Optimization of Processing Conditions Via Response Surface Methodology (RSM) of Nonwoven Flax Fibre Reinforced Acrodur Biocomposites

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Abstract

Currently, there is a significant drive to switch to more sustainable and renewable materials, whilst still reducing weight and cost and maintaining reliability. In addition, with some renewable materials, end-of-life vehicle issues are more easily addressed because the materials are biodegradable or easily recycled. Natural fibres such as flax fibre (FF) have several advantages that have made them particularly attractive to the automobile industry. These include relatively good mechanical strength, low density better thermal and acoustic insulation and low cost. The aim of this study was to produce optimised Nonwoven Flax Fibre Reinforced Acrodur (NWFA) biocomposites. Response surface methodology (RSM) was employed to study the effect of processing conditions such as moisture content, curing temperature and curing time on flexural strength and modulus. The optimized conditions was analyzed using Analysis of Variance (ANOVA) and the optimized value for the maximum flexural strength of NWFA biocomposites was found at 25% moisture content, 170°C curing temperature and 180 seconds curing time. Maximum flexural strength and modulus of 44.83 MPa and 4.70 GPa were attained. From the analysis of variance (ANOVA) technique, namely the Box–Behnken method, curing temperature significantly affects the strength of NWFA biocomposites,
followed by the moisture content and curing time. The P-value of the model of the experiment is less than 0.05 and the
determination coefficient ($R^2$) is nearly 1 suggesting that the model is significant and implies on the precision and processability
in the production.

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**Keywords:** Natural fibre; Flax; Response surface methodology; Analysis of variance; Box–Behnken

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**Nomenclature**

| Acronym | Description                  |
|---------|------------------------------|
| GF      | Glass Fibre                  |
| CF      | Carbon Fibre                 |
| FF      | Flax Fibre                   |
| RSM     | Response Surface Method      |
| NWFA    | Nonwoven Flax Fibre Reinforced Acrodur |

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**1. Introduction**

The use of natural fibre reinforced plastic composites has grown significantly in buildings, furniture
and automotive industries over the last decade because of their unique properties. Synthetic fibres, such as glass fibre
(GF) and carbon fibre (CF) have been utilized as reinforcements in composite materials in various automotive
components. However, the use of synthetic fibres has several drawbacks such as non-biodegradability and recycling
difficulty. Currently, there is a significant drive to switch to more sustainable and renewable materials, whilst still
reducing weight and cost and maintaining reliability. In addition, with some renewable materials, end of life vehicle
issues are more easily addressed because the materials are biodegradable or easily recycled. Natural fibres (NF)
such as jute, kenaf and flax have several advantages that have made them particularly attractive to the automotive
industry. These include relatively good mechanical strength, low weight, better thermal and acoustic insulation and
low cost. NF are poised to replace synthetic fibres in numerous interior parts i.e. door panels, seat backs, headliners, package trays, dashboards, seat backs, interior sunroof shields and headrests. Although many different
types of natural fibres are available, FF is a particularly attractive option due to its high strength, low density and
better environmental impact. As compared to glass fibre reinforced composite, flax composites have an advantage in
terms of specific mechanical properties. Furthermore, FF are cheap and biodegradable materials, coming from a
bio-sourced agriculture and widely available over the world.

Previously, most studies on natural fibres have been focussed on short, twisted or woven and produced either by
compression or hand lay-up technique. However, there is not much work reported on non-woven fibre mats. The
key of this study is to utilize the non-woven fibre mat-making machine, which involves the continuous processes of
fibre opening, carding, cross-lapping and needle punching to produce stitched kenaf fibre mats as natural fibre
reinforcement in composites. The term “non-woven” was created as a result of this manufacturing technique of
producing fabrics without a weaving or knitting process.

Response surface methodology (RSM) is a well-known up to date approach for constructing approximation
models to optimize a response (output variable) which is influenced by several independent variables (input
variables). Compared to conventional method the optimization properties for any industrial production of using RSM
have several advantages such as short time consuming by reducing the number of planned experiments and effective
in finding the optimum parameter particularly when it comes to the interactions of each variable. RSM has the
ability to evaluate the relationship between the responses and the independent variables as well as to define the
influence of independent variables on the responses either by each single variable or via combination in the process.

The purpose of the present work was used to investigate the effect of factors that determine the flexural
properties of NWFAbiocomposites. The factors involved are moisture content, curing temperature, curing time and
their interactions in composite fabrication process. This paper also present the optimization using RSM to quantify the effect of main processing conditions and their interactions on flexural properties of the composites and the optimum value of the moisture content, curing temperature and curing time will be suggested.

2. Experimental Procedures

2.1. Materials

Flax non-woven fibremat was supplied by EcoTechnilin SAS, France with average density value 1.37 g/cm³ and areal density of 1000 g/m². Acrodur® resin 950L was obtained from BASF. This resin is an aqueous acrylic resin based on a polycarboxylic acid and a polyalcohol free of phenol and formaldehyde.

2.2. Composites preparation

Prior to composite preparation, flax non-woven fibre mats were dried at 60 ± 5°C for 3h. The impregnation process was carried out by introducing the flax nonwoven mat to the impregnation line consisting of Acrodur® resin as illustrated in Fig 1. After impregnation the semi-finished material (prepreg) will be dried until residual moisture of approximately 15-35%. After drying process, the prepreg were then compression moulded using GOTECH GT7014-H hydraulic hot press machine. The curing time and temperature were varied from 180 s -310s and 170°C – 210°C while the pressure were kept constant at 30 bar.

![Diagram of impregnation process](image)

Fig. 1. Scheme of the impregnation process using an impregnation system.

2.3. Response surface methodology (RSM)

NWFA composite was prepared according to the experimental design. It was designed, analyzed and calculated using Minitab software 16 via RSM with three variables; moisture content (%), curing temperature (°C) and curing...
time (s) and the response are flexural properties. As three factors and three level of Box–Behnken designed were chosen, there will be 3 similar runs which is considered as centred point. Table 1 illustrates the range of variables from low (−1) and high (+1). The experiment consisted of 15 runs with an average of five replicates for each. Statistical analysis of the process was performed to evaluate the analysis of variance (ANOVA) and P-test.

### Table 1. Parameters for experimental design.

| Factor                | Level       | Low (-1) | High (+1) |
|-----------------------|-------------|----------|-----------|
| Moisture content (%)  | 15          | 35       |
| Curing temperature (°C) | 170       | 210       |
| Curing time (s)       | 180         | 310       |

2.4. Characterization of composites

The flexural strength and modulus of the composite were determined using the three-point bending test method following the ASTM D790 and maintaining a span to depth ratio of 16:1 using a universal testing machine (UTM) model Instron 3366 with a crosshead speed of 2 mm/min.

3. Results and discussion

In order to study the effect of processing conditions on the flexural properties of the NWFA composites, a full quadratic model for each response was selected based on the best fit of the experimental data. The statistical significance of the developed models was evaluated using an analysis of variance (ANOVA) and the accuracy of the models was further justified through a regression analysis, and normal plot of residuals. The experimental results obtained at different combinations of processing conditions are presented in Table 2. It can be seen that the optimum flexural strength and flexural modulus obtained are 44.83 MPa at 25% moisture content, 170°C curing temperature, 180 s curing time and 4.91 GPa at 15% moisture content, 170°C curing temperature, 270 s curing time. It is interesting to note that the highest value of each response was recorded at same curing temperature. Meanwhile, the minimum flexural strength and modulus at 19.19 MPa and 1.59 GPa, at 35% moisture content, 210°C curing temperature, 270 s curing time and 35% moisture content, 190°C curing temperature, 180 s curing time which are at the highest moisture content.

### Table 2. Actual and predicted values of flexural strength and modulus of NWFA biocomposites.

| Run | Block | Factor 1 | Factor 2 | Factor 3 | Response 1 | Response 2 |
|-----|-------|----------|----------|----------|------------|------------|
|     |       | Moisture content (%) | Curing temperature (°C) | Curing time (s) | Actual | Predicted | Actual | Predicted |
| 1   | 1     | 35       | 170      | 270      | 26.54 | 26.18 | 1.70 | 1.69  |
| 2   | 1     | 25       | 210      | 360      | 34.06 | 35.42 | 4.01 | 4.04  |
| 3   | 1     | 15       | 190      | 360      | 30.58 | 30.87 | 4.18 | 4.13  |
| 4   | 1     | 15       | 210      | 270      | 28.52 | 28.12 | 3.95 | 3.97  |
| 5   | 1     | 25       | 190      | 270      | 36.63 | 35.83 | 4.20 | 4.24  |
| 6   | 1     | 35       | 190      | 360      | 21.78 | 21.36 | 1.64 | 1.67  |
| 7   | 1     | 25       | 170      | 180      | 44.83 | 45.06 | 4.20 | 4.17  |
| 8   | 1     | 35       | 210      | 270      | 19.19 | 18.61 | 2.32 | 2.29  |
| 9   | 1     | 25       | 190      | 270      | 37.50 | 35.82 | 4.25 | 4.24  |
| 10  | 1     | 15       | 190      | 180      | 32.42 | 32.94 | 4.05 | 4.08  |
The analysis of variance (ANOVA) and estimated regression coefficient of the flexural strength and flexural modulus are tabulated in Table 3. It can be seen that the probability value (P-value) of the independent variables (moisture content as $X_1$, curing temperature as $X_2$ and curing time as $X_3$) of the processing conditions are less than 0.05 in all terms, i.e. linear, square and interaction. Meanwhile, the lacks of fit of P-value for all models are greater than 0.05, which is not significant. Both outcomes indicate that the terms in the model have significant effects on the responses\textsuperscript{16,17}. The determination coefficient, $R^2$ that fitted the model is 0.9840 for flexural strength and 0.9993 for flexural modulus. The model adequacies were justified by the $R^2$ values, in which the closer the $R^2$ value to 1, the more significant the model is to predicting the response\textsuperscript{18,19}. The $R^2$ values for each mechanical properties model was observed to approach 1. This suggests that all models were highly significant and indicate that the regression line perfectly fits the data\textsuperscript{19,20}.

Figure 2 (a) and (b) show the normal probability plots of the residual for the composites with respect to the flexural strength and flexural modulus, respectively. The residuals in each plot generate near the straight line, implying that the errors are distributed normally. Meanwhile, the residual plots versus fitted line for each response are shown in Figure 3 (a) and (b). The residuals are independently distributed with zero mean and a constant variance. The observation of the two plots of responses indicate that every single model suggested are adequate and satisfied.

| Residual Percent | Normal Probability Plot (response is Flexural Strength) | Normal Probability Plot (response is Flexural Modulus) |
|------------------|--------------------------------------------------------|-------------------------------------------------------|
| 11               | ![Normal Probability Plot](image1.png)                | ![Normal Probability Plot](image2.png)               |
| 12               | ![Normal Probability Plot](image3.png)                | ![Normal Probability Plot](image4.png)               |
| 13               | ![Normal Probability Plot](image5.png)                | ![Normal Probability Plot](image6.png)               |
| 14               | ![Normal Probability Plot](image7.png)                | ![Normal Probability Plot](image8.png)               |
| 15               | ![Normal Probability Plot](image9.png)                | ![Normal Probability Plot](image10.png)              |

Fig. 2. Normal probability plot of the residual for the composite for each mechanical response of (a) flexural strength; (b) flexural modulus.
Additionally, the significance of the model is indicated by the non-significant value of the lack of fit test of 0.175 for flexural strength and 0.252 for flexural modulus. Table 3 also reveals among the three independent variables, the effect of curing time is not significant on the flexural strength and flexural modulus of NWFA composite.

Table 3. Analysis of variance (ANOVA) for response surface quadratic models on the flexural properties of NWFA biocomposites

| Source          | Flexural Strength (MPa) | Flexural Modulus (GPa) |
|-----------------|-------------------------|------------------------|
|                 | Adj MS      | F       | P           | Adj MS      | F       | P           |
| Regression      | 132.650     | 111.02  | <0.001      | 2.445       | 1479.57 | <0.001      |
| Linear          | 101.392     | 84.86   | <0.001      | 4.053       | 2452.56 | <0.001      |
| X1              | 181.059     | 151.53  | <0.001      | 12.0898     | 7315.43 | <0.001      |
| X2              | 114.523     | 95.85   | <0.001      | 0.0649      | 39.28   | <0.001      |
| X3              | 8.595       | 7.19    | 0.025       | 0.0049      | 2.98    | 0.128       |
| Square          | 281.011     | 235.19  | <0.001      | 2.0868      | 1262.71 | <0.001      |
| X11             | 281.011     | 235.19  | <0.001      | 3.8980      | 2358.66 | <0.001      |
| X22             | 0.4418      |         |             | 267.31      |         | <0.001      |
| Interaction     | 78.063      | 65.33   | <0.001      | 0.3916      | 236.94  | <0.001      |
| X1X2            | 0.6207      |         | <0.001      | 375.56      |         | <0.001      |
| X1X3            | 70.588      | 65.33   | <0.001      | 0.1625      | 98.32   | <0.001      |
| Residual Error  | 1.195       |         | <0.001      | 0.0017      |         | <0.001      |
| Lack-of-Fit     | 1.454       | 5.05    | 0.175       | 0.0021      | 3.25    | 0.252       |
| Pure Error      | 0.288       |         |             | 0.0006      |         |             |

R² = 0.9840

R² = 0.9993
The polynomial regression equation which respectively relates the flexural strength and flexural modulus with all the variables (moisture content \(X_1\), curing temperature \(X_2\) and curing time \(X_3\)), are computed based on Minitab 16 software. The proposed equations of the model in coded units are represented by Equations 1 to 2 as follows:

\[
\text{Flexural Strength} = 158.453 + 3.8622X_1 - 0.85182X_2 - 0.4778X_3 - 0.0867X_1^2 + 0.0024X_2X_3
\]

\[
\text{Flexural Modulus} = 13.6923 + 0.0151X_1 - 0.0839X_2 + 0.0019X_3 - 0.0102X_2^2 - 0.0004X_3^3 + 0.0019X_1X_2 + 0.0001X_2X_3
\]

It should be emphasized that the equations above are only valid within the tested range conditions of \(15 < X_1 < 35\) \%, \(170 < X_2 < 210\) °C and \(180 < X_3 < 310\) sec. The regression coefficient value may signify the effect of the independent variables towards the responses. A higher regression coefficient reflects a more prominent effect of the variables to the corresponding response. Based on Equations 1 and 2, the square coefficient of the moisture content \(X_1^2\) exhibits the highest value, which indicates that this variable gives a significant effect to the flexural strength and flexural modulus of the reinforced composites.

The key features of using the RSM method is its ability to identify the combination of variable settings that jointly optimize a single response or a set of responses. In this study, the optimized combination of moisture content, curing temperature and curing time variables with the ability to provide excellent reinforcement to the non-woven composites are the main objectives. The reinforcing ability was measured by observing the highest response of flexural strength and flexural modulus. By using Minitab software, the optimization process required three factor values i.e. lower, upper and target in order to construct the desirability indices. In the optimization plane, the goals for all the responses are set to maximize and the target values are the highest values of each response obtained from the experimental results. Figure 4 depicts the optimization value of all the responses. Based on the analysis, the predicted optimum moisture content, curing temperature and curing time are 20\%, 170 °C and 180 s, respectively, with composite desirability equal to 1 for flexural strength and 0.9425 for flexural modulus.
4. Conclusions

The use of RSM via experimental design allowed the determination of optimum processing conditions for flexural strength and flexural strength of NWFA biocomposites. The response models generated by the ANOVA analysis are well satisfied and suggest that moisture content and curing temperature significantly influence the ability of NWFA biocomposites to yield highest flexural strength and flexural modulus. The determined optimum parameters are 20% moisture content, 170°C curing temperature and 180 seconds curing time. The optimization of those processing conditions results in reduced cost of production, and time and material savings.

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