Optimization of Process Parameters for Fabrication of Wool Fiber-Reinforced Polypropylene Composites with Respect to Mechanical Properties

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ABSTRACT

The present study focused optimizing the process parameters of compression molding with respect to mechanical properties for fabrication of wool fiber-reinforced polypropylene composites. An experiment was designed using the Box-Behnken method with three levels and three variables using temperature, time, and pressure, as independent variables and tensile, flexural, and impact strengths as dependent variables. The process conditions were optimized using response surface methodology with the Box-Behnken experimental design. Regression equations were obtained to analyze tensile strength, flexural strength, and impact strength and the optimum process parameters were identified. The results show that the optimum conditions for compression molding are 176°C, 7 min, and 35 bar.

INTRODUCTION

Natural fiber composites have attracted much attention from the stand point of protection of the environment from plastic disposal to conserving petroleum resources [1]. These composite materials find their way in several structural and non structural applications where they are not subjected to high loads. Poor wettability, high moisture absorption capability, insufficient adhesion between hydrophilic fiber and hydrophobic polymer, and improper processing methods and conditions are the key factors that limit the applications of these materials. Extensive research was carried out on improving the fiber matrix adhesion by various surface treatments [2-7]. However, these techniques were not cost effective and improvement in mechanical properties was limited in most of those cases. In addition to fiber/matrix adhesion, processing method and processing conditions are also the key elements that have profound influence on the mechanical properties of natural fiber composites, as natural fibers are thermally unstable.

Compression molding is a commonly used process for manufacturing thermoplastic composites. This process is effectively used for manufacturing large, thin, strong, stiff, and light weight fiber-reinforced composite materials [8]. The main advantage of compression molding is that the fibers are not subjected to any shear and it is easier to limit the damage of fibers' [9]. The quality of the final product is mainly dependent on the compression molding process parameters, because it significantly influences the properties and interfacial characteristics of the composites. Most important compression molding process parameters that influence the mechanical properties are temperature, pressure, and heating time. Therefore, suitable processing parameters must be carefully selected in order to yield the optimum composite products [10]. In practice, the optimization of the molding process variables is done on a trial-and-error basis. This depends on an engineer's experience and intuition. The trial-and-error method is time consuming and costly, because the efficiency depends greatly on the experience of the operator. Therefore, a study of optimization of compression molding process is necessary.

Currently, a vast resource of research materials is available on the potential of cellulose based-fibers obtained from renewable plant resources such as wood [11], coir [12], flax [13], sisal [14] and jute [15]. There is less literature availability on protein-fiber based composites [16-19]. Wool is a natural protein fiber possessing electrical insulating properties [20]. Hence an attempt was made in this research work to optimize the compression molding process with respect to mechanical properties of wool-fiber reinforced polypropylene composites based on the Box-Behnken design of experiment and response surface methodology of analysis.

MATERIALS AND METHODS

Materials

In this study, wool-fibers were used as a reinforcement material for the preparation of composites. Wool-fiber was procured from Tirupati Fibers, Panipat, India. Polypropylene staple fiber was used as a bonding agent for the composite fabrication which was obtained from Zenith fibers, Vadodara, India. Physical properties of these fibers are listed in *Table I*. The staple length and strength of wool-fibers were measured as per BISFA 1998 and ASTM D-3822-07 respectively.

TABLE I. Physical property of fibers.

Fiber	Staple Length (mm)	Fineness (Tex)	Strength (g/d)
Wool	45	2.17	1.5
Polypropylene	51	2.5	5.5

Fabrication of Composites

Wool-fibers were scoured using soap oil and soda ash at 60°C for 90 min at pH 8 to remove dirt, grease, and dry plant matter from the fleece. The scoured wool and polypropylene fibers were blended in 50:50 weight ratio by a laboratory scale blending machine. The Webs were prepared from wool and polypropylene blend using a Try Tex lab scale carding machine. The wool and polypropylene fiber blend was carded four times to get better fiber orientation in the carded web. The composites were produced based on the design of experiment and they were classified into three levels (namely, high [+], intermediate [0], and low [-1]) with different temperatures (165, 175, and 185°C), different time (7, 11, and 15 Min) and pressure (35, 40, and 45 Bar). The composites were produced in the compression molding machine. A sample of composite board is shown in *Figure 1*.

FIGURE 1. SEM image of wool-fiber reinforced polypropylene composites

TESTING OF SAMPLES

Tensile Strength

The tensile strength of the wool fiber-reinforced polypropylene composites was tested according to ASTM D 638. Samples were cut into dumbbell-shaped pieces. The testing was done in a standard laboratory atmosphere of 23°C ± 2°C (73.4°F ± 3.6°F) and 50 ± 5 percent relative humidity. A Universal Testing Machine (Instron 3345) was used at a cross-head speed of 50 mm/min. Specimens were positioned vertically in the grips of the testing machine. Grips were then tightened evenly and firmly to prevent any slippage keeping the gauge length at 50 mm.

Flexural Strength

Flexural testing was performed using a three-point bending method as per ASTM D790-03. Specimens were tested at the cross head speed of 50 mm/min at a temperature of 23°C and humidity of 50%. Flexural strength was determined using the following equation

$$\mathrm{FS} = \frac{3PL}{2bt^2} \quad (1)$$

where P is the maximum load, L is the span length, b and t are width and thickness of the specimen respectively.

Impact Strength

Notched Izod impact tests were carried out using a low energy instrumented Impact tester. Tests were carried out as per ASTM D256. The standard specimen for ASTM D256 is 64 x 12.7 x 3mm^3, and the depth under the notch is 10.2 mm. The pendulum impact testing machine ascertains the notch impact strength by shattering the V- notched specimen with a pendulum hammer measuring spent energy and relating to it to the cross section of the specimen.

Scanning Electron Microscope Study

The SEM study was used to identify the morphology of the wool fibers in the composite. The surface of the composites was examined using a high resolution Scanning Electron Microscope JEOL M JSM6360 with suitable accelerating voltage.

DESIGN OF EXPERIMENTS

Response surface methodology is an empirical modeling technique devoted to the evaluation of the relationship of a set of controlled experimental factors and observed results. It requires a prior knowledge of the process to achieve a statistical model.

A detailed account of this technique has been outlined [21-24]. This optimization process involves three major steps, performing the statistically designed experiments, estimating the coefficients in a mathematical model, and predicting the response and checking the adequacy of the model.

The significant variables like temperature, time and pressure were chosen as the critical variables and designated as X_1, X_2, and X_3 respectively. The low, middle, and high levels of each variable were designated as –, 0, and + respectively and depicted in *Table II.* Computation was carried out using multi regression analysis using the least squares method.

TABLE II. Levels and codes of variables for the Box-Behnken design.

Variables	Symbol		Coded levels		
	Uncoded	Coded	-1	0	+1
Temperature	X_1	X_1	165	175	185
Time	X_2	X_2	7	11	15
Pressure	X_3	X_3	35	40	45

The three test variables are coded according to the following Eq. (2):

$$Xi = \frac{Xi - Xo}{\Delta X}; i = 1,2,3 \tag{2}$$

where Xi is the coded value of the independent variable, x_i is the actual value of an independent variable; x_o is the actual value of an independent variable at the centre point and Δx is the change value of an independent variable. The Box-Behnken design consists of a set of points lying at the midpoint of each edge and the replicated centre point of the multidimensional cube. All experiments were performed in triplicate and the average of the process condition yields was taken as response.

In a system involving three significant independent variables X_1, X_2, and X_3, the mathematical relationship of the reaction on these variables can be approximated by the quadratic (second degree) polynomial equation which is shown in Eq. (3):

$$\begin{aligned} \mathrm{Y} = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2 \\ + \beta_{33} X_3^2 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 \\ + \beta_{23} X_2 X_3 + \varepsilon \end{aligned} \tag{3}$$

where Y is the predicted response, βo is the model constant, x_1; x_2 and x_3 are independent variables, β_1; β_2 and β_3 are linear coefficients, β_{12}, β_{13} and β_{23} are cross- product coefficients, and β_{11}; β_{22} and β_{33} are quadratic coefficients. The quality of fit of the polynomial model equation was expressed by coefficient of determination R^2.

The levels of variables like temperature, time, and pressure are chosen based on the melting point of the polypropylene used in this study. A multiple regression analysis is done to obtain the coefficients and the equation can be used to predict response. The degree of experiments chosen for this study was Box-Behnken [22-25], a fractional factorial design for three independent variables. It is responsible once the critical variables have been identified [21-24].

This design is preferred because relatively few experimental combinations of the variables are adequate to estimate potentially complex response functions [25]. A total of 17 experiments were necessary to estimate the 10 coefficients of the model. Using multiple linear regression analysis, the set of coefficients for its mechanical properties was calculated. The actual designs of experiments are given in *Table III.*

TABLE III. The Box-Behnken design with the actual values for three weight fractions and three levels for the mechanical properties of the wool fiber-reinforced polypropylene composites.

Run	X_1 Temp	X_2 Time	X_3 Pressure	Y_1 Tensile strength	Y_2 Flexural strength	Y_3 Impact strength
	^{0}C	Min	Bar	(Mpa)	(Mpa)	(Joules)
1	175	7	45	28	31.99	1.19
2	175	15	35	25.21	28.71	1.68
3	175	11	40	20.15	24.868	1.364
4	185	11	35	16.6	21.378	0.566
5	175	15	45	28.03	28.98	1.002
6	185	15	40	9.77	10.494	0.318
7	165	7	40	4.52	5.547	0.149
8	175	7	35	33	36.37	1.192
9	175	11	40	20.38	22.768	1.492
10	175	11	40	20.18	26.526	1.492
11	175	11	40	19.85	21.152	1.228
12	165	11	35	6.84	8.593	0.483
13	165	11	45	6.24	6.698	0.142
14	185	7	40	16.12	20.018	0.61
15	175	11	40	20.26	21.868	1.226
16	185	11	45	13.03	13.379	0.377
17	165	15	40	4.11	5.44	0.556

RESULTS AND DISCUSSION

Effect of Process Parameters on Tensile Strength

The response surface plots representing the tensile strength of wool fiber-reinforced polypropylene composites for molding temperatures ranging from 165°C to 185°C and time ranging from 7 min to 15 min are shown in *Figure 2(a). Figure 2(b)* displays the response surface plots representing the tensile strength of the wool fiber-reinforced polypropylene composites for molding pressure ranging from 35 to 45 bar and molding temperature 165-185°C. The response surface plots representing the tensile strength for pressure 35 to 45 bar and the molding time 7 - 15 minutes are shown in *Figure 2(c).*

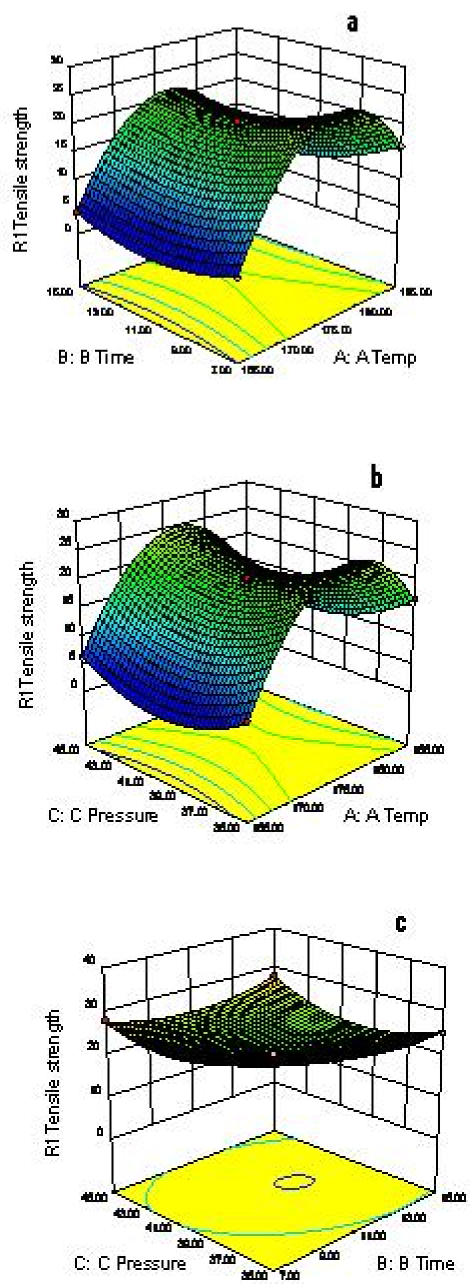

FIGURE 2. (a) Plot of effect of time and temperature on tensile strength (b) Plot of effect of pressure and temperature on tensile strength (c) Plot of effect of pressure and time on tensile strength.

It can be observed that when the compression molding temperature rises up to 175°C, the tensile strength also increases, and then it gradually decreases. This may be due to thermal degradation of the wool fibers. The thermal degradation of wool fiber leads to a non-reversible reduction of fiber strength, inevitability affecting the mechanical properties of the resultant composites.

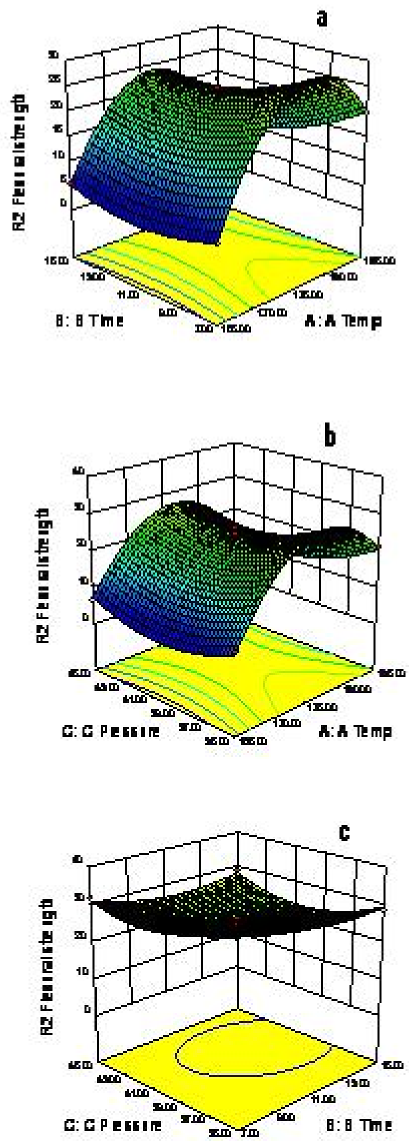

FIGURE 3. (a) Plot of effect of time and temperature on flexural strength (b) Plot of effect of pressure and temperature on flexural strength (c) Plot of effect of pressure and time on flexural strength.

In wool, the moisture evaporation by heat causes mass loss at around 55°C and thermal decomposition of fibers at around 230°C [26]. It can be observed that as the molding time increases the tensile strength reduces at the initial stage and later gradually increases. This may be the result of partial stress relaxation in the material. It can also be seen that as the molding pressure increases, the tensile strength initially decreases and then increases.

Effect of Process Parameters on Flexural Strength

The response surface plots representing the flexural strength of wool fiber-reinforced polypropylene composites for molding temperature ranging from 165 - 185°C and time ranging from 7 to 15 minutes are shown in *Figure 3(a).*

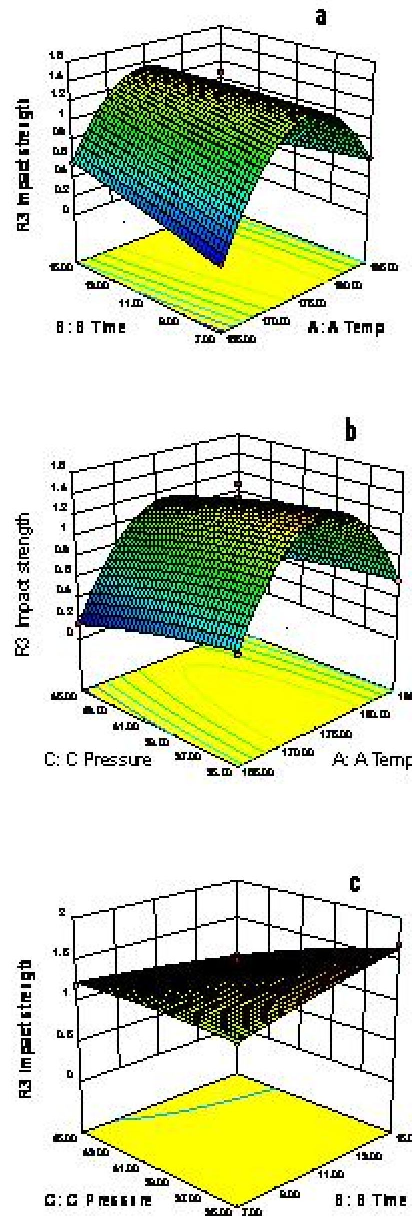

FIGURE 4. (a) Plot of effect of time and temperature on impact strength (b) Plot of effect of pressure and temperature on impact strength (c) Plot of effect of pressure and time on impact strength.

Figure 3(b) displays the response surface plots representing the flexural strength of the wool fiber-reinforced polypropylene composites for molding pressure ranging from 35 to 45 bar and molding temperature 165 - 185°C. The response surface plots representing the flexural strength for pressure 35 to 45 bar and the molding time 7 - 15 minutes are shown in *Figure 3(c).* It can be observed that as the molding temperature rises, the flexural strength also increases up to 175°C and then gradually reduces due to the thermal degradation of wool fibers. Also as the molding time increases, the flexural strength initially decreases and then increases. Similar effects of time and pressure on the flexural strength can be observed.

Effect of Process Parameters on Impact Strength

The response surface plots representing the impact strength of wool fiber-reinforced polypropylene composites for molding temperature ranging from 165 - 185°C and time ranging from 7 – 15 minutes are shown in *Figure 4(a). Figure 4(b)* displays the response surface plots representing the impact strength of the wool-fiber reinforced polypropylene composites for molding pressure ranging from 35 to 45 bar and molding temperature 165 - 185°C. The response surface plots representing the impact strength for pressure 35 to 45 bar and the molding time 7 - 15 minutes are shown in *Figure 4(c).*

The impact strength initially increases with increase in temperature then gradually reduces due to the thermal degradation of wool fibers. It can be observed that as the molding time increases the impact strength also increases. This may be a result of partial stress relaxation in the material. It can be also be seen that as the molding pressure increases, the impact strength reduces. This might be due to the fact that the increase in molding pressure leads to more compactness of the structure, which in turn, results in the formation of a higher crystalline region contributing to lower impact strength of the composite.

Structural Properties

The morphology of the wool-fiber reinforced polypropylene composite was investigated by an SEM study and the image is shown in *Figure 5.* It can be observed that the wool fibers are surrounded by the polypropylene matrix. Void content is also found in the composite samples which are manufactured with low time, temperature, and pressure leading to low tensile strength in composites.

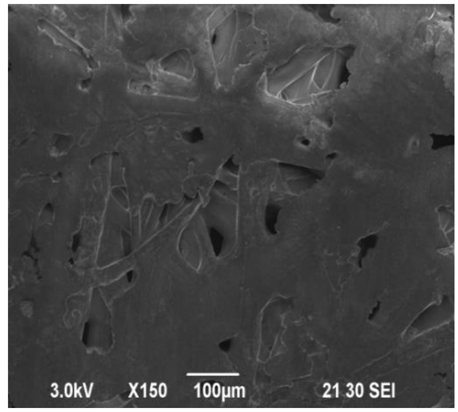

FIGURE 5. SEM Images of wool-fiber reinforced polypropylene composites.

Response Surface Models

The response surface model of tensile strength, flexural strength, and impact strength of wool-fiber reinforced polypropylene composites as functions of parameters of compression molding process, in coded value, is presented below:

$$\text{Tensile strength} = 20.16 + 4.23X_1 - 1.81X_2 - 0.79X_3 - 1.49X_1X_2 - 0.74X_1X_3 + 1.95X_2X_3 - 14.71X_1^2 + 3.17X_2^2 + 5.22X_3^2 \quad (4)$$

$$\text{Flexural strength} = 23.44 + 4.87X_1 - 2.54X_2 - 1.75X_3 - 2.35X_1X_2 - 1.53X_1X_3 + 1.16X_2X_3 - 16.03X_1^2 + 2.97X_2^2 + 5.11X_3^2 \quad (5)$$

$$\text{Impact strength} = 1.32 + 0.068X_1 - 0.052X_2 - 0.15X_3 - 0.17X_1X_2 + 0.038X_1X_3 - 0.17X_2X_3 - 0.89X_1^2 - 0.019X_2^2 + 0.036X_3^2 \quad (6)$$

The coefficients of determinations (r^2) of tensile, flexural and impact strengths are 0.99, 0.98, and 0.98 respectively. The higher correlation coefficients confirm the suitability of the models and correctness of the calculated constants. It can be seen that the relationships between tensile strength and process parameters are accounted for the variability of the data satisfactorily. The analysis of variance for the response surface model of mechanical properties is shown in *Table IV*. The relatively low P-value for the response surface models tensile, flexural and impact strengths, demonstrate their usefulness in predicting the future outcomes as functions of the chosen process parameters.

TABLE IV. Analysis of variance for the response surface model of mechanical properties.

Source	Sum of Squares	Degrees of freedom	Mean Square	F Value	P Value
Tensile strength					
Model	1224.67	9	136.07	1136.74	< 0.0001
Residual	0.84	7	0.12		
Cor Total	1225.51	16			
Flexural strength					
Model	1485.41	9	165.05	48.06	< 0.0001
Residual	24.04	7	3.43		
Cor Total	1509.45	16			
Impact strength					
Model	3.89	9	0.43	52.59	< 0.0001
Residual	0.058	7	8.230E-003		
Cor Total	3.95	16			

Optimization of Experiments

The design expert 8.1 (trial version) was used to optimize the process parameters involved in compression molding parameters for achieving better mechanical properties of wool-fiber reinforced polypropylene composites.

TABLE V. Results of verification test.

Performance measure	Predicted values	Experimental values	Error (%)
Tensile strength	33	31.32	5.0
Flexural strength	37.15	36.85	0.80
Impact strength	1.22	1.10	9.8

Good values of tensile, flexural, and impact strengths were obtained at 176°C, 7 minutes, and 35 bar based on the model equations. To test these models, experiments were performed by taking an arbitrary set of factor combinations and comparing them with the predicted values. *Table V* shows the verification of the test results. The predicted performance measures were compared with the experimental values, and a good agreement was observed. Thus the above mathematical models are important for fabrication of wool fiber-reinforced polypropylene composites for superior properties.

CONCLUSION

An integrated optimization approach based on the Box-Behnken design of experiments and response surface methodology was used for optimization of process parameters for fabrication of wool-fiber reinforced polypropylene composites with respect to mechanical properties. The results obtained in this study indicate that processing temperature has the most significant effect on all three performance measures considered in this work. Molding pressure and time were significant for tensile and flexural strengths, while they were insignificant for impact strength. Mathematical models for tensile, flexural, and impact strengths of compression molded wool fiber-reinforced polypropylene composites are successfully proposed for the selection of process parameters.

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