Journal of Multidisciplinary Engineering Science and Technology (JMEST)
ISSN: 3159-0040
Vol. 2 Issue 11, November - 2015

# Optimization Of Compounding Conditions For Adenia Lobata- HDPE Composite

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***Abstract*—This paper investigated the optimization of process factors for the compounding of HDPE - natural adenia lobata fiber using Box behnken design BBD for maximal tensile strength. The natural fiber was pretreated with sodium hydroxide, acetic anhydride, nitric acid and zinc chloride before the compounding process to improve interfacial bonding. The compounding process was achieved using two roll mill compounding machine. The optimized compounding conditions were temperature (°C), residence time (mins) and rotation speed (rpm). Quadratic models were developed for each chemical used for the pretreatment. The established optimum conditions were for untreated fiber, 99.28rpm, 175.25°C, and 11.83mins, for the sodium hydroxide treated, 90.91rpm, 173.09°C, and 14.57mins, for acetic anhydride treated, 95.85rpm, 174.87°C, and 14.5mins, for nitric acid treated, 95.85rpm, 175.56°C, and 13.33mins, for zinc chloride treated, 94.48rpm, 173.94°C, and 11.03mins. These optimum conditions were validated with little error. SEM analysis confirmed that there were intimate mixing of the fiber and polymer as a result of the compounding process.**

***Keywords—Adenia lobata fiber, compounding, treatment, HDPE, optimization,***

## I. INTRODUCTION

In recent years, the application of natural cellulose fibers as reinforcements in composite materials has gained serious improvement. The attractive features of natural fibers have been their low cost, light weight, high specific modulus and health harzards of composites reinforced with synthetic fibers such as glass, carbon and aramid fibers [1]. These advantages placed the natural fibers composites among the high performance composites having economical and environmental advantages [2-4]. However, natural fiber composites face some difficulties that prevent their widespread use. Fiber-polymer incompatibility has been the subject of previous studies [5-8]. This imcompatibility is caused by the hydrophilic nature of the fibers and the hydrophobic nature of many polymer used in this field [9]. The presence of hemicelluloses, lignin and other impurities also cause a lack of adhesion between fibers and polymer [9]. To improve the compatibility between both components, a surface modification is required. There are various chemical treatments available for surface modification of fibers [10]. Each will have its own characteristic effect on the fiber. Chemical modification is attempted to improve natural fiber hydrophobic nature, interfacial bonding between matrix and fiber, surface roughness and wettability, and also decrease moisture absorption, leading to the enhancement of mechanical properties of the natural fiber reinforced composites [11-12]. To study the effect of multiple variables on output, the response surface method (RSM) is an effective technique [13]. The RSM is a collection of mathematical and statistical techniques useful for the modelling and analyses of problems in which a response of interest is influenced by several variables and the objective is to optimize the response [14].

The aim of this study was to optimize three compounding conditions, namely temperature, time and rotation speed to obtain high tensile strength compression molded *adenia lobata* fiber reinforced composite. The fiber was treated with four chemicals, namely, sodium hydroxide, acetic anhydride, nitric acid and zinc chloride.

## II. MATERIALS AND METHODS

### A. *Natural Fibers*

Natural plant fibre; usoro (*adenia lobata*), was obtained from Olo in Ezeagu L.G.A of Enugu States, Nigeria.

### B. *Thermoplastic* (HDPE)

High density polyethylene (HDPE) pellets were obtained from De Cliff Integrated Company, Enugu. The density of HDPE is 963.3kg/m$^3$. Its melting temperature and melt index are 130-137$^0$C and 9g/10min, respectively.

### C. *chemicals*

Sodium hydroxide, Acetic anhydride, Nitric acid and zinc chloride used in this study were of analytical grade and was purchased from De Cliff integrated Company Enugu.

### *D. Extraction of fiber*

The plant fibre was extracted mainly from the plant stem using water retting extraction process, giving fibre of different length and diameters. The obtained plant stems were cut and sliced longitudinally into four pieces and each was completely submerged in water for two weeks, after which the stems were removed from the water and loosened by smashing. Bundles of fibers were further placed in water for one week at room temperature to remove impurities and facilitate their separation. The extracted fiber was finally washed thoroughly in excess tap water and air dried at room temperature for three days.

### *E. Chemical treatment of the fibers*

The fiber was subjected to chemical treatments such as mercerization with sodium hydroxide, acetylation with acetic anhydride, oxidation with nitric acid and zinc chloride treatment. This was done to improve fiber-matrix interaction for superior mechanical properties of the resulted composites.

#### *1) Alkali Treatment*

This procedure was in accordance with work done by Nural and Ishak [15] with slight modifications. The washed fibers were treated with 6% NaoH. The fibers were immersed in the alkali solution for 50 minutes, then neutralized with acetic acid and washed with distilled water repeatedly until all sodium hydroxide was eliminated. Finally, the fibers were washed with distilled water and dried at room temperature for 48h

#### *2) Acetic Anhydride Treatment*

The acetylation process was in accordance with work done by Bledzki, et al., [16] with slight modifications. The fibers were soaked in distilled water for an hour, filtered and placed in a round bottom flask containing 10% acetic acid solution for 30 minutes. After which it was placed on flask containing 14% acetic anhydride solution. The process temperature of acetylation was $30^0$C and duration was 70 minutes. After modification, the fiber was washed periodically with distilled water until acid free. Finally, modified fibers were air dried for certain time before analysis.

#### *3) Nitric Acid Treatment*

The nitric acid treatment was according to Vautard et al., [17] with modifications. The size reduced fibers were oxidized with 6% nitric acid. The prepared oxidizing solution was boiled to a temperature of $60^0$C and the fiber immersed in the solution at maintained said temperature for 50 minutes. At then, they were neutralized with NaoH solution and washed with distilled water repeatedly until all the nitric acid was eliminated. Finally, the fibers were washed again with distilled water and dried to a constant weight at room temperature.

#### *4) Zinc Chloride Treatment*

Zinc chloride treatment was done in accordance with the work done by Nadanthangam et al., [18] with modifications. The fibers were soaked in 3% zinc chloride solution for 70 minutes after which it was washed with distilled water until the washing solution became chloride free. The fibers were washed with distilled water and dried at room temperature for 48 hours.

### *F. Compounding process*

A method of Lu et al (2000) [19] was used for the compounding process. The treated and untreated fibers were separately mixed with HDPE in a blender at a specified fiber loading ratio. The mixture of the fibers and HDPE were fed into two roll mill and were mixed with heating. This step was performed to avoid the separation of fiber from the polymer during the molding process. The rotor speed, compounding temperature and the compounding time were varied to establish the optimum conditions. After compounding, all the resultant blends were segregated to smaller sizes to make them ready for compression molding.

### *G. Compression Molding Process*

This method was in accordance with the work done by Kumar and Amar (2009) [20] with slight modifications. The blend from compounding process was poured into specially made moulds. The surfaces of moulds were coated on the inside with oleic acid to avoid adhesion of the mixture and to allow easy removal of the composites. The mixture was then spread equally on the surface of the moulds. Composite sheets where prepared by compression molding technique on locally fabricated compression molding machine. In compression molding the material charge was pressed between two halves of mould and allowed to transform into a solid product. The mold was pressed with the help of jack at $170^0$C for 10mins and cooled down to room temperature. The pressure was monitored using a pressure guage attached to the machine and was kept constant at 30bar.

### *H. Determination of Tensile Strength*

The Hounsfield Tensometer testing machine (model 8889) England was used to determine the tensile strength of the composites. The samples were cut into (100x15x4) $mm^3$, after which, they were glued in between two frames to assure a good gripping and straight position in the test clamps. The tensile tests were performed on a mini tensile testing unit of the Hounsfield Tensometer machine with a load cell of 2500N where the crosshead speed was set at 1 mm/min; the load and the displacement were registered during the complete test. A rotating drum on which special graph sheet was rapped was fixed to the side of the equipment. A manually operated rotating handle was used for loading the specimen under test. With the help of an adjustable mercury level and a manually operated marker, the load and extension values of the specimen were recorded on the graph sheet at a regular interval until failure occurred. The tensile strength was calculated from this relation

$$\text{Tensile Strength (Mpa)} = \frac{\text{Maximum load (N)}}{\text{Area}(\text{mm}^2)} \qquad (1)$$

### *I. Scanning electron microscopy (SEM) and EDS analysis*

Scanning Electron microscropy (SEM) of the composites was carried out using (PHENOM PROX) analyser. Analysis was carried out on the composite to study their surface textures.

### *J. Fourier Transform Infrared Spectroscopy (FTIR)*

The surface functional groups were studied using fourier transform infrared spectroscopy [FTIR -8400S Shimadzu]. The FTIR spectra of the composites were scanned at a wavelength of 500–4000nm to obtain its spectra lines.

### *K. Statistical analysis and optimization*

Box Behnken Design (BBD) was used for the optimization of the compounding conditions. This design was used because relatively few experimental combinations of the variables are adequate to estimate potentially complex response functions. The compounding process has three numeric factors giving a total of 17 experiments for each fiber treated with four different chemicals. The factors and levels for the design are shown on table 1.

TABLE1. FACTORS AND LEVELS FOR COMPOUNDING PROCESS USING BBD

| ***Factors*** | ***Units*** | ***Levels*** | | |
|---|---|---|---|---|
| | | -1 | 0 | +1 |
| Rotation Speed | Rpm | 60.0 | 80.0 | 100.0 |
| Temp. | $^0$C | 160.0 | 170.0 | 180.0 |
| Time | Mins | 10.0 | 15.0 | 20.0 |

## III. RESULT AND DISCUSSIONS

Compounding process directly influences compounded quality of fiber-polymer blend and its resultant composites. Blending fiber with a polymer is the key step of composite production, because compounding process helps to uniformly distribute the fiber in the thermoplastic matrix, decrease pore ratio, and stabilizes the filler/matrix interaction [21]. There has been no criterion to determine optimum compounding conditions [19]. Usually, compounding condition vary with mixing machine type, compounding steps, weight ratio of fiber and the polymeric matrix, moisture content of fiber, and species of thermoplastic and fiber.

In this process, two roll mill was used for the compounding process.

The optimization of the compounding conditions was done using Box-Behnken design. Three compounding conditions were studied at three levels each with five center points giving 17 experiments each for each fiber with four different chemicals. The experimental runs were randomized to protect against an unknown bias distorting the outcome of the experiment. The experimental response was limited to the tensile strength (Mpa) of the composite. In order to identify the effect of the compounding conditions, the resultant blends were ground and used to produce a composite at constant conditions of 180$^0$C molding temperature, molding time of 10mins, pressure of 15atm and fiber loading of 40% with fiber length of 3mm. The experiment was strictly based on the design matrix.

### *A. Selection of a good predictive model*

A good predictive model is first step toward optimization. Once a good model is established, it can be optimized. Box Behnken design can fit linear, 2FI, and quadratic model. Because BBD has only three levels, it cannot predict cubic model. The best model selected was based on the lack of fit test, P-value of adjusted and predicted R-squared. The sequential model sum of squares was used to select the best model based on the highest order model that was significant (small P-value) and not aliased, no lack of fit (P-value > 0.10) and reasonable agreement between adjusted R-squared and predicted R-squared (within 0.2 of each other). Lack of fit which is the measure of risk was included because some points were replicated (center points) to produce estimate of pure error.

It was observed that the suggested model for the compounding process was quadratic model. Addition of cubic terms to the models did not improve the model but, if it does, Box Behnken design lack the design points needed to fit all terms required for the cubic model, to that effect, it labeled as being aliased. The suggested quadratic model had non significant lack of fit (P-value >0.1), and good agreement between the predicted and adjusted R-square (within 0.2 of each other).

### *B. Inspection of selected model*

Model selected using sequential sum of square was inspected for adequacy using analysis of variance (ANOVA). ANOVA was used to test the suggested model, the linear terms, interaction terms and the quadratic terms included in the model. Any term was removed from the model only when it had insignificant P-value (>0.1) or was retained in the model to support model hierarchy. From the ANOVA tables, it can be seen that time was insignificant, but was included in the final model to maintain model hierarchy since interaction of time and temperature was significant. Equally, the model was inspected based on its R-Squared, predicted R-squared and adjusted R-squared. Attention was more on the predicted and adjusted R-squared because they can give more estimate of how well the model will give good prediction for the average response. The regular R-squared can artificially rise when any model term is added whether statistically significant or not and as such, can not estimate how well the model predicts the response. There was no cut off value for R-square, the rule of the thumb is that the predicted and adjusted R-square must be within 0.2 of each other [22].

Another parameter that was used to inspect the model was adequate precision. Adequate precision measures the signal to noise ratio. It compares the range of predicted values at the design point to the average predicated error. A ratio greater than 4 is always desired for a good model [23].

TABLE 2. ANOVA TABLE FOR COMPOUNDING OF UNTREATED *ADENIA LOBATA* (USORO) FIBER.

| ***Source*** | ***Sum of Squares*** | ***df*** | ***Mean Squares*** | ***F Value*** | ***P-value Prob>F*** |
|---|---|---|---|---|---|
| Model | 141.08 | 7 | 20.15 | 57.75 | <0.0001 |
| A-Rotation Speed | 25.63 | 1 | 25.63 | 73.45 | <0.0001 |
| B-Temperature | 33.13 | 1 | 33.13 | 94.93 | <0.0001 |
| C-Time | 0.45 | 1 | 0.45 | 1.29 | 0.2849 |
| AB | 17.77 | 1 | 17.77 | 50.91 | <0.0001 |
| BC | 23.47 | 1 | 23.47 | 67.26 | <0.0001 |
| $B^2$ | 32.74 | 1 | 32.74 | 93.82 | <0.0001 |
| $C^2$ | 6.18 | 1 | 6.18 | 17.71 | 0.0023 |
| Residual | 3.14 | 9 | 0.35 | | |
| Lack of fit | 1.39 | 5 | 0.28 | 0.64 | 0.6875 |
| Pure error | 1.75 | 4 | 0.44 | | |
| Cor Total | 144.22 | 16 | | | |

R-squared = 0.9782, adj. R-square = 0.9612, pred. R- squared = 0.9386, adeq. precision = 23.488

TABLE 3. ANOVA TABLE FOR COMPOUNDING OF *ADENIA LOBATA* (USORO) TREATED WITH NaOH

| ***Source*** | ***Sum of Squares*** | ***df*** | ***Mean Squares*** | ***F Value*** | ***P-value Prob>F*** |
|---|---|---|---|---|---|
| Model | 124.43 | 7 | 17.78 | 38.18 | <0.0001 |
| A-Rotation Speed | 23.05 | 1 | 23.05 | 49.52 | <0.0001 |
| B-Temperature | 20.45 | 1 | 20.45 | 43.92 | <0.0001 |
| C-Time | 2.73 | 1 | 2.73 | 5.86 | 0.0386 |
| AB | 17.56 | 1 | 17.56 | 37.71 | <0.0002 |
| BC | 10.08 | 1 | 10.08 | 21.65 | <0.0012 |
| $B^2$ | 36.30 | 1 | 36.30 | 77.98 | <0.0001 |
| $C^2$ | 11.81 | 1 | 11.81 | 25.36 | 0.0007 |
| Residual | 4.19 | 9 | 0.47 | | |
| Lack of fit | 2.78 | 5 | 0.56 | 1.58 | 0.3383 |
| Pure error | 1.41 | 4 | 0.35 | | |
| Cor Total | 128.62 | 16 | | | |

R-squared = 0.9624, pred. R-squared = 0.8563 Adj. R-squared = 0.9420, Adeq. Precision = 19.11.

TABLE 4. ANOVA TABLE FOR COMPOUNDING OF *ADENIA LOBATA* (USORO) TREATED WITH ACETIC ANHYDRIDE.

| ***Source*** | ***Sum of Squares*** | ***df*** | ***Mean Squares*** | ***F Value*** | ***P-value Prob>F*** |
|---|---|---|---|---|---|
| Model | 133.54 | 7 | 19.08 | 65.92 | <0.0001 |
| A-Rotation Speed | 22.95 | 1 | 22.95 | 79.30 | <0.0001 |
| B-Temperature | 34.16 | 1 | 34.16 | 118.02 | <0.0001 |
| C-Time | 0.55 | 1 | 0.55 | 1.90 | 0.2009 |
| AB | 15.76 | 1 | 15.76 | 54.46 | <0.0001 |
| BC | 20.03 | 1 | 20.03 | 69.20 | <0.0001 |
| $B^2$ | 32.93 | 1 | 32.93 | 113.80 | <0.0001 |
| $C^2$ | 5.54 | 1 | 5.54 | 19.14 | 0.0018 |
| Residual | 2.60 | 9 | 0.29 | | |
| Lack of fit | 0.91 | 5 | 0.18 | 0.43 | 0.8092 |
| Pure error | 1.69 | 4 | 0.42 | | |
| Cor Total | 136.15 | 16 | | | |

R-Squared = 0.9809, Pred R-Squared = 0.9565, Adj. R-squared = 0.9660, adeq. Precision = 25.333.

TABLE 5. ANOVA TABLE FOR COMPOUNDING OF *ADENIA LOBATA* (USORO) TREATED WITH NITRIC ACID

| ***Source*** | ***Sum of Squares*** | ***df*** | ***Mean Squares*** | ***F Value*** | ***P-value Prob>F*** |
|---|---|---|---|---|---|
| Model | 132.21 | 7 | 18.89 | 59.98 | <0.0001 |
| A-Rotation Speed | 26.90 | 1 | 26.90 | 85.43 | <0.0001 |
| B-Temperature | 30.62 | 1 | 30.62 | 97.23 | <0.0001 |
| C-Time | 0.73 | 1 | 0.73 | 2.32 | 0.1617 |
| AB | 16.52 | 1 | 16.52 | 52.48 | <0.0001 |
| BC | 18.58 | 1 | 18.58 | 59.00 | <0.0001 |
| $B^2$ | 31.67 | 1 | 31.67 | 100.57 | <0.0001 |
| $C^2$ | 5.60 | 1 | 5.60 | 17.77 | 0.0023 |
| Residual | 2.83 | 9 | 0.31 | | |
| Lack of fit | 1.04 | 5 | 0.21 | 0.46 | 0.7893 |
| Pure error | 1.79 | 4 | 0.45 | | |
| Cor Total | 135.05 | 16 | | | |

R-Squared = 0.9790, Pred R-Squared = 0.9437, Adj. R-squared = 0.9627, adeq. Precision = 24.799.

TABLE 6. ANOVA TABLE FOR COMPOUNDING OF *ADENIA LOBATA* (USORO) TREATED WITH ZINC CHLORIDE

| ***Source*** | ***Sum of Squares*** | ***df*** | ***Mean Squares*** | ***F Value*** | ***P-value Prob>F*** |
|---|---|---|---|---|---|
| Model | 148.28 | 8 | 18.54 | 129.88 | <0.0001 |
| A-Rotation Speed | 35.28 | 1 | 35.28 | 247.21 | <0.0001 |
| B-Temperature | 33.13 | 1 | 33.13 | 232.14 | <0.0001 |
| C-Time | 3.25 | 1 | 3.25 | 22.78 | 0.0014 |
| AB | 14.78 | 1 | 14.78 | 103.59 | <0.0001 |
| AC | 4.43 | 1 | 4.43 | 31.05 | 0.0005 |
| BC | 20.03 | 1 | 20.03 | 140.32 | <0.0001 |
| $B^2$ | 25.84 | 1 | 25.84 | 181.04 | <0.0001 |
| $C^2$ | 9.67 | 1 | 9.67 | 67.79 | <0.0001 |
| Residual | 1.14 | 8 | 0.14 | | |
| Lack of fit | 0.56 | 4 | 0.14 | 0.96 | 0.5142 |
| Pure error | 0.58 | 4 | 0.15 | | |
| Cor Total | 149.43 | 16 | | | |

R-Squared = 0.9924, Pred R-Squared = 0.9584, Adj. R-squared = 0.9847, adeq. Precision = 36.012.

### *C. Model equations for the compounding process*

The model equation generated was mathematical representation of the compounding process. It was used to predict the response which is the tensile strength of the composite. It was presented in both actual and coded values. The coded values can only be used in predicting the response only when the factors are converted to the standard codes of -1, + 1 and "0" on the design matrix. Where "-1" represents the low value of the factorial range, "+1" represents the high value of the factorial range and "0" represents the center point. The two types of model equations can give an approximation that leads to the proper direction. The coefficients of the coded equations gave the relative contribution of each factor to the response because their units of measure have been removed, but actual equations cannot give such contributions because their coefficients depended on their unit of measure.

The predictive model equations which explain the compounding process for Adenia Lobta are shown both in actual and coded forms. The models were shown based on the chemicals used for pretreating the fiber before compounding and for the untreated fiber. The predictive models were all quadratic for all the chemicals used due to the quadratic terms included as confirmed by ANOVA. The coefficients of the factors varied because of the effect of the chemicals used on the fibers' strength.

The model equations are shown on equations 1 to 6 for both the untreated and treated fiber reinforced composites.

The Final equation in terms of coded factors for Untreated *Adenia lobata*;

Tensile strength (Mpa) = +40.21 +1.79A + 2.04B + 0.24C – 2. 11 AB –2.42 BC – 2.78$B^2$ – 1.21$C^2$ (2)

Final equation in terms of actual factors;

Tensile strength (Mpa) = - 1084.78947 + 1.88087 Rotation speed + 11.24136 Temperature + 9.73568 Time – 0.0105 Rotation speed. Temperature – 0.048450 Temperature. Time – 0.2847 Temperature$^2$ = 0.048389 Time$^2$ (3)

Final equation in terms of coded factors for *Adenia lobata* treated with Sodium Hydroxide;

Tensile strength (Mpa) = + 46.92 +1.70A + 2.60 B + 2.58C – 2.10AB -1.59BC – 2.93$B^2$ – 1.67$C^2$ (4)

Final equation in terms of Actual factors;

Tensile strength (Mpa) = - 1074.68697 + 1.8562 Rotation speed + 11.44373 Temperature + 7.52093 Time – 0.010475 Rotation speed. Temperature – 0.031750 Temperature. Time – 0.29322 Temperature$^2$ - 0.066889 Time$^2$ (5)

Final equation in terms of coded factors for *Adenia lobata* treated with Acetic Anhydride;

Tensile strength (Mpa) = + 46.56 + 1.69A + 2.07B + 0.26C – 1.98AB - 2.24BC – 2.79$B^2$ – 1.15$C^2$ (6)

Final equation in terms of Actual factors;

Tensile strength (Mpa) = - 1052.67954 + 1.77194 Rotation speed + 11.16772 Temperature + 9.03447 Time – 9.9250E - 003 Rotation speed Temperature – 0.044750 Temperature. Time – 0.02792 Temperature$^2$ - 0.045816 Time$^2$ (7)

Final equation in terms of coded factors for *adenia Lobata* treated with Nitric Acid;

Tensile strength (Mpa) = +43.65 + 1.83A + 1.96B + 0.30C – 2.03AB – 2.16BC – 2.74$B^2$ – 1.15$C^2$ (8)

Final equation in terms of Actual factors,

Tensile strength (Mpa) = - 1047.80362 + 1.81931 Rotation speed + 10.96665 Temperature + 8.76892 Time – 0.010762 Rotation speed. Temperature – 0.043100 Temperature. Time – 0.027387 Tempeature$^2$ – 0.046047 Time$^2$ (9)

Final equation in terms of coded factors for *Adenia lobata* treated with Zinc Chloride;

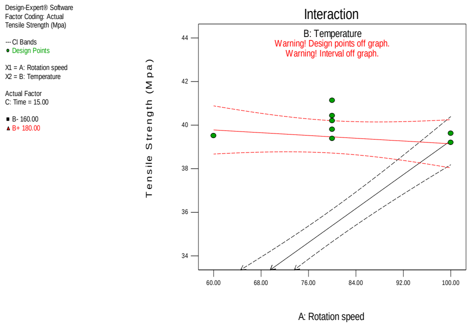
Tensile strength (Mpa) = + 40.58 + 2.10A + 2.03B + 0.64C – 1.92AB – 1.05AC – 2.24BC – 2.47$B^2$ – 1.51$C^2$ (10)

Final equation in terms of Actual factors;

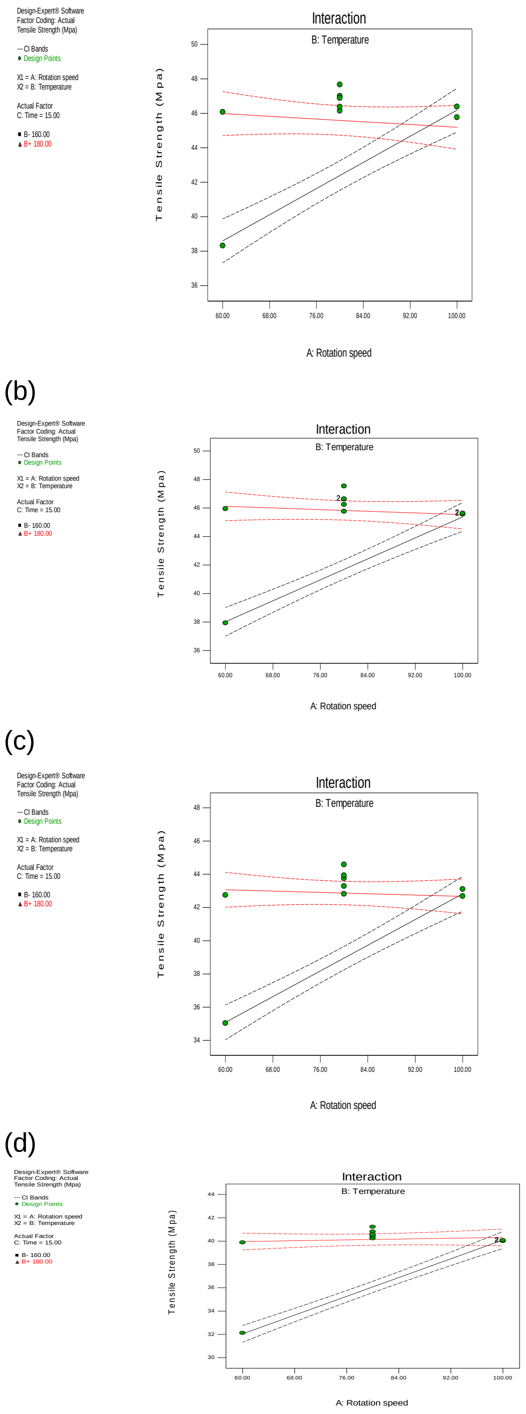
Tensile strength (Mpa) = - 990.31737 + 1.89700 Rotation speed + 10.05428 Temperature + 10.39342 Time – 9.61250E-003 Rotation speed Temperature- 0.010525 Rotation speed. Time – 0.044750 temperature time – 0.02473 temperature$^2$- 0.06547Time$^2$ (11)

### *D. Interaction effect of rotation speed and temperature*

The interaction effects were studied to ascertain the effect of a factor in combination of another. The interaction effect was studied for those factors considered significant by ANOVA. Interaction effect of temperature with rotation speed was considered significant by ANOVA for all the fibers, both untreated and treated. Interaction effect of temperature and rotation speed on the tensile strength of the resultant composite is shown on figure 1 for all the treated and untreated fiber reinforced composites.



(a)



(b)

(c)

(d)

(e)

Fig. 1. Interaction effect of temperature and rotation speed on the tensile strength of composite using *Adenia lobata* (a) untreated blend (b) treated with NaOH (c) treated with acetic anhydride (d) treated with nitric acid (e) treated with zinc chloride.

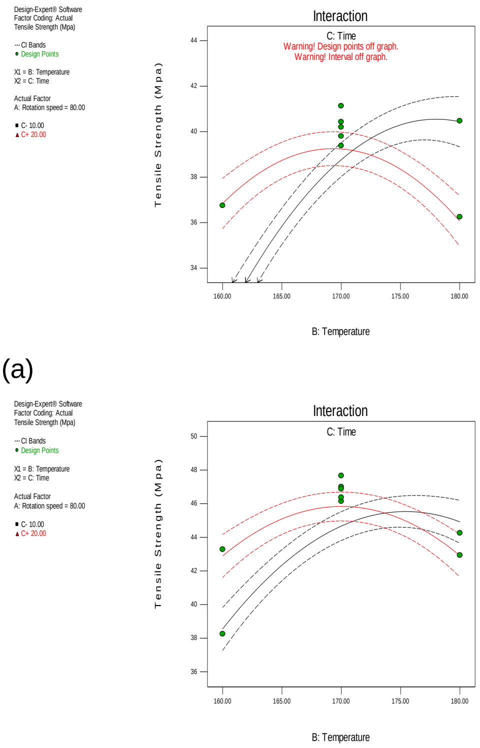
The interaction effect of temperature and rotation speed was studied at the midpoint of dispersion time of 15 minutes.

All the plots showed the same trend though with variations on the degree of the effect due to the nature of the fibers and the chemicals used for the pretreatments. The graphs showed that increase in rotation speed had no effect at higher temperature of $180^0$C, but had positive effect at lower temperature of $160^0$C. The red line showed high level of temperature ($180^0$C) while the black line showed low level of temperature ($160^0$C). At lower temperature, increase in rotation speed increased the inner temperature in the mixing chamber which increased the rheology of the polymer with consequent increase in the dispersion of the fiber on the polymer. At high temperature, increased rotation speed was not necessary because the chamber was hot enough to increase the flow of the polymer. This was why at high temperature, the increase in rotation speed did not have appreciable effect on the tensile strength of the fiber, and it led to distortion of the blend.

### *E. Interaction effect of temperature and time*

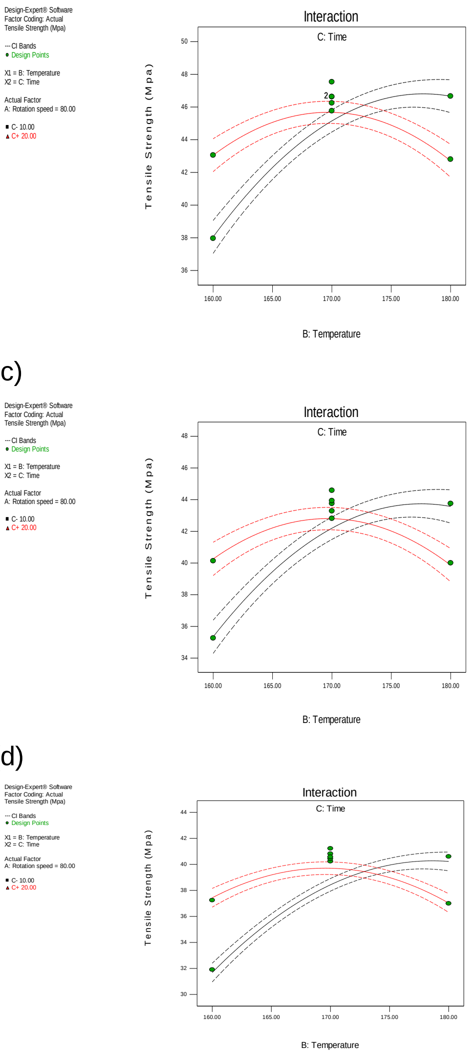
The interaction effect of temperature with time was studied to ascertain the temperature effect at both low and high dispersion time. The interaction effect was considered significant by ANOVA in all the untreated and treated fibers reinforced composite hence all were studied. The interaction effects of temperature with time for all the untreated and treated fibers are shown on figures 2. The effect was studied at the midpoint of rotation speed of 80rpm.

All the plots displayed similar behaviours for all the untreated and treated fibers with slight variation on the degree of the effect due different chemicals used for the pretreatment.



(a)

(b)



(c)

(d)

(e)

Figure 2. Interaction effect of temperature with time on the tensile strength of the composite using *adenia lobata* (a) untreated blend (b) treated with NaOH (c) treated with acetic anhydride (d) treated with nitric acid (e) treated with zinc chloride

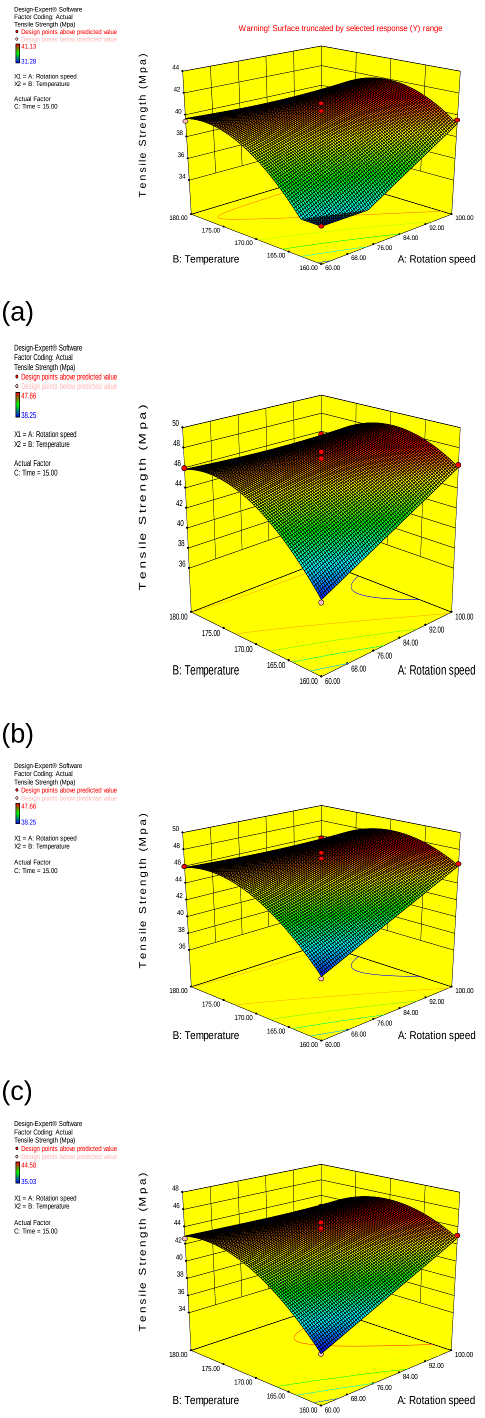
The plots showed increase in tensile strength with increase in temperature at low time of 10 minutes, with little curvature toward highest temperature of 180$^0$C. Increase in temperature at high time of 20 minutes showed pronounced curvature of decrease in tensile strength.

This showed that increase in temperature at higher time of 20 minutes slightly increased the tensile strength initially then with pronounced decrease on the tensile strength with further temperature increase. Temperature helped in melting the polymer for easier mixing with the fiber, but when the temperature was so high with prolonged mixing, it led to the decomposition and degradation of the fiber and the polymer.

At 10 minutes, tensile strength of the composites increased with increased in temperature, but decreased with increase in temperature at 20 minutes. Hence, shorter mixing time with high temperature or longer mixing time with low temperature is needed to improve the tensile strength of the resultant composites.

### *F. 3D Surface plot of interaction effect of temperature with rotation speed*

The interaction effect of temperature with rotation speed was studied using 3D surface plot. The plot for all the untreated and treated fibers is shown on figure 3. All the plots displayed the same shape for all the fibers reinforced composites. It bulged out showing maximization optimization, maximizing the response which is the tensile strength.

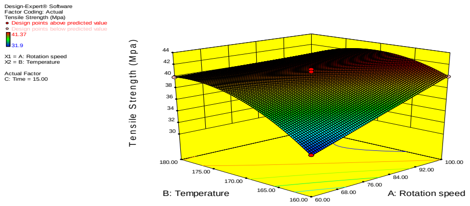


(a)

(b)

(c)

(d)



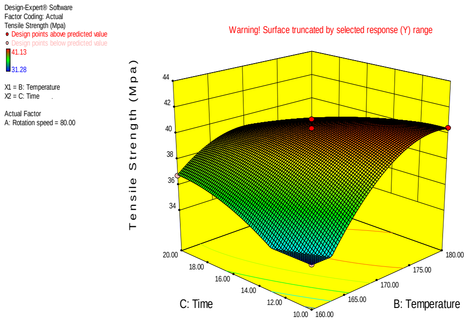
(e)

Figure 3. 3D surface plots of interaction effect of temperature with rotation speed on the tensile strength of *adenia lobata* composite (a) untreated blend (b) treated with NaOH (c) treated with acetic anhydride (d) treated with nitric acid (e) treated with zinc chloride.

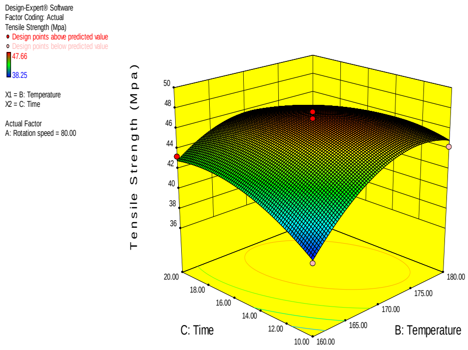
As the hill was climbed with increase in temperature and rotation speed, the tensile strength of the composites increased to the apex of the surface after which further increase in temperature and rotation speed resulted to decrease in tensile strength. Using the color coding, the colour was hottest at the apex of the hill, meaning that the tensile strength was highest at that point. Therefore the optimum conditions lied on the apex of the hill. The surface of the rotation speed was linear showing that tensile strength increased with increases in rotation speed at lower temperature range, but that of the temperature had curvature showing that temperature had limit at which when exceeded will result to decrease in tensile strength.

### *G. 3D Surface plots of interaction effect of temperature with time*

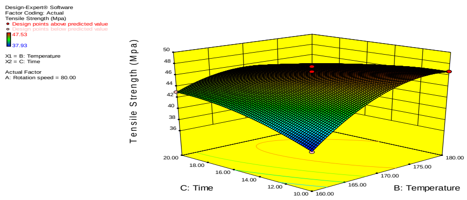
The 3D surface plot for the interaction effect of temperature with time was studied at the midpoint of rotation speed. The 3D surface plots for the untreated and treated fiber are shown in figure 4. All the plots displayed the same trend. It bulges out showing that it is a maximization optimization. The shape of the surface indicated that quadratic model explained the process well.



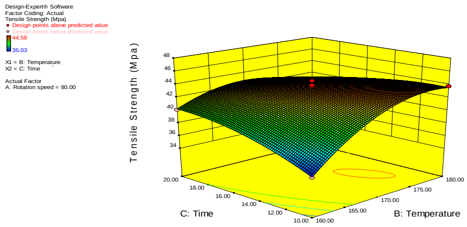
(a)



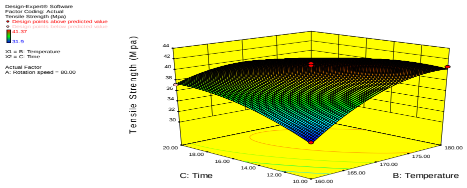
(b)



(c)



(d)



(e)

Figure 4. 3D surface plots of interaction effect of temperature with time on tensile strength of *adenia lobata* composites (a) untreated blend (b) treated with NaOH (c) treated with acetic anhydride (d) treated with nitric acid (e) treated with zinc chloride.

The color was hotter at the apex of the hill showing that tensile strength was highest there. Therefore, the optimum conditions lied on the apex. As time was increased with increase in temperature, the tensile strength of the composite increased to a point on the apex, after which further increase in time and temperature resulted to decrease on the tensile strength of the composites. This implies that high dispersion time can only lead to increase on tensile strength when done at low temperature. Higher compounding temperature required lower dispersion time to avoid degradation of the fiber and the polymer.

### *H. Optimum conditions for the compounding process*

The optimum conditions for the compounding of the polymer with the fibers were selected based on the factor settings with highest desirability. The goal that was met was the maximization of the tensile strength of the resultant composites after compounding.

The optimum conditions shown on table 7 were based on the chemicals used for the pretreatments with the untreated fibers.

TABLE 7. OPTIMUM COMPOUNDING CONDITIONS FOR *ADENIA LOBATA* FIBER-HDPE COMPOSITE

| ***Conditions*** | ***Untreated Fiber*** | ***Na0H Pretreated*** | ***Acetic Anhydride pretreated*** | ***Nitric acid pretreated*** | ***Zinc chloride pretreated*** |
|---|---|---|---|---|---|
| Rotation speed (rpm) | 99.28 | 90.91 | 96.18 | 95.85 | 94.48 |
| Temperature ($^0$C) | 175.25 | 173.09 | 174.87 | 175.56 | 173.94 |
| Time (minutes) | 11.83 | 14.57 | 14.56 | 13.33 | 11.03 |
| Desirability | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| Predicted value (Mpa) | 41.3409 | 48.4293 | 47.6806 | 44.8531 | 41.8011 |

### *I. Validation of the optimum conditions for the compounding process*

It is of paramount importance to validate the optimum conditions obtained from the model equation in order to ascertain its adequacy in predicting maximal response and to obtain its percentage deviation from the predicted optimum condition. The optimum conditions, predicted values, experimental values, and percentage errors as well as the desirability of the fibers are shown on table 8.

### *J. Scanning Electron Microscopy (SEM) and Energy-dispersive Xray Spectroscopy (EDS) Analyses*

Figure 9 shows micrographs of the surfaces of composite samples using SEM with EDS of both compounded and uncompounded composites. The morphological result in figure 9a shows that there was proper intimate mixing of the fiber with the polymer. Composites thus synthesized, revealed that the fiber was well wetted by the polymer and the surface was thick without voids. The micrographs of uncompounded composite (fig. 9b) revealed that there was no intimate mixing between the fiber and the polymer. This was evidenced by the presence of white dots which indicated the polymer separated from the fiber. This separation obviously led to lower tensile strength of the composite. The EDS analysis shows that there were no chemical interfacial reactions between the fiber and the HDPE.

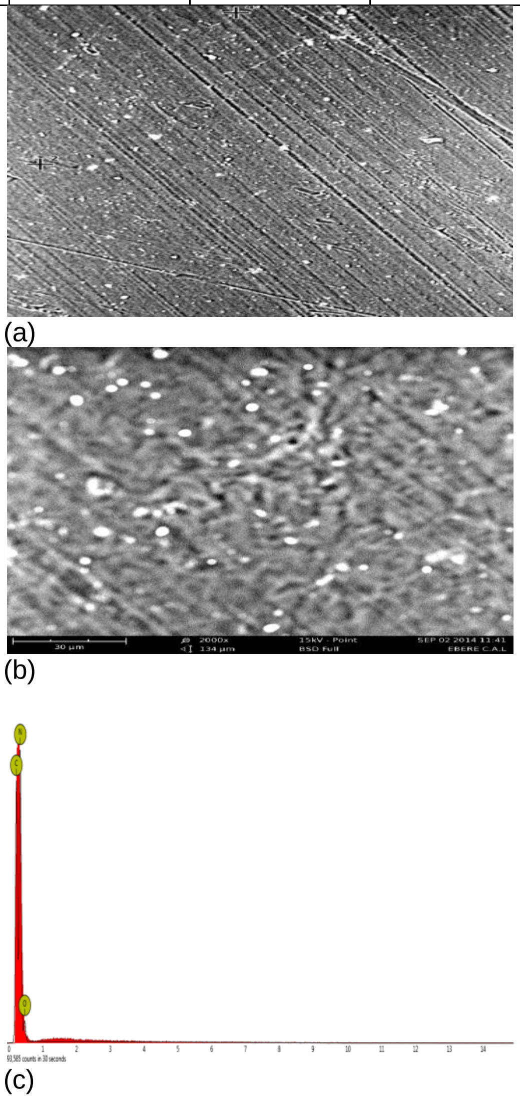

(a)
(b)
(c)

Figure 9. SEM and EDS analysis of the composite (a) compounded (b) uncompounded (c) EDS

## IV. CONCLUSION

Optimization of process conditions for the compounding of adenia lobata fiber –HDPE for compression molding process has successfully been carried out using Box behnken design (BBD).

The natural fiber was extracted from its plant stem and was treated with four chemicals; sodium hydroxide, acetic anhydride, nitric acid and zinc chloride to improve interfacial bonding with the polymer. Effect of temperature, time and rotation speed of two roll mill on the tensile strength of the resultant composite was studied. Effectiveness of the compounding process on the resultant blend was ascertained by preparing a compression molded composite from the blend, and analysing the tensile strengths. It was established that all the factors were significant except time. Quadratic model was developed for the process. The optimum conditions were validated with little error of less than 0.2%.The optimum conditions obtained were, for the untreated, 99.28rpm, 175.25°C, and time of 11.83. for NaoH treated, 90.91rpm, 173.09°C, and 14.57mins. for the acetic anhydride treated, 96.18rpm, 174.87°C, and time of 14.56mins. for nitric acid treated, 95.85rpm, 175.56°C, and time of 13.33mins. for zinc chloride treated, 94.48rpm, 173.94°C, and 11.03mins.

SEM analysis revealed that there was intimate mixing of the fiber with the polymer as a result of compounding process, while EDS revealed that there was no chemical interfacial reaction between the fiber and the matrix since the identified elements (Oxygen, carbon and nitrogen) were major elements of the fibers.

TABLE 8. VALIDATION OF THE COMPOUNDING OPTIMUM CONDITIONS FOR *ADENIA LOBATA* FIBER –HDPE COMPOSITE

| ***Model desirability*** | ***Chemical treatment*** | ***Rotation speed (rpm)*** | ***Temp. ($^0$C)*** | ***Time (minutes)*** | ***Tensile strength (Mpa)*** | | |
|---|---|---|---|---|---|---|---|
| | | | | | ***Predicted Values*** | ***Experimented Values*** | ***Error (%)*** |
| 1.0 | Untreated | 99.28 | 175.25 | 11.83 | 41.3409 | 41.3202 | 0.05 |
| 1.0 | NaoH | 90.91 | 173.09 | 14.57 | 47.4293 | 47.3818 | 0.10 |
| 1.0 | Acetic anhydride | 96.18 | 174.87 | 14.56 | 47.6806 | 47.489 | 0.4 |
| 1.0 | Nitric acid | 95.85 | 175.56 | 13.33 | 44.8531 | 44.8396 | 0.03 |
| 1.0 | Zinc chloride | 94.48 | 173.94 | 11.03 | 41.8011 | 41.7509 | 0.12 |

## ACKNOWLEDGMENT

The authors wish to thank PYMOTECH RESEARCH CENTRE AND LABORATORIES ENUGU, ENUGU STATE NIGERIA for all their facilities used throughout the research work.

## REFRENCES

[1]. Mansour Rokbi Hocine Osmani, Abdellatif Imad , Noureddine Benseddiq. "Effect of Chemical treatment on Flexure Properties of Natural Fiber-reinforced Polyester Composite". Procedia Engineering 10, 2092–2097, 2011.

[2]. Rout J, Misra M, Tripathy SS, Nayak SK, Mohanty AK. "The influence of fibre treatment on the performance of coirpolyester composites". *Compos Sci Technol.* 61:1303–1310, 2001.

[3] Herrera-Franco PJ, Valadez-González A. "A study of the mechanical properties of short natural-fiber reinforced composites".
*Compos Part B.* **36**:597–608,2005.

[4] Carmisciano S, De Rosa IM, Sarasini F, Tamburrano A, Valente M. "Basalt woven fiber reinforced vinylester composites: Flexural and electrical properties". *Mater Design,* **32**:337–42, 2011.

[5]. Bachtiar D., Sapuan S. M., Hamdan M. M. "The effect of alkaline treatment on tensile properties of sugar palm fibre reinforced epoxy composites". Materials and Design, 29, 1285–1290.DOI: 10.1016/j.matdes.2007.09.006, 2008.

[6] Bachtiar D., Salit M. S., Zainudin E., Abdan K., Mohd Dahlan K. Z. H. "Effects of alkaline treatment and a compatibilizing agent on tensile properties of sugar palm fibre-reinforced high impact polystyrene composites". BioResources, **6**, 4815–4823 (2011).

[7] Viet Cao X., Ismail H., Rashid A. A., Takeichi T., Vo- Huu T. "Mechanical properties and water absorption of kenaf powder filled recycled high density polyethylene/ natural rubber biocomposites using MAPE as a compatibilizer". BioResources, **6**, 3260–3271 (2011).

[8] Wirawan R., Sapuan S. M., Yunus R., Abdan K. "Properties of sugarcane bagasse/poly(vinyl chloride) composites after various treatments". Journal of Composite Materials, 45, 1667–1674 (2011).

[9] *Y. A. El-Shekeil, S. M. Sapuan, A. Khalina, E. S. Zainudin, O. M. Al-Shuja, (2012)* Influence of chemical treatment on the tensile properties of kenaf fiber reinforced thermoplastic polyurethane composite. *eXPRESS Polymer Letters Vol.6, No.12, 1032–1040*

[10] B. Sutharson, B., Rajendran, M., Devadasan, S.R., and Selvam, B. (2012) effect of chemical treatments on mechanical properties of jute fiber hybrid composite laminates. ARPN Journal of Engineering and Applied Sciences.

[11] H. Ku*, et al.*, "A review on the tensile properties of natural fiber reinforced polymer composites," *Composites Part B: Engineering,* vol. 42, pp. 856-873, 2011.

[12] George*, J. et al.*, "A review on interface modification and characterization of natural fiber reinforced plastic composites," *Polymer Engineering & Science,* vol. 41, pp. 1471-1485, 2001.

[13] D. Bas, I.H. Boyaci, Modeling and optimization I: usability of response surface methodology, J. Food Eng. 78 (2007) 836–845.

[14] D.C. Montgomery, Design and Analysis of Experiments, sixth ed., John Wiley & Son Inc., 2005.

[15]. Nurul Munirah Abdullah, Ishak Ahmed .(2012). Effect of chemical treatment on Mechanical and Water sorption properties of coconut fiber unsaturated polyester from Recycled PET. International scholarly Research Network. Vol 272, article ID 134683, 8 pages.

[16] Bledzki, A. K., Reihmane, S., Gassan, J. (1996). Properties and modification methods for vegetable fibers for natural fiber composites. Journal of Applied Polymer Science, 59(8), 1329-1336.

[17] Vantard, F., Fioux, P., Vidal, L. J., Dentze, J., Schultz, M., Nerdein, B., Defort (2013). Influence of an oxidation of carbon fibre surface by boiling nitric and on the adhesion strength in carbon fibre-acrylate composite cured by election beam. Surface and interface analysis Vol. 45, issue 3, pp 722-741.

[18]. Nadantangam, U., Karande, K, G. B., Hadge S.T., Mhaske, A.K., Bhari Malla (2013). Abstract No oral – 89. “Enzyme/Zncl pretreatment of short staple cotton fibers for energy reduction during neon fibrillation by refining process research conference on Technologies for property”, 2009.

[19]. Lu, J. Z, Q., Wu, H.S., McNabb. “Chemical coupling in wood fiber and polymer composites. A review of coupling agents and treatment”. Wood fiber Sci. 32(1): 88 – 104, 2000.

[20] ViJay Kumar Thakur, Amar singh Sigigha. “Physico-chemical and mechanical characterization of Natural fibre Reinforced polymer composites” Iranian polymer Journal 19(1), 3-16, 2009.

[21] Berlin, A. A., Volfson, S. A., Enikolopain, N. S., Negmaton, S. S. “Principles of Polymer composites”. Spring- Verlag, Berlin. Germany, pp 123, 1986.

[22] Mark J. Anderson, Patrick J. Whitcomb. “RSM Simplified optimizing processes using response surface methods for design of experiments” CRC press Taylor and Francis group Boca Raton London New York, 2005.

[23] Design expert software 8.0.7 guide, stat-ease, Inc East Hennepin Ave. Suite 480 Minneapolis, MN 55413, 2010.