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Optimizing the formulation of polypropylene and rubberwood flour composites for moisture resistance by mixture design

Chatree Homkhiew[1], Thanate Ratanawilai[1] and Wiriya Thongruang[2]

Abstract

D-optimal mixture experimental design was used to determine the optimal mixture of composites from rubberwood (*Hevea brasiliensis*) flour and recycled polypropylene and to systematically analyze the effects of composition, namely recycled polypropylene, rubberwood flour, maleic anhydride-grafted polypropylene, and ultraviolet stabilizer fractions. Panel samples were extruded, and their properties were characterized. The overall compositions significantly affected water absorption, thickness swelling, flexural strength and modulus, and maximum strain. Water absorption and thickness swelling increased with the fraction of rubberwood flour. At long immersion times, flexural strength and modulus decreased, but maximum strain increased with high fraction of rubberwood flour. The fraction of maleic anhydride-grafted polypropylene only slightly affected water absorption and flexural properties, while the ultraviolet stabilizer fraction had a clear negative effect increasing water absorption and decreasing flexural properties. The models fitted were used for optimization of a desirability score, substituting for the multiple objectives modeled. The optimal formulation found was 68.9 wt% recycled polypropylene, 25.0 wt% rubberwood flour, 5.0 wt% maleic anhydride-grafted polypropylene, 0.1 wt% ultraviolet stabilizer, and 1.0 wt% lubricant. This formulation of the composites can be used for most suitable applications based on the moisture resistance.

Keywords

Wood–plastic composites, rubberwood flour, recycled polypropylene, statistical method, water absorption

Introduction

In the recent decades, plastic waste has globally become a significant contributor to municipal solid waste.[1] In 2008, at least 33.6 million tons of postconsumer plastics were generated in the United States, of which 28.9 million tons went to landfills, 2.6 million tons to combustion and energy recovery, and only 2.2 million tons to recycling.[2] The plastic waste typically includes polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyethylene terephthalate (PET), and polystyrene (PS).[3] Of all these plastic types, PE and PP significantly contribute to landfills and have similarities in their structure and properties.[4] However, when PE (virgin or recycled) was blended with sawdust, it had lower stiffness and strength than similar composites with PP (virgin or recycled).[5] Due to the availability of plastic waste and increased environmental awareness, there have been many studies on natural fiber-reinforced recycled thermoplastics. For example, Cui et al.[1] fabricated composites from postconsumer high-density polyethylene and wood fiber and found that wood fiber content affected the flexural strength,

[1]Department of Industrial Engineering, Prince of Songkla University, Hat Yai, Songkhla, Thailand
[2]Department of Mechanical Engineering, Prince of Songkla University, Hat Yai, Songkhla, Thailand

Corresponding author:
T Ratanawilai, Department of Industrial Engineering, Prince of Songkla University, Hat Yai, Songkhla 90112, Thailand.
Email: thanate.r@psu.ac.th

modulus, and impact strength. Boukehili and Nguyen-Tri[6] manufactured composites from recycled polypropylene (rPP) and short bamboo fiber. They found that chemical treatment of fiber significantly improves the impermeability to helium gas and reduces the amount of water absorption. Kazemi et al.[4] produced composites from wood sawdust and blends of postconsumer PE and PP. The wood flour in composites increased tensile, flexural, and torsion moduli in comparison to the recycled plastic blends. Nourbakhsh et al.[7] also concluded that polypropylene waste and wood waste are promising alternative raw materials for making low-cost wood–plastic composites (WPCs). Hence, the use of recycled plastics may effectively dispose of plastic waste and also reduce the cost of products.[8]

The rubberwood industries of Thailand generate a large amount of wood waste in the forms of flour, sawdust, and chips at different stages of wood processing. Generally, rubberwood waste is dumped in landfills or burned, but some is used in medium-density fiberboard and particle board.[9] Rubberwood waste as reinforcement in plastic composites is of great interest both for economic and for environmental reasons. Natural fiber (wood waste)-reinforced thermoplastics also offers many advantages including biodegradability, renewable character, low cost, absence of associated health hazards, and low wear of processing equipment,[10–13] when compared with synthetic fibers.[14,15] Natural fibers have successfully improved the mechanical properties of plastic composites, as the following examples demonstrate. Homkhiew et al.[9] investigated the effects of rubberwood content on the mechanical properties of recycled PP composites and found that the modulus and hardness of composites increased linearly with an increase of wood flour loadings in the range of 25–45 wt% wood flour. Karmarkar et al.[16] also reported that the tensile strength and modulus linearly increased with an increase of wood fiber from 10 to 50 wt% in PP composites. However, the hygroscopic nature of natural fibers is a disadvantage influencing the performance of the WPCs, when exposed to environmental conditions.[17] The water-absorption characteristics WPCs limit their end-use applications,[17,18] as several mechanical and physical properties, such as dimensional stability, are affected. Likewise, water absorption (WA) is one of the key parameters in quality assessment of WPCs.[19] The WA of WPCs varies by the wood species, partly because they have different contents of cellulose, lignin, hemicelluloses, and extractants.[20] Hence, the effects of filler on the composite properties need to be characterized, and this has not been done previously for rubberwood flour (RWF) in relation to moisture resistance.

Statistical experimental design, such as mixture design, factorial design, and Taguchi method, is a well-established concept for planning and execution of informative experiments.[21] Recently, WPCs have been studied with designed experiments. For example, Jun et al.[22] used a Box–Behnken design with response surface method to determine which variables influenced board performance significantly. Matuana and Mengeloglu [23] used a four-factor central composite design to develop a response surface model and to study the foamability of rigid PVC/wood-flour composites. Stark and Matuana[24] applied a 2^4 factorial design to determine the effects of two hindered amine light stabilizers, a colorant and an ultraviolet (UV) absorber, and their interactions on the photostabilization of wood flour/high-density polyethylene composites. However, the fractions of components in a mixture cannot be changed independently since they must add up to 100%, and the methods for mixture designs have been created with this in mind.[25] Mixture designs have been successful in many applications, particularly in food and pharmaceutical industries, whereas prior studies on WPCs seem not to have used a mixture design.

Experiments with mixture design enable statistical estimation of 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.[26] Hence, a mixture experimental design was applied to model physical and mechanical characteristics of WPCs. The main objective of this work was to optimize the mixture ratios for composites made from rPP and RWF, based on experimentally determined WA and flexural failure. The new information created from this work would help better understand the properties of components, and the optimal formulation of the composites can be used for most suitable applications in condition of the water resistance.

Materials and methods

Materials

rPP pellets, WT170 with a melt flow index of 11 g/10 min at 230°C, were purchased from Withaya Intertrade Co., Ltd (Samutprakarn, Thailand) and used as the polymer matrix. They have the flexural strength and modulus about 37.02 MPa and 1.27 GPa, respectively. RWF obtained from the cutting process in local furniture industry (Songkhla, Thailand) was used as reinforcement. Maleic anhydride-grafted polypropylene (MAPP) with 8–10% of maleic anhydride was supplied by Sigma-Aldrich (Missouri, USA) and used as a coupling agent to improve the interfacial adhesion between filler and matrix. Hindered amine light stabilizer (MEUV008) was purchased from TH Color Co., Ltd (Samutprakarn, Thailand), chosen as UV stabilizer. Paraffin wax was procured from Nippon Seiro Co., Ltd (Yamaguchi, Japan) and used as lubricant (Lub).

Experimental design to optimize formulation

The region of interest for the current experiments has constraints that imposed on the component fractions,[25] and these can be incorporated in a D-optimal mixture design. The experimental results were used to statistically evaluate the effects of component fractions on WA and flexural failure by using analysis of variance (ANOVA), and the identified models were used to optimize the formulation by using response surface methodology. The experimental D-optimal mixture design and statistical analysis were done with Design-Expert software (version 8.0.6, Stat-Ease, Inc.). The formulations for the manufacture of WPCs were defined by component fractions for rPP (x_1), RWF (x_2), MAPP (x_3), UV stabilizer (x_4), and Lub (x_5). The upper and lower limits of experimental range are shown in Table 1. The ranges of rPP and RWF contents obtained from the previous preliminary study[27] and the other compositions were determined following the literature review. For example, Kuo et al.[10] reported that the optimal content of MAPP was 3–4.5 wt%. Despite the fraction of Lub being held constant, it is included as a variable because it contributes to the 100% in the mixture. The design included 15 different formulations and 5 replications to evaluate reproducibility and variances. Thus, the total number of runs was 20, as shown in Tables 2 and 3. After data collection, linear, quadratic, and special cubic models (see equations (1) to (3)) were used to model the responses.

$$Y = \sum_{i=1}^{l} \beta_i x_i \quad (1)$$

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

Table 1. Constraints for the mixture design of experiments.

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

Table 2. Experimental compositions based on mixture experimental design, and replicate averaged measured responses: WA and TS at 1, 5, and 10 weeks.

	Mixture component fraction (wt%)					Water absorption (%)			Thickness swelling (%)		
Run no.	x_1	x_2	x_3	x_4	x_5	W1	W5	W10	W1	W5	W10
1	63.9	29.9	4.5	0.7	1.0	0.97	2.97	4.77	0.24	0.70	1.28
2	70.0	25.0	3.0	1.0	1.0	0.77	2.22	3.64	0.28	0.58	0.89
3	50.0	43.0	5.0	1.0	1.0	2.65	7.28	9.87	0.49	1.63	2.72
4	54.9	38.9	4.5	0.7	1.0	1.99	6.12	8.59	0.43	1.39	2.60
5	59.5	34.5	5.0	0.0	1.0	2.06	5.13	7.56	0.30	1.00	1.99
6	55.4	39.9	3.5	0.2	1.0	1.94	5.80	8.34	0.50	1.43	2.53
7	59.5	34.5	4.0	1.0	1.0	1.06	4.67	6.90	0.40	1.11	1.91
8[a]	59.5	34.5	5.0	0.0	1.0	1.55	4.75	7.10	0.31	1.17	2.15
9	50.0	44.3	4.3	0.5	1.0	2.58	7.14	9.45	0.66	1.92	3.07
10	68.0	25.0	5.0	1.0	1.0	0.56	1.91	3.13	0.26	0.56	0.85
11	50.0	45.0	3.0	1.0	1.0	3.39	8.92	10.50	0.75	2.39	3.15
12[a]	50.0	43.0	5.0	1.0	1.0	3.08	8.05	9.70	0.43	1.92	2.63
13	60.3	35.3	3.0	0.5	1.0	1.48	4.58	6.81	0.27	0.96	1.73
14	64.9	30.4	3.5	0.2	1.0	0.97	3.39	5.28	0.29	0.89	1.60
15[a]	70.0	25.0	3.0	1.0	1.0	0.67	2.10	3.39	0.23	0.58	0.93
16	51.0	45.0	3.0	0.0	1.0	2.70	7.77	10.02	0.60	2.11	3.37
17[a]	51.0	45.0	3.0	0.0	1.0	2.91	8.12	10.31	0.61	2.27	3.47
18[a]	50.0	45.0	3.0	1.0	1.0	3.58	9.41	10.33	0.74	2.75	3.51
19	70.0	25.0	4.0	0.0	1.0	0.68	1.80	2.95	0.28	0.48	0.75
20	69.0	25.0	5.0	0.0	1.0	0.61	1.69	2.78	0.20	0.44	0.67

[a]Duplicate experiments.

$$Y = \sum_{i=1}^{l} \beta_i x_i + \sum \sum_{i<j}^{l} \beta_{ij} x_i x_j + \sum \sum_{i<j<k} \sum \beta_{ijk} x_i x_j x_k \quad (3)$$

where Y is the predicted response, i, j, k, and l mean rPP, RWF, MAPP, and UV stabilizer, respectively, β_i is the model response to a pure component in the blend, each β_{ij} scales an interaction between components, each β_{ijk} scales an interaction of three components, x_i, x_j,..., x_l are the fractions of components, terms with $x_i x_j$, $x_i x_k$,..., $x_k x_l$ are the quadratic interactions of the fractions, and the last sum in equation (3) consists of cubic interactions.

Preparation of composites

Prior to compounding, the rubberwood (*Hevea brasiliensis*) flour was sieved through a standard sieve of mesh size 80 (passing particles smaller than 180 μm) and was dried in an oven at 110°C for 8 h to minimize the moisture content. The WPCs were then manufactured in a two-stage process. In the first stage, WPC pellets were produced: rPP and RWF were dry-blended and melt-blended into WPC pellets using a twin-screw extruder (Model SHJ-36 from En Mach Co., Ltd, Nonthaburi, Thailand). The 10 temperature zones of the extruder were set to a profile in the range of 130–170°C to reduce degradation of the mixture components, while the screw-rotating speed was maintained at 70 rpm. The extruded strand passed through a water bath and was subsequently pelletized. In the second stage, WPC panels were produced: the WPC pellets were again dried in the oven at 110°C for 8 h. WPC pellets, MAPP, UV stabilizer, and lubricant compositions, indicated in Tables 2 and 3, were then dry-mixed and fed into the twin-screw extruder. The processing conditions for extruding were as follows: (a) temperature profiles: 130–190°C; (b) screw-rotating speed: 50 rpm; (c) melt pressure: 0.10–0.20 MPa depending on wood flour content; and (d) vacuum venting at nine temperature zones: 0.022 MPa. The WPC panels were extruded through a 9 mm × 22 mm rectangular die and cooled in ambient air. These specimens were machined following the standards of American Society for Testing and Materials (ASTM) for physical and mechanical testing.

Table 3. The experimental compositions and replicate averaged measured flexural properties.

	Mixture component fraction (wt%)					MOR (MPa)		MOE (GPa)		Max. ε (%)	
Run no.	x_1	x_2	x_3	x_4	x_5	W1	W6	W1	W6	W1	W6
1	63.9	29.9	4.5	0.7	1.0	38.7	36.8	1.90	1.67	3.23	3.80
2	70.0	25.0	3.0	1.0	1.0	37.2	35.8	1.76	1.65	3.03	3.81
3	50.0	43.0	5.0	1.0	1.0	37.5	29.7	2.10	1.63	2.92	3.11
4	54.9	38.9	4.5	0.7	1.0	39.7	34.2	2.03	1.62	3.12	3.66
5	59.5	34.5	5.0	0.0	1.0	41.9	32.5	1.87	1.50	3.52	3.74
6	55.4	39.9	3.5	0.2	1.0	40.7	32.5	1.96	1.59	3.41	3.61
7	59.5	34.5	4.0	1.0	1.0	36.3	32.8	1.94	1.49	2.82	3.67
8[a]	59.5	34.5	5.0	0.0	1.0	40.1	31.9	1.82	1.47	3.63	3.79
9	50.0	44.3	4.3	0.5	1.0	40.3	33.5	2.04	1.70	3.09	3.60
10	68.0	25.0	5.0	1.0	1.0	36.8	36.6	1.75	1.65	3.14	3.78
11	50.0	45.0	3.0	1.0	1.0	35.9	30.7	1.85	1.65	2.97	3.20
12[a]	50.0	43.0	5.0	1.0	1.0	37.4	28.6	2.10	1.64	2.75	2.77
13	60.3	35.3	3.0	0.5	1.0	39.9	34.2	1.94	1.52	3.15	4.14
14	64.9	30.4	3.5	0.2	1.0	40.8	37.0	1.86	1.53	3.70	4.42
15[a]	70.0	25.0	3.0	1.0	1.0	36.9	35.4	1.79	1.64	3.38	4.15
16	51.0	45.0	3.0	0.0	1.0	44.6	33.3	2.12	1.63	3.49	3.90
17[a]	51.0	45.0	3.0	0.0	1.0	43.1	34.6	2.11	1.65	3.50	3.94
18[a]	50.0	45.0	3.0	1.0	1.0	36.8	31.0	1.84	1.67	3.07	3.46
19	70.0	25.0	4.0	0.0	1.0	38.9	38.9	1.81	1.54	3.50	4.81
20	69.0	25.0	5.0	0.0	1.0	38.6	38.1	1.83	1.75	3.59	4.90

[a]Duplicate experiments.

WA and dimensional stability tests

WA and thickness swelling tests were carried out according to ASTM D570-88 specifications. Before testing, five replicate specimens of each formulation were dried in an oven at 50°C for 24 h. The weight and thickness of each specimen were measured to a precision of 0.001 g and 0.01 mm, respectively. The specimens were then submerged in water at ambient room temperature. After 1, 5, and 10 weeks, soaked specimens were removed from the water, thoroughly dried the surface with tissue papers, and immediately weighed and measured to determine the weight and thickness. The percentage of WA at any given time was calculated following

$$WA_t(\%) = \frac{W_t - W_0}{W_0} \times 100 \quad (4)$$

where WA_t is the WA at time t, W_0 is the initial dry weight, and W_t is the soaked weight of specimen at a given time t.

The percentage of thickness swelling (TS) at any given time was calculated following

$$TS_t(\%) = \frac{T_t - T_0}{T_0} \times 100 \quad (5)$$

where TS_t is the thickness swelling and T_t is the soaked thickness of specimen, both at the given time t, while T_0 is the initial dry thickness.

Flexural test of WPCs

Flexural properties of the samples were determined with an Instron Universal Testing Machine (Model 5582 from Instron Corporation, Massachusetts, USA) in accordance with ASTM D790-92. In the destructive flexural tests (three point bending), specimens with nominal dimensions of 4.8 mm × 13 mm × 100 mm, a span of 80 mm, and a cross-head speed of 2 mm/min were used. The testing was performed at ambient room temperature of 25°C with five samples of each formulation to obtain an average value. The measurements of flexural strength (MOR), modulus of elasticity (MOE), and maximum strain (max. ε) at failure were done at 1 and 6 weeks, and at the latter time, WA had reached its equilibrium so further testing was considered unnecessary.

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 12 determined responses were the values of the WA and thickness swelling at 1, 5, and 10 weeks and flexural strength, modulus, and maximum strain at 1 and 6 weeks. The results are summarized in Tables 2 and 3.

Statistical analysis of the response models

ANOVA of the alternative types of response models revealed that WA at 1, 5, and 10 weeks, TS at 1 and 5 weeks, and MOR at 1 and 6 weeks were best fit with quadratic models, instead of linear, special cubic, or cubic models, whereas MOE at both 1 and 6 weeks was best fit with special cubic model. The MOE at 6 weeks is shown as an example in Table 4. The sequential model sums of squares for quadratic and special cubic models are significant ($p < 0.05$), but not for the other model types. Moreover, the lack of fit is clearly insignificant for the special cubic model, suggesting that this model performs well. It also has the highest adjusted coefficient of determination (adj-$R^2 = 0.9726$) and predicted coefficient of determination (pred-$R^2 = 0.9484$), further indicating good fit.

The detailed ANOVAs in Tables 5 and 6 document the significant quadratic or cubic terms in models for each response, in terms of their p-values. The ANOVA shows the statistical significance ($p < 0.05$) of these terms supplementing linear models of the fractions, namely rPP, RWF, MAPP, and UV stabilizer. No interaction term was significant in models of WA, MOR, and MOE at 1 week or TS at 5 weeks. However, other response models had significant interactions, for example, between MAPP and UV stabilizer

Table 4. Fitted model summary for MOE at 6 weeks.

Source	Sequential p-value	Lack of fit p-value	Adj-R^2	Pred-R^2	
Linear	0.6971	0.0002*	−0.0884	−0.4177	
Quadratic	0.0056*	0.0023*	0.6382	−0.6377	
Special cubic	*0.0004**	*0.9391*	*0.9726*	*0.9484*	*Suggested*
Cubic	0.9391	–	0.9672	–	Aliased

*p-value less than 0.05 is considered significant.

Table 5. ANOVA and model adequacy for WA and TS responses.

	Water absorption			Thickness swelling		
Source	W1	W5	W10	W1	W5	W10
Model	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
Linear mixture	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
x_1x_2	0.1380	0.9034	0.0042*	0.0005*	0.0925	–
x_1x_3	0.1107	0.3851	0.4548	0.0007*	0.6560	–
x_1x_4	0.7164	0.0701	0.0246*	0.1941	0.2073	–
x_2x_3	0.1097	0.3679	0.5153	0.0010*	0.7388	–
x_2x_4	0.7422	0.0746	0.0242*	0.1839	0.2088	–
x_3x_4	0.2698	0.0205*	0.0065*	0.2339	0.1190	–
Lack of fit	0.4366	0.4456	0.2319	0.1650	0.5991	0.2126
R^2	0.9703	0.9906	0.9959	0.9810	0.9749	0.9727
Adj-R^2	0.9435	0.9821	0.9921	0.9639	0.9524	0.9675
Pred-R^2	0.8245	0.9526	0.9800	0.8801	0.8898	0.9584
C.V. (%)	13.29	6.65	3.54	8.14	11.79	8.39

ANOVA: analysis of variance.
*P-value less than 0.05 is considered significant.

Table 6. ANOVA and model adequacy for flexural properties.

	MOR		MOE		Max. strain	
Source	W1	W6	W1	W6	W1	W6
Model	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*	<0.0001*
Linear mixture	<0.0001*	<0.0001*	<0.0001*	0.0018*	<0.0001*	<0.0001*
x_1x_2	0.9169	0.0067*	0.1091	0.5197	–	–
x_1x_3	0.2973	0.0726	0.3808	0.2568	–	–
x_1x_4	0.1474	0.0124*	0.2373	0.0007*	–	–
x_2x_3	0.3236	0.0952	0.3308	0.5825	–	–
x_2x_4	0.1700	0.0127*	0.2523	0.0008*	–	–
x_3x_4	0.0968	0.0020*	0.8386	0.7737	–	–
$x_1x_2x_3$	–	–	0.5757	0.3760	–	–
$x_1x_2x_4$	–	–	0.0619	0.0039*	–	–
$x_1x_3x_4$	–	–	0.7685	0.5580	–	–
$x_2x_3x_4$	–	–	0.6719	0.3347	–	–
Lack of fit	0.6701	0.1192	0.7600	0.9391	0.6122	0.2335
R^2	0.9499	0.9514	0.9936	0.9913	0.8354	0.8382
Adj-R^2	0.9048	0.9077	0.9799	0.9726	0.8045	0.8079
Pred-R^2	0.8310	0.8449	0.7753	0.9484	0.7360	0.7354
C.V. (%)	1.86	2.51	0.91	0.78	3.86	5.90

ANOVA: analysis of variance.
*p-value less than 0.05 is considered significant.

for WA at 5 weeks and between rPP and RWF, rPP and UV stabilizer, RWF and UV stabilizer, and MAPP and UV stabilizer for MOR at 6 weeks. The frequent interactions with UV stabilizer might indicate that it reacted chemically with the other components or affected their distribution and interactions. In addition, the ANOVA also showed that lack of fit was not significant for any of the response surface models at 95%

confidence level. The regression models fit the data in a statistically sound manner.

Tables 5 and 6 also include the coefficient of determination (R^2), adj-R^2, pred-R^2, and coefficient of variation (C.V.). The R^2 values of the 12 response fits are in the range from 0.8354 to 0.9959. The R^2 values of maximum strain at 1 week (0.8354) and WA at 10 weeks (0.9959) indicate that only 16.46% and 0.41%, respectively, of the total variability in observations are not explained by the models; R^2 values close to 1 indicate good fits.[28] Also, the adj-R^2 values in the range from 0.8045 to 0.9921 suggest good fits and the same goes for pred-R^2 values. The pred-R^2 value of WA at 10 weeks was 0.9800 meaning that the fitted model would explain about 98% of the variability in new data. The coefficients of variation of all response fits based on the replications of experiments show low values in the range from 0.78 to 13.29%. The low-C.V. values indicate that the determinations of material characteristics had a good precision and can serve the fitting of parametric models.

Model adequacy checking

Model adequacy checking is performed to verify the appropriate approximation of the fitted model.[29] Figure 1(a) displays the normal probability plots of the residuals for WA at 10 weeks (WAW10). The good linear fit in this plot indicates that the residuals (approximation errors remaining in the model) are close to normally distributed. Basically, normally distributed residuals are a requirement for the validity of least squares regression, so the model is adequate. Likewise, there is no indication of possible outliers, such as faulty experiment cases with particularly large residuals.[25] The plot of residuals versus predicted values in Figure 1(b) exhibits no obvious patterns that would suggest adding a term to the model, to account for that pattern. If the residuals had such structure, the model would not be appropriate.[25] Figure 1(c) shows model predictions versus observations. The model outputs fit the actual observations quite well, with WAW10 model deviating from actual by less

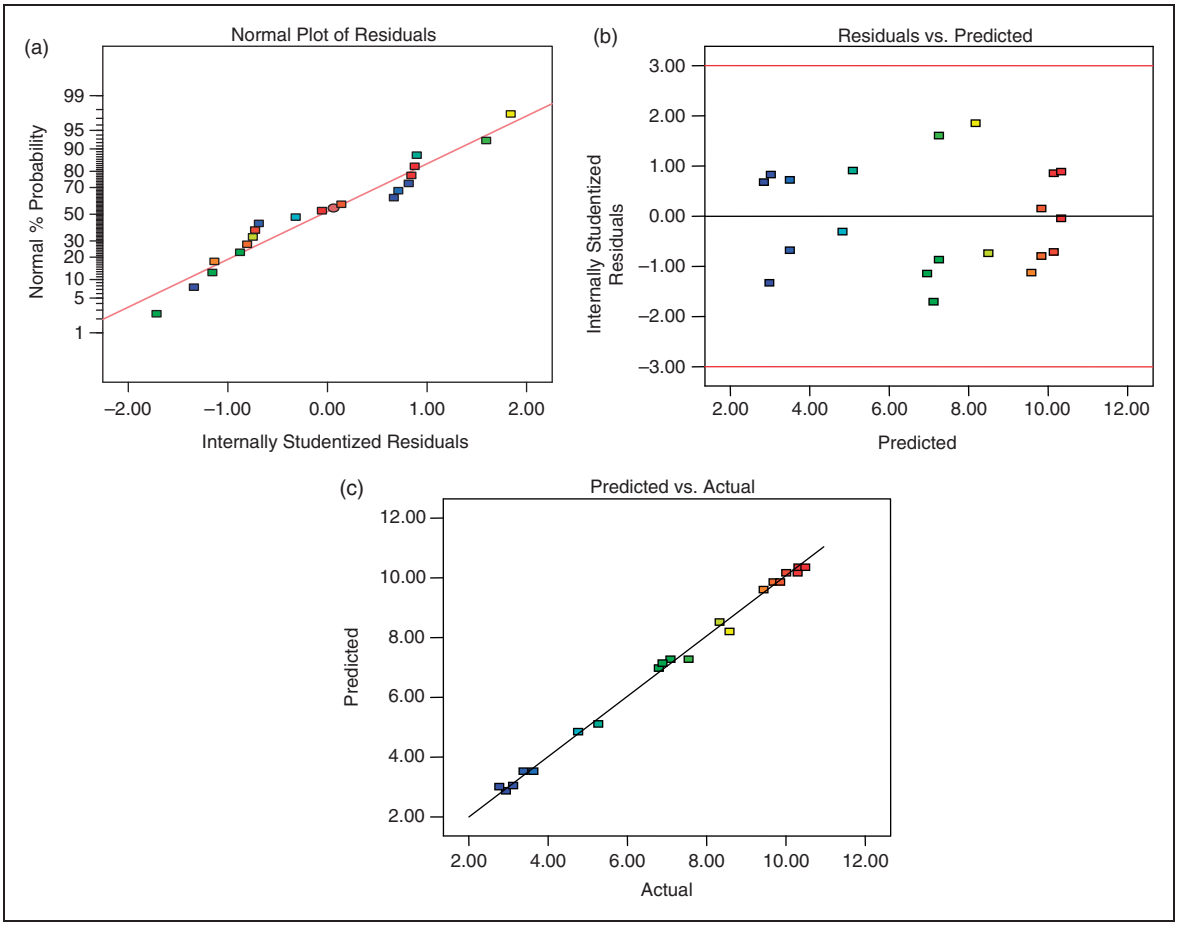

Figure 1. Model adequacy checking for water absorption at 10 weeks; (a) normal probability plot of residuals, (b) plot of residuals versus predicted values, and (c) plot of predicted versus actual values.

than about 5%. These adequacy checks of the WAW10 response model indicated accuracy with the model type or its fit to data. Similar checking for the other modeled responses gave no indications of problems with the fitted models either. This type of checking cannot guarantee predictive capability, but suggests that the models are sound approximations for interpolating within the experimental range.

Effect of composition on WA and optimal formulation

The quadratic regression models for WAW1, WAW5, and WAW10 were

$$\begin{aligned}\text{WAW1} = {} & 0.73x_1 + 2.99x_2 + 120.23x_3 + 115.96x_4 \\ & - 1.18x_1x_2 - 129.92x_1x_3 - 121.13x_1x_4 \\ & - 129.82x_2x_3 - 109.74x_2x_4 - 346.98x_3x_4 \quad (6)\end{aligned}$$

$$\begin{aligned}\text{WAW5} = {} & 1.8x_1 + 8.23x_2 + 91.78x_3 + 907.58x_4 \\ & + 0.13x_1x_2 - 96.75x_1x_3 - 942.92x_1x_4 - 100.05x_2x_3 \\ & - 926.36x_2x_4 - 1171.62x_3x_4 \quad (7)\end{aligned}$$

$$\begin{aligned}\text{WAW10} = {} & 2.99x_1 + 10.38x_2 + 57.33x_3 + 861.54x_4 \\ & + 2.80x_1x_2 - 60x_1x_3 - 890.01x_1x_4 - 51.83x_2x_3 \\ & - 894.58x_2x_4 - 1056.74x_3x_4 \quad (8)\end{aligned}$$

These equations show a positive coefficient for all the individual components, namely rPP (x_1), RWF (x_2), MAPP (x_3), and UV stabilizer (x_4), and the coefficients of rPP and RWF increased with immersion time. The rPP has the smallest coefficient in the fit for the WA due to hydrophobicity of this matrix polymer.[19] Contour plots of WAW1 and WAW10 are shown in Figure 2(a) 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, which include the experimental observations, present the WAW1 and WAW10 regression fits varying from 0.5 to 2.5% and 2.9 to 9.5%, respectively. Both WAW1 and WAW10 clearly increase with an increase in the RWF content: the free OH groups of RWF cellulose increase the WA of the composites.[30,31] Likewise, the acceleration of water uptake of WPCs could be concluded on four factors such as lumen, hydrophilicity of wood cellulous, microcracks in wood flour, and adhesion between wood flour and polymer matrix.[31] Increasing the MAPP addition from 3 to 5 wt% slightly affects the WA. This is because the coupling agent increases bonding in WPCs, by improving interfacial adhesion between the wood particles and the polymer. Then the plastic can cover more of the wood surfaces, resulting in a lower WA.[32,33] Besides, Adhikary et al.[8] also concluded that the addition of MAPP significantly reduced the WA, when compared with the composites without MAPP. The composites, consisted of 50 wt% wood flour and 3 wt% MAPP, decreased the WA from 4.1% to 1.31% for the 24-h immersion tests.[8] Furthermore, adding 1 wt% UV stabilizer increased the moisture content in the rPP/RWF composites. This may be attributed to the nonuniform spatial distribution of wood flour, polymer, and UV stabilizer.[9,34] When WPCs experienced with the nonuniform spatial distribution and poor interfacial adhesion between the compositions, it allows easier access of water into the structure. Figure 3 displays the numerically optimized composition, based on these model fits. Since three models are optimized simultaneously, the software

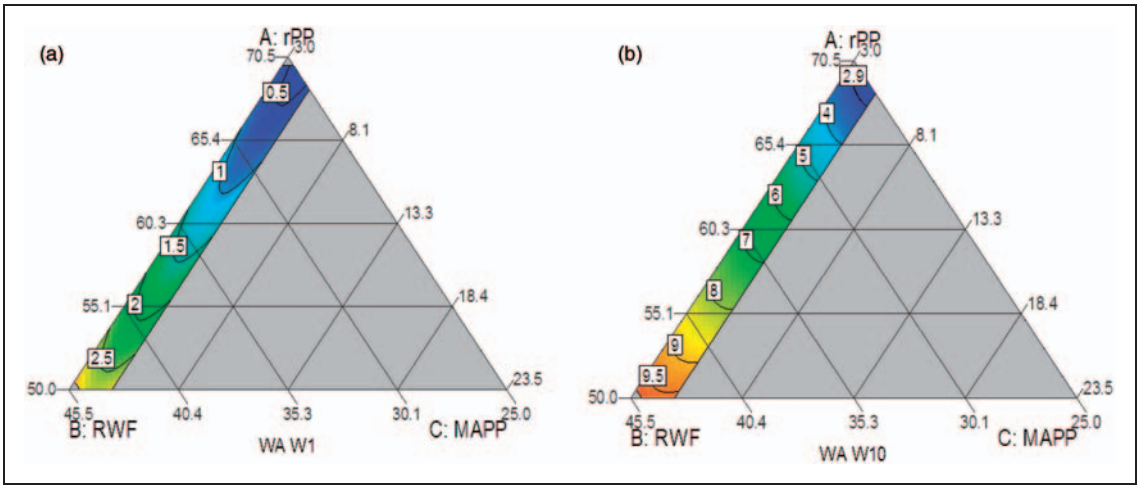

Figure 2. Triangular contour plots for effects of the compositions on water absorption at (a) 1 and (b) 10 weeks, with UV stabilizer fixed at 0.5 wt% and Lub at 1 wt%.

actually uses a single surrogate called "desirability" to balance them. This is reasonable because the three characteristics (WA at 1, 5, and 10 weeks) are not competing, but are in a good mutual agreement. The model-based optimal formulations are shown in Table 7, and the "overall" WA represented by the desirability was minimized by 69.3 wt% rPP, 25.0 wt% RWF, 4.6 wt% MAPP, 0.1 wt% UV stabilizer, and 1 wt% Lub, with a high-desirability score of 0.992 that indicates the agreement of the multiple objectives.

Effect of composition on thickness swelling and optimal formulation

The regression fits for the thickness swelling (TS) at 1, 5, and 10 weeks were

$$\begin{aligned}\text{TSW1} = {} & 0.093x_1 + 0.67x_2 - 43.45x_3 + 63.68x_4 \\ & - 0.52x_1x_2 + 49.74x_1x_3 - 63.11x_1x_4 \\ & + 47.51x_2x_3 - 64.78x_2x_4 - 52.62x_3x_4 \quad (9)\end{aligned}$$

$$\begin{aligned}\text{TSW5} = {} & 0.33x_1 + 2.33x_2 - 17.17x_3 + 273.76x_4 \\ & - 0.87x_1x_2 + 21.95x_1x_3 - 281.51x_1x_4 \\ & + 16.33x_2x_3 - 280.74x_2x_4 - 326.17x_3x_4 \quad (10)\end{aligned}$$

$$\text{TSW10} = 0.95x_1 + 3.45x_2 - 0.71x_3 - 1.10x_4 \quad (11)$$

The equations of TS at all immersion times show positive coefficients for fraction of rPP (x_1) and RWF (x_2) and negative coefficient for fraction of MAPP (x_3) due to the decrease of WA and moisture penetration in the composite systems[35] when MAPP was added. Likewise, when the positive coefficients between rPP and RWF were compared, the RWF showed higher coefficients than the rPP due to nature of the hydrophilic filler.[19] Figure 4 shows that TS at 10 week (in range of 1 to 3%) increases with an increase in the RWF fraction. The wood flour expands and keeps absorbing water until the cell walls are saturated.[18] The addition of MAPP from 3 to 5 wt% affected the thickness swelling

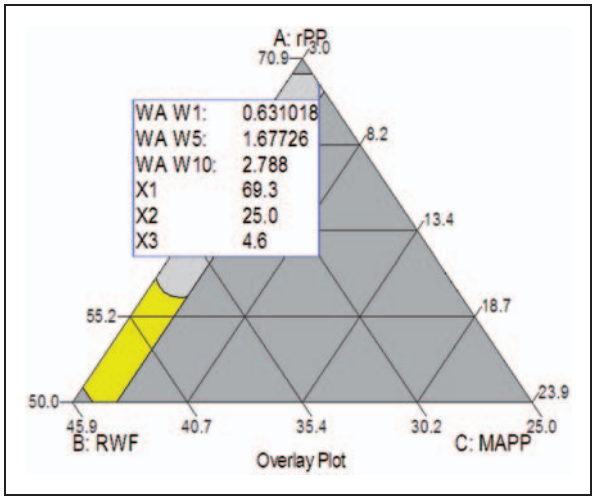

Figure 3. The optimal formulation for water absorption.

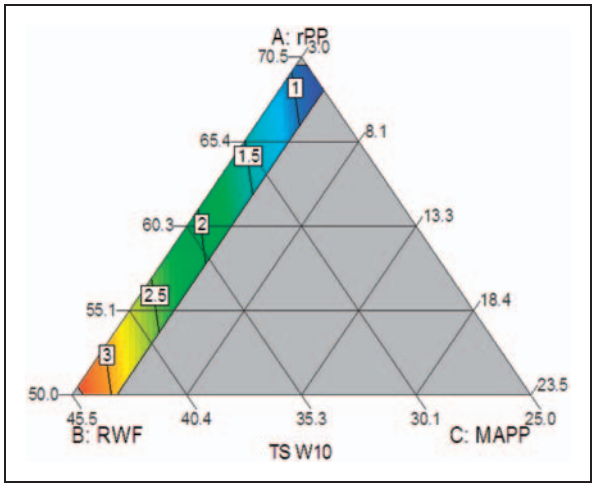

Figure 4. Triangular contour plots for effects of composition on thickness swelling at 10 weeks, with UV stabilizer fixed at 0.5 wt% and Lub at 1 wt%.

Table 7. Predicted optimal formulations and their responses, from multiobjective optimizations.

	Mixture component fraction (wt%)					Predicted response				
Property	x_1	x_2	x_3	x_4	x_5	W1	W5	W6	W10	Desirability
WA (%)	69.3	25.0	4.6	0.1	1.0	0.63	1.67	–	2.78	0.992
TS (%)	68.9	25.0	5.0	0.1	1.0	0.22	0.49	–	0.78	0.969
MOR (MPa)	69.4	26.4	3.1	0.1	1.0	40.4	–	37.7	–	0.678
MOE (GPa)	50.0	44.1	4.2	0.7	1.0	2.03	–	1.74	–	0.862
Max. strain (%)	69.8	26.2	3.0	0.0	1.0	3.70	–	4.77	–	0.968

Note: For example, the formulation in first row is optimal for a desirability score that balances WA at times W1, W5, and W10.

of composites, so that the swelling decreased with MAPP fraction. The reason is probably similar to what was discussed in relation to WA. The optimal formulation based on these numerical models, combined by a desirability score for optimization, is also included in Table 7.

Effect of composition on flexural strength and optimal formulation

The quadratic regression models fitted for the flexural strength MOR at 1 and 6 weeks were

$$\begin{aligned}\text{MOR W1} = {} & 40.35x_1 + 43.78x_2 + 247.09x_3 \\ & - 1498.46x_4 + 0.24x_1x_2 - 247.39x_1x_3 \\ & + 1542.49x_1x_4 - 232.7x_2x_3 + 1453.86x_2x_4 \\ & + 1649.2x_3x_4 \end{aligned} \quad (12)$$

$$\begin{aligned}\text{MOR W6} = {} & 37.82x_1 + 33.70x_2 - 437.66x_3 - 3328.49x_4 \\ & - 8.79x_1x_2 + 526.16x_1x_3 + 3484.81x_1x_4 \\ & + 481.2x_2x_3 + 3468.82x_2x_4 + 4344.19x_3x_4 \end{aligned} \quad (13)$$

The coefficients of rPP (x_1), RWF (x_2), MAPP (x_3), and UV stabilizer (x_4) decrease with the immersion time. The UV stabilizer fraction has the largest negative coefficient in the model fits, so it should be minimized. UV stabilizer in WPCs is known to reduce the flexural properties due to nonhomogeneous spatial distribution of wood flour, polymer, and UV stabilizer.[34] Therefore, ability of the stress transfer in the composite structure is reduced. The triangular contour plots in Figure 5(a) and (b) illustrate that an increase of wood flour loading slowly increased MOR at 1 week but greatly decreased MOR at 6 weeks, respectively. The water molecules reduced interfacial adhesion between RWF and polypropylene.[18] When water molecules infiltrate into the composite, the wood flour tends to swell, resulting in localized yielding of the polymer matrix and loss of adhesion between the wood flour and matrix.[18,36,37] Furthermore, the addition of MAPP about 3 wt% is close to optimal for MOR, based on the regression fit. Because MAPP acts as a compatibilizer providing a hydrophobic-rich layer attached to wood flour.[38] Thus, MOR increased with the RWF content after immersing for 1 week. Similar results were found in the work of Kuo et al.[10] who reported that the optimal content of MAPP was 3–4.5 wt% because the interfacial adhesion weakens at higher MAPP contents. The optimal composition based on the quadratic regression models is shown numerically in Table 7.

Effect of composition on flexural modulus and optimal formulation

The special cubic models fitted for the flexural modulus MOE at 1 and 6 weeks were

$$\begin{aligned}\text{MOE W1} = {} & 1.88x_1 + 2.15x_2 + 19.67x_3 - 64.10x_4 \\ & - 0.43x_1x_2 - 20.28x_1x_3 + 66.89x_1x_4 \\ & - 21.35x_2x_3 + 62.9x_2x_4 - 149.38x_3x_4 \\ & + 1.61x_1x_2x_3 + 16.71x_1x_2x_4 + 235.95x_1x_3x_4 \\ & + 332.38x_2x_3x_4 \end{aligned} \quad (14)$$

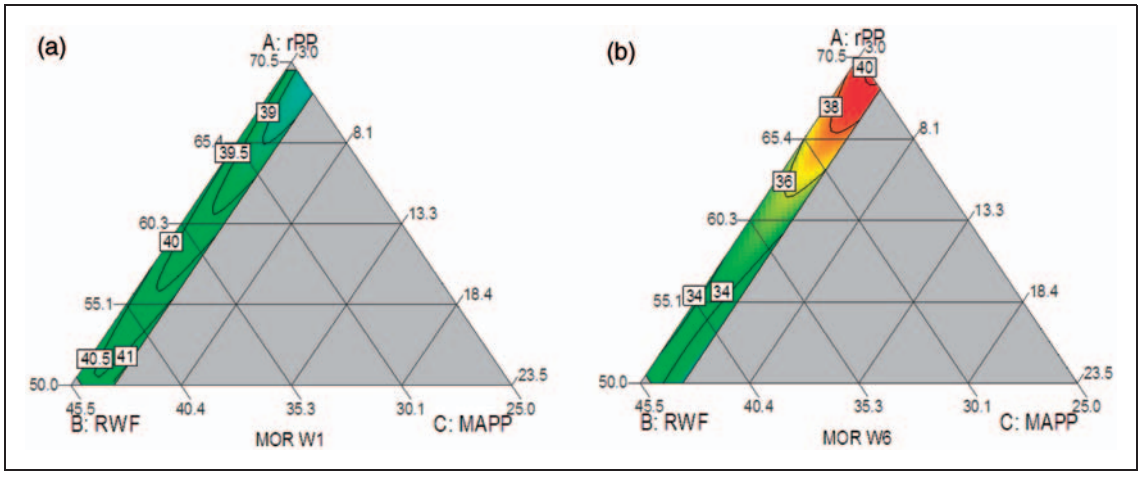

Figure 5. Triangular contour plots for effects of composition on MOR at (a) 1 and (b) 6 weeks, with UV stabilizer fixed at 0.5 wt% and Lub at 1 wt%.

$$\begin{aligned}\text{MOE W6} = {} & 1.24x_1 + 1.67x_2 - 10.92x_3 - 211.79x_4 \\ & - 0.11x_1x_2 + 19.33x_1x_3 + 232.57x_1x_4 \\ & + 8.43x_2x_3 + 224.01x_2x_4 - 151.98x_3x_4 \\ & + 1.87x_1x_2x_3 - 23.9x_1x_2x_4 + 341.82x_1x_3x_4 \\ & + 563.41x_2x_3x_4 \end{aligned} \quad (15)$$

These equations show kind of coefficients as flexural strength (MOR), decreasing with immersion time. Figure 6(a) shows that MOE at 1 week (in range of 1.85–2.05 GPa) increased for high fractions of wood flour. Since RWF is a high-modulus material compared to the plastic matrix, composites with higher wood flour concentration require a higher stress for the same deformation.[39] Likewise, fractions of MAPP about 3–4 wt% gave high-flexural modulus. This is because MAPP can improve the interfacial adhesion between wood flour and rPP matrix, leading to improve the stress transfer from polymer to wood particles.[40] However, too much MAPP relative to wood flour will cause self-entanglement, resulting in slippage with the PP molecules.[40] In Figure 6(b), when composites were soaked in water for 6 weeks, MOE at high 45 wt% RWF fraction was comparable to composites with 25 wt% RWF. The wood flour as hard filler, in comparison to the plastic matrix, increased the stiffness of the composites. With moisture wood flour plasticizes, becoming ductile, this decreased the stiffness of composites.[36] Figure 7 shows the optimal formulation based on the special cubic models for MOE and a desirability score combining their outputs. The optimal formulation is also included in Table 7.

Effect of composition on maximum strain and optimal formulation

The linear regression models for the maximum strain (max. ε) at 1 and 6 weeks were

$$\text{Max.}\varepsilon\ \text{W1} = 3.72x_1 + 3.48x_2 + 2.63x_3 - 7.75x_4 \quad (16)$$

$$\text{Max.}\varepsilon\ \text{W6} = 4.82x_1 + 3.87x_2 + 1.23x_3 - 9.29x_4 \quad (17)$$

The fraction of rPP (x_1) has the largest coefficients in the fit, so the maximum strain increased with high fraction of rPP. This is because rPP has high ductility and viscosity, resulting an increase of the maximum strain. In contrast, the maximum strain decreased with the

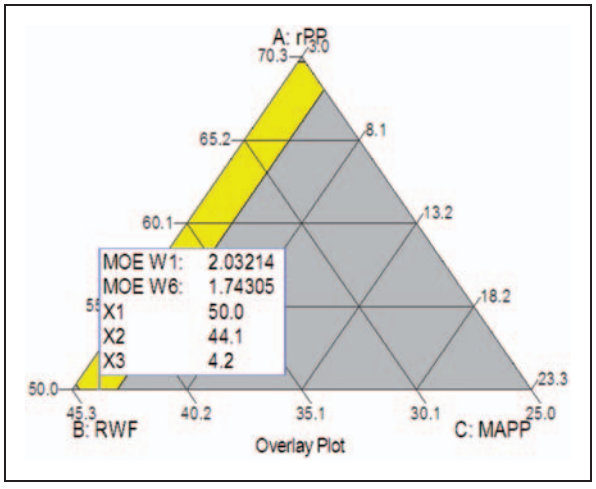

Figure 7. The optimal formulation for MOE.

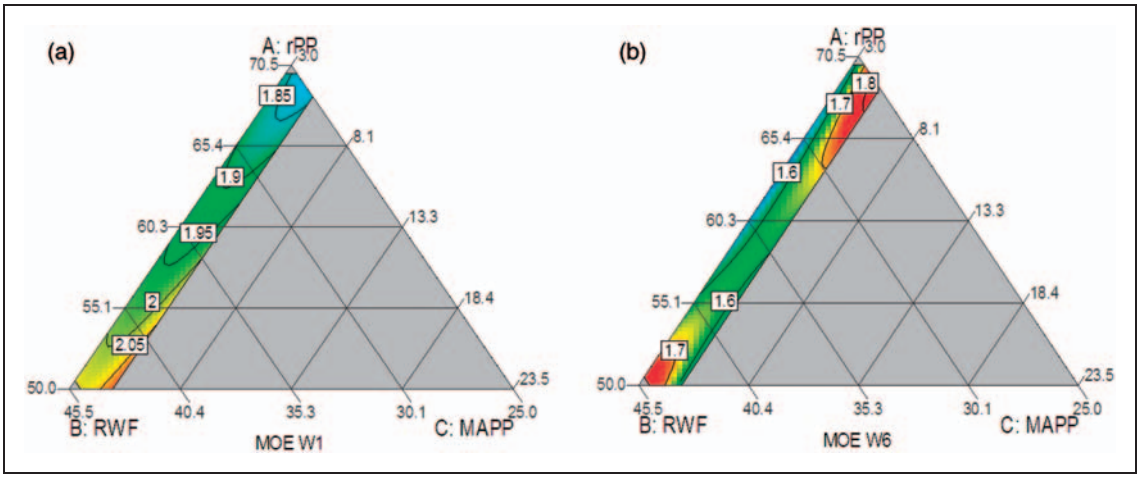

Figure 6. Triangular contour plots for effects of composition on MOE at (a) 1 and (b) 6 weeks, with UV stabilizer fixed at 0.5 wt% and Lub at 1 wt%.

fraction of UV stabilizer (x_4) that has a negative coefficient. The reason for this phenomenon is probably similar to that shown in the flexural strength. Using 1 wt% of UV stabilizer may be unnecessary and to reduce the negative effects on the mechanical properties, the amount of UV stabilizer should be minimized.[34] Again, the maximum strain increases with the WA and immersion time. The reason for this is probably similar as described earlier that ductile wet wood increases the maximum strain. Figure 8 shows that the maximum strain decreased with the RWF content. This is due to the increase in the stiffness and brittleness reducing the maximum strain. The stress concentrations at the fiber ends have been recognized as the leading cause for embrittlement.[8] The optimized composition based on these linear regression models is shown numerically in Table 7.

Optimal formulation of the overall properties based on WA

An optimal formulation for rPP/RWF composites was determined to minimize WA and thickness swelling and maximize flexural strength, modulus, and maximum strain. This multiobjective optimization, using all of the regression models, was performed with the Design-Expert software by constructing a desirability score that balances all of the fitted models in Figure 9. The optimal formulation was 68.9 wt% rPP, 25.0 wt% RWF, 5.0 wt% MAPP, 0.1 wt% UV stabilizer, and 1.0 wt% Lub. The optimal formulation is given in Table 8, along with the model-based responses. The overall formulations in Table 7 closely agree with this optimum.

Conclusions

Mixture experimental design, statistical model, and optimization were used to quantify the effects of rPP/RWF composite formulation and to optimize the formulation for moisture resistance. ANOVA revealed that all the component fractions experimentally varied, namely rPP, RWF, MAPP, and UV stabilizer, statistically significantly affected the WA, thickness swelling, flexural strength and modulus, and maximum strain. In general, a high fraction of RWF increased the WA and TS across immersion times due to the free OH

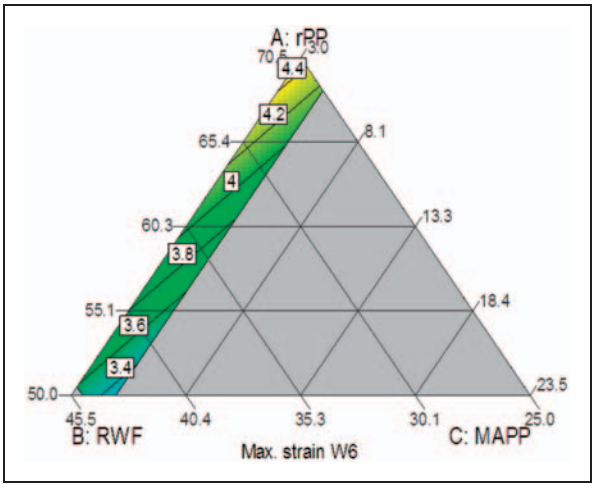

Figure 8. Triangular contour plots for effects of composition on the maximum strain at 6 weeks, with UV stabilizer fixed at 0.5 wt% and Lub at 1 wt%.

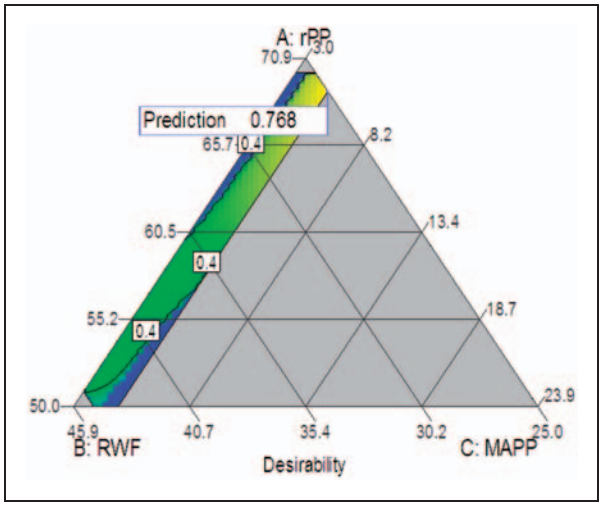

Figure 9. The optimal formulation for overall desirability.

Table 8. Predicted responses with the formulation optimized jointly for all properties.

	Mixture component fractions (wt%)					Predicted response			
Property	x_1	x_2	x_3	x_4	x_5	W1	W5	W6	W10
WA (%)	68.9	25.0	5.0	0.1	1.0	0.85	1.81	–	2.82
TS (%)						0.22	0.50	–	0.78
MOR (MPa)						38.9	–	38.5	–
MOE (GPa)						1.84	–	1.79	–
Max. strain (%)						3.56	–	4.41	–

groups in wood flour contributing WA. When the composites were soaked in water for 1 week, high fractions of RWF increased MOR and MOE but reduced maximum strain. In contrast, at 6 weeks, the MOR and MOE decreased but maximum strain increased with RWF loading. At the longer immersion time, water reduced the interfacial adhesion between RWF and rPP, and moisture plasticized the wood flour making it ductile. This decreased the strength and stiffness but increased the maximum strain of composites. Therefore, the optimum found had 25 wt% RWF, which was the minimum in the experimental design. The compatibilizer MAPP slightly affected WA and TS, which decreased with the MAPP content. The fraction of UV stabilizer also had negative effects on the WA, MOR, and maximum strain. This study demonstrated design and analysis of mixture experiments as an efficient tool to optimize the formulation of rPP/RWF composites for minimum WA and for maximum flexural properties.

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References

1. Cui Y, Lee S, Noruziaan B, et al. Fabrication and interfacial modification of wood/recycled plastic composite materials. *Composites Part A* 2008; 39: 655–661.
2. Themelis NJ, Castaldi MJ, Bhatti J, et al. Energy and economic value of nonrecycled plastics and municipal solid wastes that are currently landfilled in the fifty States. EEC study of non-recycled plastics, Earth Engineering Center, Columbia University, 2011.
3. Selke SE and Wichman I. Wood fiber/polyolefin composites. *Composites Part A* 2004; 35: 321–326.
4. Kazemi Y, Cloutier A and Rodrigue D. Mechanical and morphological properties of wood plastic composites based on municipal plastic waste. *Polym Compos* 2013; 34: 487–493.
5. Najafi SK, Hamidinia E and Tajvidi M. Mechanical properties of composites from sawdust and recycled plastics. *J Appl Polym Sci* 2006; 100: 3641–3645.
6. Boukehili H and Nguyen-Tri P. Helium gas barrier and water absorption behavior of bamboo fiber reinforced recycled polypropylene. *J Reinf Plast Compos* 2012; 31: 1638–1651.
7. Nourbakhsh A, Ashori A, Tabari HZ, et al. Mechanical and thermo-chemical properties of wood-flour/polypropylene blends. *Polym Bull* 2010; 65: 691–700.
8. Adhikary KB, Pang S and Staiger MP. Dimensional stability and mechanical behaviour of wood-plastic composites based recycled and virgin high-density polyethylene. *Composites Part B* 2008; 39: 807–815.
9. Homkhiew C, Ratanawilai T and Thongruang W. Composites from recycled polypropylene and rubberwood flour: effects of composition on mechanical properties. *J Thermoplast Compos Mater*. Epub ahead of print 14 February 2013. DOI: 10.1177/0892705712475019.
10. Kuo PY, Wang SY, Chen JH, et al. Effects of material compositions on the mechanical properties of wood–plastic composites manufactured by injection molding. *Mater Des* 2009; 30: 3489–3496.
11. Favaro SL, Lopes MS, Neto AGVC, et al. Chemical, morphological, and mechanical analysis of rice husk/post-consumer polyethylene composites. *Composites Part A* 2010; 41: 154–160.
12. Petchwattana N and Covavisaruch S. Influences of particle sizes and contents of chemical blowing agents on foaming wood plastic composites prepared from poly(vinyl chloride) and rice hull. *Mater Des* 2011; 32: 2844–2850.
13. Mirzaei B, Tajvidi M, Falk RH, et al. Stress-relaxation behavior of lignocellulosic high-density polyethylene composites. *J Reinf Plast Compos* 2011; 30: 875–881.
14. Khalid M, Ratnam CT, Chuah TG, et al. Comparative study of polypropylene composites reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose. *Mater Des* 2008; 29: 173–178.
15. Mishra S and Aireddy H. Evaluation of dielectric behavior of bio-waste reinforced polymer composite. *J Reinf Plast Compos* 2011; 30: 134–141.
16. Karmarkar A, Chauhan SS, Modak JM, et al. Mechanical properties of wood–fiber reinforced polypropylene composites: effect of a novel compatibilizer with isocyanate functional group. *Composites Part A* 2007; 38: 227–233.
17. Najafi SK, Kiaefar A, Hamidina E, et al. Water absorption behavior of composites from sawdust and recycled plastics. *J Reinf Plast Compos* 2007; 26: 341–348.
18. Law TT and Ishak ZAM. Water absorption and dimensional stability of short kenaf fiber-filled polypropylene composites treated with maleated polypropylene. *J Appl Polym Sci* 2011; 120: 563–572.
19. Khanjanzadeh H, Tabarsa T and Shakeri A. Morphology, dimensional stability and mechanical properties of polypropylene-wood flour composites with and without nanoclay. *J Reinf Plast Compos* 2012; 31: 341–350.
20. Li B, Jiang H, Guo L, et al. Comparative study on the effect of manchurian ash and larch wood flour on mechanical property, morphology, and rheology of HDPE/wood flour composites. *J Appl Polym Sci* 2008; 107: 2520–2530.

21. Martinello T, Kaneko TM, Velasco MVR, et al. Optimization of poorly compactable drug tablets manufactured by direct compression using the mixture experimental design. *Int J Pharm* 2006; 322: 87–95.
22. Jun Z, Xiang-Ming W, Jian-Min C, et al. Optimization of processing variables in wood–rubber composite panel manufacturing technology. *Bioresour Technol* 2008; 99: 2384–2391.
23. Matuana LM and Mengeloglu F. Manufacture of rigid PVC/wood-flour composite foams using moisture contained in wood as foaming agent. *J Vinyl Addit Technol* 2002; 8: 264–270.
24. Stark NM and Matuana LM. Ultraviolet weathering of photostabilized wood-flour-filled high-density polyethylene composites. *J Appl Polym Sci* 2003; 90: 2609–2617.
25. Montgomery DC. *Design and analysis of experiments*, 7th edn. Hoboken, NJ: John Wiley & Sons, Inc., 2009.
26. Khosrowshahi YB and Salem A. Influence of polyvinyl alcohol and carboxymethyl cellulose on the reliability of extruded ceramic body: application of mixture design method in fabricating reliable ceramic raschig rings. *Int J Appl Ceram Technol* 2011; 8: 1334–1343.
27. Homkhiew C, Ratanawilai T and Thongruang W. Effect of wood flour content and cooling rate on properties of rubberwood flour/recycled polypropylene composites. *Adv Mater Res* 2012; 488–489: 495–500.
28. Amini M, Younesi H, Bahramifar N, et al. Application of response surface methodology for optimization of lead biosorption in an aqueous solution by *Aspergillus niger*. *J Hazard Mater* 2008; 154: 694–702.
29. Myers RH, Montgomery DC and Anderson-Cook CM. *Response surface methodology: Process and product optimization using designed experiments*, 3th edn. Hoboken, NJ: John Wiley & Sons, Inc., 2009.
30. Shakeri A and Ghasemian A. Water absorption and thickness swelling behavior of polypropylene reinforced with hybrid recycled newspaper and glass fiber. *Appl Compos Mater* 2010; 17: 183–193.
31. Sathishkumar TP, Navaneethakrishnan P, Shankar S, et al. Mechanical properties and water absorption of snake grass longitudinal fiber reinforced isophthalic polyester composites. *J Reinf Plast Compos* 2013; 32: 1211–1223.
32. Adhikary KB, Pang S and Staiger MP. Long-term moisture absorption and thickness swelling behaviour of recycled thermoplastics reinforced with pinus radiata sawdust. *Chem Eng J* 2008; 142: 190–198.
33. Mohebby B, Younesi H, Ghotbifar A, et al. Water and moisture absorption and thickness swelling behavior in polypropylene/wood flour/glass fiber hybrid composites. *J Reinf Plast Compos* 2010; 29: 830–839.
34. Wechsler A and Hiziroglu S. Some of the properties of wood–plastic composites. *Build Environ* 2007; 42: 2637–2644.
35. Nourbakhsh A and Ashori A. Influence of nanoclay and coupling agent on the physical and mechanical properties of polypropylene/bagasse nanocomposite. *J Appl Polym Sci* 2009; 112: 1386–1390.
36. Tamrakar S and Lopez-Anido RA. Water absorption of wood polypropylene composite sheet piles and its influence on mechanical properties. *Constr Build Mater* 2011; 25: 3977–3988.
37. Masoodi R and Pillai KM. A study on moisture absorption and swelling in bio-based jute-epoxy composites. *J Reinf Plast Compos* 2012; 31: 285–294.
38. Ashori A and Nourbakhsh A. Mechanical behavior of agro-residue-reinforced polypropylene composites. *J Appl Polym Sci* 2009; 111: 2616–2620.
39. Rahman MR, Huque MM, Islam MN, et al. Improvement of physico-mechanical properties of jute fiber reinforced polypropylene composites by post-treatment. *Composites Part A* 2008; 39: 1739–1747.
40. Mohanty S, Verma SK, Nayak SK, et al. Influence of fiber treatment on the performance of sisal–polypropylene composites. *J Appl Polym Sci* 2004; 94: 1336–1345.