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Flexural creep behavior of composites from polypropylene and rubberwood flour

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Abstract. The effects of plastic grades and composition contents on creep behavior of extruded composites from polypropylene and rubberwood flour were investigated. Virgin polypropylene gave lower creep strain than recycled polypropylene, both in composites and as unfilled plastic. An increase of rubberwood flour content reduced the creep deformation of the composites, both virgin and recycled plastics. Maleic anhydride-grafted polypropylene as a coupling, 5 wt% addition increased the creep strain of the composite materials. Likewise, an addition of 1 wt% ultraviolet (UV) stabilizer content significantly enhanced the creep deformation. The results recommend that the amount of UV stabilizer should be as small as possible to limit its negative effects. Four-element Burger model offered a good fitting on the creep behavior of each composite formulation.

Introduction

Natural organic fibers have been often filled to plastic matrix for improving their strength and stiffness, increasing their durability and thermal stability. Recent advances, they create opportunities for improved materials from renewable resources, supporting global sustainability [1]. Polypropylene (PP) is one of the most well-know plastics that have been widely used in wood-plastic composite industries in the past few decades [2]. However, its use in some applications is limited by several disadvantages, such as low creep stability in long-term loading applications [3]. Reinforcement of natural fibers has demonstrated to be an effective response to increase the creep stability of PP. For example, wood flour was used to decrease the creep strain of PP [4], while addition of wood flour from *Eucalyptus saligna* resulted in an increase of the creep resistance of PP [5]. Despite the composites between the natural fiber and the PP were acceptably described for improving the creep behavior, there is no prior report of the positive effect of rubberwood flour (RWF) reinforced PP on the final creep deformation.

As recently reported, the natural organic fibers can considerably improve the mechanical, physical, and thermal properties of polyolefins, such as polypropylene [2, 6]. Therefore, the objective of the recent work was to assess the effects of material compositions (including different grades of plastic and the contents of rubberwood flour, coupling agent, and ultraviolet stabilizer) on the creep behavior of PP/RWF composites.

Experimental

Materials

Recycled polypropylene (rPP) pellets were purchased from Withaya Intertrade Co., Ltd (Samutprakarn, Thailand), with a melt flow index of 11 g/10 min at 230 ^{o}C. Virgin polypropylene (vPP) granules were procured from Mitsui Petrochemical Industries Co., Ltd (Tokyo, Japan), with a melt flow index of 7 g/10 min at 230 ^{o}C. Rubberwood flour, used as the reinforcement, was supplied by a local furniture factory (Songkhla, Thailand). The wood flour was sieved through a standard sieve of 80 mesh size and was dried in an oven at 110 ^{o}C for 8 h before compounding. Maleic anhydride-grafted polypropylene (MAPP), with 8-10% of maleic anhydride, used as a coupling agent was supplied by Sigma-Aldrich (Missouri, USA). Hindered amine light stabilizer additive, chosen as the ultraviolet (UV) stabilizer, was supplied by TH Color Co., Ltd (Samutprakarn, Thailand) under the trade name MEUV008. A paraffin wax was purchased from Nippon Seiro Co., Ltd (Yamaguchi, Japan), used as lubricant (Lub).

Sample preparation

Wood-plastic composites (WPCs) were produced in a two-stage process. In the first stage to produce WPC pellets, RWF and PP were mixed into WPC pellets by using a twin-screw extruder (Model SHJ-36 from En Mach Co., Ltd, Nonthaburi, Thailand). The extruding temperature ranged from 130^{o}C to 170 ^{o}C to reduce degradation of the compositions. In the second stage to produce WPC panels, WPC pellets, MAPP, UV stabilizer, and lubricant were dry-mixed and added into feeder of the twin-screw extruder according to the compositions given in Table 1. The 10 temperature zones of extruder were set to profile in the range of 130-190 ^{o}C, while the screw rotating speed was maintained at 50 rpm. The samples were then extruded through a 9 mm × 22 mm rectangular die and cooled in atmospheric air. Consequently, the specimens were machined following the standards of ASTM for creep and flexural tests.

Table 1 Wood-plastic composite formulation (percent by weight) and creep strain of PP and PP/RWF composites (T = 25 ^{o}C, σ = 19 MPa)

Composite sample	Composition (wt%)						Creep strain (%)		
code	rPP	vPP	RWF	MAPP	UV	Lub	C_e	C_{ve6000}	C_{t6000}
rPP100	100						1.27	0.52	1.79
vPP100		100					1.07	0.44	1.51
rP70R25M3U1	70		25	3	1	1	1.03	0.40	1.43
vP70R25M3U1		70	25	3	1	1	0.94	0.38	1.32
rP60R35M3U0.5	60.3		35.3	3	0.5	1	0.88	0.34	1.22
vP60R35M3U0.5		60.3	35.3	3	0.5	1	0.78	0.27	1.05
rP50R45M3U1	50		45	3	1	1	0.70	0.25	0.95
vP50R45M3U1		50	45	3	1	1	0.68	0.23	0.91
rP68R25M5U1	68		25	5	1	1	1.07	0.43	1.50
rP69R25M5U0	69		25	5	0	1	0.96	0.33	1.29

Note; The selected formulations from the mixture experiment design were carried out. The rP70R25M3U1 means 70 wt% rPP, 25 wt% RWF, 3 wt% MAPP, and 1 wt% UV.

Characterization

Three-point bending creep tests of PP and PP/RWF composites were carried out on an Instron Universal Testing Machine (Model 5582 from Instron Corporation, Massachusetts, USA) in Figure 1a, according to ASTM D2990 standard. Testing specimens of dimensions 4.8 mm × 13 mm × 100 mm and a span of 80 mm were employed for studying. All the tests were conducted under a constant load of 19 MPa and performed at ambient conditions of 25 ^{o}C. The creep test duration is 100 min (6000 sec). Modulus of elastic (MOE) was also measured in a three-point flexural test at a cross-head speed of 2 mm/min, according to ASTM D790 standard. Five replications of each formulation were tested.

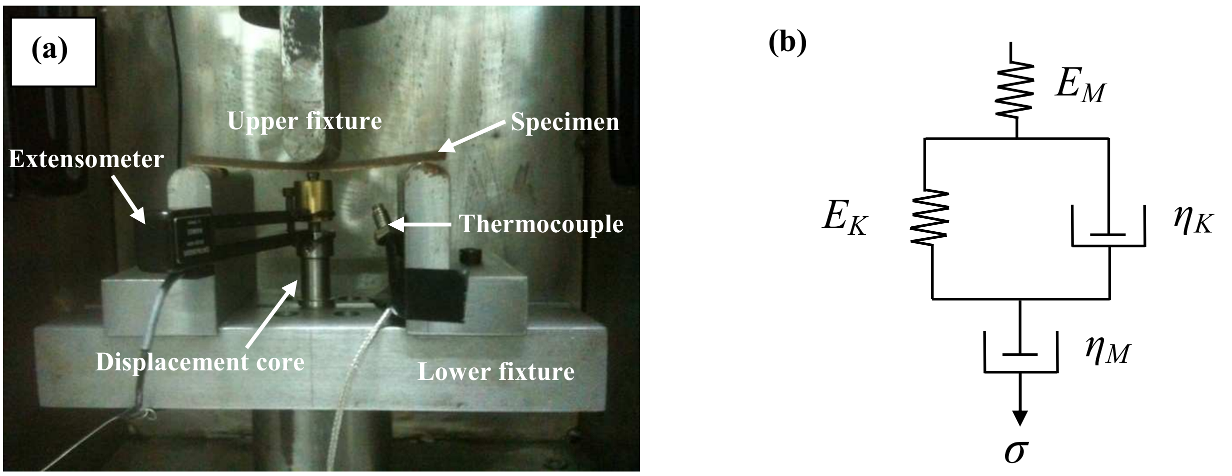

Fig. 1 (a) Test apparatus of creep and (b) schematic of the four-element Burger model.

Creep modeling

Basically, creep strain, $\varepsilon(\sigma, t, T)$, depends on stress ($\sigma$), time ($t$), and temperature ($T$) [7]. It consists of three main elements: (1) elastic deformation (stress-temperature dependence) $\varepsilon_e(\sigma, T)$; (2) viscoelastic deformation (stress-time-temperature dependence) $\varepsilon_{ve}(\sigma, t, T)$; and (3) viscoplastic deformation (stress-time-temperature dependence) $\varepsilon_p(\sigma, t, T)$ [7]:

$$\varepsilon(\sigma,t,T) = \varepsilon_e(\sigma,T) + \varepsilon_{ve}(\sigma,t,T) + \varepsilon_p(\sigma,t,T) \quad (1)$$

To describe and predict the short-term creep behavior, many models were developed and applied by using the constitutive relation of polymeric materials [8]. The four-element Burger model (Figure 1b) is a mathematical model that has been revealed to give a satisfactory prediction and description [8-10]. This model is combinations of Maxwell and Kelvin-Voigt models, which consists of elastic and viscous elements [8, 10]. The mathematical equation for Burger model can be expressed as follows:

$$\varepsilon(t) = \frac{\sigma}{E_M} + \frac{\sigma}{E_K}\left[1 - \exp\left(-t\frac{E_K}{\eta_K}\right)\right] + t\frac{\sigma}{\eta_M} \quad (2)$$

where ε is strain accumulated following time (t), when a certain stress (σ) is employed. E_M represents the elastic modulus of the spring in the Maxwell element, which defines the instantaneous elastic deformation that can be immediately recovered when stress is removed. E_K and η_K represent the elastic modulus of the spring and the viscosity of the dashpot, respectively, in the Kelvin element, which associate with the stiffness and viscous or oriented flow of amorphous polymer chains in the short term. η_M represents the viscosity of the dashpot in the Maxwell element, which defines the viscous flow [11].

Results and Discussion

Effect of composition on creep behavior

The short-term flexural creep behavior of PP and PP/RWF composites with different RWF and additive contents is shown in Figure 2, while the values of the instantaneous creep strain (C_e), of the viscoelastic creep strain after 6000 s (C_{ve6000}), and of the total creep strain after 6000 s (C_{t6000}) are also exhibited in Table 1. As can be seen in Figure 2a, the neat PP (both virgin and recycled) presented the highest creep in the duration of testing, and an increase of rubberwood flour content in the composites showed the decreased creep tendency. This behavior is probably due to an increase in modulus of elastic (MOE) of composites with high wood flour content [12], as shown in Table 2. The MOE of composites (both virgin and recycled plastics) increased with wood flour loadings. Since RWF is a high modulus material compared to the plastic matrix, composites with higher wood flour content require a higher stress for the same deformation [13, 14]. MOE thus has positive effect on decreasing deformation and effective improvement in creep behavior. In addition, unfilled rPP and composites based on rPP show higher creep strain than those based on vPP, for the same plastic to wood ratio. This is probably because of the virgin plastic being stiffer than recycled plastics [13]. However, the two types of plastic with 45 wt% RWF seem to have the same creep behavior, in good agreement with the values of MOE.

The effects of different amounts of MAPP and UV stabilizer on the creep strain are shown in Figure 2b. The effects of 3 wt% MAPP (rP70R25M3U1) and 5 wt% MAPP (rP68R25M5U1) additions on the creep behavior of rPP/RWF composites with 25 wt% RWF show that an increase in the coupling agent content insignificantly increases the creep strain. Generally, the addition of coupling agent in the wood-plastic composites reduced the creep deformation because of the improved filler dispersion and the stronger interfacial adhesion between wood flour and polymer matrix [5, 15, 16]. However, too much MAPP relative to wood flour will causes self-entanglement, resulting in slippage with the PP molecules [13, 17]. Furthermore, addition of 1 wt% UV stabilizer affects the creep strain of the composites with 25 wt% RWF so that the creep strain is significantly increased. This is due to non-homogeneous spatial distribution of wood flour, polymer, and UV stabilizer [13, 18]. Using 1 wt% of UV stabilizer may be unnecessary, and to decrease the negative effects on the creep behavior the amount of UV stabilizer should be as small as possible [13, 18].

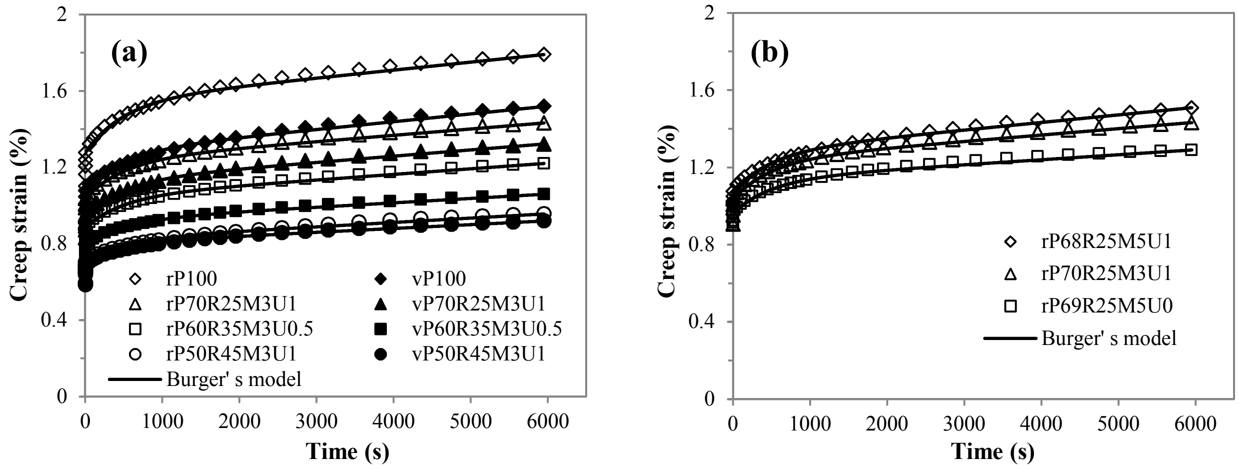

Fig. 2 Creep strain (dot) as a function of times at 25 °C, affected by (a) rubberwood flour contents and (b) additive contents; solid lines represent Burger fit.

Creep modeling analysis

Figures 2a and 2b also show fit of creep curves using Burger model with the solid lines. It can be seen that Burger model provided a good fitting with the experimental data of each formulation. Similar results were found in the work of Liu et al. [9] and Tamrakar et al. [10] who reported that the Burger model offered a good fitting for the creep curves of the composites. The first instantaneous creep arises from the elastic modulus or the spring (E_M) and later time-dependent deflection participates with the spring (E_K) and dashpot (η_K), and last time-dependent deformation comes from the viscous dashpot flow (η_M) [8]. The short-term creep curves were modeled with the Burger equation and the parameters are summarized in Table 2. According to these results, viscosity increased with an increase of the rubberwood flour content and a decrease of the UV stabilizer concentration, and lower flow occurred in the dashpot and permanent deflection also reduced. In the Maxwell spring part, the modulus (E_M) of composites from both virgin and recycled plastics exhibited the enhanced values with the wood flour loading. This is attributed to the stiffness of the composite materials with higher RWF content, and thus reduced the instantaneous elastic deformation during creep experiments. The viscous flow (η_M) values tend to also enhance with the increase of the wood flour content. This is caused by the fact that increasing additions of wood flour content reduced the amount of polymer chains in the plastic composites, resulting in the increase of the viscosity. The retardant elasticity (E_K) and viscosity (η_K) revealed a similar trend on wood flour, coupling agent, and UV stabilizer content, increasing with wood flour content and with a decrease of coupling agent and UV stabilizer content. It could be concluded that the deformation of the Kelvin-Voigt element decreased with increasing wood flour content and enhanced with increasing MAPP and UV stabilizer content.

Table 2 Modulus of elastic and Burger's model parameters

Composite sample code	MOE (GPa)	E_M (MPa)	E_K (MPa)	η_M (MPa·s)	η_K (MPa·s)
rPP100	1.27 (0.06)*	14.89	71.42	4.55E + 05	3.57E + 04
vPP100	1.67 (0.06)	17.64	95.47	4.67E + 05	4.77E + 04
rP70R25M3U1	1.76 (0.03)	18.32	95.95	5.73E + 05	4.79E + 04
vP70R25M3U1	1.93 (0.06)	20.12	103.26	5.82E + 05	5.16E + 04
rP60R35M3U0.5	2.17 (0.08)	21.46	118.01	6.49E + 05	5.90E + 04
vP60R35M3U0.5	2.31 (0.02)	24.20	138.68	8.25E + 05	6.93E + 04
rP50R45M3U1	2.68 (0.08)	27.02	162.39	8.31E + 05	8.11E + 04
vP50R45M3U1	2.66 (0.05)	27.73	165.21	9.50E + 05	8.26E + 04
rP68R25M5U1	2.01 (0.04)	17.67	94.52	4.85E + 05	4.72E + 04
rP69R25M5U0	1.93 (0.09)	19.70	110.46	7.34E + 05	5.52E + 04

*The values in parentheses are standard deviations from five replications.

Conclusions

The effects of plastic grades and contents of wood flour, coupling agent, and UV stabilizer on the creep behavior of PP/RWF composites were investigated. The plastic grades and rubberwood flour contents showed a large impact on the creep behavior of the composites. The neat vPP and composites based on vPP exhibited lower creep deformation than those based on rPP, for the same plastic to wood ratio. The unfilled PP (both virgin and recycled) demonstrated the highest creep strain in range of time studied. The creep strain reduced as the wood flour level increased, due to the resulting increase in stiffness [4]. It was clearly revealed that the addition of rubberwood flour in PP composites can be efficiently improved the poor creep stability of polyolefin. The MAPP and UV stabilizer content also affected the creep deformation of such materials. The additions of 5 wt% MAPP content increased the creep strain of composites, because of the resulting slippage of the PP molecules [13, 17]. Likewise, the creep strain was significantly increased by an addition of 1 wt% UV stabilizer content. To reduce negative effects on the creep behavior the amount of UV stabilizer should be as small as possible. Besides, the short-term flexural creep behavior could be well fitted by using the Burger model, and the data of modeling offered an understanding of the deformation mechanism for three elements: elastic, viscoelastic, and viscoplastic deformation.

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