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Parameter optimization: Effect of Humidity and Fabrication Process on Flexural Strength of Kenaf/Polylactic Acid Biocomposite

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ABSTRACT

Biocomposite from kenaf reinforced polylactic acid (Kenaf/PLA) has the potential to replace a synthetic reinforcement and matrix in polymer composite applications. Kenaf is well known as a natural fiber that can be replaced with synthetic fiber and reinforced with synthetic resin, whether thermoset or thermoplastic. An increase in environmental impact drives increased usage of a biodegradable polymer such as PLA in the application of biocomposite. However, moisture uptake from the humidity and elevated temperature exposure during fabrication of composite is mostly a factor that affects the properties of biocomposite. Therefore, this study aims to optimize the factors consisting of humidity exposure, temperature and time holding during hot press using the design of experiment (DOE) by Box-Behnken Design (BBD) approach. The kenaf/PLA was exposed to the humidity range from 40% up to 80% before undergoing composite fabrication. Then produce the composite using a hot press molding where the parameter consists of elevated temperature (range from 160 °C up to 200 °C) and time holding (range from 3 minutes up to 10 minutes) that possibly affects the mechanical properties of kenaf/PLA. These three factors were evaluated based on optimizing maximum results on flexural properties, where all the combination factors were assessed using DOE. Using analysis of variance (ANOVA) revealed that humidity exposure and hot press temperature are essential factors affecting kenaf/PLA flexural strength. The results indicated that the selected humidity and hot press parameters were 40 %RH, 160 °C press molding temperature, and a 3-minute heating duration for optimal flexural strength. The parameter will obtain 111.61 MPa of flexural strength. Implementing the chosen factor can produce the kenaf/PLA biocomposite with an optimum flexural strength of 111.61 MPa.

Keywords: Biocomposite; Kenaf; Polylactic acid, flexural properties

INTRODUCTION

Biocomposite is an alternative material for reducing pollution and depleting petroleum-based polymer sources. The good properties of natural fibers, such as low specific weight ratio, low cost, and thermal solid and acoustic insulating capabilities compared to synthetic fibers, further contribute to the use of biocomposite (S. Christian and S.Billington 2009; F.Vilaplana et al. 2010). Biocomposite has increased use in the automotive and consumer products industries as an alternative material (Md. S. Islam et al. 2020; F.Hassan et al. 2017; M. Murariu and P. Dubois 2016).

Hence, kenaf reinforced polylactic acid (kenaf/PLA) is a possible biocomposite combination to replace synthetic composite. Kenaf as a reinforcement to provide the strength and stiffness, and PLA as a matrix to distribute the load to the kenaf fiber and hold it in a biocomposite form (R.B.Yusuf et al. 2016).

Even though kenaf/PLA biocomposite offers numerous advantages over synthetic composite, some critical difficulties still need to be resolved, particularly during biocomposite manufacture. Bioplastics offer significant unsolved biocompatibility, strength, heat resistance, performance, and processing ease compared to petroleum-based composites. Because PLA is thermoplastic and requires heat to mold into the necessary shape, hot press molding is the best method for fabricating laminated unidirectional kenaf yarn/PLA(O.A. Khondker et al. 2006; S. Shibata et al. 2007; S.Ochi 2008; G.Ben et al. 2007). Meanwhile, the fundamental issue with kenaf yarn/PLA is heat resistance, which means that when heat is present, the qualities of the biocomposite are reduced (S.Ochi 2008; N.A.A. Hassan et al. 2019; M. W. Czabaj and B. D. Davidson 2015). PLA's thermal characteristics are notorious for their low heat resistance when exposed to heat.

Reduced factors will influence the biocomposite and lead it to fail. Furthermore, PLA has low thermal properties, which are known to the range to 60 °C in the glass transition temperature. Exposure of composite components to elevated temperature can cause degradation of the reinforcement and depolymerization of resin (M. S. Meor Sha and R. Zulkifli 2019).

Instead of improving, the main drawback of biocomposite application is environmental exposure (O. Faruk et al. 2012; N. Graupner and J. Müssig 2011). This weakness causes an impact on the properties of biocomposites. Among the factors, exposure to humidity and temperature most affected the mechanical properties of natural fiber (O. Faruk et al. 2012; N. Ezekiel et al. 2011; M. S. Huda et al. 2008; C. A. Fuentes et al. 2016). A study conducted by D. Zhang (2013) showed at 70 %RH starts to drop sharply in flexural properties and interfacial shear strength of the natural fiber, and the reduction continues until 90 %RH. To produce optimum properties of biocomposite, the selected humidity exposure of reinforcement should be maintained before undergoing composite fabrication.

This study focuses on biocomposite material that combines unidirectional kenaf yarn fiber reinforced with polylactic acid (PLA). The objective of this study is to observe the flexural behavior of the biocomposite while the reinforcement exposes to selected humidity and thermal during fabrication using the hot press method. Hence, optimization parameters have been used to prevent strength reduction due to thermal degradation (S.Ochi 2008; I. Tharazi et al 2017). This study came out with suitable humidity and the unidirectional kenaf/PLA biocomposite fabrication factor. Humidity has varied from 40 %RH, 60 %RH, and 80 %RH, with temperatures ranging from 160 °C up to 200 °C and holding times ranging from 3 min to 10 min. In the end, the relevant factor that gives the optimum mechanical properties of the biocomposite was founded.

METHODOLOGY

MATERIALS

Unidirectional kenaf reinforcement was supplied by Innovative Pultrusion Sdn. Bhd. Unidirectional kenaf in yarn was treated with a 5% NaOH solution to increase the adhesion. The PLA matrix is shown in Figure 2 in the form of granules of grade Ingeo biopolymer 2003D supplied by Nature Works.

COMPOSITE FABRICATION

Unidirectional kenaf fiber was conditioned in a humidity chamber, ranging from 40 %RH to 80 %RH, for 24 hours before fabricating the composite panel. Then, kenaf/PLA biocomposite was fabricated using the hot press machine with 30%wt of the kenaf as reinforcement. Figure 1 showed the Differential Scanning Calorimeter (DSC) result when the sample of PLA was melted and then cooled at 0.1 °C/min. As seen in the Figure 1 glass transition, Tg occurred around 65 °C, and melting occurred with endothermic peaks starting at 153 °C. During these peaks and forward, the PLA is in the melting phase; it was a suitable temperature for mixing with kenaf during the pressing process. The temperature range used by other research in fabricating composites reinforced bio-resin is based on table 1. Therefore, from the DSC and previous study, the compression was used with the variation of the parameter; 160 °C up to 200 °C of temperature and time for 3 to 10 minutes at a constant pressure of 3 MPa.

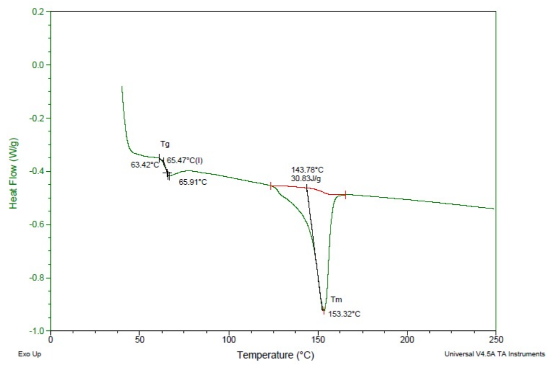


FIGURE 1. DSC results of PLA

TABLE 1. Parameter of temperature and holding time to fabricated composite reinforced with PLA from previous research

Material	Temperature	Holding Time	Ref.
Kenaf/PLA	170 °C	6 min	A.Manral et. al 2020
Kenaf/PLA	190 °C	5 min	I.Tharazi et. al 2017
Kenaf/PLA	185 °C	8 min	W.Jaafar et. al 2012
Kenaf/PLA	150 °C	5 min	N.Ibrahim et al 2010
Kenaf/PLA	160 °C	10 min	Ochi 2008
Kenaf sheet/PLA	185 °C	20 min	G.Ben et al. 2007
Kenaf/Corn starch Bamboo/Corn Starch	160 °C	10 min	Shibata et al. 2008
Jute/PLA	170 °C	5 min	B.Goriparthi et al. 2012

FLEXURAL

Flexural or three-points bending test was conducted according to ASTM D-790 standard, were performed using a Universal Testing Machine by Instron. The average dimensions of the specimens were 12 mm width 2.5 mm of thickness with 45mm of support span as in Figure 2. All the results were taken as the average value of 5 samples replication.

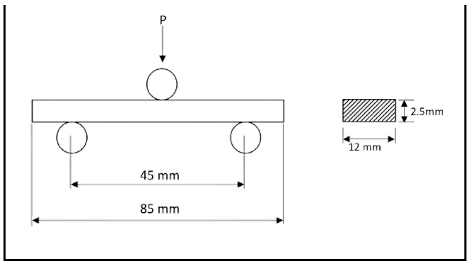


FIGURE 2. Dimension of the sample for flexural test

OPTIMIZATION THROUGH RESPONSE SURFACE METHODOLOGY

Response Surface Methodology (RSM) was used to study humidity and hot press parameters using Box Behnken Design (BBD) for optimization study. The work was completed using Design Expert software version 13, with a total of 17 experimental runs were carried out, including three replicates for each run. The influence of humidity exposure to kenaf and process factors on kenaf/PLA biocomposite properties was investigated using an experimental approach. The selection of humidity and hot press parameters and levels is shown in Table 2 below.

TABLE 2. Process variables and their actual values for the coded values in the experimental design

Parameter	Level		
	Low (-1)	Medium (0)	High (+1)
A: Temperature (^{o}C)	160	180	200
B: Humidity (%RH)	40	60	80
C: Holding time (min)	3	5	10

RESULT AND DISCUSSION

The experimental design and results obtained at various combinations of humidity and hot press parameters are shown in Table 3. To evaluate the influence of parameters on flexural strength of the unidirectional Kenaf/PLA biocomposites, the main effect and interaction of process parameters on response, analysis of variance (ANOVA), and regression analysis were studied. The best combination of process parameters was determined based on the contour analysis. Furthermore, a mathematical model for the variation of flexural strength with significant process factors was generated using multiple linear regression analysis.

TABLE 3. Experimental Design Runs and response result for Unidirectional Kenaf/PLA

Runs	Level			Response
	A: Humidity (%RH)	B: Temperature (^{o}C)	C: Holding Time (min)	Flexural Strength (MPa)
1	40	180	3	105
2	80	180	3	51.6
3	60	160	3	85.8
4	60	180	7	82
5	40	180	10	81.8
6	40	160	7	113
7	60	160	10	85.7
8	60	180	7	82
9	80	160	7	44.5
10	60	180	7	82
11	80	180	10	76.05
12	60	200	10	51.2
13	60	200	3	66.1
14	60	180	7	82
15	80	200	7	32.6
16	60	180	7	82
17	40	200	7	80.7

ANALYSIS OF VARIANCE (ANOVA)

Table 4 shows the ANOVA table that calculated and summarized the experimental factor. The information can conclude the influence of each factor obtained from experiment results in Table 3.

TABLE 4. ANOVA linear model for flexural strength

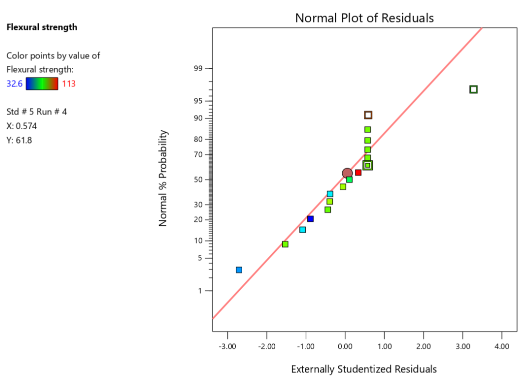
Source	Sum of Squares	df	Mean Square	F-value	p-value
Model	5094.36	3	1698.12	12.82	0.0004
A-Humidity	3861.14	1	3861.14	29.15	0.0001
B-Temperature	1210.32	1	1210.32	9.14	0.0098
C-Time	22.90	1	22.90	0.1729	0.6843
Residual	1722.00	13	132.46		
Lack of Fit	1722.00	9	191.33		
Pure Error	0.0000	4	0.0000		
Cor Total	6816.36	16			

The Model F-value of 12.82 implies the model is significant. There is only a 0.04% chance that this large F-value could occur due to noise. P-values less than 0.0500 indicate model terms are significant. In this case, A B are significant model terms. Values of P-Value show greater than 0.1000 indicate the model terms are not significant.

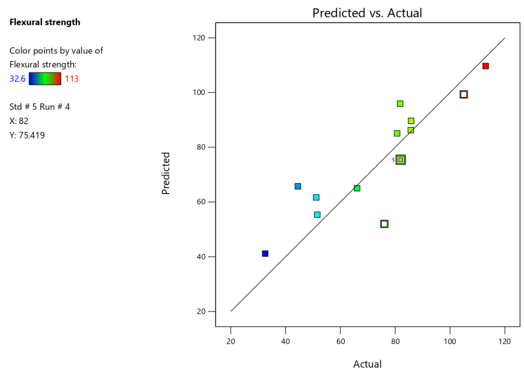
By applying multiple regression analysis on the experimental data, the equation in terms of actual factor (A; humidity, B; temperature C; holding time) can be used to predict the maximum value of flexural strength for given levels of each factor. The equation obtained as follows;

$$\text{Flexural Strength} = 75.66 - 21.97*A - 12.30*B - 1.68*C$$

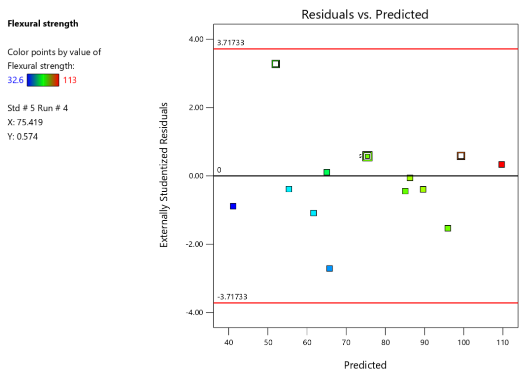
This model is supported by diagnostic plot including normal plots, residual vs. predicted, predicted vs. actual, and residual vs. the run, as shown in figure 6. Figures 3(a) and (b) verified that normal plots and predicted vs. actual are nearer to the straight line. Hence, it offers a good correlation that exists between the developed model and the experimental values. Figures 3(c) and (d) below also strengthen the results, showing that the residuals are running positively and negatively with the experiment's efficacy.



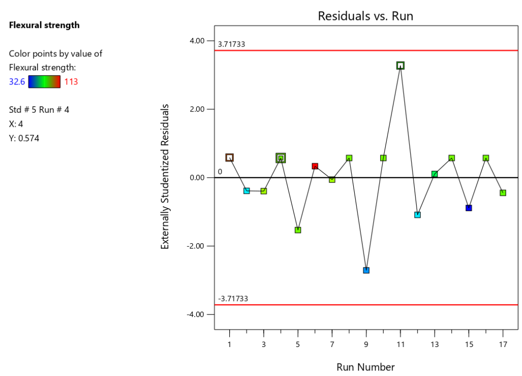
(a)



(b)



(c)

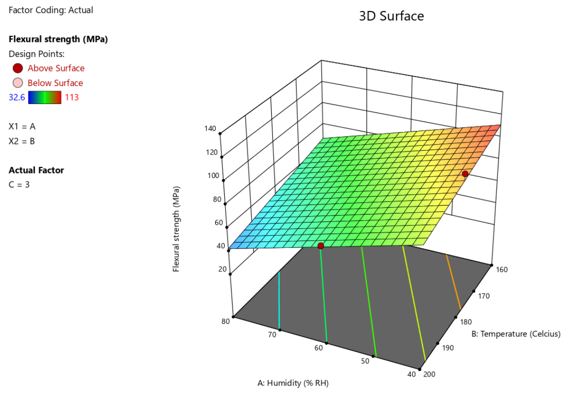


(d)

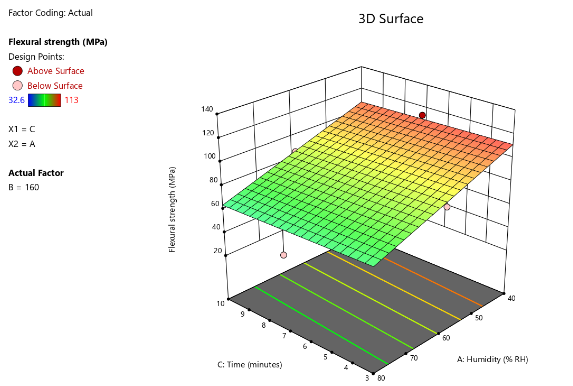
FIGURE 3. Diagnostic Plot (a) Normal Plot Residuals (b) Predicted vs. actual (c) Residuals vs. Predicted (d) Residuals vs. Run

OPTIMIZATION OF THE FLEXURAL STRENGTH OF UNIDIRECTIONAL KENAF/PLA

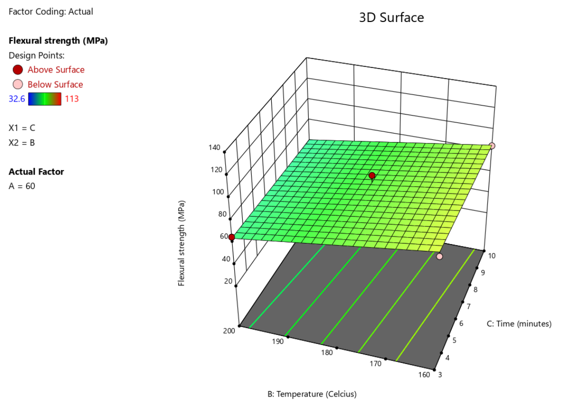
Optimizing the parameters that influence the flexural strength of the biocomposite materials generates the response surface plots from the regression model and optimizes the variables. For this purpose, the humidity, temperature, and holding time were set in range while the output(flexural strength) was placed at the maximum level. This will show the effects on the modulus by the humidity, pressing temperature, and the holding time on the flexural strength of the composite.



(a)



(b)



(c)

FIGURE 4. Response surface for flexural strength (a) Humidity vs. Temperature (b) Humidity vs. Time (c) Temperature vs. Time

From Figure 4, the contour showed the relationship between flexural strength and three factors of humidity, temperature, and holding time. Figure 7(a) shows the relation between humidity vs. temperature, whereas flexural strength improved when using low humidity at 40 %RH with the elevated temperature at 160 °C. Then, the connection between humidity and temperature, as shown in figure 7(b) 3 minutes of holding time, obtained the maximum result of flexural strength at the same humidity. In figure 7(c), the relation between temperature and holding time shows a slight difference between the shorter time at lower temperature and vice versa. However, holding time causes exposure to the elevated temperature is takes longer also affects the flexural strength. The suggestion parameter agrees with this result by Table 5, where the optimized parameter for the maximum range of flexural strength is obtained.

TABLE 5. Optimized parameter for maximum flexural strength

Humidity %RH	Temperature °C	Holding time min
40	160	3

Summary from table 5 shows the optimized hot press parameter for flexural properties with the optimal set of processing parameters kenaf/PLA was 40 %RH of exposure humidity with 160 °C molding temperature and 3 minutes of holding time. It was observed from the analysis that the temperature of press molding and humidity are the main significant affecting the flexural strength. Still, the time depends on these two factors and can be selected based on the desired range of flexural properties. The result showed the effect of humidity also agreed by C.A Fuentes et al. 2016, kenaf as a hydrophilic material, meaning it can absorb a lot of water proportional to increasing humidity. In addition, during fabrication, instead of mixing the composite presence of temperature can be attributed to oxidation, dehydration, and decreased thermal stability of the components. Hence, if the manufacturing process occurs in a moisture environment and uses the elevated temperate can cause increases the probability of failure during bending.

CONCLUSION

The objective of this paper was to use RSM and a Box Behnken design (BBD) to investigate the mechanical properties of a kenaf/PLA biocomposite. The ANOVA findings indicate that the factors significantly impacted the flexural strength.

As shown in the experimental results, the humidity of kenaf and hot press processing temperature are the significant parameters influencing the flexural strength of the kenaf yarn/PLA biocomposite material. Even though holding time showed no significance to the flexural strength, it was observed that interaction between holding time and other factors gradually increased when using lower holding time.

After running the experiment and the analysis, ANOVA was give a suggested combination of the parameters to obtain the higher flexural strength are at 40 %RH of humidity exposure, 160 °C of hot press temperature, and 3 minutes of holding time.

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DECLARATION OF COMPETING INTEREST

None

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