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Time–temperature and stress dependent behaviors of composites made from recycled polypropylene and rubberwood flour

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HIGHLIGHTS

- Creep of composites between recycled polypropylene and rubberwood flour is studied.
- Burger and Power law models were both able to fit well the creep data.
- At high temperature and stress levels, Power law gave poorer fit than Burger model.
- HRZ model fitted data almost as well as Power law fits of individual curves.
- Master curves from TTS and TSS were in good agreement.

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ABSTRACT

The effects of time, temperature, and stress on the flexural creep of composites from recycled polypropylene (rPP) and rubberwood flour (RWF) were experimentally investigated and numerically modeled. Creep of rPP/RWF composites increased with an increase of time, temperature, and stress. A critical temperature of rPP composites containing 44.5 wt% RWF is 65 °C. Burger, Power law, and HRZ models fit the creep profiles well in general, but at high temperature and stress levels the Power law and HRZ models performed poorly. However, the HRZ model interpolated creep well across the applied stresses, or across the temperatures. The time–temperature superposition (TTS) and time–stress superposition (TSS) principles were used to model long-term creep. The master curves from TTS and TSS principles were in good agreement with each other. They predicted that the lifetime limitation by long-term creep exceeds 10 years for 15 MPa stress at 25 °C. All these results pertain to a specific formulation of rPP/RWF composites.

1. Introduction

In recent decades, reinforcing thermoplastics with inorganic fibers such as carbon, glass, graphite, and talc, has successfully produced high performance composites [1]. Also the reinforcement use of organic fibers in plastic composites, particularly use of wood flour, is of great interest due to several potential advantages, such as low cost, low density, biodegradability, low health hazard during handling, and non-abrasive nature [2,3]. Therefore the use of wood flour to replace inorganic filler has an increasing trend in the plastic composite industries. Ashori and Nourbakhsh [4] manufactured composites from recycled high density polyethylene (rHDPE) or recycled polypropylene (rPP) and old newspaper fiber. They found that recycled materials can be used to manufacture value-added panels. Boukehili and Nguyen-Tri [5] produced composites from short bamboo fiber and rPP, and found that chemical treatment of fiber significantly reduces the amount of water absorption and improves the impermeability to helium gas. Ashori and Sheshmani [6] made hybrid composite materials from rPP, recycled newspaper fiber (RNF) and poplar wood flour. Composites containing high fraction of RNF exhibited the highest water absorption during the whole duration of immersion. Nourbakhsh and Ashori [7] investigated effects of fiber content on mechanical properties and water absorption of composites based on rHDPE and poplar fibers (*Populus deltoids*). They found that flexural and tensile strengths and the water absorption increased with fiber loading. Furthermore, wood-plastic composites (WPCs) may have good moisture resistance and dimensional stability because of the continuous thermoplastic matrices [8]. They have been largely

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used as a replacement for softwood lumber as decking, railings, door and window frames, and other outdoor applications, where they have better durability than softwood lumber [9,10].

Rubber tree (*Hevea brasiliensis*) is widely planted in Thailand for the production of latex, and is cut down when it becomes unproductive at about 25 years of age [11]. Rubberwood lumber is mainly used to produce furniture, toys, and packing materials. In rubberwood industries, a large amount of wood waste in the forms of flour, sawdust, and chips, is generated at different stages of processing. Generally, rubberwood waste is dumped in landfills or burned, but some of the waste is also used to produce medium-density fiberboard and particleboard [12]. The utilization of rubberwood waste as a filler in polymer composites could decrease environmental impacts from the waste, as well as add value when contributing to the composite properties.

The mechanical characteristics of WPCs include creep; time-dependent deformation under loading due to their continuous thermoplastic matrices, and such creep is a critical issue in many engineering applications including biomedical, aerospace, and civil engineering infrastructure applications [13]. Creep being an important characteristic relates to load-bearing capacity of wood-plastic composite products [14,15]. Likewise, durability and lifetime limitations of products, from large deformations, should be estimated at the design stage [16]. Environmental parameters, such as temperature and humidity, influence the creep of WPCs [17], due to their influences on the polymer and wood [18]. Long term evaluation of creep would be prohibitively costly. Therefore, accelerated testing methods and mathematical models are used instead [18]. Acha et al. [13] investigated the effects of modifying the interfacial adhesion between jute and polypropylene on the creep behavior, and used the time–temperature superposition (TTS) to predict long-term creep deformation. Mosiewicki et al. [19] experimentally evaluated the creep of composites made from linseed oil-based polyester thermoset, and compared the results with Power law and Burger models; both models fitted the data well. Chevali et al. [20] studied flexural creep behavior of nylon 6/6, polypropylene, high-density polyethylene, and their long fiber thermoplastic composites. The HRZ (for Hadid, Rechak, and Zouani, [21]) model provided an excellent fit to the experimental data, and the master curves obtained from TTS were able to predict the long-term creep. Banik et al. [22] investigated the influence of unidirectional and cross-ply polypropylene composites on the creep behavior, and both Burger and Findley Power law models were satisfactory for fitting short-term creep behavior. Subramanian and Senthilvelan [23] experimentally evaluated the creep of leaf springs from glass–fiber-reinforced thermoplastic composites at various loads, and compared the results with the HRZ model. Research projects related to inorganic fiber and natural fiber reinforced plastics were carried out, only a few studies on creep behavior have used rubberwood flour (RWF) to reinforce virgin plastics, and there is no prior report on the creep of RWF-reinforced postconsumer plastics that we focus on.

In our earlier work, the formulation of recycled polypropylene/RWF composites was optimized for select mechanical properties [24], but creep deformation was not investigated. The evaluation of creep is necessary for developing a new product subjected to long-term loading, when the materials are known to have viscoelastic time-dependent behavior. Therefore, creep should affect the design of WPCs and selecting their end-use applications. The objective of this work was to investigate the creep characteristics of composites made from recycled polypropylene and rubberwood flour, in particular the effects of temperature and stress levels. The Burger, Power law, and HRZ models were employed to fit short-term creep data. The time–temperature superposition and the time–stress superposition principles were used to construct master curves of creep deformation, for predicting long-time creep.

2. Materials and methods

2.1. Materials

Rubberwood flour collected from local furniture factory was used as reinforcing filler. Raw material was screened through a standard sieve of mesh size 80 (passing particles smaller than 180 μm) and dried in an oven at 110 °C for 8 h to minimize the moisture content. The main chemical constituents of rubberwood are: cellulose (39%), hemicellulose (29%), lignin (28%), and ash (4%) [25]. The matrix polymer was recycled polypropylene with properties as displayed in Table 1, which purchased as pellets with a melt flow index of 11 g/10 min at 230 °C from Withaya Intertrade Co., Ltd (Samutprakarn, Thailand). Maleic anhydride grafted polypropylene (MAPP) with 8–10% of maleic anhydride, used as a coupling agent to improve interfacial bonding between wood flour and plastic matrix, was supplied by Sigma–Aldrich (Missouri, USA). TH Color Co., Ltd (Samutprakarn, Thailand) supplied hindered amine light stabilizer additive, under the trade name MEUV008, chosen as the ultraviolet (UV) stabilizer. Paraffin wax used as lubricant (Lub) in processing was procured from Nippon Seiro Co., Ltd (Yamaguchi, Japan). The composite formulation was held constant at 50.3 wt% rPP, 44.5 wt% RWF, 3.9 wt% MAPP, 0.2 wt% UV stabilizer, and 1.0 wt% Lub. In our prior work, this formulation had the maximal 47.28 MPa flexural strength, and modulus 2527 MPa [24].

2.2. Composite samples

The WPCs were produced in a two-step process. In the first step to produce WPC pellets, RWF and rPP were blended and formed into composite pellets using a twin-screw extruder (Model SHJ-36 from En Mach Co., Ltd, Nonthaburi, Thailand). The temperature of ten processing zones in the extruder was 160, 165, 165, 170, 130, 150, 155, 160, 160, and 170 °C, respectively, while the screw rotating speed was controlled at 70 rpm, and melt pressure is 0.10–0.20 MPa. The extrudate was passed through a water bath and subsequently pelletized. In the second step to produce WPC panels, the WPC pellets were again dried prior to use in an oven at 110 °C for 8 h. The WPC pellets, MAPP, UV stabilizer, and lubricant were dry-mixed, and fed to the twin-screw extruder. The extruding conditions were as follows: (1) temperature profiles of ten processing zones: 170, 175, 180, 170, 130, 150, 160, 170, 180, and 190 °C, respectively; (2) screw rotating speed: 50 rpm; (3) vacuum venting at 9 temperature zones: 0.022 MPa; and (4) melt pressure: 0.10–0.20 MPa. The samples were extruded through a rectangular 9 × 22 mm^2 die and cooled in atmospheric air. Consequently, the specimens were machined, following the flexural creep testing standard.

2.3. Characterization of the samples

Short-term creep tests of rPP/RWF composites were carried out using an Instron Universal Testing Machine (Model 5582 from Instron Corporation, Massachusetts, USA) with three-point bending as shown in Fig. 1. In all tests, the flexural strain was measured by an Instron extensometer with a travel distance of 5 mm and a gauge length of 10 mm. The specimens were 13 × 4.8 × 100 mm^3 (width × thickness × length), and the test span was 80 mm in the direction of extrusion. To evaluate the effects of various stress levels, creep tests were conducted at 25 °C ambient temperature at ten different stress levels: 3, 7, 11, 15, 19, 23, 27, 31, 35, and 39 MPa. Five levels of temperature in the range from 25 to 65 °C, were used with constant 19 MPa stress to assess temperature effects. This constant stress was approximately 40% of the ultimate flexural strength, from quasi-static tests at 25 °C. Before each creep test, the specimens were equilibrated in an environmental chamber for 15 min. The loading duration of a test was 6000 s (100 min), and there were five replications at each test condition.

Table 1
Mechanical and thermal properties of recycled polypropylene.

	Tensile (MPa)		Flexural (MPa)		Melting temperature (°C)	Crystallization temperature (°C)
	Strength	Modulus	Strength	Modulus		
Recycled polypropylene	25.9	317	44.8	1446	160.2	119.9