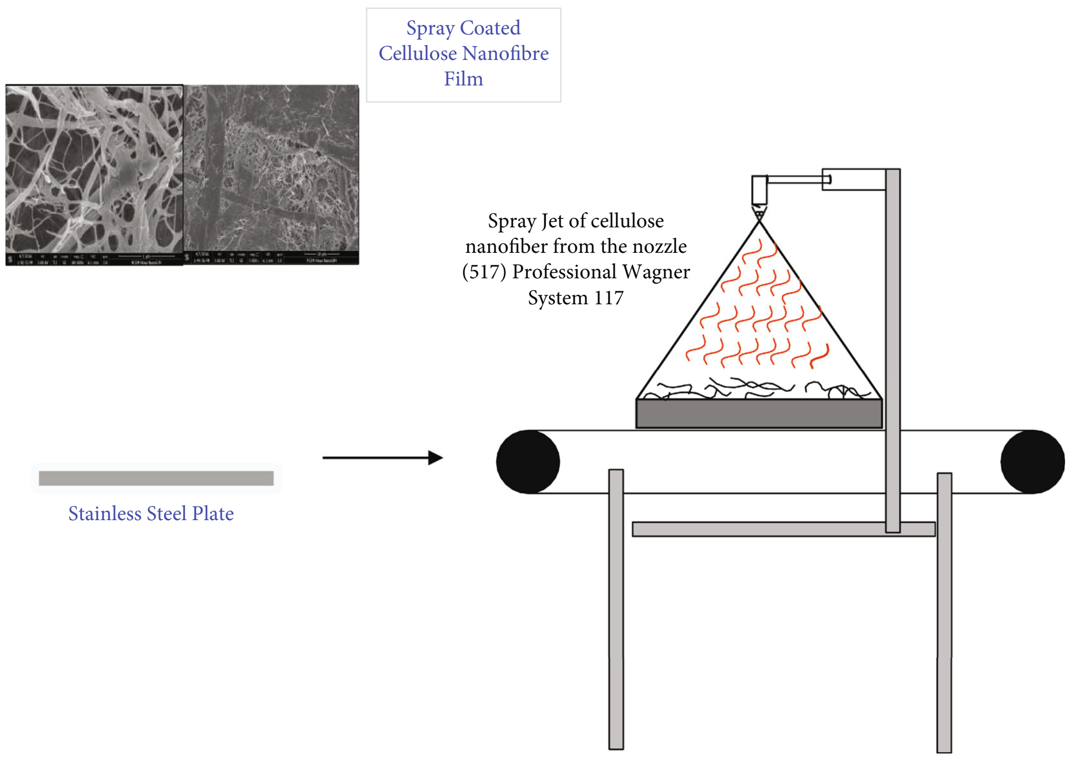

Figure 1: Preparation of CNF film via spray coating. The basis weight and thickness of the film were tailored by either fixing the suspension consistency or varying the velocity of the conveyor. The spray distance is also one of the important factors in the thickness and basis weight of the CNF film.

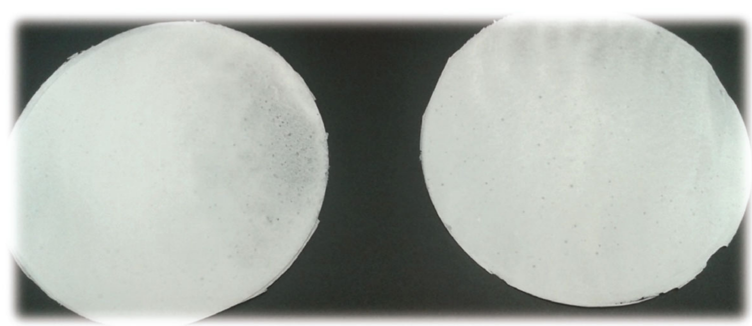

Figure 2: Spray coated cellulose nanofiber film.

technique for a design of experiments. By this method, the effect of dependent and independent variables can be determined for spraying process. This method is commonly applicable in process design where input variable impacts on the formation of CNF film via spraying. The model from this study depends on various factors influencing the basis weight and thickness of CNF films. In the optimization method, the linear or quadratic models are used to express the relation between various responses with input variables [15, 16].

The correlation between input variables ($x_1, x_2, \cdots \cdots, x_k$) and responses (y) can be mentioned as the following correlation:

$$y = f(x_1, x_2, \cdots \cdots, x_k) + €, \tag{1}$$

where y = responses from the effect of input variables and € means statistical error.

The central composite design was implemented to the effect of variables on the spraying process to fabricate the CNF films. The expected linear model is in the form of the following correlation with considerations of main variables and their interaction between them:

$$y = \beta o + \beta 1 \times 1 + \beta 2 \times 2 + \beta 3 \times 3 + \cdots \cdots ..\beta n \times n. \tag{2}$$

For getting a good regression fit from response surface to actual surface, the range of input variables is carefully considered and listed in Table 1. Generally, the lowest and highest range of variable can be designated by -1 and +1, respectively. The CNF suspension concentration, velocity

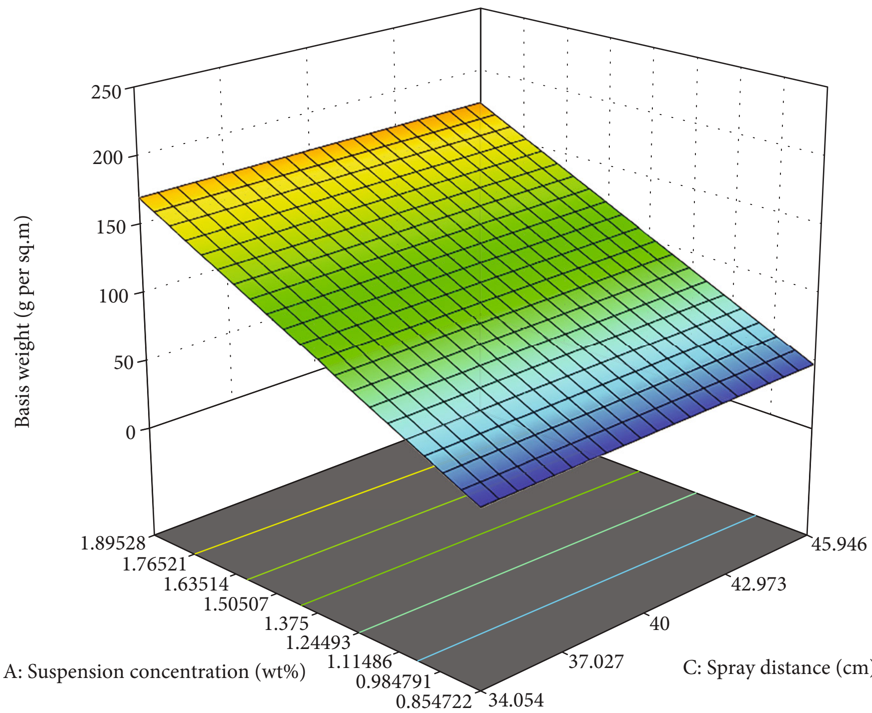

Figure 3: 3D response surface plot for the effect of CNF suspension concentration and spray distance on the basis weight of the film. This concludes the interaction effect between CNF suspension and spray distance on the basis weight of the film.

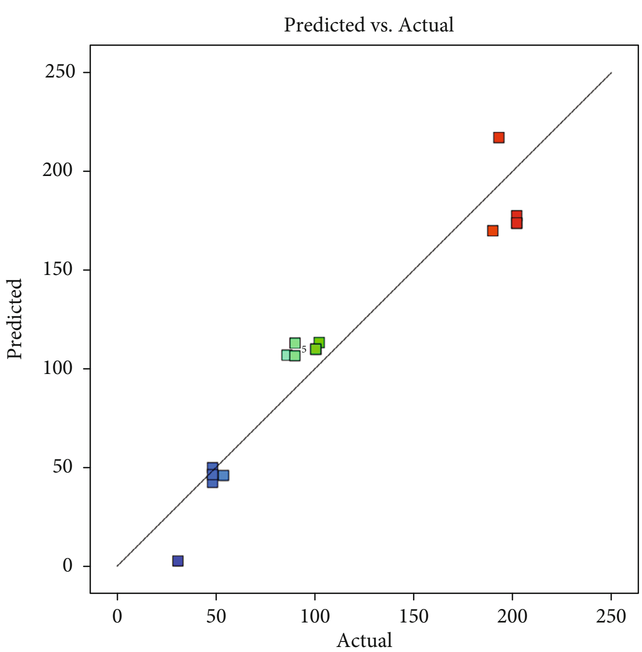

Figure 4: The predicted and actual plot for basis weight of the CNF film.

of the conveyor, and spray distance were the important variables to impact on the basis weight and thickness of the CNF film [12].

This paper deals with the optimization of the process variables in the spraying process for the preparation of CNF films and evaluating the effect of dependent and independent variables on the spraying process, and the developed model is validated with the experimental data.

2. Materials and Methods

Cellulose nanofiber (CNF) also called as microfibrillated cellulose (MFC) supplied from DAICEL Chemical Industries Limited (Celish KY-100S, diameter of cellulose nanofibers around ~73 nm with average length of 8 μm) was used for spraying CNF on the polished meta surface to fabricate the free-standing CNF films. Celish cellulose nanofiber was produced via microfibrillation of refined cellulose macrofibers. During this process, the width of cellulose fiber was reduced from several microns into 1/100th of microns. This is why it is commercially called as microfibrillated cellulose. The solid content in KY100S was varied from 23 wt. % to 27 wt. %.

2.1. Methods for Optimization. For optimizing the process conditions in the spraying process, CCD is used as a RSM method. The Design-Expert 8.0.5 software (State-Ease Inc., Minneapolis MN, USA) was used for this investigation. The effect of three main variables on spraying was CNF suspension concentration, velocity of the conveyor, and distance between the spray tip and the base. The variables are optimized and their interaction evaluated via this method. The level and ranges chosen for the factors are shown in Tables 1 and 2. The complete design consisted of 19 experimental points. The 19 samples were prepared in random order. In each experiment, the basis weight and thickness of CNF film were measured and the trial was performed in triplicates. The basis weight

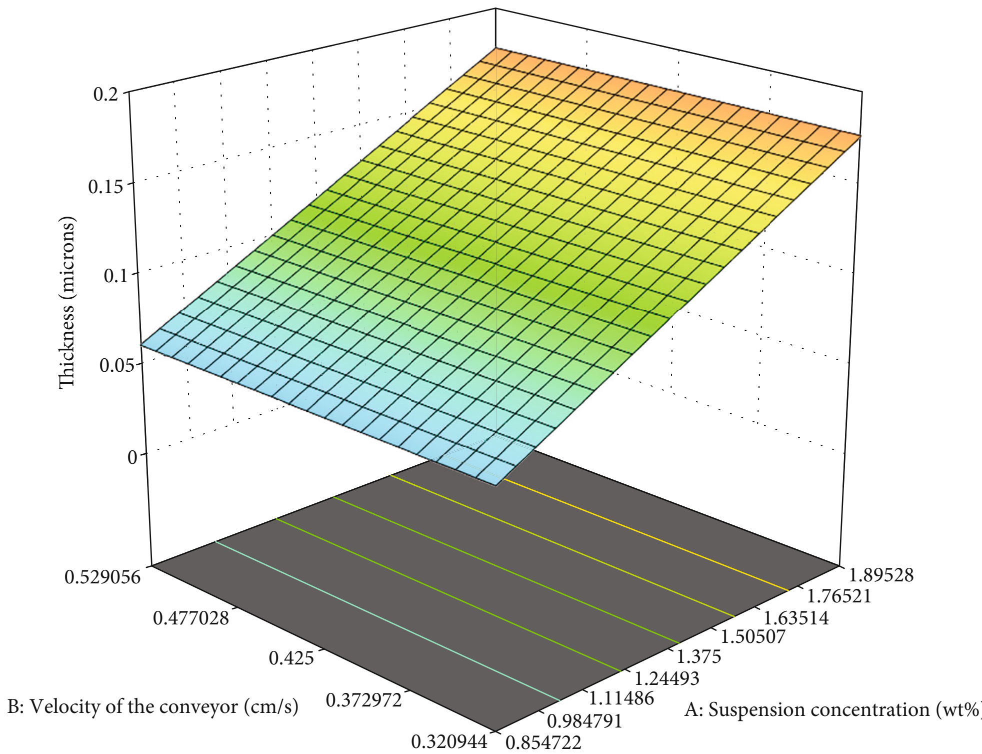

Figure 5: Effect of velocity of the conveyor and CNF suspension concentration on the thickness of the film.

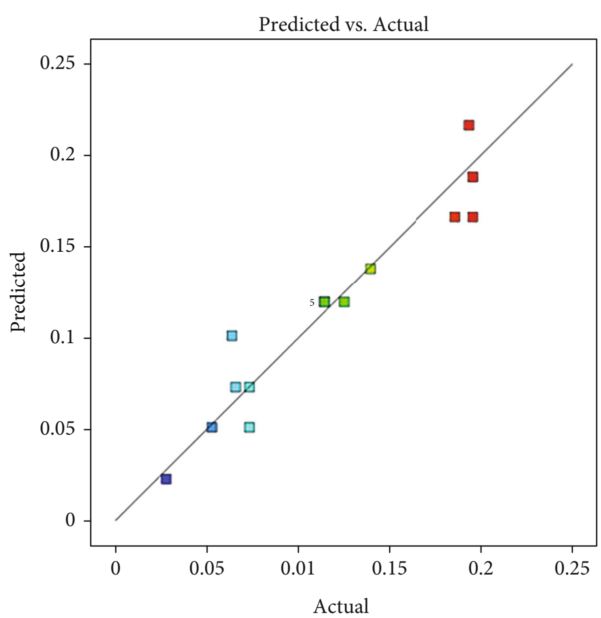

Figure 6: The predicted and actual plot for thickness of the CNF film.

and thickness of the CNF film were taken mean values as responses. Table 2 shows the experimental design matrix and consists of corresponding outputs as responses.

2.2. Experimental Works. The optimized conditions obtained from the developed model were compared with the data from real-time experimental works. The developed spray coating prototype for fabrication of CNF film is shown in Figure 1. The DIACEL CNF (25 wt. %) was used as a feedstock for preparation of CNF film via spraying after disintegration of fibers. 25 wt. % CNF was diluted with double distilled water and disintegrated to the CNF suspension varies from 1 wt. % to 2 wt. %. These suspensions are used for spraying on the polished metal surface. The circular stainless steel plate is used as base surface, and it is kept on the variable speed conveyor in the spray coating experimental setup. The Professional Wagner spray system (model number 117) was used for spraying operations and was operated at a pressure of 200 Bar. The elliptical spray jet was developed by the type 517 spray tips, and the angle and beam of the spray jet are 50° and 22.5 cm, respectively. 30.0 ± 1.0 cm from the spray nozzle was achieved to coat 59 cm diameter of the circular steel plate.0.32 cm/sec was fixed to run the conveyor for the spraying of CNF on the base surface. During the operation of spray coating prototype, the Wagner spray system was allowed to spray for 30 seconds before spray deposition of CNF on the base surface. By this way, the spray system was reached to equilibrium. The wet CNF film on the circular stainless steel plate was dried under the standard laboratory conditions. The drying of wet CNF film consumed 24 hours. The well-dried CNF film was peeled from the stainless steel plate and kept at 23°C and 50% RH for further characterizations [10]. The basis weight (g/m^2) of the CNF film was calculated by dividing the weight of the film, after 4 hours drying in the oven at a temperature of 105°C, by the film's area [12, 14].

Table 3: Response 1: basis weight.

Source	Sum of squares	df	Mean square	F value	p value	
Model	55507.95	3	18502.65	46.98	<0.0001	Significant
A: suspension concentration	55409.46	1	55409.46	140.68	<0.0001	
B: velocity of the conveyor	44.16	1	44.16	0.1121	0.7424	
C: spray distance	54.33	1	54.33	0.1379	0.7155	
Residual	5907.89	15	393.86			
Lack of fit	5907.89	11	537.08			
Pure error	0.0000	4	0.0000			
Cor. total	61415.84	18				

Factor coding is *coded*. Sum of squares is *type III-partial*.

Table 4: Response 2: thickness.

Source	Sum of squares	df	Mean square	F value	p value	
Model	0.0469	3	0.0156	57.84	<0.0001	Significant
A: suspension concentration	0.0453	1	0.0453	167.44	<0.0001	
B: velocity of the conveyor	$4.028E-11$	1	$4.028E-11$	$1.490E-07$	0.9997	
C: spray distance	0.0016	1	0.0016	6.09	0.0261	
Residual	0.0041	15	0.0003			
Lack of fit	0.0041	11	0.0004			
Pure error	0.0000	4	0.0000			
Cor. total	0.0510	18				

Factor coding is *coded*. Sum of squares is *type III-partial*.

3. Results and Discussion

Spraying is the fastest process in the fabrication of CNF films and a potential for scale-up in an industry, and the formation of CNF film consumes less than a minute when compared to the conventional method like vacuum filtration [12]. Figure 2 shows the spray coated cellulose nanofiber film which is compact and consists of two unique surfaces, namely, the rough sides exposed to air and the smooth side exposed to stainless steel surface. To optimize the variables in the process design for spraying, the optimization and mathematical modelling will be assisted to find the effect of input variables in the CNF film formation [12, 14].

Figure 3 reveals that the CNF suspension concentration is the most important parameter for controlling the basis weight of the film. The variation of spray distance between CNF suspension and velocity of the conveyor does not much influence on the basis weight of the film. At lower suspension concentration, spray distance does affect the basis weight and thickness of the CNF film via the reflecting of the CNF jets from the spray pattern. The viscosity of CNF suspension is high and higher CNF suspension which inhibits the reflection of fluids from the spray jets. Spray distance is a hidden parameter for controlling the basis weight and thickness of the film [14].

Figure 4 shows the predicted and actual plot of the basis weight of the CNF film. The actual response data are experimental values for various runs, and it is compared with predicted values from mathematical correlations. The determination coefficient (R^2) was evaluated as 0.9038, and the value of adjusted R^2 (R^2 adj) was 0.885. This confirms that there is a good correlation between actual and the predicted data in the spraying process. "The predicted R^2 of 0.8269 is in reasonable agreement with the adjusted R^2 of 0.8846: i.e., the difference between these two values is less than 0.2."

Figure 5 reveals the effect of velocity of the conveyor and CNF suspension concentration on the thickness of the film. CNF suspension concentration is the main variable for controlling the thickness of the CNF film. As discussed earlier, the spraying of diluted CNF suspension causes the reflection of spray jet and resulting poor uniform film in thickness. The thickness and basis weight of the film are directly proportional to the CNF suspension concentration. The impact of other variables such as velocity of the conveyor and spray distance has a little influence in the CNF film thickness and also on the basis weight compared to CNF suspension concentration.

Figure 6 shows the plot between predicted and actual values for the thickness of the CNF film. The determination coefficient R^2 is found to be 0.9204, and the adjusted R^2 is found to be 0.9045. "The predicted R^2 of 0.8544 is in reasonable agreement with the adjusted R^2 of 0.9045: i.e., the difference between these two values is less than 0.2." The predicted values are less within the deviation of 5% which is in great agreement with the expected values. This also implies that the developed model is significant.

3.1. ANOVA for Linear Model (Basis Weight). "The model F value of 46.98 implies that the developed model is significant

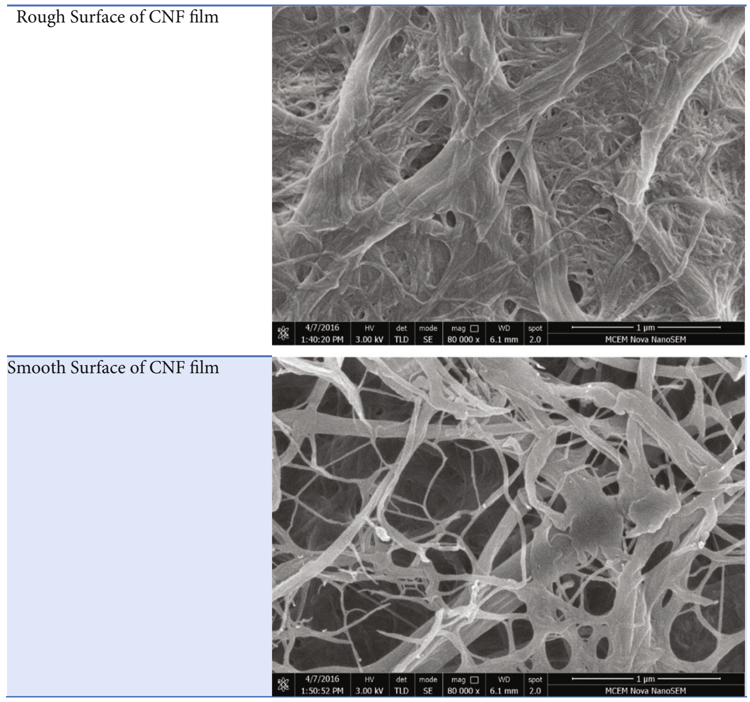

Figure 7: SEM micrographs of CNF film.

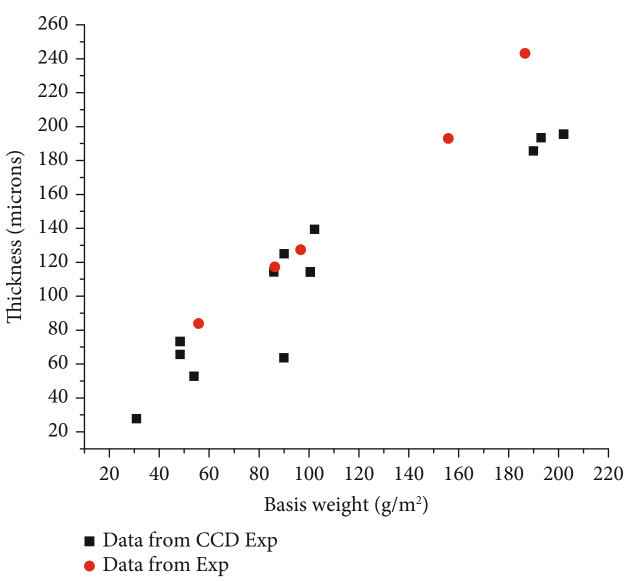

Figure 8: Linear relation between basis weight and thickness.

(Table 3). There is only a 0.01% chance that an F value this large could occur due to noise. p values less than 0.0500 indicate that model terms are significant. In this case, CNF suspension concentration is a significant model term. Values greater than 0.1000 indicate that the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model."

3.2. ANOVA for Linear Model (Thickness). "The model F value of 57.84 implies that the model is significant (Table 4). There is only a 0.01% chance that an F value this large could occur due to noise. p values less than 0.0500 indicate that model terms are significant. In this case, A, 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 your model."

As per the ANOVA table for basis weight and thickness, larger "F" values indicate that the models were statistically anyone of the input variables have significant and significant effect on their responses. In both models, p values for CNF suspension concentration were less than 0.0001 confirming the strong effect on the basis weight and thickness of the CNF film. p values for velocity and spray distance are 0.1121 and 0.1329, respectively, for basis weight of the film, 0.99 and 0.026 for thickness of the film confirms these effects have not much impact on the bulk properties of the film than that of CNF suspension consistency.

4. Model Equation

The following linear models (Equations (3) and (4)) were developed from CCD. From the experimental results, it was observed that basis weight and thickness of the CNF film were strongly dependent on the CNF suspension concentration rather than velocity of the conveyor and spray distance. The mathematical developed using CCD ensures the strong correlation between input factors and output responses which was affirmed by R^2 and its associated values. p values of velocity of conveyor and spray distance for each response basis weight and thickness of film were greater than 0.1 as shown in Tables 2 and 3 which indicates that these parameters did not show significant effect on the

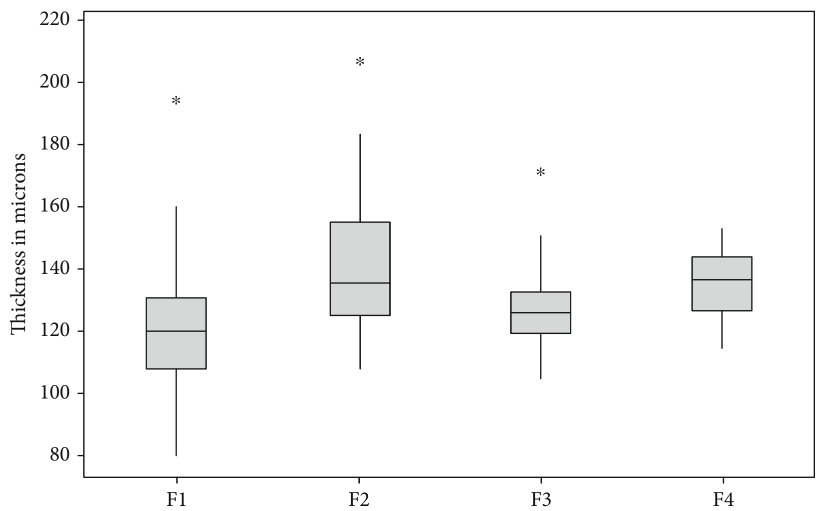

FIGURE 9: Thickness of the CNF film from 1.5 wt. % CNF sprayed on the stainless steel. Thickness variation observed due to the poor handling of spray coated wet CNF film.

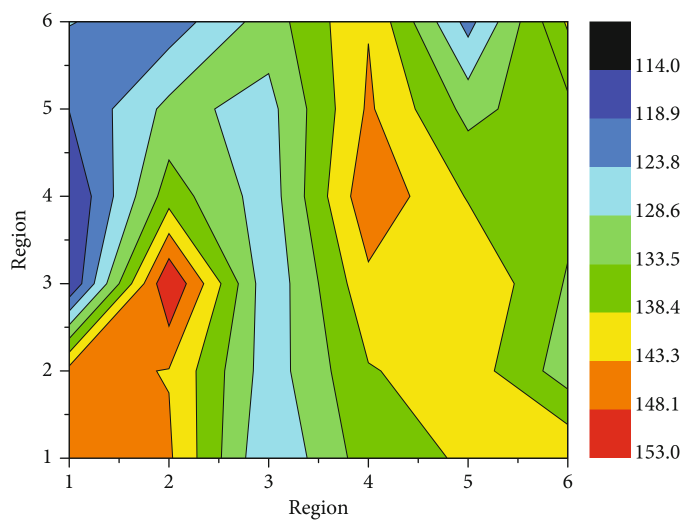

FIGURE 10: Thickness mapping of the 1.5 wt. % sprayed CNF film.

response.

$$\text{Basis weight} = -64.45 + 122.43 * \text{CNF suspension concentration} - 17.28 * \text{velocity of the conveyor} + 0.34 * \text{spray distance}, \quad (3)$$

$$\text{Thickness} = -0.106 + 0.111 * \text{CNF suspension concentration} - 0.000017 * \text{velocity of the conveyor} + 0.002 * \text{spray distance}. \quad (4)$$

The above models are linear models confirming the relation between basis weight and thickness which is responses and CNF suspension concentration as one of the most important input variables.

5. Experimental Confirmation

The spraying of CNF suspension on the stainless steel plate produces a unique film having two surfaces, namely, the rough surface exposed to air side and the smooth surface exposed to stainless steel plate [12].

The spray coated film was highly compact and fibrous matrix which provides barrier against air. The air permeance of the film is <0.003 μm/Pa·s confirming that the film was impermeable [12] and can be used as barrier in packaging films [17]. In addition to that, the CNF film can be used as a substrate for flexible electronics, constructing the drug delivery vehicle [7] and tissue engineering films for various biomedical applications [7]. Figure 7 shows the rough and smooth side of spray coated cellulose nanofiber film. The rough side is very porous and high surface roughness due to the various size distributions of cellulose nanofibrils [18]. The smooth side of CNF film was peeled from the

Table 5: Comparison of ordinary experimental data and data from models.

Input variables	Responses	Experimental	Statistical models
CNF suspension consistency (1.5 wt. %) Velocity 0.32 cm/sec Spray distance 40 cm	Basis weight (g/m^2) Thickness (μm)	108.7 $g.m^{-2}$ 127.85 μm	127.063 $g.m^{-2}$ 133.45 μm

stainless steel surface and partially replicated the smoothness of the stainless steel plate to the CNF film. The SEM micrographs of CNF film reveal the compact structure of the film and various interconnectivity pores on the surface of the film. In addition to that, the complex tortuous pathway was observed on the surface of the film. This helps the barrier performance of the CNF film as an impermeable barrier against air, oxygen, and water vapor [19].

5.1. Basis Weight and Thickness Relation. Figure 8 confirms the linear relation between basis weight and thickness of the CNF film and compared with data collected from CCD experimental design and normal experiments. It shows that the model from CCD design is a good match with the practical experimental data and capacity to use for scaling up the spraying process [14].

5.2. Thickness Mapping. The thickness of the CNF film prepared via ordinary laboratory experiments was mapped and compared with the experiments designed via RSM. The thickness mapping of the CNF films from both conditions is the same and uniform in thickness of all parts of the CNF film. There was no difference between two films in thickness mapping confirming that the films from both conditions are good in uniformity. Figures 9 and 10 show the thickness variation between the CNF films and thickness mapping. Figure 9 reveals that there is a minimal variation in thickness of the films. These variations come due to improper handling of wet film on the stainless steel plates. This can be resolved by the spraying of high concentration of CNF on the stainless steel plates. Therefore, the film of wet suspension is very dense and has resistance to flow on the stainless steel plates.

In comparison with experimental conditions, data are summarized in Table 5. It was observed that the experimental data are exactly matched with the data derived from the mathematical models. The responses obtained from mathematical model for optimized input variable were compared with experimental results, and it is summarized in Table 5. Table 5 reveals that there is a deviation in the value of basis weight of CNF film between experimental and statistical models. The reason for this deviation was the improper handling of wet spray coated CNF suspension on the stainless steel plate. After spraying, the sprayed CNF suspension on the plate was wet and with good fluidity to move around when handling the CNF wet film on the plate and may result in the spillage of CNF suspension from the stainless steel plate. As a consequence, the basis weight and mean thickness of the CNF film could be reduced. It is an operational problem and can be resolved by the reengineering of the experimental spray system.

6. Conclusion

In order to scale up the spraying process for rapid production of CNF film, a successful mathematical model was developed via central composite design in RSM. The main variables involved in the spraying process which impacts the bulk properties of the CNF film were CNF suspension consistency, velocity of the conveyor, and spray distance in the experimental setup. The developed linear RSM model exhibits the relation between the input variables and the output responses for spray coating process. The significance of models was confirmed and investigated via ANOVA. The model was experimentally verified with the optimized input variables, and the affirmation between both the results implies that the model is statistically significant. The relation between basis weight and thickness of the film was linear and verified both experimentally and mathematically. p value > F confirmed that the linear model for spraying process is significant. These RSM studies confirmed that CNF suspension concentration is a strong parameter for controlling the basis weight and thickness of the CNF film than velocity of the conveyor and spray distance from the spray tip to the base in the experimental setup. Given this correspondence, RSM optimization of spraying process can be a good base for scaling up the process for industrial applications.

Data Availability

All the data supporting the results of this study were included in this article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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