ROSLAN, S.A., HASSAN, M.Z., RASID, Z.A., BANI, N.A., SARIP, S., DAUD, M.Y.M. and MUHAMMAD-SUKKI, F. 2019. Mode I fracture toughness of optimized alkali-treated Bambusa Vulgaris bamboo by Box-Behnken design. In Awang, M., Emamian, S.S. and Yusof, F. (eds.) *Advances in material sciences and engineering: selected papers from the proceedings of the 4th international conference on mechanical, manufacturing and plant engineering (ICMMPE 2018)*, 14-15 November 2018, Melaka, Malaysia. Lecture notes in mechanical engineering. Singapore: Springer [online], pages 565-575. Available from: https://doi.org/10.1007/978-981-13-8297-0_58

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2019

Mode I Fracture Toughness of Optimized Alkali-Treated *Bambusa Vulgaris* Bamboo by Box-Behnken Design

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Abstract

Alkaline treatment is widely being used to treat natural fibres and it improves the fibre surface for better bonding with the polymer matrix. The aim of this study is to optimize the alkaline treatment variables such as sodium hydroxide (NaOH) concentration, soaking and drying time that influence the strength of natural fibres, including bamboo. In this study, Box-Behnken design (BBD) of the response surface method was employed to set an experimental parameter of alkaline treatment for the bamboo specimen. In order to investigate the effect of treatment conditions on crack propagation behaviour of the bamboo along the longitudinal direction, Mode I interlaminar fracture toughness (G_{IC}) test was carried out. It can be suggested from the statistical analysis approach (ANOVA) that bamboo treated with 1 wt.% concentration of NaOH is able to reach fracture toughness value up to 365.86 J/m^2, which differs by only 0.82% from the experimental finding. It is also shown that all proposed variables for treatment in this study i.e. the concentration of the NaOH is highly significant with the soaking and drying time.

Keyword: Mode I Fracture Toughness, Box-Behnken Design, Bamboo

1.0 Introduction

Natural fibres can simply be defined as non-synthetic fibres. They can be obtained from animal, plant or mineral resources. Among these types, the combination of plant-based fibres with polymer matrices to form natural fibre reinforced polymer composites (NFPCs) are extensively being employed. This is because their combination exhibited a great mechanical strength and stiffness to weight ratio [1]. The increase in the utilisations of NFPCs in the automotive industries [2], marine [3] and construction [4] are good demonstrations to this affirmation. Bamboo is one of the most researched natural fibres of late due to its specialties of having low density and high specific strength, which is up to 4 times higher than that of the mild steel. These two characteristics show that bamboo fibre is on par with glass fibre properties.

A major drawback from using natural fibres in polymeric composites is the incompatibility between the natural fibre and polymer matrix due to hydrophilic and hydrophobic interaction. The high moisture content in natural fibres makes them difficult to bond properly with the polymer matrix and thereby degrading the mechanical properties of NFPCs. Thus, chemical treatment on natural fibre is being promoted to increase the interfacial bonding compatibility between the fibre and polymer matrix [5]. Among the chemical treatments that have been obtained, alkaline treatment is frequently used. This method is carried out by immersing the natural fibre in a particular concentration of the alkaline solution, like sodium hydroxide (NaOH), for a certain period of time. It is reported that this treatment improves the strength of fibre and increases the surface roughness for better interlocking between fibre and polymer matrix [6-7]. Jacob *et. al.* [8] studied the effect of NaOH concentrations of 0.5, 1, 2, 4 and 10% on sisal fibre reinforced composites. They reported that the highest tensile strength composite laminate has been found at 4% of NaOH concentration tested at room temperature. In contrast, Mishra *et. al.* [9] reported that sisal fibre reinforced polyester composites which were treated with 5% concentration of NaOH exhibited good tensile strength properties to those treated with 10% of concentration. They also highlighted that the higher alkali concentration would cause excess delignification of the natural fibre and thus weaken it. Zhang *et. al.* [10]

treated bamboo fibres with 4% concentration of NaOH for one hour. It was discovered that the treatment increased the effective surface area for better bonding of the fibre with the matrix by removing chemical components such as hemicellulose and lignin. They observed from SEM images that the treated bamboo fibre surface seems to be smoother than those untreated fibre due to the removal of chemical components and impurities. Phong *et. al.* [8] suggested that 1% concentration of NaOH treated on bamboo/epoxy laminate resulted in higher mechanical characteristics compared to 2% and 3% concentration. In their study, bamboo fibres were immersed in NaOH concentrations of 1, 2 and 3% for 10 hours at 70°C and allowed to dry for a day at 105°C in the air circulation oven. It was found that bamboo fibre with 1% concentration of NaOH showed the highest tensile strength and Young's modulus among those treated with different concentrations. The result was in good agreement with Rao *et. al.* [11] who reported that 1% concentration of NaOH is the best to treat bamboo polymeric composite. On the contrary, few studies claimed that a higher NaOH concentration is the best in treating bamboo fibre and capable of removing excess moisture thoroughly. In spite of that, the NaOH concentration is the most dominant factor that gives effect on the natural fibre. In addition, treatment variables such as soaking time, drying period and temperature could also influence the end results.

Many attempts have been made, including applying of Box-Behnken design (BBD) in the optimization of chemical treatment for natural fibres, as conducted by Aly *et. al.* [12] in NaOH treatment parameters of flax fibre. They reported that the BBD is an accurate tool for optimizing chemical treatment to obtain the outstanding mechanical properties of the fibre matrix composite. Vardhini *et. al.* [13] employed BBD tool to determine the optimum treatment conditions in treating banana and kenaf fibres, where they found that treatment conditions of 11g/L NaOH concentration, 2.5 hours of treatment time and temperature of 90°C are able to remove lignin from the banana fibre at higher rates. Thus, these previous works proved that the BBD is an efficient tool in optimization of work processes that involve more than two variables.

In this study, the Box-Behnken design (BBD) of the response surface method was employed to set an experimental design of alkaline treatment conditions for the *bambusa vulgaris* bamboo. Mode I interlaminar fracture toughness (G_{IC}) test was conducted to investigate the effect of alkaline treatment conditions on crack propagation behaviour along the longitudinal direction of the bamboo. Three conditions are considered for the concentration of NaOH, soaking and drying time. The results are subsequently analysed statistically using the Analysis of Variance (ANOVA).

2.0 Experimental Method

2.1 Box-Benhken Design Iteration

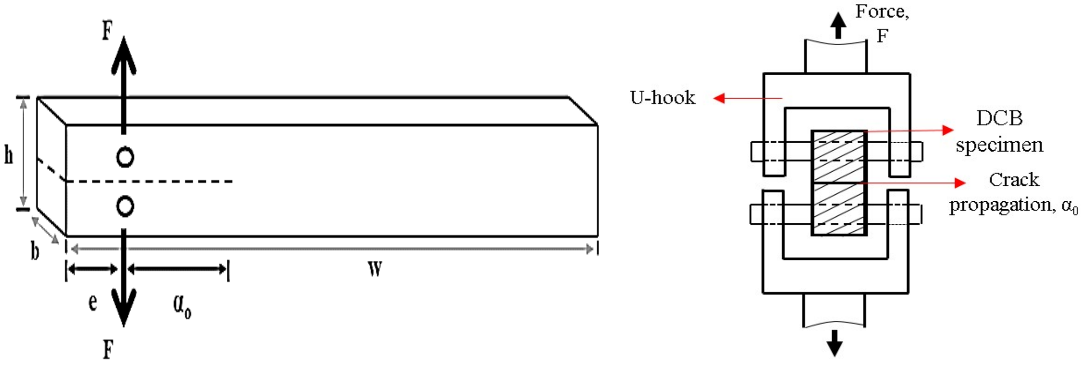
In order to determine the optimum chemical treatment condition, an experimental design composed of three variables was built using Box-Behnken design (BBD). **Table 1** shows the design points for low, middle and high levels of each variable condition. This input data then was randomized and modelled by BBD using the Design-expert (6.0.8) software. It offers a total number of 17 experimental runs that consist of 12 runs and 5 replication runs of the centre point.

Table 1 The initial setting of Box-Behnken design (BBD)

Factors / Independent Variables	**Symbols**	**Coded and Actual Levels**		
		Low (-1)	**Middle (0)**	**High (+1)**
Concentration of NaOH (%)	X_1	1	2	3
Soaking duration (hours)	X_2	3	6	9
Drying duration (hours)	X_3	2	48	72

2.2 Material and Testing

The bamboo material that was used in this study belongs to *bambusa vulgaris* family. The raw bamboo was cut approximately 5 metres above the ground. The age of bamboo was approximately four years old and taken from Jeli, Kelantan at the north-eastern state of Malaysia. Mode I testing specimens are prepared as a double cantilever beam (DCB) according to the ASTM D5528 standard [14]. The dimension of the DCB specimen is, longitudinal direction, $w = 200$ mm; tangential direction, $h = 20$ mm; and radial thickness, $b = 9$ mm with initial crack length, $\alpha_0 = 40$ mm. Two loading holes of 5 mm in diameter were made at the point crack initiation (about 20 mm before the end). **Figure 1 (a)** shows a schematic diagram of the DCB specimen. After that, the specimens were chemically treated with an alkali solution. The concentration of this solution, which was sodium hydroxide (NaOH), was prepared by weight per volume (*w/v*) percentage. In order to obtain 1 wt.% concentration of NaOH, 1 gram of NaOH pellets was diluted in 100 ml of distilled water. After undergoing alkali treatment and drying process, an initial crack was cleaved along the middle-line of the bamboo DCB specimen parallel to grain by a stiff razor. Following that, the bamboo DCB specimen was fitted to the U-shaped hook steel which connected to a 10 kN load cell on a Shidmazu universal testing machine as illustrated in **Figure 1(b)**.



(a) (b)

Figure 1 Schematic diagram of DCB specimen (a) connected to U-hook (b) following tensile test

2.3 G_{IC} Analysis

According to the compliance method, the reciprocal slope from load-displacement traces is the corresponding compliance (*Ci*) of the DCB specimen with a certain crack length (α_i). The relationship between C and α can be described as stated in the following Equation 1.

$$C = q\alpha^m \tag{1}$$

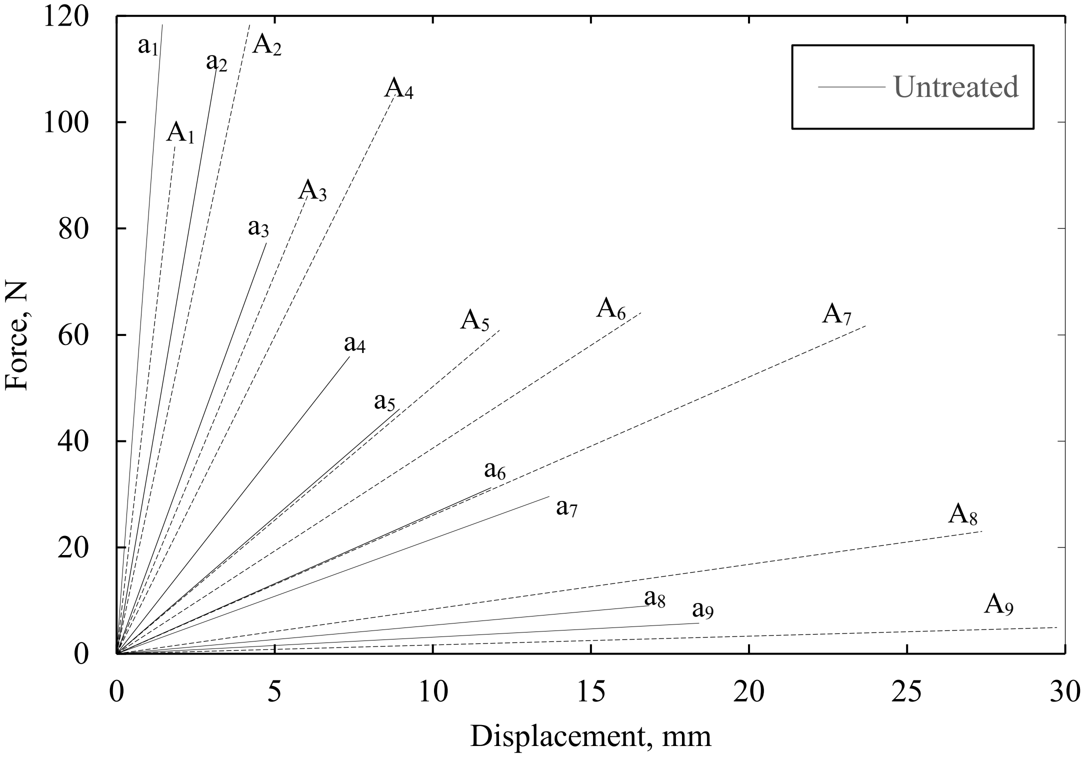
where q and m are the fitting coefficients of the compliance curve of the DCB specimen. So, after taking a logarithm of Equation 1, the equation meets the linear model as stated in Equation 2. Following the Mode I test, the results were analysed and evaluated statistically using the Analysis of Variance (ANOVA).

$$\lg C = \lg q + m \lg \alpha \tag{2}$$

3.0 Result and Discussion

3.1 G_{IC} Characterization of Bamboo DCB Specimen

Figure 2 depicts the force-displacement traces corresponding to different crack lengths, α, of untreated and treated bamboo specimens. Here, a_1, a_2, a_3,…, a_9 and A_1, A_2, A_3,…, A_9 are the sum of the measured crack length, α, for untreated and treated specimens respectively. It can be seen that the slope of traces for both conditions decreased with the increase of crack lengths. However, it seems that the treated specimen exhibited an inconsistent trend of force and displacement traces compared to untreated, in which the traces occasionally dropped on lower or higher forces with the larger of displacement. It can be proposed that the alkaline treatment has caused the specimen to be rougher on the crack surface due to the forming of peaks and valleys. These peaks and valleys disrupted the distribution of applied force along the fibre and caused the load to concentrate on a particular point on the fibre. Thereby, it causes the specimen to yield at an uneven load. On the other hand, Islam [15] and Bledzki *et. al.* [16] reported that alkali-treated fibre tends to break at a higher displacement value as a resultant of the softening of the inter-fibrils matrix. This softening effect has negatively affected the transfer of stress between the fibres, thus disturbing the overall stress development in fibre during tensile deformation.



a_1= 42 mm, a_2= 56 mm, a_3= 66 mm, a_4= 83 mm, a_5= 78 mm, a_6= 97 mm, a_7= 105 mm, a_8= 114 mm, a_9= 122 mm, a_{10}= 163 mm

Figure 2 The typical force-displacement traces of untreated and treated bamboo

3.2 The BBD Analysis of G_{IC} for Treated Bamboo

The G_{IC} results of treated bamboo were further investigated using ANOVA in order to determine which variables significantly affect the G_{IC} value of treated bamboo. By applying a multiple regression analysis of the responses, the outcome proposed the highest order polynomial in which the additional terms were significant and the model was not aliased. Following that, backwards elimination method was applied to exclude insignificant terms automatically. The ANOVA for the reduced quadratic models summarised in the G_{IC} value, is shown in Equation 2.

$$G_{IC} = 573.235 - 160.801\, X_1 - 14.743\, X_2 - 1.229\, X_3 + 27.696\, X_1^2 - 0.679\, X_2^2 + 0.033\, X_3^2 + 3.936\, X_1 X_2 - 1.095\, X_1 X_3 \tag{3}$$

Equation 3 represents the relationship between NaOH concentration (X_1), soaking time (X_2) and drying time (X_3) towards G_{IC} value of treated bamboo. This equation was also used to generate predictions of the response for a given level of each variable. **Table 2** depicts the predicted G_{IC} values using Equation 8 and the experimental finding value. It is in good agreement between the yield predicted and the experimental responses, with a small difference in G_{IC} value measured as observed in **Table 2**. The percentages of error are also calculated to determine the precision of calculations. Here, the error percentage of each run is less than 4%, which is considered effective [17]. The bamboo treated with 1 wt.% concentration of NaOH offers the fracture toughness value up to 365.86 J/m^2, with 0.82% of error from the testing result. The G_{IC} value of treated bamboo specimens is declined with the increasing NaOH concentration and soaking time. Again, the higher NaOH concentration and soaking time could worsen mechanical properties of fibre due to the softening effect and weakens the fibre. Longer drying time, however, has a positive correlation with the response. The model afterwards was inspected statistically using the F-test and regression coefficient, R^2 for validity purposes.

Table 2 The BBD analysis predicted and experimental G_{IC} values

Run	**Independent Variables**						**G_{IC} (J/m^2)**			
	Coded values			**Actual values**			**Experimental**	**Predicted**	**Residual**	**Error (%)**
	X_1	**X_2**	**X_3**	**X_1**	**X_2**	**X_3**				
1	0	+1	-1	2	9	24	186.11	182.40	3.71	2.03
2	0	0	0	2	6	48	205.64	208.42	-2.78	1.33
3	0	-1	-1	2	3	24	270.83	272.58	-1.75	0.64
4	0	0	0	2	6	48	207.19	208.42	-1.23	0.59
5	0	+1	+1	2	9	72	163.97	169.94	-5.97	3.51
6	+1	-1	0	3	3	48	185.06	184.32	0.74	0.40
7	0	0	0	2	6	48	207.63	208.42	-0.79	0.38
8	0	-1	+1	2	3	72	264.13	260.12	4.01	1.54
9	-1	+1	0	1	9	48	251.32	252.06	-0.74	0.29
10	+1	0	+1	3	6	72	142.71	143.60	-0.89	0.62
11	0	0	0	2	6	48	208.73	208.42	0.31	0.15
12	-1	-1	0	1	3	48	362.85	365.86	-3.01	0.82
13	-1	0	+1	1	6	72	356.94	354.09	2.85	0.80
14	-1	0	-1	1	6	24	314.88	313.99	0.89	0.28
15	0	0	0	2	6	48	212.91	208.42	4.49	2.15
16	+1	+1	0	3	9	48	120.76	117.76	3.00	2.55
17	+1	0	-1	3	6	24	205.77	208.62	-2.85	1.37

Table 3 presents the results acquired following executing the analysis of variance (ANOVA). The significance of the coefficient terms is determined by the F and p values. As shown in **Table 3**, this regression model is highly significant with F-value of 551.22. The interactions among the NaOH concentration (X_1), soaking time (X_2) and the drying period (X_3) with a probability value ("Prob > F" > 0.05) indicate that the model terms are highly significant. According to "Prob > F", the most significant model terms that affect G_{IC} values of the treated bamboo are NaOH concentration (X_1), soaking time (X_2), drying time (X_3), second-order NaOH concentration (X_1^2), second-order soaking time (X_2^2) and second-order drying time (X_3^2). On the other hand, the interaction between NaOH concentration and soaking time ($X_1 X_2$) and interaction between NaOH concentration and drying time ($X_1 X_3$) are the subsequent significant factors in this study.

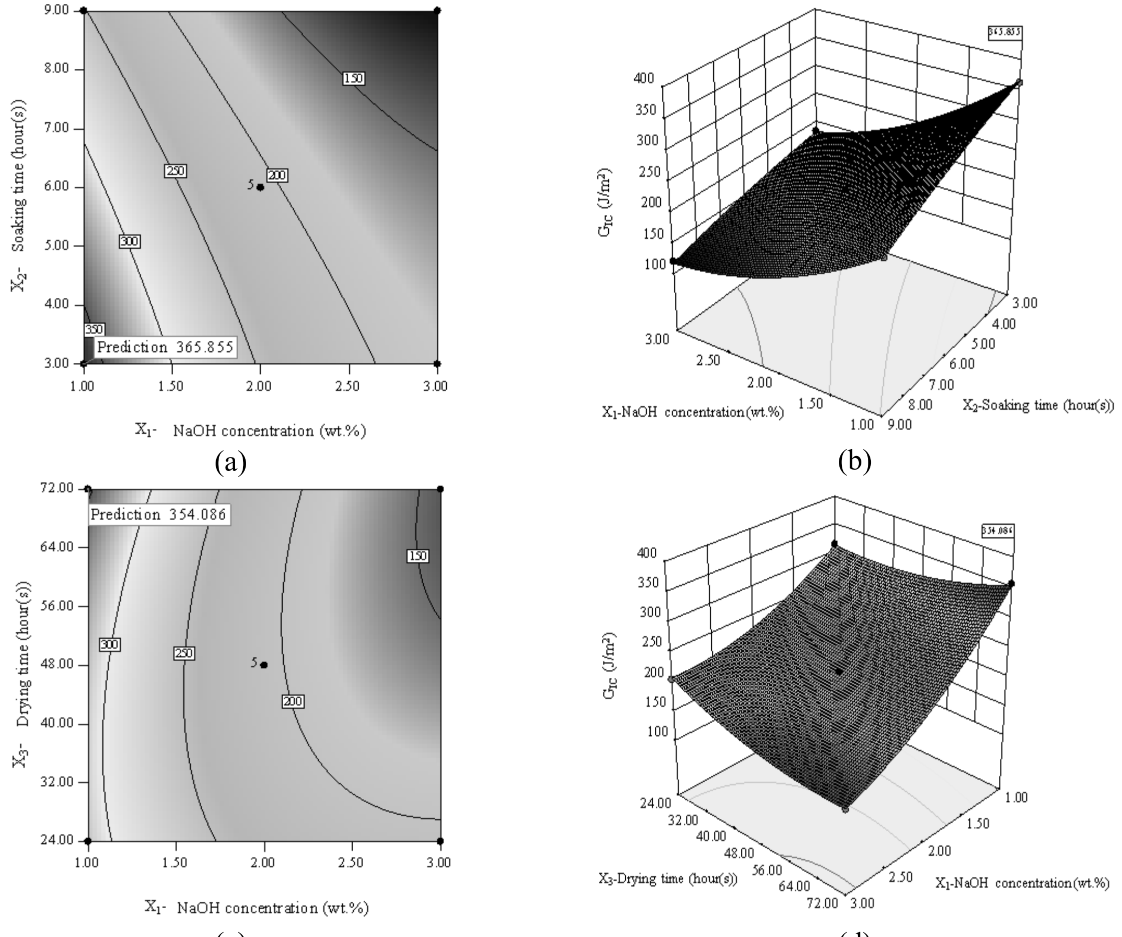
Table 3 The ANOVA of quadratic model for alkaline treatment of bamboo

Source	Sum of squares	Degree of freedom	Mean square	F-value	p-value (Prob > F)
Model	74816.17	8	9352.02	551.22	< 0.0001*
X_1- concentration	49879.03	1	49879.03	2939.93	< 0.0001*
X_2- soaking	16263.96	1	16263.96	958.62	< 0.0001*
X_3- drying	310.50	1	310.50	18.30	0.0027*
X_1^2	3229.82	1	3229.82	190.37	< 0.0001*
X_2^2	157.64	1	157.64	9.29	0.0159*
X_3^2	1513.41	1	1513.41	89.20	< 0.0001*
$X_1 X_2$	557.67	1	557.67	32.87	0.0004*
$X_1 X_3$	2762.55	1	2762.55	162.83	< 0.0001*
Residual	135.73	8	16.97		
Lack of Fit	105.61	4	26.40	3.51	0.1259**
Pure Error	30.12	4	7.53		
Cor total	74951.90	16			

* Significant **Not significant

3.3 Response Surface Plots

The predicted models can be visualised as two-dimensional (2D) contour plot and three-dimensional (3D) surface graph. Each plot shows the effects of two variables within the studied ranges, while the other variables are fixed at their zero-coded level value. In contour plots, the contour lines display the extent of the interactions between two independent variables. It is easy to spot the optimum levels and it is convenient for the user to present the shape of the response of a two-dimensional projection. The 3D-surface graph visualises the tendency of each variable to influence the response in a graphical view. The curvature in the 3D graph is formed based on the quadratic dependence of response and parameters. Contour plot and response surface for the interaction effect of NaOH concentration (X_1) and soaking time (X_2) on G_{IC} values of bamboo at 48 hours of drying time (X_3) are presented in **Figure 10 (a-b)**. The highest G_{IC} value of 365.86 J/m^2 was recorded at the lowest NaOH concentration and soaking time. The G_{IC} value dropped to the minimum when treated with the highest NaOH concentration for 9 hours. **Figure 10 (c-d)** presents the response surface and corresponding contour plots for the interaction effect of NaOH concentration (X_1) and drying time (X_3) on G_{IC} values of bamboo for 6 hours of soaking time (X_2). The highest G_{IC} value of 354.09 J/m^2 was recorded when treated with the lowest NaOH concentration and be dried at the maximum drying hour. However, G_{IC} value seemed to be dropped to 143.22 J/m^2 at the maximum NaOH concentration even though the soaking and drying time was kept in the same condition. Based on the finding, the proposed variables were found to significantly influence the G_{IC} values of treated bamboo. The analysis suggested that the bamboo needs to be treated with low NaOH concentration for a short soaking period and dried at a longer drying time to obtain the optimum G_{IC} value of bamboo at room temperature.



(a) (b)

(c) (d)

Figure 10 The response surface plots of X_1X_2 in 2D contour and 3D model

4.0 Conclusion

Based on Mode I test results, it was found that the untreated bamboo specimen exhibited greater G_{IC} values compared to the treated specimen. This may be attributed to the softening effect caused by alkaline treatment which influences the stress transfer process along the specimen during the test. For treated bamboo, NaOH concentration (X_1) was recorded as the most significant parameter that affected the G_{IC} values of bamboo. The highest G_{IC} value for treated bamboo was predicted at 365.86 J/m^2 when the bamboo was treated with 1 wt.% concentration of NaOH for 3 hours and dried for 72 hours at room temperature (1wt.%-3hr-72hr). G_{IC} value gradually dropped to 120 J/m^2 when the bamboo was treated to higher than 1 wt.% concentration of NaOH. Such results may be influenced by the softening effect of alkaline treatment which disturbs the overall performance of the specimen during Mode I of loading.

Acknowledgements

The appreciation is given to Universiti Teknologi Malaysia and Ministry of Higher Education under Fundamental Research Grant Scheme, R.K130000.7840.4F880 and "*Geran Universiti Penyelidik*' (GUP) Tier 1, Q.K130000.2540.15H70 and Q.K130000.2540.16H95 for financially support provided throughout the course of this research project.

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