Evaluation of water absorption of polyvinyl alcohol-starch biocomposite reinforced with sugarcane bagasse nanofibre: Optimization using Two-Level Factorial Design

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Abstract. Global concern on petroleum based plastics which is non-degradable in our environment has led researchers to develop biodegradable plastics. However, biodegradable plastics have poor barrier properties because of their hydrophilic character of biopolymers. It is known that incorporation of nanocellulose extracted from plant sources to improve barrier properties of biocomposites because of its nanoscopic structure. This study aims to develop biodegradable film based from PVA/Starch and nanocellulose from sugarcane bagasse. Investigation on the effect of sugarcane bagasse nanofibre (SCB-NF) content, PVA content, starch content and water content to the water absorption property of polyvinyl alcohol (PVA)/Starch (S) composites reinforced with sugarcane bagasse nanofibre (SCB-NF) was carried out using Design Expert Version 9.0 with a two-level factorial design (2-FI). Composition of SCB-NF, PVA, Starch and water content was varied from the range of 1 to 9%, 3-8 gram, 1-4 gram, and 80 to 100mL. The nanofibre content was found to have significant effect ($p=0.0099$) on the water absorption of biocomposite film, which is in parallel to the theory of nanofillers to decrease water absorption of biocomposite. As expected, the individual and interaction effects of the compositions can have effect on the water absorption of the biocomposite because of the chemical bonds interaction that they form during the synthesis of biocomposite film.

1.   Introduction

Presently, food packaging industry predominantly use petroleum based plastic materials. The most commonly used petroleum based plastics in food packaging includes low density polyethylene (LDPE), polystyrene (PS), and polyethylene terephthalate (PET). However, the problem with petroleum based plastics in food packaging is their non-degradable nature, and the limited resources [1].One of the attempts to overcome the problem of petroleum based plastic waste is by developing a biodegradable polymer. Polyvinyl alcohol (PVA) is one of the promising polymers since it is the only vinyl type polymer with biodegradable characteristics. However, the application of PVA materials is limited due to the high cost and slow degradation process especially under anaerobic condition [2]. Considering this limitation, PVA is often blended with other cheap biodegradable polymers [3].

The best known renewable resources capable of making biodegradable plastics are starch and cellulose. Starch is well known abundant raw material and relatively cheap biodegradable polymer. Previous studies have reported that blending of PVA and starch can enhance their tensile strength, elongation and toughness [4]. Furthermore, the presence of hydroxyl group in both PVA and starch makes it form a strong hydrogen bond and relatively good compatibility [5]. The disadvantages of

using these PVA/starch blends for packaging is limited due to their poor water barrier properties, hydrophilic nature due to presence of large number of hydroxyl groups. Thus, several studies have proposed the incorporation of nanofillers into PVA/starch blends in order to improve water barrier property [3].

Water barrier property is important in developing film for food packaging. Nanocellulose has been used as nanodimension filler in composite material for reinforcing strength, improving biocompatibility, controlling water sorption, and barrier properties [6]. Nanocellulose is an emerging sector in the food packaging industry as nanoreinforcment for polymers matrix. It is known that nano size cellulose is one of the strategies to improve the polymer composite adhesion and compatibility [7]. Morphology of the nanocomposites, bearing tortuous mixing nanofillers with a macromolecular matrix, leads to higher barrier properties and thus lowering the permeability towards water [8]. In addition, it is known that gas or water molecules penetrate in crystalline ordered structure of nanocellulose, and their dense percolating network held together by strong inter-particle bonds idealize them as good candidate for use as barrier films [9]. This phenomenon was noted by Yoon et al. [10], where water resistance of the composites improved by 70% by addition of nanosized poly (methyl methacrylate-co-acrylamide) particles.

The structure of nanocomposites is important in having remarkable barrier properties. Properties of nanocomposite film depend on many factors. According to Kumar [11], the choice of the filler is important in determining the adhesions between the matrixes and the filler. The strength of bonding will be stronger if the filler is functionalized meaning more availability of OH bonds presence on the surface to bind to the matrix and the homogenize dispersion of the filler in the matrix. Another important factor is the strong adhesion between filler and matrix, which affect the percolation thresholds, and lessen empty spaces (voids) between the polymer matrix interfaces.

By understanding various factors that affect water absorption of nano composite film, the best approach for the optimization of various factors is a 2 level factorial design by Design Expert software. Two- level factorial is designed to study the effects of factors and their interactions simultaneously at two levels over a range of chosen factor levels. The range of loadings chosen was estimated based from previous researchers. Lani, 2014 concluded that with a 7: 3 ratio of PVA and starch is the best combination for optimum properties. Besides, Lani also found that nanocellulose from EFB fibre loading of 5% showed the best water resistance properties of the nanocomposite film. Further studies by Naguib et al. [12], showed that with 5% nanocellulose reinforcement from bagasse fiber, the composite showed the least water absorption.

In developing a new material which is biodegradable and have potential in food packaging this study aims to investigate the effect of factors that determine the water absorption property of the PVA/starch/nanocellulose biocomposite. It is hypothesized that amount of constituents of polymer matrix composites and their interaction may have effect on the final property of water absorption of biocomposite film. Using 2 level factorial design (2-FI) the factors involved are PVA content, starch content, water content (solvent), and nanocellulose loading, and their interaction are optimized that give the least water absorption of the biocomposite film. Nanocellulose is chosen as nanoreinforcment for the PVA/starch blend matrix. Nanocellulose having the characteristics of three hydroxyl groups laterally along surface of cellulose chain, and the high surface area makes it an ideal candidate to functionalize with the polymer matrix of PVA/starch matrix. Furthermore, the advancement of nanocellulose fibre derived from sugarcane bagasse is not widely exploited yet [13].

2. Experimental

2.1. Materials
Sugarcane bagasse originated from green canes (*Saccharum officinarum, Gramineae* family) was collected from night market, Taman Melati, Kuala Lumpur, Malaysia. All chemical reagents were of analytical reagent grade. Polyvinyl alcohol (MW 89,000) with degree of hydroxylation 98 % was purchased from Sigma Aldrich (Malaysia). Pure potato starch was obtained from Systerm (Malaysia). Sodium hydroxide and hydrogen peroxide were purchased from Friendemann Schmidt. Sulphuric acid was purchased from Laboratory reagent.

2.2. Extraction of nanocellulose from sugarcane bagasse

2.2.1 Alkali treatment. Sugarcane bagasse undergoes alkali treatment to remove the lignin and hemicellulose structure from the sugarcane bagasse fibres. The sugarcane bagasse was treated with an alkali solution (2%w/v NaOH) placed in water bath at 80 °C. The solid residue was then filtered and washed several times using deionized water under vacuum pump.

2.2.2. Bleaching. Following pulp disintegration process, the bleaching process comes with aqueous hydrogen peroxide solution in the ratio of 1:1 at 75 °C. The mixture was also washed and filtered several times to ensure proper washing.

2.2.3. Hydrolysis. Bleached fibres were hydrolysed with a 1% v/v H_2SO_4 solution at 80 °C.

2.2.4. Ultra sonication. Hydrolyzed sugarcane bagasse fibres were then sonicated to break down the individualized fibres by using microcentrifuge tubes filled with ice packed in a beaker. Hydrolyzed fibres was filled with distilled water, vortexed, and ultrasonicated with 80 Hertz. Following ultrasonication, the microtube was centrifuged for 20 minutes at 10,000 rpm, and kept in refrigerator at 4 °C.

2.3 Preparation of PVA/Starch/Nanocellulose biocomposites using Two factor Factorial design
Preparing the biocomposite film was done after extracting the nanocellulose from previous experiment. In order to determine the significance of the parameters, experiments were designed in determining the best parameters and amount needed for the preparation of PVA/Starch/nanocellulose.

A two factor factorial design from statistical software Design Expert version 9 (Stat-Ease, Minneapolis, USA) was used to optimize the conditions for the preparation of the composites. The ranges and levels of the factors investigated in this study are shown in Table 1. Process condition was carried out for 4 hours at 130 °C with rotor speed at 450 rpm, and the water absorption test was measured using ASTM D570. Samples of 20 mmx 20 mm in dimensions were immersed in 10mL of distilled water in room temperature for 24 hours. Before immersion, samples were dried in an oven for 50 °C for 24 hour and initially weighed. According to standard test method for water absorption of plastics ASTM D570, following immersion for 24 hour, water accumulated on the sample surface was gently wiped and samples were reweighed for the nearest 0.001. Water absorption was calculated using the following equation.

$$\text{Percentage water absorption}, \% = \frac{\text{final weight-initial weight}}{\text{final weight}} \times 100 \tag{1}$$

Four independent variables were employed by factor factorial design (FFD). The variables used were nanocellulose loading (A), PVA(B), starch(C) , and water (D). The design consists of 19 runs including 3 center points. The response measured is the water absorption.

Table 1: Values and coded levels of each variable in 2 factor factorial design.

Variables		Coded levels	
		Low level (-1)	High level (+1)
A	Nanocellulose loading (%)	1	9
B	PVA (g)	3	8
C	Starch (g)	1	4
D	Water (mL)	80	100

3.   Results and discussion

The nanofibre dimension from sugarcane bagasse was confirmed by FESEM analysis (JEOL JSM 5600). Figure 1 illustrates the even surface morphology of sugarcane bagasse fibres at 100k mmagnification. From the figure, fibres are clearly seen in individualized forms covering the area. The fibres is between 20 -50 nm in diameter and several micrometer in length.
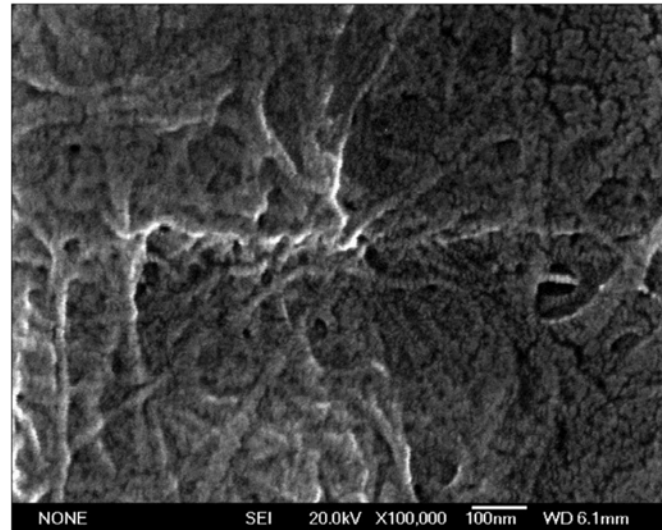

Figure 1: FESEM analysis of nanocellulose from sugarcane bagasse.

Optimization test was done after isolating nanocellulose from sugarcane bagasse. The influence of PVA, starch, solvent i.e. water and nanocellulose loading for the water absorption of the resulting biocomposite film was investigated by 2 level factorial design. An ANOVA analysis (Table 2) is performed to determine the effect of the factors and the interactions. The most significant range of amount for PVA, starch, water, and nanocellulose content were optimized to obtain the optimum mixture for the production of PVA/Starch/Nanocellulose film. Nineteen experiments were conducted as designed by 2 level factorial design while the water absorbing behavior were evaluated and acted as the response to the design. The relationship between the experimental results and independent variables was expressed as second order polynomial regression model which relates the water absorption (response) with all the variables (nanocellulose content, PVA content, starch content, and water content was calculated using design expert and represented as Eq. (2).

$$water\ absorption\ (Y) = \beta_0 + \beta 1A + \beta 2B + \beta 3C + \beta 4D + \beta 12AB + \beta 13AC + \beta 14AD + \beta 23BC + \beta 24BD + \beta 1234ABCD + \in \quad (2)$$

Table 2: The experimental results of the parameters used in 2 factor factorial design.

Trials	Independent variables				Response
	Nanocellulose (g)	PVA (g)	Starch (g)	Water (mL)	Water absorption (%)
1	1	8	4	80	73.3
2	1	3	1	80	64.7
3	9	3	1	100	81.25
4	9	8	1	80	93.1
5	5	5.5	2.5	90	81.57
6	1	3	4	100	86.4
7	1	8	1	100	78.8

8	1	8	1	100	77.4
9	9	3	4	80	83.3
10	1	3	1	80	73.6
11	9	8	4	100	75
12	9	8	4	100	74.2
13	1	8	4	80	78.43
14	9	3	1	100	88.23
15	5	5.5	2.5	90	88.04
16	1	3	4	100	82
17	9	8	1	80	91.37
18	5	5.5	2.5	90	79.24
19	9	3	4	80	76

Table 2 tabulates the experimental values of responses in accordance to experimental design, out of 19 experiments, run 17 achieved the highest water absorption while run 2 had the lowest water absorption. There are many factors that can affect the final properties of the nanocomposite. One of the key parameters in achieving an improved performance of nanocomposites film is by having an evenly distributed dispersion of the materials. Processing PVA/Starch nanocomposites filled with nanocellulose is like mixing the materials with different scale dimension, which depends on many different interacting factors including the strong adhesion/bonding strength of the nanofiller with the matrix, type of material used, and geometry and orientation of the fibres [11].

Barrier properties of nanocomposites are sensitive to the microstructure and the interface between the matrix and filler. The interactions between the organic and inorganic phase at the interface determine the final behavior of the barrier performance of the composites [14]. The presence of void or free volumes in nanocomposite film increases the diffusion of water through these empty areas. Unlike oxygen molecules, water not only interacts with themselves, but also with the polymer matrices where they can form hydrogen bonds. The ability to form strong rigid structure is important in resisting water molecules to diffuse in the film.

By nature, PVA, starch, and nanocellulose are polar molecules that are hydrophilic, meaning water absorbing material. However, with the concept of nano scale dimension of fibres that has high surface area to volume ratio makes the nanocomposite film to have complex strong rigid network. This strong network may be attributed to the chemical interaction that involves PVA/starch/nanocellulose in that strong hydrogen bonding might occur among the nanocellulose structure of three hydroxyl groups on the surface with the structure of the starch and PVA in presence of water. The aspect ratio in terms of length to width ratio of the nanocellulose also plays a role in determining how strong is the forces between the chemical structures.

Table 3: Analysis of variance (ANOVA) for quadratic model of water absorption of PVA/S/SBNF.

Source	Sum of squares	DF	Mean squares	F-value	p value
Model	713.66	6	118.94	7.78	0.0014*
A	142.92	1	142.92	9.35	0.0099*
B	2.34	1	2.34	0.15	0.7024
C	24.55	1	24.55	1.61	0.2291
D	5.62	1	5.62	0.37	0.5557
AC	315.77	1	315.77	20.66	0.0007*
AD	222.46	1	222.46	14.55	0.0025*
Residual	183.45	12	15.29		
Lack of fit	25.63	2	12.81	0.81	0.4713
Pure error	157.82	10	15.78		
Cor total	897.11	18			

Standard deviation= 3.91, mean= 80.31, R-square= 0.7955, and adeq. precision= 9.727
*p value < 0.05, indicates significant factor

Table 3 summarizes the statistical results obtained after analyzing by Design Expert software. Analysis of variance (ANOVA) was performed to find the effect and interactions of each variable. Both F-value and p-value were used to confirm the significance of each variable. The greater F- value indicates the estimated variation data is real and p values less than 0.05 suggested that the variable is significant. Based from Table 3 above, it can be seen that the model had a significant effect on the responses. The model F-value 7.78 and p-value 0.0014 implies the model is significant. The variables A, B, C and D are the independent variables nanocellulose, PVA, starch, and water content, respectively. AC and AD represents the interaction effects. As it is observed, the lowest p-values are obtained by nanocellulose loading interaction with amount of water (AD), followed by nanocellulose loading (A), and nanocellulose loading interaction with amount of starch (AC). Insignificant lack of fit, with p-value of 0.4713 is desirable and implies that the test is insignificant relative to pure error. High values of R^2 on the model of nanocomposite film (0.7955) clarifying the relation involving the response with the independent variables is highly correlated and that the model is highly significant.

Pareto chart in Figure 2 was used to distinguish between the factors that determine the importance of an effect and the greatest effects, considering the line on the chart at t-value of effect. There are two different t- value of effects which are Bonferroni limit line (t-value of effect= 3.294) and t-value limit line (t-value of effect= 2.200). Coefficients with t-value of effect above Bonferroni line are designated as certainly significant coefficients, and coefficients with t-value of the effect between Bonferroni line and t-limit are termed as likely to be significant, and coefficients below the t limit line is statistically insignificant.   As depicted from the graph, factor A (nanocellulose loading) shows to have the highest effect on the water absorption, followed by D (amount of water), and B (amount of PVA). This shows that increasing number of nanocellulose will decrease the water absorption of the nanocomposites. The negative effects of the factors are the interaction AC (interaction of nanocellulose loading and starch), AD (interaction of nanocellulose loading and water), and C (starch).
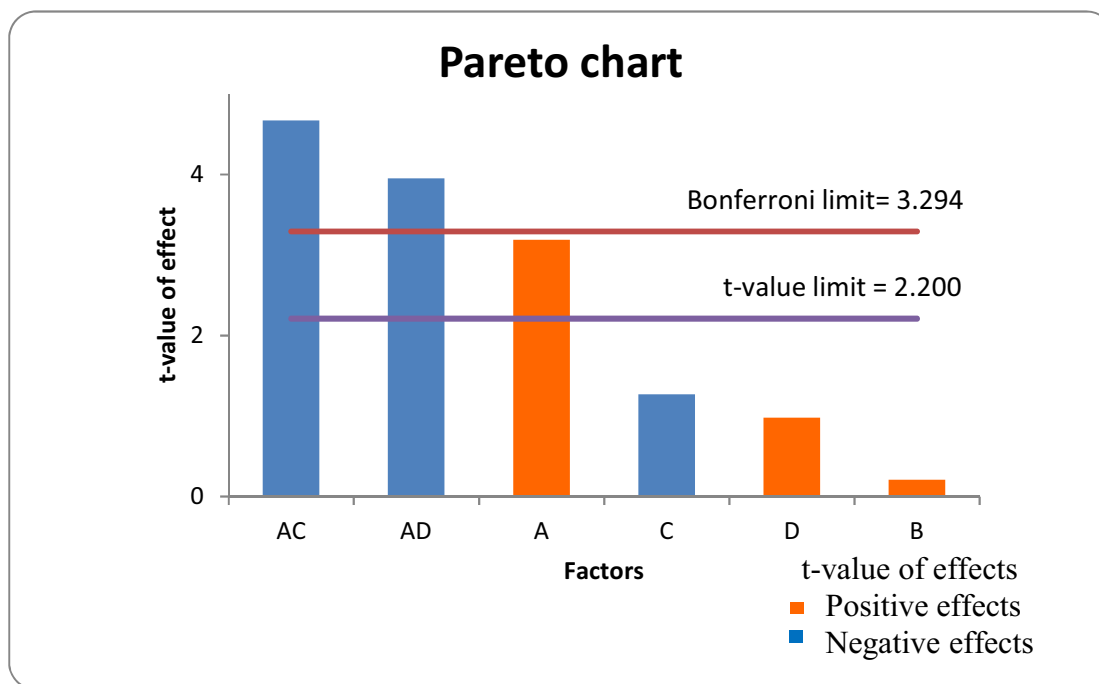

Figure 2: Pareto chart of four factors and their interaction effects on water absorption properties of PVA/starch/nanocellulose film; orange-coloured bar indicates positive effects of factors; the blue-coloured bar indicates negative effects of factors.

The reason for the negative effects of the interactions between nanocellulose with water and starch could be attributed by the chemical structure of each constituents and nanocellulose fibre morphology. Nanocellulose fibres morphology plays an important role in the mechanism of water absorption of film. The nanofibrils that form complex dense structure and smaller pores in nanometer range, increases the tortuosity within the film and thus decreases the permeability within the film [15]. Tortuosity is intended to describe the effort of a permeate e.g. oxygen or water in transmitting through the blocking material [16]. Nair et al., [17] reported that nanocellulose has a strong reducing effect on the water vapour diffusion due to its nanoscale dimension and rigid framework compared to cellulose fibres. Chemical structure of starch also plays a role in water absorption of the composite; this is supported by Zou et al., [18] reported with an increased content of starch, water absorption of PVA/starch composites increased. This may be attributed to the structure of starch with higher presence of OH group thereby increasing the site for water to bind to the OH groups.
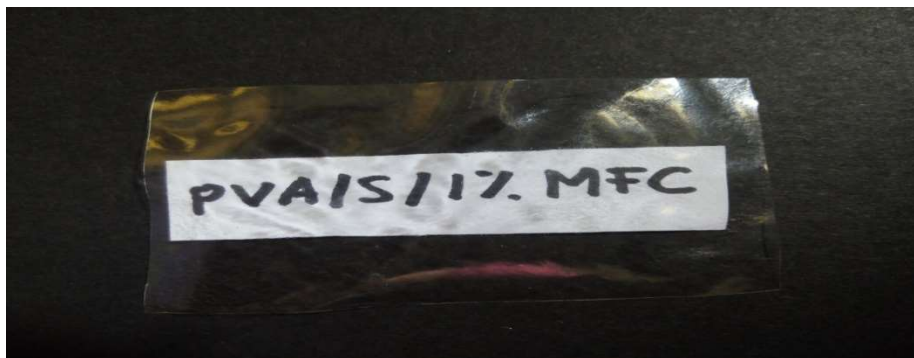
Nanocellulose fibres bearing three hydroxyl groups present on the surface which makes it an interesting property. The relation between nanocellulose fibres and water absorption of the biocomposites has been reported by other researchers as well. The structures of the fibres play a role in diffusivity of the water to pass through. Kiani et al., [19] studied PVC reinforced with pulp fibres and concluded that water absorption increases with increasing fibre content due to the porous tubular structure, accelerating the diffusion of water into the polymer membrane. This shows porous structure of the fibres might accelerate the water permeability to the material. In contrast to a study by Abdul Khalil et al., [20] that incorporation of two fibre; woven jute fibre and EFB fibre in an epoxy composite film decreased the water absorption of the hybrid composites. This may be due to the packaging and arrangement of these fibres that filled up the voids during the formation of the composite film. As compared with only EFB fibres/epoxy composite the water absorption increases, due to presence of higher EFB fibres voids (8.6%) as compared to jute fibre (2.6%). Thus another factor that affects water absorption might be due to the void content in the polymer matrix. The greater voids presence, more water is absorbed into the composite film. Pothan [21] explained the water absorption of the fibres mechanism may be due to formation of micro-channels, providing a way for water to pass through the pores on the surface of the fibres. This shows those fibres with structure that gave higher amount of voids leads to higher amount of water absorption.

Physical structure of nanocellulose in terms of crystallinity also affects the water diffusion into the film. This statement is supported by Dufresne [22] where he reported that water transmission is affected by the degree of crystallinity, as crystalline phases block more transmission than amorphous phases. Sjöholm [23] also supported this statement, that the greater the degree of crystallinity the higher the barrier properties. The concept of having a rigid network that pack perfectly is like modelling the polymer chains forming a concise structure that impermeates the penetrating molecules [24]. Other factors such as lumen size, and adhesion of matrix fibre also effect water absorption behavior of nanocomposites [25]. Svagan et al. [26] studied the influence of nanocellulose on water sorption of starch/nanocellulose polymer. Nanocellulose was found to have strong effect on the diffusivity of water attributed by characteristics and geometry of cellulose, rigid fibre netwroks, and strong interactions between nanocellulose and starch matrix.

The goal of biocomposite film was to improve water resistance of the film, therefore the target values of response of water absorption percentage was the lowest values obtained from the experimental results. The acceptable values of desirability function were the values close to 100%. In this study, the water absorption property of the biocomposite with 1 % nanocellulose, 3 g PVA, 1 g starch, and 80 mL water had 83 % desirability. These levels of independent variables yield the lowest responses of water absorption with 69.41 %.

Table 4: Results for target values of the response that yields the lowest value of water absorption of nanocomposites film.

Constituents amount	Nanocellulose (%)	PVA (g)	Starch (g)	Water (mL)	Water absorption (%)	Desirability (%)
	1%	3	1	80	69.41	83

Figure 3: Photograph of optimized PVA/starch/nanocellulose nanocomposites placed on a background paper for demonstrating transparency.

Validation experiments were carried out to verify the accuracy of the model. Three set of experiments were carried out by using the optimal values of the factors. The experimental result showed water absorption of 69.9; while DOE software predicted the response to be at 69.41 %. It was found that experimental result differed with only 0.7 % error more than the predicted response. This reflects the accuracy 99.3% between the experimental and predicted results. This shows that 2 level factorial design model is accurate in predicting the water absorption of biocomposite film.

Table 5: Results of the validation experiments for water absorption of PVA/starch/nanocellulose film.

Validation	Nanocellulose (%)	PVA (g)	Starch (g)	Water (mL)	Water absorption		% Error
					Predicted	Actual	
	1	3	1	80	69.41	69.90	0.7

4.   Conclusion
This study showed that statistical design can be used in evaluating the water absorption properties of the PVA/Starch/Nanocellulose thin film composites. Nanocellulose loading was identified by 2 level factorial designs as an important parameter for improving water absorption of nanocomposite film. Nanocellulose loading showed positive effect, whereas amount of starch, water, and PVA had a negative main effect. This suggests that the optimized content of PVA of 3g, content of starch 1g, content of water 80mL, and nanocellulose loading of 1% could be used for further characterization. This optimized condition shows strong adhesion and interfacial bonding between polar molecules of PVA starch blend and nanocellulose could be achieved resulting in minimum water absorption (64.1%). Optimization of these variables resulted in reduced cost, time, and material. Further studies could be focused on modeling the mechanism of water absorption studies of nanocomposites film.

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