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The optimal formulation of recycled polypropylene/rubberwood flour composites from experiments with mixture design

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ABSTRACT

A mixture design was used in experiments, to determine the optimal mixture for composites of rubberwood flour (RWF) and reinforced recycled polypropylene (rPP). The mixed materials were extruded into panels. Effects were determined of the mixture components rPP, RWF, maleic anhydride-grafted polypropylene (MAPP), and ultraviolet (UV) stabilizer, on the mechanical properties. The overall composition significantly affected flexural, compressive, and tensile properties. The fractions of recycled polypropylene and rubberwood flour increased all the mechanical material properties; however, increasing one fraction must be balanced by decreasing the other, and the rubberwood flour fraction had a higher effect size. The fraction of MAPP was best kept in mid-range of the fractions tested, while the UV stabilizer fraction overall degraded the mechanical properties. Our results suggest that the fraction of UV stabilizer should be as small as possible to minimize its negative influences. The models fitted were used for optimization of a desirability score, substituting for the multiple objectives modeled. The optimal formulation found was 50.3 wt% rPP, 44.5 wt% RWF, 3.9 wt% MAPP, 0.2 wt% UV stabilizer, and 1.0 wt% lubricant; the composite made with this formulation had good mechanical properties that closely matched the model predictions.

1. Introduction

Wood waste is generated when wood is processed for various applications, such as in sawmills and in furniture making. The waste in the forms of flour, sawdust, and chips, has primarily been used as inexpensive filler in plastic industries, to reduce raw material costs and to increase the strength and modulus of various thermoplastics. Likewise, the wood particles show high specific strength and modulus that allow the production of low-density composites with higher filler content [1,2], and advantages associated with wood particles include their non-abrasive nature, low energy consumption, and biodegradability. Hence, these natural plant-based fillers offer several benefits over synthetic fillers [1]. Recent advances in natural fillers may lead to improved materials using renewable resources; this trend would also support global sustainability [3]. The mechanical properties of environmentally friendly plastic composites have been improved with wood waste from various tree species including eastern red cedar [4], maple [5], oak [4], pine [6], and rubberwood [7]. In addition, the increasing worldwide production and consumption of plastics has caused serious public concerns about effective and safe disposal [8]; however, plastic waste could be a promising raw material source for wood plastic composites (WPCs) [9]. The use of recycled plastics for producing WPCs would not only decrease the consumption of energy and natural resources, but also offers an effective and safe way to dispose of plastic waste [10]. Therefore, increasing the use of wood and plastic waste could reduce solid waste, lessen the amounts going to landfills, and decrease the cost of making WPCs [6,8].

A D-optimal mixture experimental design is a special type of statistical approach to experimentally find the individual effects and interactions of components in a mixture, and the fitted models can be used to find the optimal formulation of a composite material [11]. A D-optimal design can considerably reduce the number of experiments needed for scientific and technical information on the composition effects. It allows restricting the ranges of component fractions, and within this range of formulations helps fit the mathematical models, used to improve the characteristics of final goods [11,12]. Moreover, this method is appropriate for non-linear models [13].

The fractions of components in wood–plastic composites, such as polymer, filler and coupling agent, significantly affect their mechanical properties. Recently, several publications have assessed the effects of each material component on the thermal and mechanical properties. Mixture designs and factorial designs have been used in experiments on WPCs. Matuana et al. [14] used a four-factor central composite design to develop a response

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surface model and to study the foamability of rigid PVC/wood-flour composites. Stark and Matuana [15] applied a 2^4 factorial design to determine the effects of two hindered amine light stabilizers, a colorant, an ultraviolet absorber, and their interactions on the photo stabilization of wood flour/high-density polyethylene composites. Jun et al. [16] used a Box-Behnken design with response surface method to determine which variables influenced board performance significantly. Prior studies on the component effects and interactions, and optimization of the formulation for WPCs, seem not to have used a D-optimal mixture design. Here, a D-optimal mixture design was applied to model mechanical characteristics of WPCs. The main objective of this work was to optimize the mixture ratios for composites made from recycled polypropylene and rubberwood flour, based on mechanical properties determined experimentally. The new information will facilitate informed decisions regarding manufacture of such composites.

2. Materials and methods

2.1. Materials

Rubberwood flour (RWF) collected from a local furniture factory was used as lignocellulosic filler, and the size of the wood flour particles was smaller than 180 μm, after sieving through a standard sieve of 80 mesh. The chemical composition of RWF was, by weight: cellulose 39%; hemicellulose 29%; lignin 28%; and ash 4% [17]. Withaya Intertrade Co., Ltd. (Samutprakarn, Thailand) supplied recycled polypropylene (rPP) pellets with a melt flow index of 11 g/10 min at 230 °C, under the trade name WT170. The interfacial adhesion between wood flour and polymer was improved using maleic anhydride grafted polypropylene (MAPP), supplied by Sigma–Aldrich (Missouri, USA), with 8–10% of maleic anhydride (M_w = 9100, M_n = 3900) as a coupling agent. The ultraviolet (UV) stabilizer used was hindered amine light stabilizer additive, purchased from TH Color Co., Ltd. (Samutprakarn, Thailand) under the trade name MEUV008. Paraffin wax chosen as the lubricant (Lub) was supplied by Nippon Seiro Co., Ltd. (Yamaguchi, Japan).

2.2. Experimental design to optimize formulation

The responses of a process to various factors and parameters are effectively explored with designed experiments, using approaches such as the Taguchi method, factorial design, and mixture design [18,19]. The fractions of components in a mixture cannot be changed independently, and for this situation the mixture designs are appropriate. The nonnegative fractions must add up to 100%. For example, if $x_1, x_2, \ldots, x_l$ denote the fractions of l components of a mixture, then [18]

$$0 \leqslant x_i \leqslant 1 \quad i = 1, 2, \ldots, l$$

$$\text{and} \quad x_1 + x_2 + \ldots + x_l = 1 \ \text{(i.e., 100\%)}$$

The region of interest for the current experiments is not this simplex but has additional constraints added [18], so a D-optimal design was used to statistically evaluate the effects of component fractions on the mechanical properties, and the identified models were used to optimize the formulation. The optimized experimental design had mixture compositions for the manufacture of WPCs, the components being rPP (x_1), RWF (x_2), MAPP (x_3), UV (x_4), and Lub (x_5). The upper and lower limits of experimental range for the fractions are shown in Table 1. Despite the fraction of Lub being held constant, it is included as a variable because it contributes to the 100% in the mixture. The experimental design and analysis were done with Design-Expert software (version 8.0.6, Stat-Ease, Inc.), according to D-optimal mixture design. The design included 15 different formulations and 5 replicates to check the lack of fit. Thus, the total number of runs was 20, as shown in Table 2. After data collection, linear and quadratic models following Eqs. (1) and (2), respectively, were used to model the responses.

Table 1
Selected components and their constraints for the mixture design of experiments.

Component	Fraction restriction (wt%)
rPP (x_1)	$50 \leqslant x_1 \leqslant 70$
RWF (x_2)	$25 \leqslant x_2 \leqslant 45$
MAPP (x_3)	$3 \leqslant x_3 \leqslant 5$
UV stabilizer (x_4)	$0 \leqslant x_4 \leqslant 1$
Lub (x_5)	=1

$$Y = \sum_{i=1}^{l} \beta_i x_i \tag{1}$$

$$Y = \sum_{i=1}^{l} \beta_i x_i + \sum \sum_{i<j}^{l} \beta_{ij} x_i x_j \tag{2}$$

where Y is the predicted response, β_i is the model response to a pure component in the blend, each β_{ij} scales an interaction between components, $x_i, x_j, \ldots, x_l$ are the fractions of components, and $x_i x_j, x_i x_k, \ldots, x_k x_l$ are the quadratic interactions of the fractions. Note that mixture models differ in appearance from the general polynomials applied in response surface work, because the constraint $\sum x_i = 1$ enables elimination of terms quadratic in a single fraction [18]. Because of this, Eq. (2) has the same power to fit data from mixtures as a general quadratic polynomial; such a polynomial can be rewritten in this form.

2.3. Composites processing

To minimize its moisture content, the rubberwood flour was carefully dried prior to use; in an oven at 110 °C for 8 h. WPCs were then manufactured in a two-stage process. In the first stage to produce WPC pellets, rubberwood flour and recycled polypropylene were dry-blended, and then melt-blended into wood–plastic composite pellets using a twin-screw extruder machine (Model SHJ-36 from En Mach Co., Ltd., Nonthaburi, Thailand). The 10 temperature zones of the extruder were set to a profile in range 130–170 °C, to reduce degradation of the mixture components, while the screw rotating speed was controlled at 70 rpm. The extruded strand passed through a water bath and was subsequently pelletized. In the second stage to produce WPC panels, the WPC pellets were again dried at 110 °C for 8 h. WPC pellets, MAPP, UV stabilizer, and lubricant compositions indicated in Table 2 were then dry-mixed, and added into the feeder of a twin-screw extruder. The processing conditions for extruding were as follows: (1) barrel temperatures: 130–190 °C; (2) screw rotation speed: 50 rpm; (3) melt pressure: 0.10–0.20 MPa depending on wood flour content; and (4) vacuum venting at nine temperature zones: 0.022 MPa. The samples were extruded through a 9 mm × 22 mm rectangular die and cooled in atmospheric air. Consequently, the specimens were machined following the standards of American Society for Testing and Materials (ASTM) for flexural, compressive, and tensile tests.

2.4. Mechanical properties

Flexural properties were measured in a three-point bending test at a cross-head speed of 2 mm/min, with nominal dimensions of 4.8 mm × 13 mm × 100 mm, and a span of 80 mm in accordance with ASTM D790-92. For compressive properties, prism specimens were used to determine the compressive strength and modulus.

Table 2
Experimental compositions and responses based on mixture experiment design.

Experiment run no.	Mixture component fraction (wt%)					Response (MPa)					
						Flexure		Compression		Tension	
	x_1	x_2	x_3	x_4	x_5	MOR	MOE	CS	CM	TS	TM
1	63.9	29.9	4.5	0.7	1.0	39.38	2120	12.43	978	24.86	833
2	70.0	25.0	3.0	1.0	1.0	36.84	1807	9.45	763	23.89	787
3	50.0	43.0	5.0	1.0	1.0	36.91	2429	8.89	1133	23.37	1058
4	54.9	38.9	4.5	0.7	1.0	41.62	2387	14.43	1018	26.17	967
5	59.5	34.5	5.0	0.0	1.0	42.51	1965	14.82	830	26.32	872
6	55.4	39.9	3.5	0.2	1.0	43.97	2472	16.44	1123	28.10	997
7	59.5	34.5	4.0	1.0	1.0	36.64	2119	9.03	945	23.79	961
8[a]	59.5	34.5	5.0	0.0	1.0	41.41	2040	15.61	915	27.42	867
9	50.0	44.3	4.3	0.5	1.0	40.44	2569	15.02	1287	26.84	1067
10	68.0	25.0	5.0	1.0	1.0	37.04	2007	8.21	826	23.29	738
11	50.0	45.0	3.0	1.0	1.0	39.66	2685	13.59	1202	23.97	993
12[a]	50.0	43.0	5.0	1.0	1.0	37.85	2485	10.23	1236	24.00	1083
13	60.3	35.3	3.0	0.5	1.0	40.23	2175	15.73	1151	25.38	879
14	64.9	30.4	3.5	0.2	1.0	41.01	1969	13.02	832	25.20	765
15[a]	70.0	25.0	3.0	1.0	1.0	36.94	1760	8.25	711	23.00	649
16	51.0	45.0	3.0	0.0	1.0	46.24	2601	17.96	1449	28.36	1087
17[a]	51.0	45.0	3.0	0.0	1.0	47.63	2740	18.20	1418	28.33	1074
18[a]	50.0	45.0	3.0	1.0	1.0	39.49	2676	11.74	1262	24.70	1024
19	70.0	25.0	4.0	0.0	1.0	38.95	1902	10.55	1006	24.65	760
20	69.0	25.0	5.0	0.0	1.0	38.44	1929	8.96	789	25.01	785

[a] Duplicate experiments.

Table 3
Tabulation of *p*-values from analysis of variance, for the quadratic and linear models, and for the individual interaction terms included in the quadratic models.

Resource	MOR	MOE	CS	CM	TS	TM
Model	Quadratic	Quadratic	Quadratic	Quadratic	Quadratic	Linear
	<0.0001*	<0.0001*	<0.0001*	0.0002*	<0.0001*	<0.0001*
Linear mixture	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
x_1x_2	0.5289	0.0072*	0.1054	0.0958	0.9599	–
x_1x_3	0.8167	0.3759	0.3675	0.9867	0.3210	–
x_1x_4	0.6484	0.0844	0.0171*	0.4518	0.1583	–
x_2x_3	0.7577	0.5301	0.3433	0.9665	0.3374	–
x_2x_4	0.7047	0.0841	0.0196*	0.4440	0.1918	–
x_3x_4	0.5885	0.0195*	0.0273*	0.1605	0.1815	–
Lack of Fit	0.0628	0.4678	0.2521	0.0631	0.5874	0.6260

* *P*-value less than 0.05 is considered significant.

The displacement rate was a constant 0.5 mm/min, following ASTM standard D6108-97. Type-IV tensile bar specimens with dimensions of 115 mm × 19 mm × 4 mm were cut and machined from the extruded composite panels. The crosshead speed of tensile test was 5 mm/min, according to ASTM standard D638-99. The flexural, compressive and tensile measurements were carried out on an Instron Universal Testing Machine (Model 5582 from Instron Corporation, Massachusetts, USA) and performed at ambient conditions of 25 °C. Five replicates of each composite formulation were tested. Extrusion is directional and orients the fibers and polymer chains. The composite will not be similar in all directions (isotropic); instead, it has a preferred direction. The span in flexural testing was in the extrusion direction, and the same for tensile testing. The compression tests, however, compressed normal to the extrusion direction.

2.5. Morphological analysis

The interfacial morphology and phase dispersion of the wood flour in the polymeric matrix were assessed by imaging with a scanning electron microscope (SEM). The surfaces were prepared by sputter coating with gold, to prevent electrical charging, and were imaged with a FEI Quanta 400 microscope (FEI Company, Oregon, USA) at an accelerating voltage of 20 kV. Magnifications of 150× and 1000× were used.

Table 4
Model adequacy indicators for each modeled response of rPP/RWF composites.

Response	R^2	Adj-R^2	Pred-R^2	CV
MOR	0.9390	0.8841	0.5496	2.63
MOE	0.9838	0.9693	0.9237	2.51
CS	0.9490	0.9031	0.6751	8.15
CM	0.9258	0.8589	0.6135	7.94
TS	0.9533	0.9112	0.8004	2.04
TM	0.9153	0.8995	0.8577	4.72

3. Results and discussion

The D-optimal mixture design of experiments, with five fractions as (mutually dependent) variables (that sum to one), had 20 runs in a randomized order. The six determined responses were flexural strength (MOR) and modulus (MOE), compressive strength (CS) and modulus (CM), and tensile strength (TS) and modulus (TM). The results are summarized in Table 2.

3.1. Statistical analysis of the response surface model

Analysis of variance (ANOVA) of the response surface models indicated the quadratic model as the best fit with MOR, MOE, CS, CM, and TS, while TM was best fit with a linear model. These best

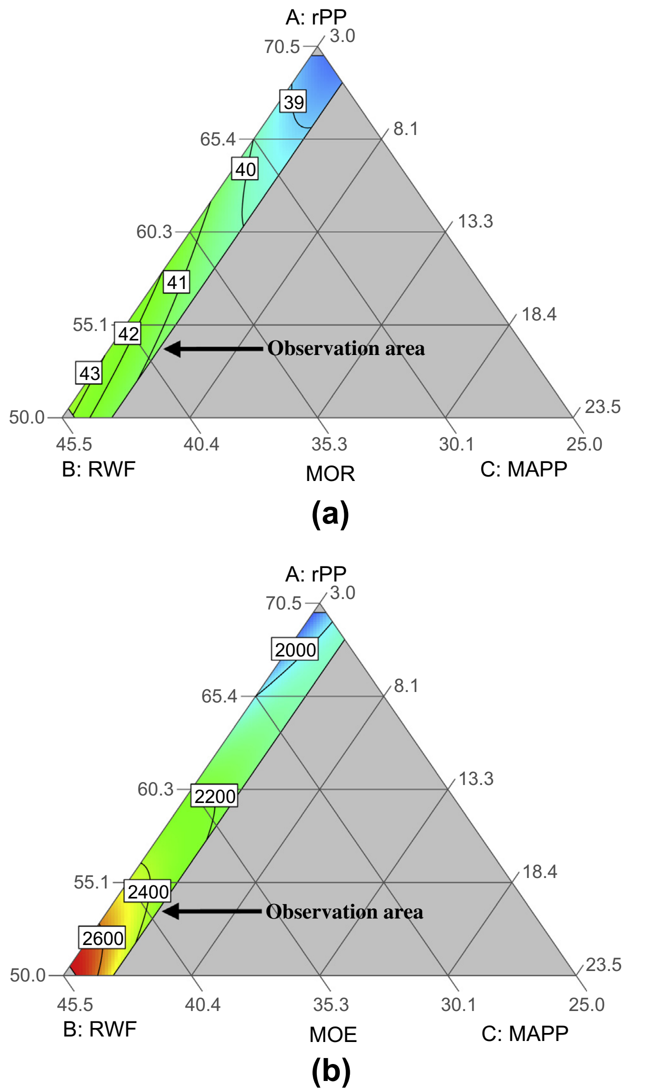

Fig. 1. Triangular contour plots for composition effects at fixed UV stabilizer fraction of 0.5 wt%, and Lub fraction 1 wt%: (a) MOR, and (b) MOE. The contours represent the models fit to experimental data.

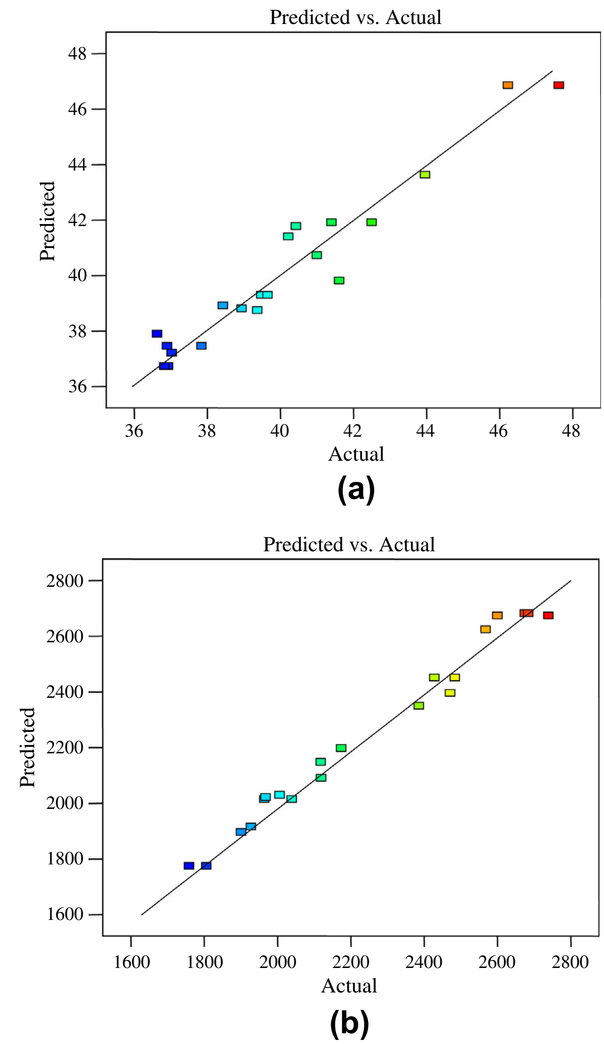

Fig. 2. Comparisons of model outputs to the fitted observed values for rPP/RWF composites. Model output was (a) MOR and (b) MOE.

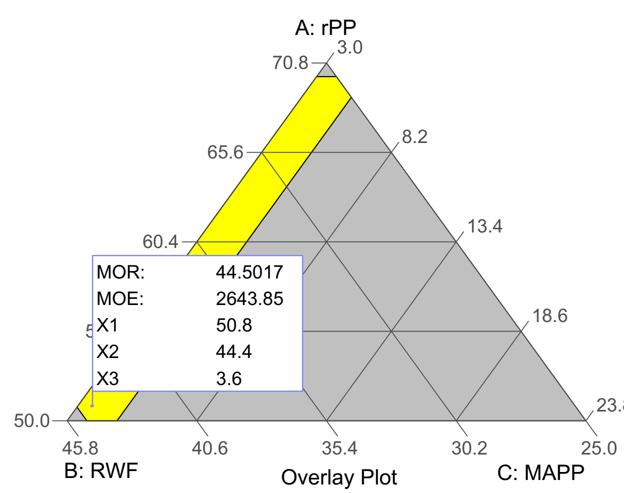

Fig. 3. The optimal formulation for flexural properties.

fit models had insignificant lack of fit and high coefficients of determination (both adjusted adj-R^2, and predicted pred-R^2). For example the quadratic model for the TS response had insignificant lack of fit with p-value 0.5874, and the coefficients of determination adj-$R^2 = 0.9112$ and pred-$R^2 = 0.8004$. The ANOVA analysis in Table 3 also indicates statistically significant quadratic terms in these models by p-values less than a significance threshold α (significance level $\alpha = 0.05$ was used for markings in the table). In the linear models, fractions of rPP, RWF, MAPP, and UV stabilizer significantly influence ($P < 0.0001$) all the mechanical properties. No significant interaction effects were indicated on MOR, CM, or TS; while for modeling MOE there were significant interactions between rPP and RWF, and between MAPP and UV stabilizer. The CS was affected by significant interactions between rPP and UV stabilizer, RWF and UV stabilizer, and MAPP and UV stabilizer. The frequent interactions with UV stabilizer might indicate it reacted chemically with the other components. The insignificant p-values for lack of fit, at 95% confidence level, also suggest that the fits of the data are appropriate.

The fit of these empirical models was also checked by the coefficient of determination (R^2), the adj-R^2, the pred-R^2, and the coefficient of variation (CV); see Table 4. The R^2 values of the six response fits are in the range from 0.9153 to 0.9838. The extreme R^2 values of TM (0.9153) and MOE (0.9838) indicate that only 8.47% and 1.62%, respectively, of the total variability in observa-

Table 5
Predicted responses with optimized formulation of each property.

Property	Mixture component proportion (wt%)					Predicted response (MPa)		
	x_1	x_2	x_3	x_4	x_5	Strength	Modulus	Desirability
Flexure	50.8	44.4	3.6	0.2	1.0	44.50	2643	0.803
Compression	51.2	44.2	3.4	0.2	1.0	17.51	1333	0.886
Tension	50.0	44.8	4.0	0.2	1.0	28.47	1065	0.975

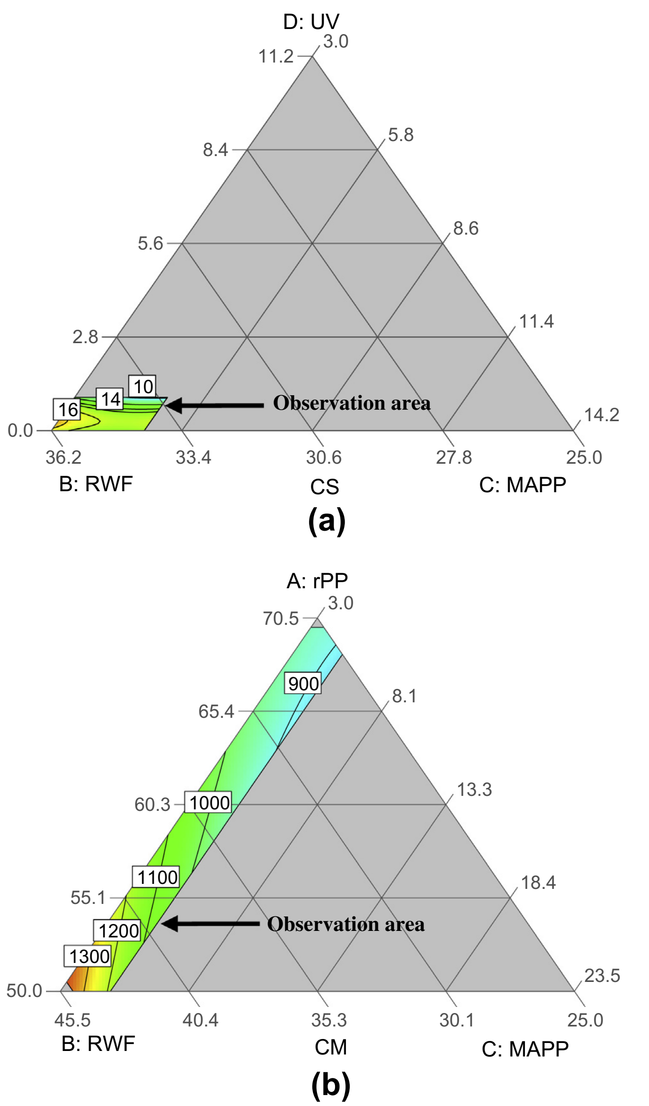

Fig. 4. Triangular contour plots for effects of the compositions on (a) CS with rPP fixed at 59.8 wt% and Lub at 1 wt%; and (b) CM with UV stabilizer fixed at 0.5 wt%, and Lub at 1 wt%.

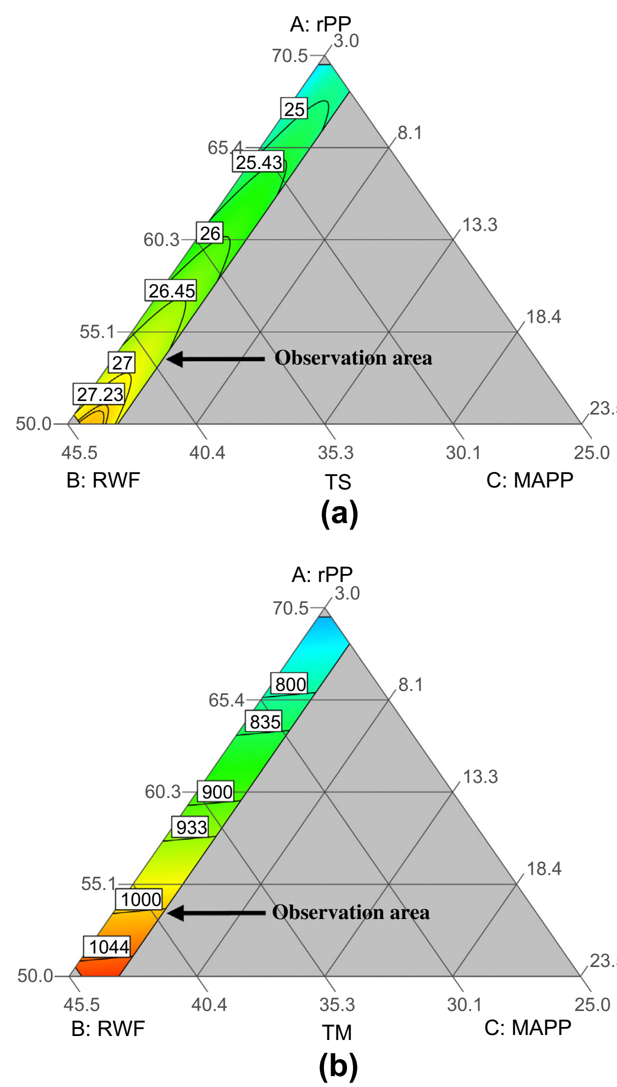

Fig. 5. Composition effects on (a) TS and (b) TM. The fractions held fixed were UV stabilizer at 0.5 wt% and Lub at 1 wt%. The contours represent the numerical models fitted to experimental observations.

tions was not explained by the models; R^2 values close to 1 indicate good fits [20]. The pred-R^2 value of MOE was 0.9237, meaning that the full model is estimated to explain about 92.37% of the variability in new data. The coefficients of variation, of MOR, MOE, CS, CM, TS, and TM, were estimated at 2.63%, 2.51%, 8.15%, 7.94%, 2.04%, and 4.72%, respectively, based on the replicates of experiments. The low CV values indicate that the determinations of material characteristics had a good precision, and can serve the fitting of parametric models. Basically, the coefficient of variation was used to measure the residual variation in the data [18].

3.2. Effect of composition on the flexural properties, and optimal formulation

The quadratic regression models fitted to experimental MOR and MOE values were:

$$\begin{aligned}\mathrm{MOR} = {} & 39.04x_1 + 47.14x_2 + 107.71x_3 - 646.43x_4 + 2.09x_1x_2 \\ & - 77.5x_1x_3 + 668.48x_1x_4 - 102.86x_2x_3 + 555.13x_2x_4 \\ & + 728.26x_3x_4 \end{aligned} \tag{3}$$

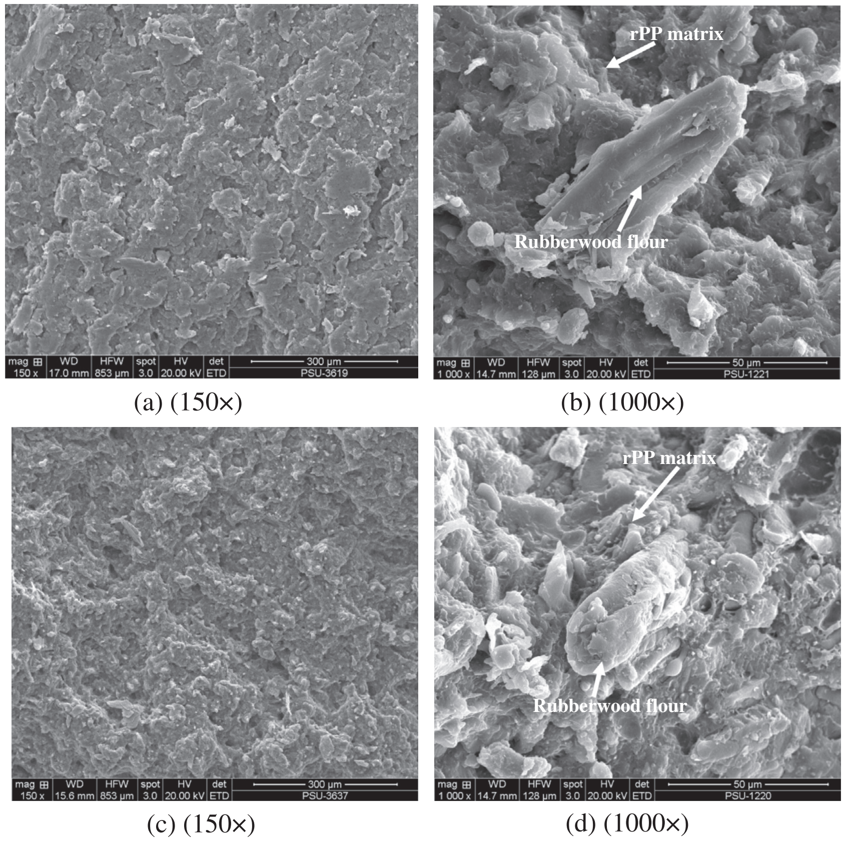

Fig. 6. Scanning electron micrographs of rPP/RWF composites with (a), (b) 25 wt% RWF and (c), (d) 45 wt% RWF. Magnifications were 150× and 1000×, from left to right.

$$\begin{aligned}\mathrm{MOE} = {} & 1803.43x_1 + 2743.06x_2 - 11575.14x_3 - 136983x_4 \\ & - 573.4x_1x_2 + 16071.92x_1x_3 + 145070x_1x_4 \\ & + 11237.53x_2x_3 + 145344.6x_2x_4 + 192792.7x_3x_4 \end{aligned} \quad (4)$$

The equation of MOR shows a negative coefficient for fraction of UV stabilizer (x_4), and MOE shows negative coefficients for MAPP (x_3) and UV stabilizer (x_4). However, since these are quadratic models, also the quadratic interaction terms must be inspected, for example at some reasonable values of the other fractions. This is why linear models are much more interpretable, and even on inspecting them, the dependency between the fractions (they must sum to one) makes model interpretation difficult. The addition of UV stabilizer in the wood–plastic composites is known to reduce the flexural properties due to non-homogeneous spatial distribution of wood flour, polymer, and UV stabilizer [21]. The covered experimental regions of MOR and MOE are shown in Fig. 1a and b, respectively. In these triangular plots the three pure components (rPP, RWF, and MAPP) are represented by the corners, while the additive levels were fixed (UV stabilizer at 0.5 wt% and Lub at 1 wt%). The contours in the colored areas, that include the experimental observations, present the MOR and MOE regression fits varying from 39 to 43 MPa and 2000 to 2600 MPa, respectively. MOR and MOE clearly increase with the rubberwood flour content, and its good interfacial adhesion to recycled polypropylene contributes to this. MAPP acts as a compatibilizer providing a hydrophobic rich layer attached to wood flour [22]. Generally, the strength and modulus of wood flour reinforced composites depend on the properties of constituents and the interfacial adhesion [22]. The MAPP addition of about 3–4 wt% is close to optimal for MOE, based on the regression fit. Similar results were found in the work of Kuo et al. [23] who reported that the optimal content of MAPP was 3–4.5 wt% because the interfacial adhesion weakens at higher MAPP contents.

Fig. 2a and b shows the MOR and MOE model predictions vs. observations. The model outputs fit the actual observations quite well, with MOR model deviating from actual by less than about 5%, and MOE model being slightly more accurate. These correlations verified that the Eqs. (3) and (4) are adequate to predict the MOR and MOE responses. The numerically optimized composition, based on these model fits, is shown in Fig. 3. Since two models are optimized simultaneously, the software actually uses a single surrogate called "desirability" to balance them. The model-based optimal formulation is included in Table 5.

3.3. Effect of composition on the compressive properties, and optimal formulation

The quadratic regression models for the compressive properties CS and CM were:

$$\begin{aligned}\mathrm{CS} = {} & 9.76x_1 + 18.28x_2 + 287.82x_3 - 3776.11x_4 + 5.56x_1x_2 \\ & - 299.68x_1x_3 + 3956.09x_1x_4 - 314.6x_2x_3 \\ & + 3852.86x_2x_4 + 3278.65x_3x_4 \end{aligned} \quad (5)$$

$$\begin{aligned}\mathrm{CM} = {} & 1014.25x_1 + 1461.58x_2 - 1406.56x_3 - 87880.43x_4 \\ & - 462.14x_1x_2 + 435.39x_1x_3 + 87388.18x_1x_4 \\ & - 1096.7x_2x_3 + 89032x_2x_4 + 155014.6x_3x_4 \end{aligned} \quad (6)$$

Again these equations do not lend themselves to easy interpretation, due to interaction terms and dependencies between the model

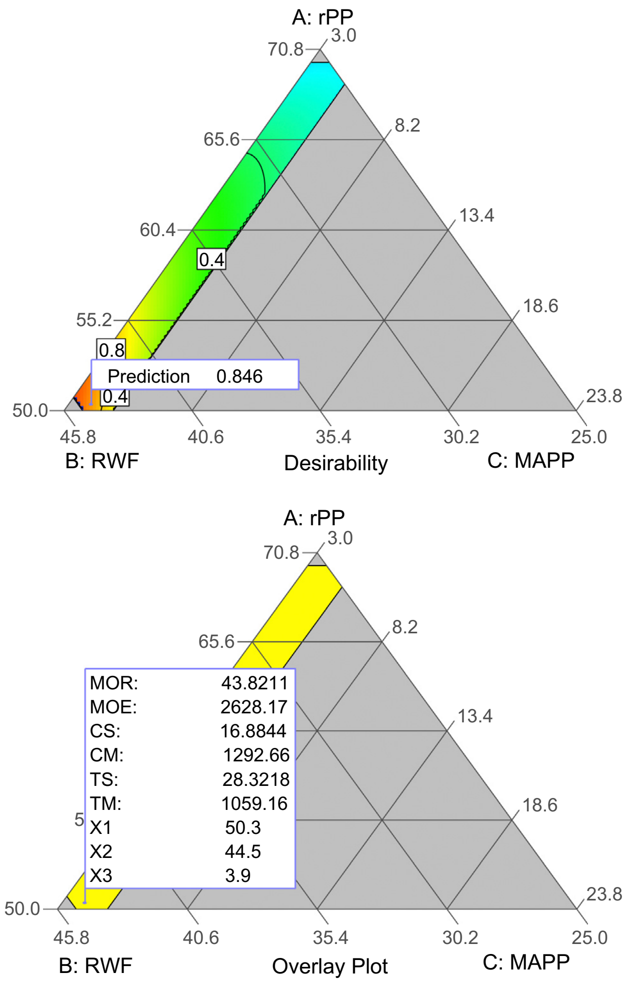

Fig. 7. The optimal formulation for overall desirability.

input variables. We resort to inspecting plots of the model outputs. Fig. 4a shows that CS (in range of 16–10 MPa) decreases for high fractions of the UV stabilizer. The reason for this phenomenon is probably similar to what was discussed in relation to flexural properties. In Fig. 4b, the CM values vary in range of 900–1300 MPa and increase with wood flour loading, since wood flour is stiffer than neat plastic [24]. Likewise, the optimal addition of MAPP for the compressive modulus is approximately 3–4 wt%. Too much MAPP relative to wood flour will cause self-entanglement, resulting in slippage with the PP molecules [25]. The optimal formulation based on these regression models is also included in Table 5.

3.4. Effect of composition on the tensile properties, and optimal formulation

The regression fits for the tensile strength (TS) and modulus (TM) were:

$$\begin{aligned} TS = {} & 23.54x_1 + 28.64x_2 - 112.44x_3 - 989.02x_4 + 0.081x_1x_2 \\ & + 166.14x_1x_3 + 1059.37x_1x_4 + 159.72x_2x_3 \\ & + 973.37x_2x_4 + 914.15x_3x_4 \end{aligned} \tag{7}$$

$$TM = 717.6x_1 + 1067.03x_2 + 1114.53x_3 + 687.04x_4 \tag{8}$$

By these equations, rPP (x_1) and RWF (x_2) increase the tensile properties; all terms containing these variables have positive coefficients. Of these two, RWF has the larger coefficient in the fit for TS and TM, so it should be maximized. The fractions of MAPP (x_3) and UV stabilizer (x_4) each have both positive and negative coefficients in the model for tensile strength, but both increase the tensile modulus. Fig. 5a and b show that TS and TM increase with the rubberwood flour content. The SEM micrographs in Fig. 6 show that the composites with 25 and 45 wt% of RWF had low porosity, good contact between the wood flour and the PP matrix, and good dispersion of wood flour. Stress transfer was therefore supported at these high rubberwood flour contents. The composition optimized based on these regression models is shown numerically in Table 5.

Table 6
Predicted and observed responses with the formulation optimized jointly for all the mechanical properties.

	Mixture component proportion (wt%)					Response (MPa)					
						Flexure		Compression		Tension	
	x_1	x_2	x_3	x_4	x_5	MOR	MOE	CS	CM	TS	TM
Predicted	50.3	44.5	3.9	0.2	1.0	43.82	2628	16.88	1292	28.32	1059
Observed						47.28	2527	17.11	1369	27.68	1024
						(2.92)[a]	(112)	(2.72)	(109)	(2.41)	(128)

[a] The values in parentheses are standard deviations from five replicates.

3.5. Optimal formulation of the overall mechanical properties

Multiobjective optimization using all of the regression models was performed with the Design-Expert software, using its default settings to construct a desirability score that balances all of the fitted models. The plot in Fig. 7 shows the formulation that was considered optimal, along with contours of the desirability score. The optimal formulation is given in Table 6, and can be compared with the formulations in Table 5: all the previous optima were at practically the same formulation, so a reasonable desirability score must also give this formulation. Table 6 also shows the model predicted responses for this formulation. Test samples with five replicates were prepared with this formulation, and the average material properties along with their standard deviations are included in Table 6. The maximum deviation between model prediction and experimental average occurs for MOR and is of the order 10%.

4. Conclusions

Design and analysis of a D-optimal mixture experiment were used to obtain the optimal formulation of an rPP/RWF composite. The formulation provides high values for all the material characteristics modeled. Analysis of variance revealed that all the component fractions experimentally varied, namely of rPP, RWF, MAPP, and UV stabilizer, statistically significantly affected every one of the mechanical properties (MOR, MOE, CS, CM, TS, and TM). In general, a high fraction of RWF improved all of these, and the optima found had close to 45% RWF that was the maximum in the experimental design. At this wood flour loading stress transfer was still supported by good dispersion and surface contact with the polymer, and the wood flour is much stiffer than the rPP matrix. The compatibilizer MAPP had negative effects on MOE and CM, while for TS a middle of the range value seemed optimal (Fig. 5a). The fraction of UV stabilizer overall degraded the mechanical properties. While the actual optimal composition may depend on a variety of factors, including the quality of raw materials and processing conditions, we have demonstrated the applicability of particular techniques to optimizing properties of composites. In this case, the optima for various mechanical properties agreed well, while in general the joint optimization of multiple responses will depend on their prioritization.

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