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Assessing the effect of wood acetylation on mechanical properties and extended creep behavior of wood/recycled-polypropylene composites

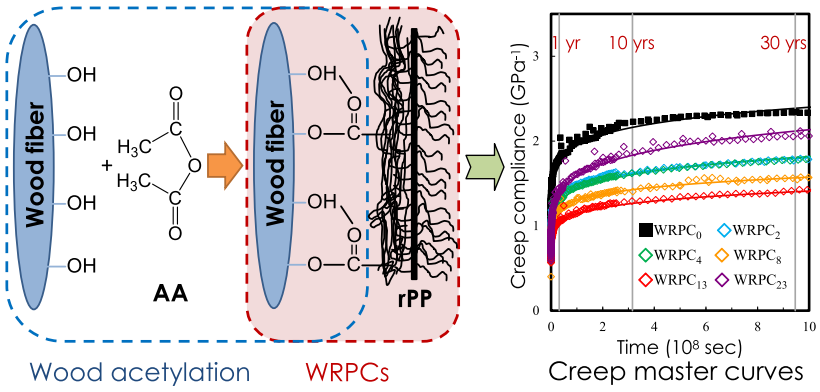
Ke-Chang Hung, Tung-Lin Wu, Yong-Long Chen, Jyh-Horng Wu *

Department of Forestry, National Chung Hsing University, Taichung 402, Taiwan

HIGHLIGHTS

- Mechanical and creep properties of WRPCs with acetylated wood particles were studied.
- The TTSP-predicted creep compliance curve fit well with the long-term experimental data.
- The creep resistances of all acetylated WRPCs were improved 11–41% over a 30-year period.
- WRPC with 13% WPG of acetylated wood particles exhibited the best performance.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates the effect of wood acetylation on the mechanical properties and creep resistance of wood/recycled-polypropylene composites (WRPCs) using the time–temperature superposition principle (TTSP). The results revealed that the flexural and tensile strength of WRPCs increased with increasing weight percent gain (WPG) of acetylated wood particles up to 13%. Additionally, the TTSP-predicted creep compliance curve fit well with the long-term experimental data. The creep resistance of WRPCs with acetylated wood particles was greater than that of WRPCs with unmodified wood particles, especially for the WRPC with 13% WPG of acetylated wood particles.

1. Introduction

Over the past decade, wood–plastic composites (WPCs), in which renewable and low-cost natural fibers are added as reinforcement to the plastic composite, have gained popularity and found important applications in recovery, reuse and recycling of a variety of byproducts from industrial use of natural resources. These WPCs are of great interest in various applications due to such advantages as low-density, low equipment abrasiveness, high stiffness and strength, low maintenance requirements, and biodegradability [1]. However, incompatibility between hydrophilic lignocellulosics and hydrophobic thermoplastics results in poor interfacial interaction. Thus, to overcome this problem, several physical and chemical approaches have been used to modify the lignocellulosic materials by increasing their hydrophobicity [2,3] and improving their dimensional and thermal stabilities [4,5].

* Corresponding author.
E-mail address: eric@nchu.edu.tw (J.-H. Wu).

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Among these various approaches, wood acetylation with acetic anhydride has received the most attention.

The conventional acetylation process includes impregnation of dried wood with a liquid-phase acetic anhydride before external heat is applied. However, the downside to this method is that the process is time-consuming and requires large quantities of modification agent. Therefore, the chemical reagent is used in the vapor phase because the reactivity of the reagent in the vapor phase is higher than in the liquid phase. As a result, acetylation with acetic anhydride *via* a vapor phase reaction offers certain benefits, such as reduced reagent consumption, decreased reaction time, and minimized environmental impact [6]. The best-known virgin thermoplastics used as the matrix in WPC products are high and low density polyethylene (HDPE and LDPE) and polypropylene (PP) [7,8]. In addition, it is well known that all of the recycled plastics that can be melted and processed below the degradation temperature of wood or other lignocellulosic fillers are commonly suitable for manufacturing WPCs [9]. Thus, in the past decade, the use of recycled thermoplastics also has been considered for manufacturing of WPCs [10–12]. However, one of the disadvantages of this approach is changes in the WPC mechanical properties with temperature, which leads to limits in wider applications. Therefore, it is also important to investigate the temperature sensitive properties of WPCs, e.g., creep behavior, because WPCs exhibit a strong time–temperature dependent response. Moreover, it is time-consuming and expensive to conduct full-scale creep tests on a normal time scale. In this study, an accelerated creep test based on the time–temperature superposition principle (TTSP) was implemented to predict the long-term creep response. Methods that use this principle have been employed to confirm that TTSP is applicable to various WPCs [13–17].

To date, investigations into WPCs have primarily focused on the effects of various attributes on the thermal and mechanical properties of the composites (e.g., fiber type, fiber loading, functional additives, and fiber modification) to increase the compatibility between the hydrophilic natural fiber and hydrophobic polymeric matrix [3,18–21]. However, little information is available with respect to the effect of wood acetylation on the creep behavior of WPCs. Therefore, in addition to qualification of the physical properties and static mechanical properties of these materials, an objective of the current study was to investigate the time–temperature dependent response and extended creep behavior of WPCs with unmodified and various acetylated wood particles using the TTSP. In addition, different extents of acetylated wood particles were prepared using the vapor phase reaction method in this study.

2. Materials and methods

2.1. Materials

Japanese cedar (*Cryptomeria japonica* D. Don) sapwood was sourced from the experimental forest of the National Taiwan University. Wood particles were prepared by hammer milling and sieving, and particles between 30 and 60 mesh (250–595 μm) were investigated. Recycled-polypropylene (rPP) with a melt index of 3.7 g/10 min and a density of 910 kg/m^3 was purchased from Orbit Polymers Co., Ltd. (Taichung, Taiwan). Acetic anhydride was purchased from the Sigma–Aldrich Chemical Co. (St. Louis, MO, USA). The other chemicals and solvents used in this experiment were of the highest quality available.

2.2. Acetylation

Wood particles were acetylated with acetic anhydride using the vapor-phase reaction method reported in our previous study [6]. Before the reaction, the wood particles were Soxhlet-extracted with acetone for 8 h, and then dried at 105 °C for 12 h. The reaction conditions for different extents of acetylated particles are shown in Table 1. At the end of the reaction, the acetylated wood particles were washed with distilled water and Soxhlet-extracted with acetone for 8 h. Finally, the acetylated wood particles were dried at 105 °C for 12 h. The weight percent gain (WPG) of the wood materials was calculated based on the oven-dried method.

Table 1
Reaction conditions for different extents of acetylated wood particles.

Wood particles	Wood:acetic anhydride (g:mmol)	Reaction time (min)	Particle weight gain (%)
WPG 2	1:2	120	1.6 ± 0.2
WPG 4	1:4	120	4.4 ± 0.8
WPG 8	1:7	120	7.6 ± 0.1
WPG 13	1:20	120	12.7 ± 0.6
WPG 23	1:40	120	23.3 ± 0.3

2.3. Composite panel manufacture

Manufacture of wood/rPP composites (WRPCs): The flat platen pressing process was applied as reported in our previous papers [22,23]. The weight ratio of the oven-dried wood particles (moisture content <3%) to rPP powder was 10:90. The expected density of the WRPCs was 900 kg/m^3. The dimensions of the WRPC samples were 300 mm × 200 mm with a thickness of 4 mm. All of the WRPCs were produced in a two-step pressing process described as follows: (1) hot pressing (2.5 MPa) at 200 °C for 3 min and (2) finishing with cold pressing until the temperature of WRPCs decreased to 30 °C.

2.4. Mechanical properties

Flexural and tensile tests were performed according to the ASTM D790-10 [24] and ASTM D638-14 [25] standards, respectively. A specimen size of 80 mm × 16 mm with a thickness of 4 mm was used to determine modulus of rupture (MOR) and modulus of elasticity (MOE) in a three-point static bending test with a loading speed of 1.7 mm/min and a span of 64 mm. A dumbbell-shaped (Type I) test specimen with a thickness of 4 mm was used for tensile test at a tensile speed of 5 mm/min. Five specimens of each blend were tested at 20 °C. The samples were conditioned at 20 °C and 65% relative humidity (RH) for two weeks before testing.

2.5. Creep test

The short-term accelerated creep tests on the WRPCs were measured in three-point bending mode *via* dynamic mechanical analysis (DMA) (DMA 8000, PerkinElmer) at a frequency of 1 Hz. The dimensions of the sample were 30 mm × 10 mm with a thickness of 4 mm. With a real-time short-term creep response at elevated temperatures, TTSP is used to predict the long-term creep performance of the composites. The creep compliance is given by $S(T_{ref}, t) = S(T_{elev}, t/\alpha_T)$, where S is the creep compliance as a function of temperature and time, T_{ref} is the reference temperature, T_{elev} is the elevated temperature, and α_T is the shift factor. The master curve of creep compliance was determined by DMA. Creep and creep recovery cycles were conducted at isotherms between 20 °C and 70 °C at intervals of 5 °C. A three-point bending mode with a span of 40 mm was used. For each isotherm, 20% of the average flexural strength was applied for 1 h followed by a 1 h recovery period. In addition, long-term creep tests in bending were conducted to serve as a basis of comparison with the results from short-term accelerated creep tests. The tests were conducted under a conditioned environment (20 °C and 65% RH) for 775 days. The dimensions of the sample were 80 mm × 16 mm with a thickness of 4 mm. The applied loading was 20% of the average flexural strength, and the data for creep displacement were recorded by a data logger.

2.6. X-ray diffraction measurement

The X-ray diffractograms were obtained with a MAC science MXP18 analyzer (Japan), and wood samples were prepared by powdering. The diffraction patterns were measured from $2\theta = 2°$ to 35° using CuKα_1 radiation at 40 kV and 30 mA. The crystallinity index (*CrI*) of the wood particle was calculated according to the following equation [26]:

$$CrI(\%) = \frac{I_{002} - I_{am}}{I_{002}} \times 100 \quad (1)$$

where I_{002} is the maximum intensity of the 002 lattice reflection of the cellulose crystallographic form at $2\theta = 22°$, and I_{am} is the intensity of diffraction of the amorphous material at $2\theta = 18°$.

2.7. Scanning electron microscopy

Scanning electron microscopy (SEM) was used to examine the morphology of wood particles and plastics in the composites. After the tensile test, the fracture surfaces of composites were dried and sputtered with gold. A JEOL JSM-6330F SEM (Japan) instrument equipped with a field emission gun and an acceleration voltage of 2.8 kV was used to collect SEM images for the composite specimen. The samples were viewed perpendicular to the fractured surface.