JOURNAL OF Applied Polymer SCIENCE

Optimal Formulation of Rice Husk Reinforced Polyethylene Composites for Mechanical Performance: A Mixture Design Approach

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ABSTRACT: Rice husk (RH) and linear medium density polyethylene (LMDPE) were used along with maleic anhydride grafted polyethylene (MAPE) to study the effects of component composition on the mechanical properties of the composites. Ten different blends along with four replicated blends were prepared with different selected percentages of RH, MAPE and LMDPE using mixture design approach. Trace and contour plots were used to examine the effects of RH, MAPE and LMDPE on the mechanical properties of the manufactured composites. Regression coefficients were also estimated for each fitted response (mechanical property). The results show that tensile and flexural properties of the composites improved with an increase in amount of RH, whereas Charpy impact strength decreased with increasing fibre loading. Tensile strength, flexural strength and Charpy impact strength increased with an increase in MAPE loading up to a certain percentage of MAPE, beyond which any further increase decreased these properties. The effect of MAPE on tensile and flexural modulus was not significant. The fitted models were used to optimise formulation of RH, MAPE and LMDPE for multiple responses for overall "best" mechanical properties. The optimal formulation for the overall "best" mechanical properties were found to be 50 wt% for RH, 4.1 wt% for MAPE and 45.9 wt% for LMDPE. The mechanical properties of the composite manufactured with this formulation closely matched the values predicted by the models. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40647.

KEYWORDS: thermoplastics; composites; mechanical properties

Received 3 January 2014; accepted 27 February 2014
DOI: 10.1002/app.40647

INTRODUCTION

Lignocellulosic thermoplastic composites (LTCs) are manufactured by combining agro-wastes with thermoplastics.[1] LTCs are generally produced from semi-crystalline polymers like polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC).[2] Recently, these composites have come forward as a new addition to composites manufacturing field.[2,3]

Among such lignocellulosic materials, rice husk (RH) is an agro-waste which is received as a by-product in bulk quantity during rice milling.[4–6] Rice is a source of primary food for over 40% of world population.[4] The annual production of rice in 2012 was approximately 718 million tons according to the Food and Agriculture Organization of the United Nation,[7] and almost 22 wt % of the rice paddy is yielded as husk during the milling process.[6] RH is either used as animal bedding, land filling, or burnt.[5] It is therefore important to look into a valuable usage of RH in the manufacturing of composites rather than treating it as a waste.

RH as fillers in polymer-based composite materials can be a good option, if a good compatibility between RH and base polymer matrix is ensured.[6] Like other lignocellulosic materials, RH is hydrophilic and its use with hydrophobic thermoplastics results in poor compatibility and adhesion between the counter parts. Fibre–matrix adhesion and interfacial bond strength can be improved by introducing compatibilizers,[8–12] which chemically link with the hydrophilic lignocellulosic fiber on one side, and facilitate the wetting of the hydrophobic polymer chain on the other side.[13] A strong interfacial bonding results in an efficient load transfer from the matrix to the fibers. Previous research proves that the compatibilizers improve mechanical properties of RH-reinforced composites.[13–15] RH has been used in different percentages by weight ranging from 5 wt % to 70 wt % with different polymers. The optimum mechanical properties resulted with RH loading between 30 wt % and 40 wt %.[16–18]

The main objective of this study was to conduct a complete parametric study on the manufacturability of RH-reinforced composites based on the resulting mechanical properties. This was probed by varying the percentages of each component/constituent material (RH, linear medium density polyethylene (LMDPE), and maleic anhydride grafted polyethylene (MAPE)) to (i) develop predictive models for each measured response, (ii) investigate the effects of each individual component/

constituent material on the measured properties, and (iii) optimize the formulation of components/constituent materials for overall "best" mechanical properties.

MATERIALS AND METHODS

Materials

RH was imported from Pakistan. The aspect ratio of RH was 4.0 and the mean length of the fibers was 5 mm. Before blending, the RH were dried at 100°C for 24 h to remove as much moisture as possible and the final moisture content after drying was 3.7%.

The thermoplastic polymer used in this research was LMDPE which was supplied by ICO Polymers in New Zealand with a melt flow index (MFI) of 6.0 g/10 min (190°C/ 2.16 kg), and a density of 0.932 g/cm^3.

MAPE supplied by Dupont in the name of Fusabond® A560 was used as compatibilizer. The MFI of MAPE was 5.6 g/10 min (190°C/2.16 kg), and the density was 0.93 g/cm^3.

Composites Manufacturing

RH was thoroughly mixed with LMDPE and MAPE in a mechanical mixer before compounding in a Lab-Tech co-rotating twin screw extruder. The speeds of the extruder and feeder were 50 rpm and 1.1 rpm, respectively. The temperatures of nine different heating zones of the extruder ranged from 170°C to 180°C, whereas the die temperature was 185°C. The extruded strands were pelletized and dried overnight at a temperature of 70°C before injection molded into appropriate test specimen for tensile, flexural, and Charpy impact tests according to relevant ASTM standards. The nozzle temperature and the injection pressure of the injection molding machine were 200°C and 75 bar, respectively.

Characterization

The tests for tensile properties (tensile strength (TS) and tensile modulus (TM)) of the composites were performed on a universal testing machine (Instron 1185) according to ASTM D638. The crosshead speed was kept at 5 mm/min. Each value obtained represented the average of five samples.

Flexural strength (FS) and flexural modulus (FM) tests were also conducted using Instron 1185 in accordance with ASTM D790. The crosshead speed was 1.3 mm/min and an average of five samples was taken for each blend.

Charpy impact tests were conducted on a Ceast pendulum impact tester according to ASTM D 6110. The energy of the hammer was 1 J and an average of five samples was taken for each blend.

Scanning electron microscopy (SEM) of the fractured surfaces was performed using FEI Quanta 200 environmental SEM, to study the fiber distribution, fiber pull out, and fiber–matrix adhesion within the composites.

Design of Experiments

Mixture design has been selected to design experiments and analyze the effects of component proportions on mechanical properties of manufactured RH/LMDPE composites.

Suppose that q represents the number of components of a composite blend and let x_i represent the fractional proportion of the ith component in the blend, then

$$0 \le x_i \le 1, \; i = 1, 2 \ldots, q \tag{1}$$

and;

$$\sum_{i=1}^{q} x_i = x_1 + x_2 + \ldots + x_q = 1.0 \tag{2}$$

The design for this study was a three-component mixture with lower and upper bound constraints. The three components were RH (X_1), MAPE (X_2), and LMDPE (X_3). The lower and upper limits were selected as 15 wt % and 50 wt %, 1 wt % and 6 wt %, and 44 wt % and 84 wt % for the three components respectively, known as "actual" components. These limits were chosen for three reasons. First, no composite could be manufactured with either RH or MAPE alone or by mixing just these two components. Therefore, there was a restriction on the component proportions which did not allow exploration of the entire mixture space. Secondly, the initial results and previous research work in the area of RH/PE composites provided the guidelines for selecting the upper and lower limits of the components, where an increase in RH loading increased mechanical properties of the composites. Third, the composites that having RH percentage of more than 50 wt % reduced the MFI of the blends which made it impossible to perform injection molding with above 50 wt % of RH.

According to eqs. (1) and (2), the lower and upper limits for each component were designated to be 0.15 and 0.50 for RH, 0.01 and 0.06 for MAPE, and 0.44 and 0.84 for LMDPE. These are known as the "real" component values. In addition to that, in mixture problems having lower and upper limits, the "pseudo" component values (a set of values from 0 to 1 over the mixture design region) were introduced and used in the mixture design calculations. This transformation from "real" to "pseudo" component values makes coefficients for different components comparable in size,[19] since the use of "actual" or "real" component values in model fitting could have serious effects on least square estimators of regression coefficients in a sense that the variances of these coefficient estimators are inflated.[20] The pseudo component values (PCVs) were employed to fit the models and construct the trace and contour plots.

Ten different blends along with four replicate blends were selected to manufacture and test the composites using mixture design approach. The replicate blends were used to check the repeatability of the experiments.

Data Analysis

The experiments were designed and analyzed in Design Expert® version 8.0. Data from mechanical results were used to fit to the Scheffe's canonical special cubic equation:

$$Y = \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{123} X_1 X_2 X_3 \tag{3}$$

where Y is the measured response (mechanical property), β_s are the coefficients estimated for each linear and cross-product term of the model for RH (X_1), MAPE (X_2), and LMDPE (X_3), respectively. The models were subjected to analysis of variance (ANOVA) to assess the level of significance. The selected model

Table I. Mechanical Properties of the Composites

	Real components			Pseudo components[a]			Measured responses				
Blends	RH	MAPE	LMDPE	RH	MAPE	LMDPE	Tensile strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Charpy impact strength (J/m)
1	0.50	0.060	0.440	0.875	0.125	0.00	17.0	1863	28.0	1,233	79.2
2	0.50	0.010	0.490	0.875	0.00	0.125	16.3	1923	28.5	1,253	64.8
3	0.413	0.022	0.565	0.656	0.031	0.313	16.6	1663	26.3	1,072	92.6
4	0.325	0.060	0.615	0.437	0.125	0.438	15.5	1295	22.1	798	113.2
5	0.325	0.010	0.665	0.437	0.00	0.563	14.9	1318	22.6	814	100.4
6	0.267	0.043	0.690	0.292	0.083	0.625	15.1	1141	20.2	676	130.7
7	0.238	0.022	0.740	0.219	0.031	0.750	14.7	1094	19.9	668	134.9
8	0.150	0.060	0.790	0.00	0.125	0.875	13.4	775	15.7	507	188.0
9	0.150	0.035	0.815	0.00	0.063	0.938	13.7	784	16.3	519	188.8
10	0.150	0.010	0.840	0.00	0.00	1.00	12.8	798	16.1	541	162
11[b]	0.50	0.060	0.440	0.875	0.125	0.00	17.1	1837	27.8	1,222	78.1
12[b]	0.50	0.010	0.490	0.875	0.00	0.125	16.4	1941	28.7	1,270	64.4
13[b]	0.150	0.060	0.790	0.00	0.125	0.875	13.5	788	15.4	475	194.4
14[b]	0.150	0.010	0.840	0.00	0.00	1.00	13.0	840	16.0	533	168.5

[a] Calculated as $X_i' = \frac{X_i - L_i}{1 - \sum L_i}$, where X_i' is the ith pseudo-component; X_i is the real component value; L_i is the lower limit for the ith component and $\sum L_i$ is the sum of all lower limits for all the components in the design.
[b] Replicate blends.

should have significant *P*-value (less than 0.05), an insignificant lack-of-fit *P*-value (greater than 0.10) and coefficient of determination (R^2) close to 1.0.[19–21]

RESULTS AND DISCUSSION

The results for the mechanical properties for the 14 blends (in terms of real and PCVs) are shown in Table I. Table II shows regression coefficients and analysis of variance (ANOVA) of mixture design models for the measured responses. All the developed models were statistically significant as they all had significant *P*-values, insignificant lack-of-fit *P*-values, and R^2 reasonably close to 1.0, which suggested that these models could adequately predict the responses. The statistical model for FM (Y_4) shown in Table II has been simplified after eliminating insignificant terms (β_{12} and β_{23}), whose *P*-value of more than 0.10 is deemed to be insignificant.[22]

Table II. Regression Coefficients and ANOVA of Mixture Design Models[a]

	Responses[b]				
Coefficients	Y_1	Y_2	Y_3	Y_4[c]	Y_5
β_1	16.7	2074.3	30.25	1402.54	65.33
β_2	−125.3	361.1	−108.65	71.16	−1491.34
β_3	12.89	821.52	16.06	542.0	165.95
β_{12}	165.48	–	137.33	–	1896.70
β_{13}	1.6	–	1.7	−317.51	−102.83
β_{23}	162.57	–	137.39	–	2115.82
β_{123}	–	–	–	–	–
Model *P*-value	<0.01	<0.01	<0.01	<0.01	<0.01
Lack-of-fit *P*-value	0.15	0.23	0.11	0.12	0.19
R^2	0.9	0.9	0.9	0.9	0.9

[a] β_1 = Rice husk, β_2 = MAPE, β_3 = LMDPE.
[b] Y_1 = Tensile strength, Y_2 = Tensile modulus, Y_3 = Flexural strength, Y_4 = Flexural modulus, Y_5 = Charpy impact strength.
[c] Simplified model.

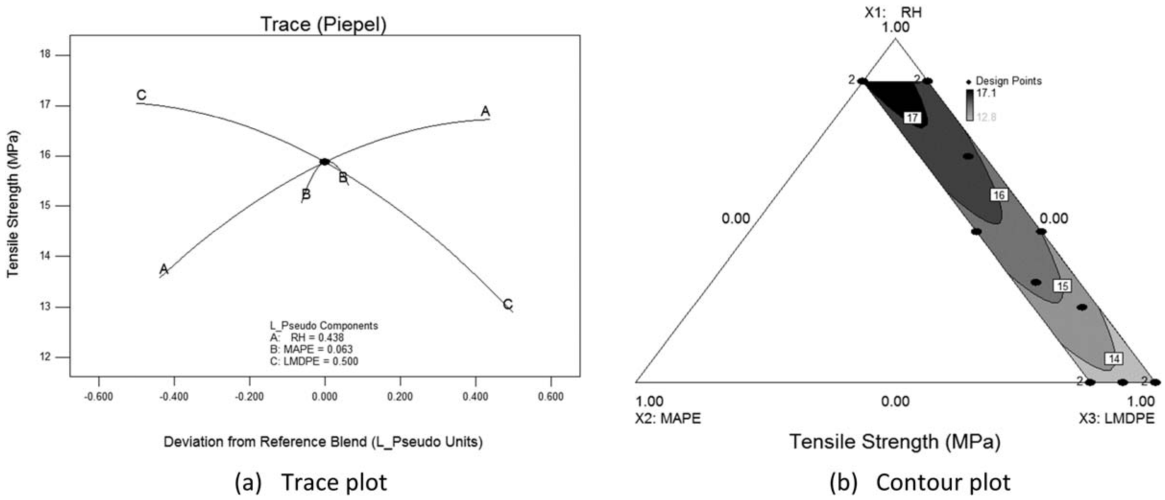

Figure 1. Trace plot and contour plot for TS.

In the following sections, the effects of the individual components (RH, MAPE, and LMDPE) on each mechanical property have been discussed in detail using trace and contour plots.

Tensile Strength

The effects of components on TS are shown in Figure 1. From the trace plot [Figure 1(a)] and contour plot [Figure 1(b)], it can be seen that TS increased with an increase of the RH percentage (Component A). As the percentage of RH increased, the stress transfer from LMDPE to RH improved which in turn improved the TS of the composites. There was an increase of 26% in TS when RH loading was increased from 15 wt % to 50 wt %.

MAPE, Component B, improved the TS with an increase in loading up to around 4 wt %, beyond which any further increase started to decrease the TS of the composites. MAPE was varied from 0.0 to 0.13 in terms of PCVs, and the maximum increase in the TS was achieved around 0.07, which corresponds to around 4 wt % in terms of actual components. This trend can be seen in the contour plot [Figure 1(b)]. Effective mechanical reinforcement by incorporation of fibers cannot be achieved without good interfacial bond strength. Through the utilization of compatibilizers, a bridge can be made between the hydrophilic fibers and hydrophobic matrix system, which results in substantial improvements in strength.[23,24] The compatibilizer chemically bonds to available OH groups on the fiber surface and then adheres to the matrix through molecular chain entanglement.[25] As the percentage of the MAPE was increased beyond 4 wt %, the strength of the composites started to decrease. Owing to the low molecular weight of MAPE, the chain entanglement of LMDPE and MAPE was nearly implausible, hence resulting in the interaction between the two dominated primarily by van der Waal forces.[26] The anhydride units in MAPE couple with the cellulose in the lignocellulosic fibers with equal probability and maintain the loop conformation within the fiber/matrix system. Any excess MAPE not coupled with RH helps fiber–matrix adhesion, therefore, proved harmful for the composites. The effect of different percentages of MAPE on the composites is shown in SEM micrographs of composites fractured surfaces (Figure 2) with 15 wt % of RH in each case. The SEM micrographs clearly show a better bonding between RH and LMDPE at MAPE loading of 3.5 wt % as compared to 1 wt % and 6 wt %. The composites with a low percentage of MAPE [Figure 2(a)] have voids and poor interfacial adhesion than those having 3.5 wt % of MAPE. At a high percentage of MAPE (6 wt %), the creation of voids between RH and LMDPE [Figure 2(c)] is probably due to the presence of excess anhydride units of MAPE which could not couple with RH.

Tensile Modulus

The effects of Components A, B, and C are shown in the trace and contour plots [Figure 3]. The effect of Component A, RH, was positive for TM. The TM increased by around 140% with an increase in RH loading from 15 wt % to 50 wt %. The main purpose of incorporating fibers into the polymer matrix is to improve the modulus of the composites.[27] As expected, the TM, which indicates the material stiffness, increased steadily with an increase in the fiber content. This is a common behavior when rigid fibers are incorporated into softer polymer matrices.[28] The increase in the TM is attributed to the reinforcing effect created by RH, which increased with an increase in RH loading. Similar results have been observed with PP where an increase in RH increased the TM of the composites.[29]

The variation of MAPE loading, Component B, as shown in the trace and contour plots, did not display any noticeable effect on the TM of the manufactured composites. Similar results have been reported,[23,30,31] as the TM is mainly dependant on the initial strain of the compound and is measured as the slope of the stress–strain curve at the initial stage and, therefore, is practically not much influenced by the existence of compatibiliser.[32] TM decreased to only around 3% with an increase in MAPE loading from 1 wt % to 6 wt %.

Flexural Strength

Figure 4 shows the effects of RH, MAPE, and LMDPE on FS. The results remain similar to those of TS. FS increased with increase in RH loading (Component A). Similar to TS, the increase in FS with increase in RH loading could be attributed to an improved stress transfer from LMDPE to RH. There was an increase of

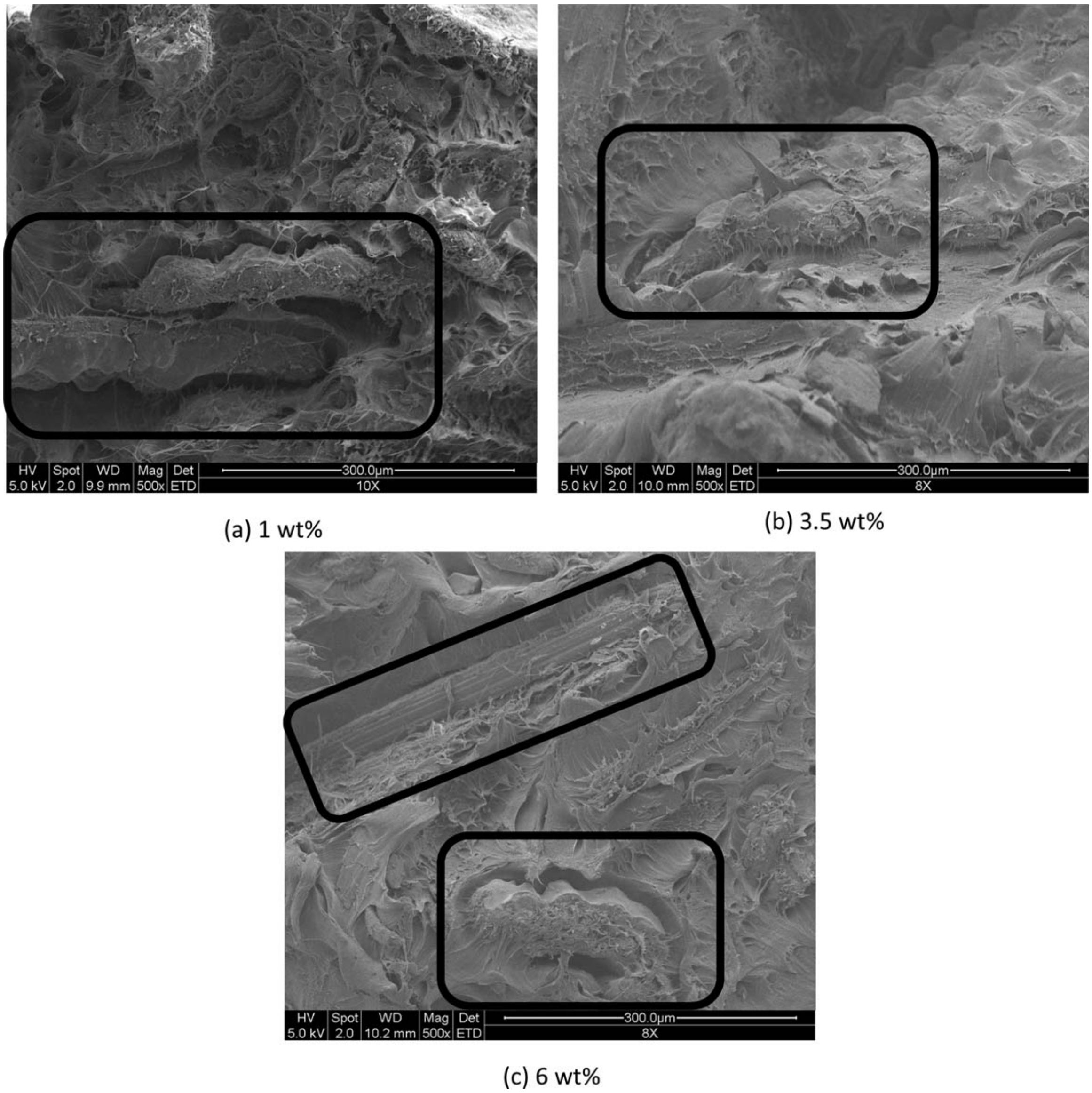

Figure 2. Effect of MAPE on composites.

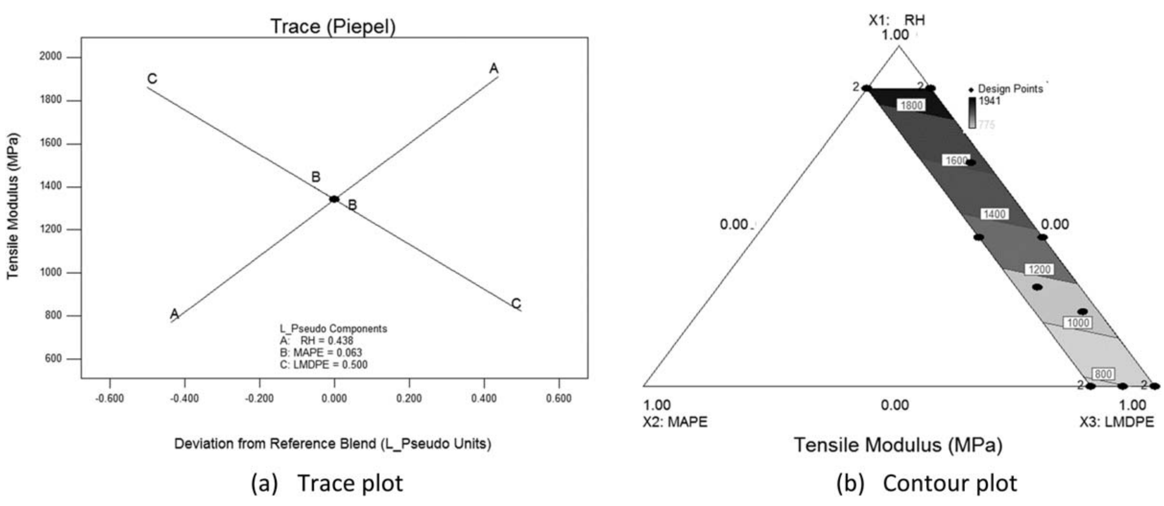

Figure 3. Trace plot and contour plot for TM.

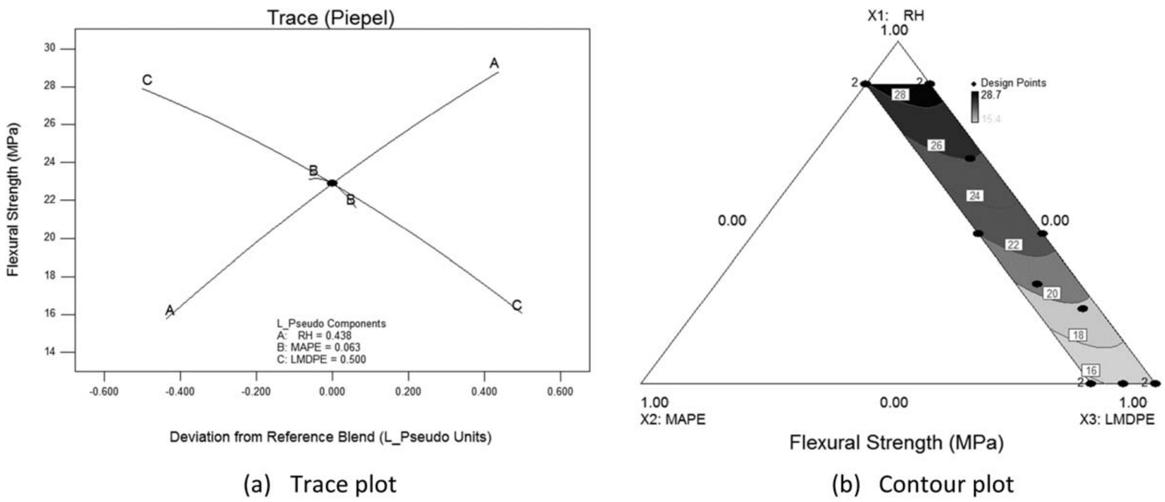

Figure 4. Trace plot and contour plot for FS.

around 78% in the FS of the composites with RH loading of 50 wt % as compared to 15 wt % of RH loading.

The effect of MAPE (Component B) was also similar to TS. The FS of the composites increased with an increase in MAPE until a plateau region was achieved (around 3 wt %, 0.005 in terms of PCVs), beyond which any further increase started to decrease the FS. This increase in FS can be attributed to an improved fiber–matrix interfacial adhesion.[33] This improved adhesion is due to the ability of anhydride groups to react with the hydroxyl groups of the fibres.[34] MAPE chains became more involved in inter-chain entanglements and contributed to an increased strength by ensuring the mechanical continuity of the system. At higher percentage of MAPE, beyond 3 wt %, the strength started to decrease with further addition of MAPE since there were more anhydride groups available than required to couple with the hydroxyl group of RH which proved harmful for the composites.

Flexural Modulus

The results of the effects of RH, MAPE, and LMDPE on FM (Figure 5) were similar to those of TM. FM increased with RH loading (Component A). Modulus is one of the basic properties of the composites generally considered as a linear function of the filler volume fraction.[32] FM increased with increase in the fiber loading and this increase generally depends upon fiber characteristics like aspect ratio and fiber wetting. This enhancement is the result of the fibers exerting a resistance against the plastic deformation of the matrix, which restricts polymer chain elongation.[23] The increase in the FM by an increase in RH loading has been reported in the literature.[29] The FM increased to 135% when the RH loading was increased from 15 wt % to 50 wt %.

Figure 5 shows that FM is more or less independent of the MAPE loading (Component B). FM decreased to just 3% with an increase in MAPE loading from 1 wt % to 6 wt %. As mentioned earlier, MAPE improved fiber–matrix interface, but this improvement in fiber–matrix interface did not pose any significant effect on the value of FM. The previous literature also shows no particular dependence of FM on the compatibilizer.[35,36]

Charpy Impact Strength

The effects of components on Charpy impact strength (CIS) are shown in Figure 6. CIS decreased with an increase in RH loading. As the content of RH increased, the composites got brittle which

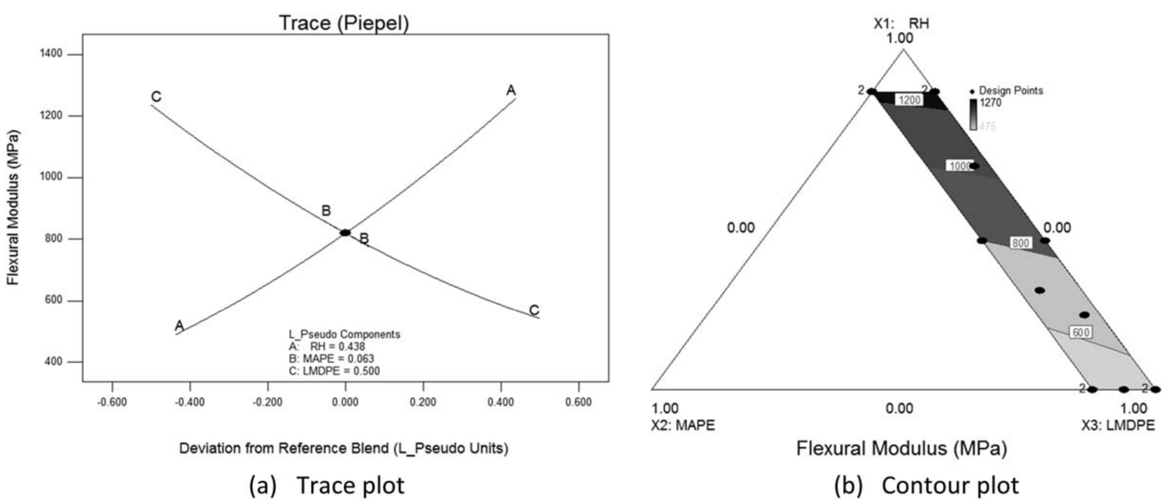

Figure 5. Trace plot and contour plot for FM.

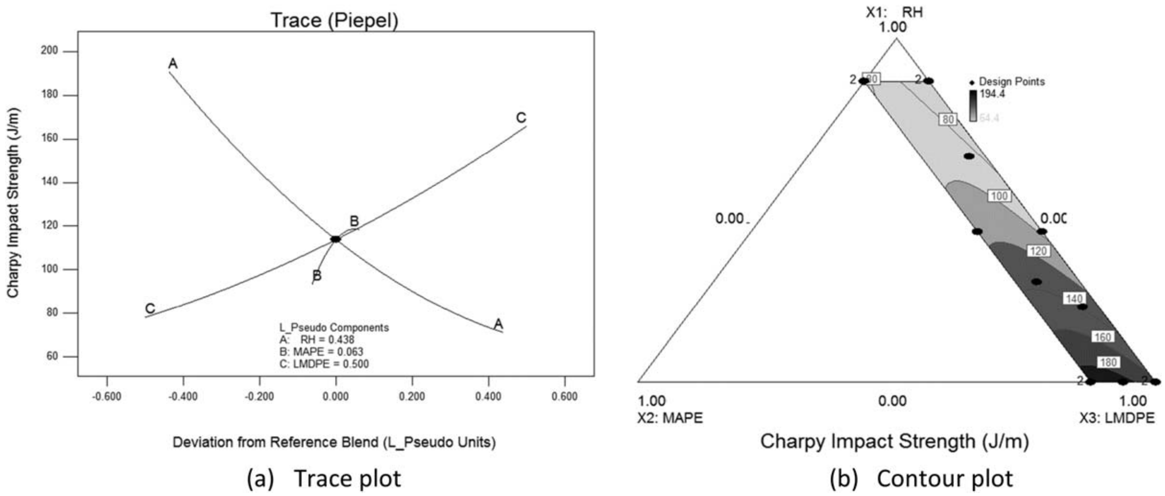

Figure 6. Trace and contour plot for CIS.

allowed for cracks to propagate more easily and with less energy than in the composites having low percentage of RH. The points of stress concentrations increase as the fiber content is increased which easily propagate the crack growth along the relatively weak interphase between the fibers and the matrix.[23] CIS decreased to around 65% when RH content was increased from 15 wt % to 50 wt %. The failure mechanism of the composites at high percentage of RH (50 wt %) is shown in the SEM micrograph in Figure 7. The micrograph shows that the failure of the composites occurred due to crazing of the polymer, fiber pull out, and debonding.

MAPE played an important role in determining the composites failure due to impact energy (Figure 6). The impact strength increased with an increase in MAPE loading up to around 5 wt % (around 0.9 in terms of PCVs) and then started to decrease [Figure 6(b)]. Too much interaction between the fiber and the matrix as well as a very poor fiber–matrix interaction leads to poor impact strength. Too much interaction leads to brittle failure, whereas a poor interaction leads to easy fiber pull out.[32] The incorporation of MAPE improved fiber distribution and wettability within the composites which led to an increase in the energy required for crack initiation, fiber de-bonding as well as fiber pull-out. At high MAPE content, the strength started to decrease. This was due to the migration of excess amount of compatibilizer around the fibers rather than the polymer matrix causing self-entanglement among the compatibilizer chains.[36,37]

Optimal Formulation of Components for Mechanical Properties

The regression models for TS, TM, FS, FM, and CIS in terms of actual components are:

$$TS = 0.164X_1 - 9.26X_2 + 0.0994X_3 + 0.103X_1X_2 + 0.001X_1X_3 + 0.102X_2X_3 \quad (4)$$

$$TM = 34.95X_1 - 7.88X_2 + 3.632X_3 \quad (5)$$

$$FS = 0.418X_1 - 7.987X_2 + 0.094X_3 + 0.0858X_1X_2 + 0.00106X_1X_3 + 0.0859X_2X_3 \quad (6)$$

$$FM = 31.246X_1 - 10.77X_2 + 3.976X_3 - 0.198X_1X_3 \quad (7)$$

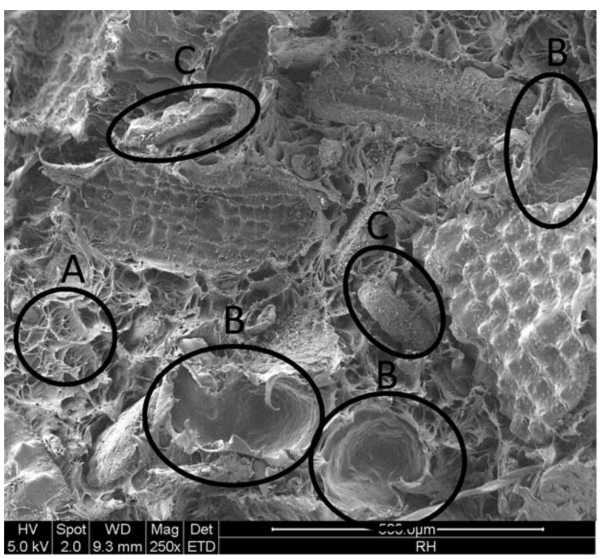

Figure 7. Failure mechanism of composites (A) crazing of polymer, (B) fiber pull out, (C) debonding.

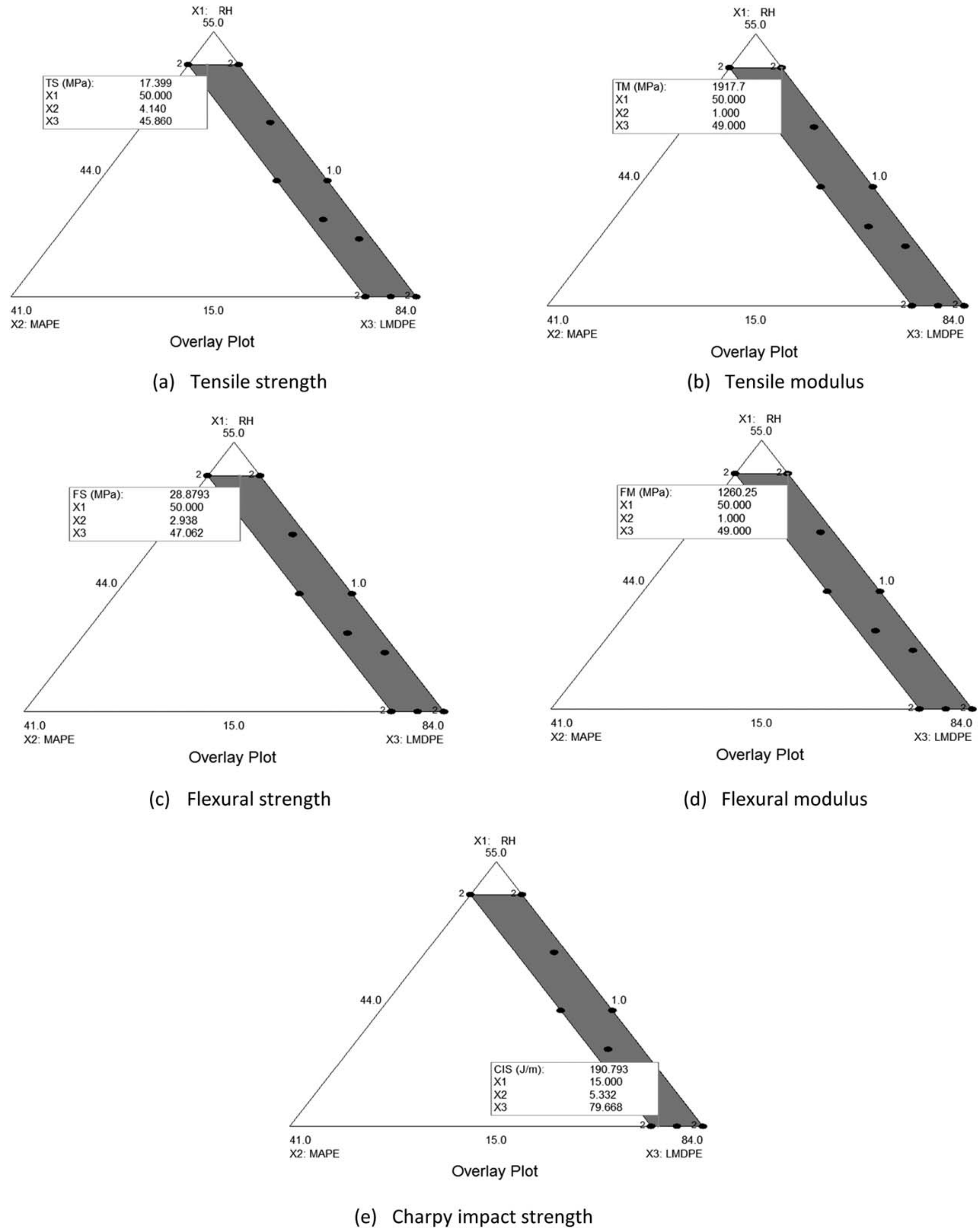

Figure 8. Overlay plots of mechanical properties.

$$CIS = 1.913X_1 - 114.61X_2 + 2.428X_3 + 1.185X_1X_2 - 0.064X_1X_3 + 1.322X_2X_3$$

The numerically optimized formulations for each mechanical property are shown in terms of actual components (RH, MAPE, and LMDPE) in the overlay plots in Figure 8. The goal was to maximize each mechanical property by searching for those combinations of components that could produce a composite with the best of each mechanical property. The optimization was done in Design Expert 8.0. The plots show that the TS was maximum

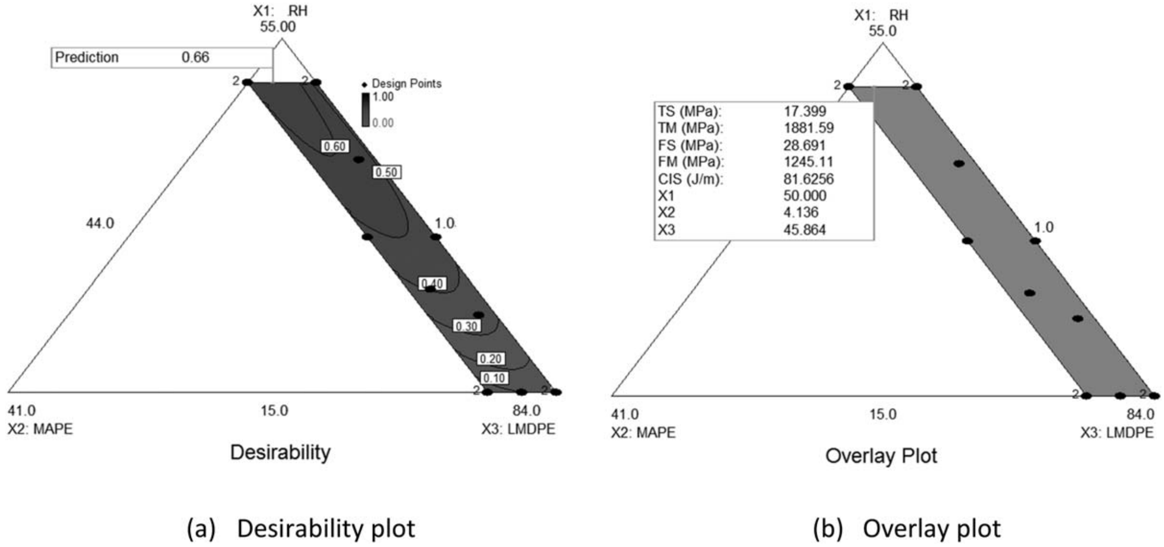

Figure 9. Optimal formulation for overall "best" mechanical properties.

Table III. Predicted and Observed Responses for Optimal Formulation of Components for Overall "Best" Mechanical Properties

	Responses				
	TS	TM	FS	FM	CIS
Predicted values	17.34	1,881.59	28.69	1245.11	81.63
Observed values[a]	17.12 (0.32)	1,756.76 (61.47)	27.83 (0.41)	1369.5 (72.97)	83.41 (2.25)

RH = 50 wt %; MAPE = 4.1 wt %, LMDPE = 45.9 wt %.
[a] Standard deviation shown in parenthesis.

(17.4 MPa) at 50 wt% RH, 4.14 wt % MAPE, and 45.86 wt % LMDPE. The FS was maximum (28.88 MPa) at 50 wt % RH, 2.9 wt % MAPE, and 47.1 wt % LMDPE. Both the tensile and flexural moduli were maximum at RH 50 wt %, MAPE 1 wt %, and LMDPE 49 wt % at 1917.7 MPa and 1260.25 MPa, respectively. The CIS was maximum at 190.79 J/m with RH at 15 wt %, MAPE at 5.33 wt %, and LMDPE at 79.67 wt %.

Multiple response optimization using all the regression models was also performed to maximize all the responses (mechanical properties) simultaneously. This was done in Design Expert, by generating a desirability function that balanced all the fitted models. The numerical optimization finds a point that maximizes the desirability function. Desirability is an objective function that ranges from zero outside of the limits to one at the goal (to maximize all the mechanical properties). For several responses, all goals get combined into one desirability function. The optimized point of mixture ratio on desirability and overlay plot is represented in Figure 9(a,b), respectively. The desirability plot shows a point that maximized the desirability function to 0.66. The obtained desirability depicts that the optimized component values for overall "best" mechanical properties lie on this point. Likewise, the overlay plot in Figure 9(b) shows the point of optimal formulation of components. The optimal formulation of the components was 50.0 wt % of RH, 4.1 wt% of MAPE, and 45.9 wt % of LMDPE for overall "best" mechanical properties. The optimal formulation and the corresponding predicted values for each response are shown in Table III. Test samples from the optimal formulation were manufactured and tested according to relevant ASTM standards with five replicates. The results are also shown in Table III along with the predicted values. The experimental results closely matched with the values predicted by the model. The maximum deviation between the predicted values and the experimental values occurred for FM with a difference of almost 10%.

CONCLUSIONS

The following conclusions are drawn from the aforementioned results in this study:

1. Tensile and flexural properties (strength and modulus) of RH/LMDPE composites increased with an increase in RH loading. CIS decreased with an increase in RH loading.
2. MAPE increased the TS, FS, and CIS of the composites until a maximum was reached, beyond which any further increase in MAPE loading decreased these properties. MAPE did not influence the tensile and flexural moduli.
3. The optimum value of RH for tensile and flexural properties (strength and modulus) was 50 wt %, whereas, for CIS the optimum percentage was 15 wt %. The optimum percentage of MAPE was 4.14 wt % for TS, 2.9 wt % for FS, 1 wt % for both tensile and flexural moduli and 5.3 wt % for CIS.

4. The optimal formulation for overall "best" mechanical properties was found to be 50 wt % for RH, 4.1 wt % for MAPE, and 45.9 wt % for LMDPE with an overall desirability of 0.66.
5. The incorporation of RH and MAPE improved the overall mechanical properties of the composites.

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