Application of Response Surface Methodology for Parameters Optimization in Hot Pressing Kenaf Reinforced Biocomposites

Izdihar Tharazi[1, 2], Abu Bakar Sulong[1], Farrahshaida Mohd Salleh[2], Abdul Hakim Abdullah[2], Nur Farhani Ismail[1]*
[1]Department of Mechanical and Material Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia

[2]Faculty of Mechanical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia
**izdihar.tharazi@gmail.com*

ABSTRACT

Processing parameters of composite materials have a significant influence on the mechanical properties of the final product. Typically, the most important processing parameters are temperature, heating time and pressure but there are cases where more or fewer parameters are involved depending on the type of the materials, fabrication techniques including the capabilities of the processing equipment. The traditional approach in experimental work is costly and time consuming due to evaluation of multiple dependent variables. In this paper, design of experiments (DOE) technique based on statistical analysis has been employed to optimize hot press parameters on tensile properties of unidirectional kenaf fibres reinforced polylactic-acid (PLA) composite. The kenaf/PLA composite samples were fabricated using hot press method by stacking the aligned kenaf fibres with the PLA films. The stacked materials were hot pressed at varying processing parameters specifically the temperature, pressure and heating time. The Box-Behnken Design (BBD) through Response Surface Methodology (RSM) was employed to identify the cause and effect of relationship between the processing parameters with the composite's tensile strength and Young's modulus. Results from ANOVA showed that all three parameters and interactions significantly affect the composite's tensile strength. For Young's modulus, pressure and heating time are the significant parameters. Optimal processing parameters for composite

ISSN 1823-5514, eISSN 2550-164X

Received for review: 2020-09-15
Accepted for publication: 2020-10-08
Published: 2020-10-15

fabrication for optimum tensile properties are at 200 MPa, 3MPa and 7 minutes.

Keywords: *kenaf fibres, polylactic-acid composite, tensile properties, optimization, response surface methodology*

Introduction

Nowadays, there has been an increasing demand from industrial around the world for natural fibre polymer composites due to the advantages they offer in terms of biodegradability, renewability as well as reduced emission of green house gas. The most important and widely-used natural fibres is plant-based which can be derived from the seed, bast, leaf, stalk, wood and fruits of a plant [1]. Fibres obtained from the bast are the strongest and are often used as reinforcement in polymer composites for automotive application [2]. Some of the plants that can be used to provide bast fibres are kenaf, flax, hemp and jute. Kenaf or known as Hibiscus Cannabinus L. is from the Malvaceae family and has existed since 4000 years ago. Kenaf is a short-term plant and can grow up to a height of five meters and has a short maturity of three to six months under wide range of weather [3].

Green composites produced by a combination of natural fibres and bio-polymers are gaining more attention with new regulations, environmental concerns and the need for sustainability. One of the most promising bio-polymersis polylactic-acid (PLA) due to its biodegradability and eco-friendly material with comparable properties to those of synthetic polymers such as polypropylene (PP), polyethylene(PE) and polystyrene(PS) [4]. PLA can be processed by injection moulding, compression moulding, extrusion, thermoforming, or spinning in a similar manner to synthetic polymers [5]. PLA exhibits good mechanical performance, good aesthetics, biocompatibility and easy processing with most equipment [6]. It is usually used in consumer packaging, biomedical, automotive as well as electrical applications [7].

Processing parameter is one of the important things to consider in order producing composites with good mechanical and physical properties [8]. For every types of polymer, the processing parameters are different, depending on the structure, type and shape of the material including the capabilities of the processing equipment [9]. Numerous studies have successfully carried the processing and fabrication of biocomposites [8, 9, 13, 14], but there is still lack of studies on the optimization of biocomposites processing and formation methods, especially for biopolymer reinforced with natural fibres. According to Mukherjee and Kao [14], the processing conditions directly affect the mechanical properties of the fibre composites. In order to obtain the optimum composite properties according to its intended application, the suitable processing parameters must be cautiously selected. In the traditional approach, one factor is designated as the dependent variable while keeping all the other factors fixed; the experiments are repeated with different factors as the dependent variable each time. This method is time consuming and does not produce reasonable results suitable for a wide range of experimental settings [15]. Therefore, design of experiments (DOE) technique based on statistical analysis has been introduced so that an effective result and valid conclusions can be produced from experimental data.

Response surface methodology (RSM) is one of the available DOE techniques to optimize the operating parameters or conditions in a system [16]. RSM is particularly applied in circumstances where a number of potential input variables affect a performance or characteristic of the process or product [17]. Kumar and Balachandar [18] used the Box-Behnken Design (BBD) technique and demonstrated that the optimum hot press parameters for high flexural strength of glass reinforced with polypropylene (PP) composites is at high temperature but with low holding time. Wu [17] also used BBD and found that the optimization parameters for moulding conditions for foam material and cup depths of mould are moulding temperature and dwell time length. Kiran et al. [19] used Taguchi method and found that press forming temperature has the most significant effect on the composite’s mechanical properties. Hence, this paper attempts to study the effects of hot pressing method on tensile strength of unidirectional long kenaf fibre/PLA composite.

In our study, Box-Behnken design (BBD) was implemented as experimental strategy to design and analyze the influence of the processing parameters. The BBD was chosen among the other RSM methods because fewer number of experimental runs is required than the Central-Composite design (CCD) and full-factorial design (FFD). In addition, quadratic model fitting is improved because of the final matrix is completed with several replications of the central point as well as there are no extreme values in experimental points in all of the factors. The main objective was to identify the optimized processing parameters for fabrication of kenaf/PLA composite with the highest tensile properties and simultaneously to create a mathematical model which could predict the variation of tensile properties within the range of the investigated parameters.

Materials and Method

Materials

Continuous long fibres from kenaf bast were supplied by Innovative Pultrusion Sdn. Bhd. Kenaf fibre bundles were then combed and cut to the desired length of 175 mm (same length with the hot press mould). PLA polymer in form of pellets and microfine powders with 20 μm size were purchased from Shenzun Esun Ltd. China. Kenaf fibres, PLA pellets and powders were dried in a vacuum oven for 10 hours at 80 ºC before usage to remove any moisture. The fibres weight content was set to 50% wt with 50% wt matrix and used throughout the experiment [20]. Based on preliminary study, ratio of polymer was fixed to 70:30 portions for pellets and powders.

Composite Fabrication and Characterization

Prior to fabrication of the composites, the PLA pellets were pressed at 190ºC with 5 MPa pressure for 7 minutes to produce PLA film. Kenaf fibres were placed and shaken manually in a container with PLA powders to improve adhesion and distribution of the fibres. Next, the fibres were evenly aligned and stack between PLA films in a hot press mould. Stacked materials were pressed at temperature range between 190 °C to 210 ºC, pressure range between 3 to 7 MPa and heating time of 5 to 15 minutes. A similar cooling time approach was employed to ensure the cooling and solidification of the final composites. A shearing machine was used to cut the composite plates into rectangular shape for tensile tests. The sample thickness was 2 mm with 115 mm length and 20 mm width.

ZwickRoell tensile machine with 100 kN load was used to performed the tensile tests. An average of five samples were tested according to the ASTM D3039 standard with 5 mm/min cross-head velocity.

Response Surface Methodology with Box-Behnken design

A response surface methodology (RSM) using the Box-Behnken Design was utilized for the experimental investigation. A total of 15 experimental runs including 3 replicates of centre points were performed. The settings of the parameters were as shown in Table 1. These parameter settings were considered based on a preliminary study. Design Expert software version 10 (US, Stat-Ease Inc.) was used as the tool for data statistical analysis.

Table 1: List of parameters and levels

Parameter (Unit)	Levels		
	Low (-1)	Medium (0)	High (+1)
A: Temperature (°C)	190	200	210
B: Pressure (MPa)	3	5	7
C: Heating Time (mins)	5	10	15

The influence of these three parameters on the tensile properties can be approximated by following second-order polynomial model equation:

$$\hat{Y} = \beta_0 + \sum_{j=1}^{k}\beta_j x_j + \sum_{i<j}\sum\beta_{ij}x_i x_j + \sum_{i=0}^{k}\beta_{jj}x^2_j + e \tag{1}$$

where $\hat{Y}$ is the predicted response value (tensile strength and young modulus), β_0 is the model constant, β_j is the linear coefficient, β_{jj} is the quadratic coefficients, β_{ij} is the interaction coefficient, x is the independent variables in coded values, and e is the experimental error.

Results and Discussion

Experimental results

Table 2 shows the design and results obtained from the experimental runs. The average tensile strength ranged from a minimum of 72 MPa to a maximum of 185 MPa while for Young's modulus, the values obtained were ranged between 9.4 GPa and 14.6 GPa.

Table 2: Experimental design runs and responses results

Runs	Variables			Responses	
	A: Temperature (°C)	B: Pressure (MPa)	C: Heating Time (min)	Tensile Strength (MPa)	Young's Modulus (GPa)
1	190	3	10	153	11.7
2	200	3	15	162.8	14
3	200	3	5	184.8	14.3
4	210	3	10	172.1	14.4
5	210	5	15	97.3	12
6	190	5	5	158.8	12.7
7	200	7	15	99.2	11.4
8	200	5	10	172	14.3
9	200	7	5	136.1	12.4
10	210	5	5	146.6	14.6
11	200	5	10	177	14.6
12	200	5	10	178	14.6
13	210	7	10	71.6	9.4
14	190	7	10	161.9	13.7
15	190	5	15	154.3	12.5

Analysis of Variances (ANOVAs)

Table 3 and Table 4 show the analysis of variances (ANOVAs) for tensile strength and Young's modulus respectively. The ANOVA table is commonly used to make conclusions and to provide case statistics for further interpretation.

From the ANOVA in Table 3, the model was found to be significant with P-value less than 0.05 and F-value of 111.84. The process parameters with P-values less than 0.05 indicate that the terms of the model significantly affected the response in the design space. In this case A, B, C, AB, AC, A^2, B^2, and C^2 were significant model terms, except for BC. The values exceeding 0.1000 indicate that the model terms were insignificant. However, the term BC was maintained in the model to avoid significant changes to the mathematical equation. Furthermore, the determination coefficient R^2 of 0.9951 and adjusted R^2 of 0.9862 indicate that there is a relationship between the variables under consideration and they are represented adequately by the model.

Table 3: ANOVA results for tensile strength

Source	Sum of Squares	DF	Mean Square	F Value	P Value Prob> F
Model	15698.09	9	1744.23	111.84	0.0001
A-Temperature	2464.02	1	2464.02	157.99	0.0001
B- Pressure	5196.90	1	5196.90	333.22	0.0001
C-Heating Time	1587.66	1	1587.66	101.80	0.0002
AB	2992.09	1	2992.09	191.85	0.0001
AC	501.76	1	501.76	32.17	0.0024
BC	55.50	1	55.50	3.56	0.1179
A^2	1666.65	1	1666.65	106.87	0.0001
B^2	805.58	1	805.58	51.65	0.0008
C^2	849.80	1	849.80	54.49	0.0007
Residual	77.98	5	39.21		
Lack of Fit	57.31	3	67.75	1.85	0.3699
Pure Error	20.67	2	17.80		
Cor Total	15776.07	14			
R^2	0.9951				

The ANOVA result in Table 4 shows that the model was found to be significant with a P-value of less than 0.05 and F-value of 10.82. The significant model terms were B, AB, A^2, and B^2. The coefficient R^2 of 0.9511 and adjusted R^2 of 0.8632 also indicate that the model adequately represents the real relationship between the variables under consideration.

Table 4: ANOVA results for Young's modulus

Source	Sum of Squares	DF	Mean Square	F Value	P Value Prob > F
Model	31.88	9	3.54	10.82	0.0087
A-Temperature	$1.25x10^{-3}$	1	$1.25x10^{-3}$	$3.18x10^{-3}$	0.9531
B- Pressure	7.22	1	7.22	333.22	0.0054
C-Heating Time	2.10	1	2.10	101.80	0.0523
AB	12.60	1	12.60	191.85	0.0016
AC	1.44	1	1.44	32.17	0.0901
BC	0.12	1	0.12	3.56	0.5675
A^2	4.67	1	4.67	106.87	0.0129
B^2	4.07	1	4.07	51.65	0.0168
C^2	0.67	1	0.67	54.49	0.2129
Residual	1.64	5	1.64		
Lack of Fit	1.58	3	1.58	1.85	0.0545
Pure Error	0.060	2	0.060		
Cor Total	33.52	14	33.52		
R^2	0.9511				

Development of second order equation

Next, the second order statistical equation model as in Equation (2) and Equation (3) were developed based on the experimental results in Table 3. Since the R^2 was above 90%, this model equation could be applied to predict the values of the responses within the limited range of the investigated parameters.

$$X\ (\text{Tensile Strength}) = 175.67 - 17.55 *A - 25.49 *B - 14.09 *C - 27.35 *AB - 11.20 *AC - 3.73\ BC - 21.25 *A^2 - 14.77 *B^2 - 15.17 *C^2 \quad (2)$$

$$Y\ (\text{Young's Modulus}) = 14.5 + 0.20 *A - 1.16 *B - 0.51 *C - 2.20 *AB - 0.61*AC - 0.17\ BC - 0.91*A^2 - 0.84*B^2 - 0.64*C^2 \quad (3)$$

where *A* is the temperature, *B* is the pressure and *C* is the heating time.

Model accuracy check

Figure 1 and Figure 2 depict the normal probability plot of residuals for the tensile strength and Young's modulus of the composite respectively. They reveal that the residuals followed normal distribution and generally fell on a least-square line closely with no reasonable outliers. In addition, all the data was within the limit with no unusual structure in the model.

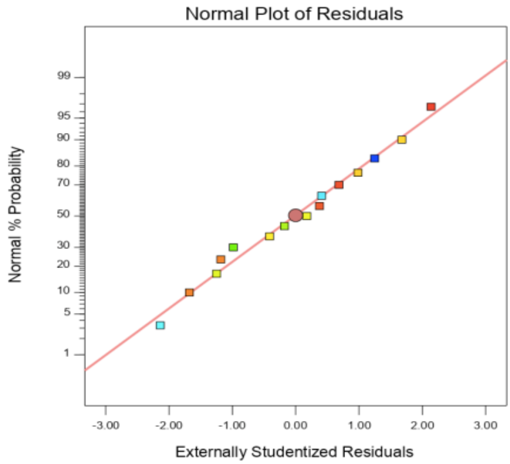

Figure 1: Normal probability plot of residuals for tensile strength.

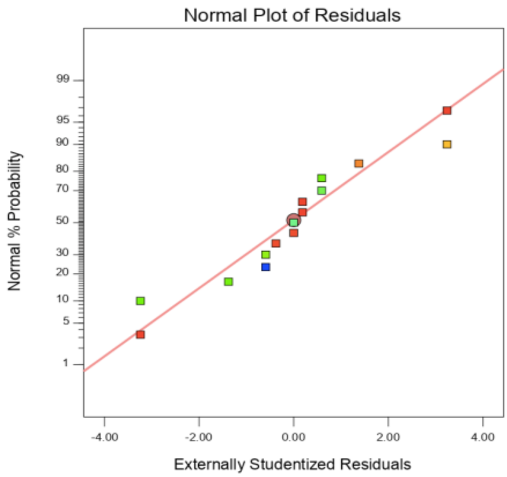

Figure 2: Normal probability plot of residuals for Young's modulus.

Optimization of Parameters

The maximum and optimal values predicted for tensile strength and Young's modulus from the experiment could be determined by the equation from the response surface. The relationships between tensile strength and Young's modulus with interaction of the parameters involved have been presented in 3D surface plots shown in Figure 3, Figure 4 as well as Figure 5. Each of the 3D surface plots shown had been generated by keeping one factor constant at centre point and varying the others within the experimental range.

As can be seen from the plot in Figure 3, at 10 minutes heating time and minimum pressure of 3 MPa, the tensile strength increased with increasing temperature from 190 °C to 200 °C. On the other hand, increasing temperature beyond 200 °C caused a slight decrease in tensile strength and the lowest value occurred at the highest pressure of 7 MPa. Therefore, the temperature and pressure needs to be carefully controlled. The temperature and pressure need to be at certain values so that polymer matrix can flow easily between the fibres but at the same time the values cannot be too high or fibre degradation will occur [19].

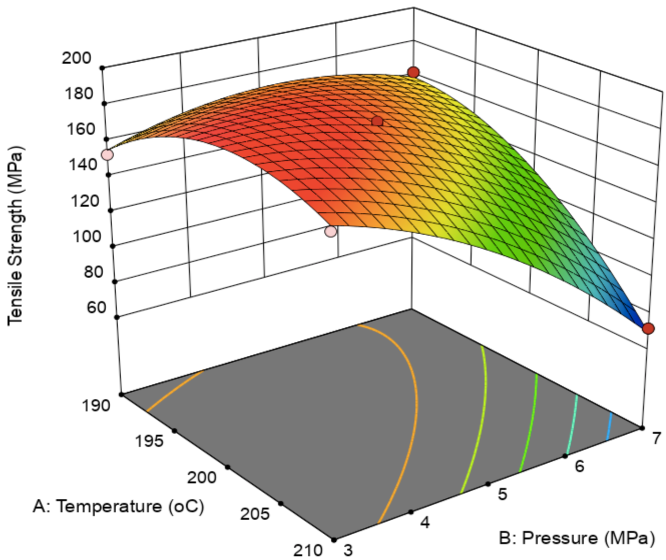

Figure 3: 3D Response surface plots representing the significant interaction on tensile strength - interaction temperature and pressure when heating time was 10 minutes.

With respect to the interaction between temperature and heating time at constant pressure of 5 MPa as depicted in Figure 4, it was clear that the tensile strength increased with increasing heating time from 5 to 9 minutes. Conversely, beyond 9 minutes, the tensile strength started to decrease and reached the lowest value at maximum temperature and heating time of 210 °C and 15 minutes respectively. The heating time must be suitable to allow for good wetting as it leads to better impregnation of the fibres into the matrix.

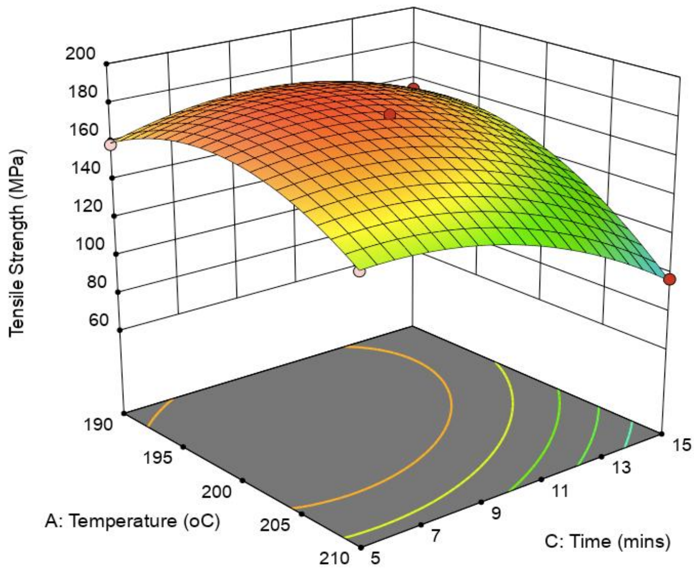

Figure 4: 3D response surface plots representing the significant interaction on tensile strength - interaction temperature and heating time when pressure was at 5 MPa.

Figure 5 indicates the changing response surface for Young's modulus at heating time of 10 minutes. The plot shows an ascending trend when the temperature was increased from 190 °C to around 200 °C at pressure below 4 MPa. However, the Young's modulus started to decrease when the temperature and pressure exceeded the above values simultaneously. In addition, an apparent interactive effect existed between the temperature and pressure as shown by the elliptic contours pattern. This could have been due to the high processing pressure which may have caused damage to the fibres and also lead to matrix starvation [21]. According to Kiran [19], high temperature causes thermal degradation to the fibres. In general, thermal degradation of natural fibres occurs in three stages where the first stage occurs as a result of waxes decomposition at lower temperature of 120 °C. It is followed by the second stage at approximately 180 °C where it leads to pectin decomposition and the last stage occurs at approximately 230 °C where decomposition of cellulose takes place. Consequently, some compromise must be made where the range of temperature is limited in order to obtain good wetting and at the same time avoiding fibre degradation. Bodros [11] suggested that viscosity of polymer especially for thermoplastic has to be close to 100 Pa, for better impregnation of the fibres to the matrix which will lead to the enhancement of the mechanical properties.

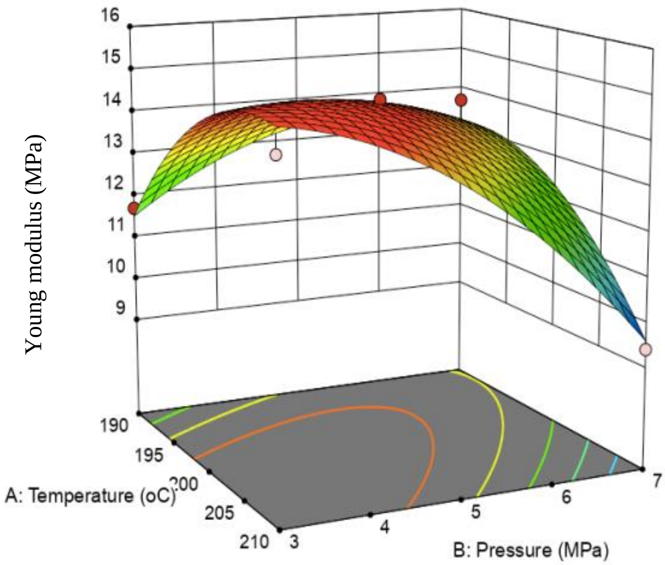

Figure 5: 3D response surface plots representing the significant interaction of temperature and pressure on Young's modulus when heating time was10 minutes.

Confirmation test

The maximum predicted values for tensile strength and Young's modulus as well as experimental optimum values for maximizing both responses could be gained from the model equations. By using point prediction from the numerical optimization available in Design-Expert of RSM, the optimal parameters setting was at a temperature of 200 °C, a pressure of 3 MPa and a heating time of 7 minutes with desirability of 1. Confirmation runs were conducted to verify whether the predictions were accurate. This was to ensure that nothing had changed and that the response values were close to the predicted values. Under the optimal parameter setting conditions, the values of tensile strength and Young's modulus were predicted and the values from the confirmation run were obtained as shown in Table 5. The percentage error was calculated to compare the predicted and the actual values. The average error was 4.04% for tensile strength and 6.25% for Young's modulus.

Table 5: Confirmation run results with optimal setting parameters

Temperature (°C)	Pressure (MPa)	Heating Time (mins)	Tensile Strength (MPa)		Young's Modulus (GPa)	
			Predicted	Actual	Predicted	Actual
200	3	7	190	198	14.4	15.3

Conclusion

The influence of hot pressing parameters on tensile strength and Young's modulus of the green composites were investigated through response surface methodology. Hot press parameters such as temperature, pressure and heating time as well as the temperature-pressure and temperature-heating time interactions were found to have significant effect on the tensile properties of the green composite. However, the main factor that contributes greatly for the Young's modulus was the pressure and the interaction between temperature and pressure. The optimum area in the designated range of process parameters was found using 3D response surface plots and optimum hot press parameters were achieved at temperature of 200 °C, 3 MPa pressure and heating time of 7 minutes. Validity of the model was verified through confirmation runs results and the errors yield was as low as 10%. The highest tensile properties achieved through the optimized parameters were 198 MPa for the tensile strength and 15.3 GPa for the Young's modulus. In this study, it is evident that the parameters of hot pressing is important for the fabrication of green composites and highly influence their tensile properties. Future research can include more parameters as well as adding more responses such as flexural properties, impact and hardness.

Acknowledgement

The authors would like to express appreciation to Universiti Kebangsaan Malaysia and Kementerian Pengajian Tinggi Malaysia for the financial support under the Long Term Research Grant Scheme (LRGS/TD/2012/USM-UKM/PT/05).

References

[1] J. K. Pandey, S. H. Ahn, C. S. Lee, A. K. Mohanty and M. Misra, "Recent advances in the application of natural fiber based composites,"

Macromolecular Materials and Engineering, vol. 295, no. 1, pp 975–989, 2010.

[2] M. Hughes, "Defects in natural fibres : Their origin, characteristics and implications for natural fibre-reinforced composites," *Journal of Materials Science*, vol. 47, pp 599–609, 2012.

[3] H. M. Akil, M. F. Omar, A. A. M. Mazuki, S. Safiee, Z. A. M. Ishak and A. Abu Bakar, "Kenaf fiber reinforced composites: A review," *Materials & Design*, vol. 32, pp 4107–4121, 2011.

[4] G.Koronis, A. Silva and M. Fontul, "Green composites: A review of adequate materials for automotive applications," *Composites Part B: Engineering*, vol. 44, no. 1, pp 120–127, 2013.

[5] M. Murariu and P. Dubois, "PLA composites: From production to properties," *Advanced Drug Delivery Reviews*107, pp 17–46, 2016.

[6] P. K. Bajpai, I. Singh and J. Madaan, "Development and characterization of PLA-based green composites: A review," *Journal of Thermoplastic Composite Materials*, vol. 27, no. 1, pp 52–81, 2014.

[7] J. Holbery and D. Houston, "Natural-Fiber-Reinforced Polymer Composites in Automotive Applications," *J. Miner. Met. Mater. Soc.*, pp 80–86, 2006.

[8] Y. Zhou, M. Fan, and L. Chen, "Interface and bonding mechanisms of plant fibre composites: An overview," *Composites Part B: Engineering*, vol. 101, pp 31–45, 2016.

[9] B. Madsen and H. Lilholt, "Physical and mechanical properties of unidirectional plant fibre composites-an evaluation of the influence of porosity," *Composites Science & Technology,* vol.63, no. 9, pp 1265–1272, 2003.

[10] H. Ku, H. Wang, N. Pattarachaiyakoop, and M. Trada, "A review on the tensile properties of natural fiber reinforced polymer composites," *Composites Part B: Engineering*, vol. 42, pp 856–873, 2011.

[11] E. Bodros, I. Pillin, N. Montrelay and C. Baley, "Could biopolymers reinforced by randomly scattered flax fibre be used in structural applications ?," *Composites Science and Technology*, vol. 67, pp 462–470. 2007.

[12] M. Ho, H.Wang, J. -H. Lee, C. Ho, K. Lau, J. Leng & D. Hui, "Critical factors on manufacturing processes of natural fibre composites," *Composites Part B: Engineering*, vol. 43, no. 8, pp 3549–3562, 2012.

[13] Z. N. Terzopoulou, G. Z. Papageorgiou, E. Papadopoulou, E. Athanassiadou, E. Alexopoulou, and D. N. Bikiaris, "Green composites prepared from aliphatic polyesters and bast fibers," *Ind. Crops Prod.* vol. 68, pp 60 – 79, 2014.

[14] T. Mukherjee and N. Kao, "PLA Based Biopolymer Reinforced with Natural Fibre : A Review," *Polymer. Environment,* vol. 19, pp 714–

725, 2011.

[15] N. H. A. Halim, C. H. C. Haron, J. A.Ghani and M. F. Azhar, "Prediction of cutting force for milling of Inconel 718 under cryogenic condition by response surface methodology," *Journal of Mechanical Engineering*, vol. 16, no. 1, pp 1 – 16, 2019.

[16] N.Badroush, C. H. C. Haron, J. A.Ghani, M. F. Azhar and N. H. A. Halim, "Performance of coated carbide tools when turning Inconel alloy 718 under cryogenic condition using RSM," *Journal of Mechanical Engineering*, vol. 5 (Special issue 3), pp 73 – 87, 2018.

[17] L. Wu, K. L. Yick, S. P. Ng, and J. Yip, "Application of the Box-Behnken design to the optimization of process parameters in foam cup molding," *Expert System Application*, vol. 39, no. 9, pp 8059–8065, 2012.

[18] B. S. Kumar and S. Balachandar, "A Study on the Influence of Hot Press Forming Process Parameters on Flexural Property of Glass/PP Based Thermoplastic Composites Using Box-Behnken Experimental Design," *ISRN Material Science*, vol. 3, no 4, pp 1–6, 2014.

[19] G. B. Kiran, K. N. S. Suman, N. M. Rao and R. U. M. Rao, "A study on the influence of hot press forming process parameters on mechanical properties of green composites using Taguchi experimental design," *Int. J. Eng. Sci. Technol.* vol. 3, no. 4, pp 253–263, 2011.

[20] I. Tharazi, N. Muhamad, N.F. Ismail, M. K. F. M.Radzi, Z.Razak, D. Tholibon and M. N Akhtar, "Effects of fiber content and processing parameters on tensile properties of unidirectional long kenaf fiber reinforced polylactic-acid composite," *Journal of Mechanical Engineering* SI, vol 3, no. 1, pp 65 – 76, 2017.

[21] K. L. Pickering, M. G. A. Efendy and T. M. Le, "A review of recent developments in natural fibre composites and their mechanical performance," *Composites Part A*, vol.83, pp 98 – 112, 2015.