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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.