

# Environmental effects on the mechanical and thermomechanical properties of aspen fiber–polypropylene composites

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## Abstract

The mechanical properties of newly developed aspen fiber–polypropylene composites (APC) were experimentally explored and numerically predicted at the temperatures and humidity that are typical for domestic housing applications. The mechanical properties of APCs with five different fiber-loadings were evaluated at the room temperature, 4 °C, and 40 °C. Environmental effects on the mechanical properties of APCs were experimentally quantified after conditioning the APCs with two different fiber-loadings in the following temperature and humidity for over 7000 h: (1) hot/dry at 40 °C and 30% relative humidity (RH), (2) hot/wet at 40 °C and 82% RH, (3) cold/dry at 4 °C and 30% RH, and (4) cold/wet at 4 °C and 82% RH. The tensile moduli, flexural moduli, and the flexural strength increased as the woodfiber content increased in the composites. However, the tensile strength decreased as the fiber content increased. The tensile strength was shown to slightly improve with an addition of a coupling agent between the aspen fibers and polypropylene. The simple empirical micromechanics Halpin–Tsai model for randomly distributed short fiber reinforced composites was employed to predict the homogenized elastic moduli of APC, by optimizing the interfacial model parameter. Scanning electron microscopy (SEM) micrographs confirmed that an addition of the adhesion promoter maleated anhydride polypropylene (MAPP) between the aspen fibers and polymeric matrix improved the interfacial bonding.

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## 1. Introduction

Woodfiber plastic composites (WPCs) have received considerable attention in the forest product industry for civil engineering applications due to their many merits such as increased durability of woods, low maintenance during service, low cost, and ease of fabrication using traditional plastic/composite processing techniques [1,2]. Knowledge of mechanical properties of WPCs under special environmental conditions for domestic housing industry is essential for their applications as housing structural materials [3–5]. Substantial research has been conducted on the inter-

actions between cellulosic wood fibers and amorphous polymers [6]. The mechanics of woodfiber composites is not only a function of fiber tensile strength, fiber volume fraction, and matrix strength, which can be demonstrated by using micromechanics theory [7], but also depends on the chemical and physical bonding between the woodfiber and the polymer matrix [6–10]. Currently, the major limitation of using wood fibers for reinforcement is the poor interfacial adhesion between the polar-hydrophilic wood fibers and the non-polar-hydrophobic polymers. Maleated anhydride polypropylene (MAPP) has been used in WPCs to increase compatibility between polypropylene and the woodfiber and thus improves the microscopic interfacial bonding between the woodfiber and plastic matrix [7–10]. Aspen fiber–polypropylene composites (APCs) are newly manufactured wood-based composites used to prolong

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and enhance the longevity of some wood structures [11–13]. The addition of a small amount of MAPP has shown considerable improvements in the mechanical strength of certain WPCs but no quantification of the effect of MAPP on APCs exists.

Numerous micromechanics-based modeling approaches have been developed to evaluate the elastic properties of short fiber reinforced composites. The first studies of discontinuous or short fiber reinforced composites were studies of the paper manufacturing process [14] and whisker-reinforced metals [15]. Rigorous theoretical analyses on composites of randomly oriented fibers with high volume fractions were subsequently performed [16,17]. Halpin–Tsai [18,19] developed a simple and effective semi-empirical model to study randomly oriented short fibers in polymer matrix composites based on micromechanics analysis and finite element simulation. A multi-inclusion unit cell was employed to study the elastic and elastoplastic behavior of metal matrix composites reinforced by randomly oriented short fibers [20]. The Halpin–Tsai empirical model was recognized as providing a good prediction of elastic moduli compared to the many modified Eshelby model and modified shear-lag models [20]. Thus, the Halpin–Tsai model was used in this study on WPCs with the statistical analysis on the randomly distributed fibers with an orderly pattern.

The aim of this research was to systematically study the structure-property relations of APCs with or without MAPP with the intention to optimize the mechanical properties of APCs. Furthermore, we employed the Halpin–Tsai model to determine the homogenized elastic moduli of the APCs, which had not been accomplished to date, to the authors' knowledge. To this end, APCs were prepared with five fiber-loadings ranging from 0 to 60 wt% in a polypropylene (PP) matrix with and without MAPP. Tensile and flexural tests were performed to evaluate the tensile and flexural moduli and strengths of the composites under various temperatures. Temperature and humidity effects on the mechanical properties were also experimentally quantified by evaluation after conditioning the APCs in the following temperatures and humidity for over 7000 h: (1) hot/dry at 40 °C and 30% relative humidity (RH) that mimics the summer in Arizona, (2) hot/wet at 40 °C and 82% RH that mimics the summer in Florida, (3) cold/dry at 4 °C and 30% RH that mimics most of the weather in Montana, and (4) cold/wet at 4 °C and 82% RH that mimics weather conditions in Wisconsin. Microstructures of cross sections of failed specimens were examined using a scanning electron microscope (SEM) to analyze the failure mechanisms of the composites.

## 2. Experiments

### 2.1. Materials and specimens

Harvested from mature trees, aspen filaments of approximately 1 cm in length with 6–9% moisture content were

Table 1  
Compositions of aspen fiber–polypropylene composites

Material #	aspen fiber (wt%)	Polypropylene (wt%)	MAPP (wt%)
PP-00	0	100	0
PP-02	0	98	2
APC-30-00*	30	70	0
APC-30-02*	30	68	2
APC-40-00	40	60	0
APC-40-02	40	58	2
APC-50-00*	50	50	0
APC-50-02*	50	48	2
APC-60-00	60	40	0
APC-60-02	60	38	2

\* APCs environmentally conditioned and tested at elevated (40 °C) and near frozen (4 °C) temperatures.

used along with a polypropylene (PP) homopolymer, Solvay 1602, to fabricate the APCs. For a special batch, the wood fiber was first treated with the coupling agent MAPP. The blending was accomplished at 4600 rpm, which resulted in a blade tip speed of about 30 m/s. The blended mass was then automatically discharged at 190 °C. The total residence time of the blending averaged about 2 min. The discharged mass was cooled in a cold press and granulated. The granules were then injection molded into ASTM standard specimens using a 33-ton Cincinnati Milacron injection-molding machine at 190 °C. The specimens were then stored at 20% humidity and 32 °C for at least three days. It should be noted that APCs that are fabricated via extrusion or in-jet molding of the melt plastic with fine short solid wood fibers exhibit preferred fiber orientations along the extrusion directions [21].

APCs were prepared with woodfiber weight percentages of 0, 30, 40, 50, and 60. The 2 wt% of MAPP was introduced to the composite by replacing the corresponding PP in the composite in the special batch. All the composite compositions are listed in Table 1.

### 2.2. Temperature and humidity conditionings

APCs of 30 wt% and 50 wt% aspen fiber contents with and without MAPP were subjected to four environmental conditioning for over 7000 h prior to testing, according to temperature and humidity conditions that are typical for domestic housing applications, which are: (1) 40 °C and 30% relative humidity (RH) that mimics the summer in Arizona (hot/dry), (2) 40 °C and 82% RH as the summer in Florida (hot/wet), (3) 4 °C and 30% RH as the weather in Montana (cold/dry), and (4) 4 °C and 82% RH as those in Wisconsin (cold/wet). The isothermal 'hot/dry' environmental condition was achieved using a convection oven with a digital temperature controller. The 'hot/wet' environment exposure was accomplished by exposing the specimens in a Tenney Bench-Top Environmental Chamber. The isothermal 'cold/dry' and 'cold/wet' environment exposure was conducted in a refrigerator with temperature and humidity functions. No load was applied to the

specimens during the environmental exposure. The mechanical tests were conducted immediately after the exposure. It should be noted that after exposure to the ‘hot/wet’ and ‘cold/wet’ conditions, the APC specimens showed slight weight gains due to moisture absorption. The weight gain after 7000 h exposure was less than 0.1% of the total weight of the specimen.

### 2.3. Mechanical measurements

Tensile properties were obtained according to ASTM D3039 under uniaxial tension while flexural properties were tested according to ASTM D 790 using a three-point bend setup. The tests were conducted at atmospheric pressure in laboratory air (23 °C, 50% RH) using a 10 kN servo-hydraulic test frame equipped with a digital controller and computer data acquisition. All of the experiments were under displacement-control at a loading rate of 1.25 mm/min, corresponding to an average strain rate of approximately 0.01 per minute. Extensometers of a 1-in. span were used to measure the strain during tensile tests. To reduce experimental error, at least five replicate tests were performed. Selected composites of interests, denoted by \* in Table 1, were tested for properties at the near frozen (4 °C) temperature and the elevated (40 °C) temperature, according to typical conditions in domestic housing applications. The specimens after environmental conditioning were also tested at the three temperatures. The elevated and near frozen temperature tensile and flexural tests were conducted inside an environmental chamber mounted on the test frame. Prior to testing, the specimens were kept in the chamber for at least 25 min to ensure that the temperature of the specimens reached equilibrium.

## 3. Results and discussion

### 3.1. Tensile and flexural properties

APCs with and without MAPP additives were tested with the aspen fiber contents from 0 wt% to 60 wt%. Fig. 1 shows the tensile moduli and strength of the APCs as a function of the weight fraction of the woodfiber. The statistical deviation of the measurement is less than 5% for APCs of different fiber contents. We observed that as the woodfiber content increased the tensile modulus increased. The maximum tensile modulus was over 3.6 times of that of neat polypropylene (PP) with 60 wt% woodfiber content. In contrast, the composite tensile strength decreased as the woodfiber contents increased when MAPP was not present. With MAPP, the tensile strength was slightly higher than that of PP with the maximum strength approximately 1.5 times the strength of neat PP. It is probable that the stress concentration at the wood-fiber tip induce a poor fiber/matrix adhesion that led to poor APC tensile strengths. Fig. 2 shows the flexural modulus and strength of the composite as a function of the weight fraction of aspen fiber. The flexural modulus of

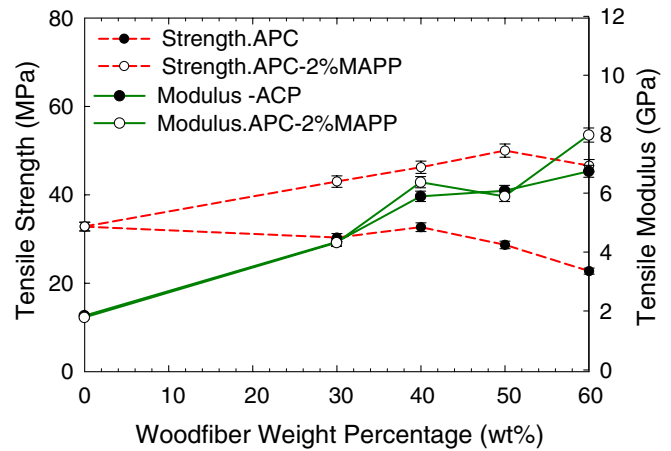


Fig. 1. Tensile modulus and strength of aspen fiber-polypropylene composites with the fiber weight fraction from 0% to 60% and with 2% maleated anhydride polypropylene replacing the same amount of polypropylene.

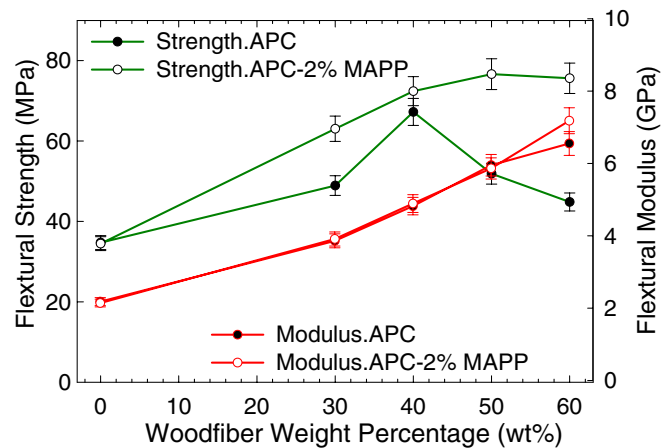


Fig. 2. Flexural modulus and strength of aspen fiber-polypropylene composites with the fiber weight fraction from 0% to 60% and with 2% maleated anhydride polypropylene replacing the same amount of polypropylene.

the composite increased almost linearly as the woodfiber content increased. The maximum flexural modulus of the APC was approximately two and half times that of neat PP. The flexural strength increased almost in a parabolic manner with respect to the woodfiber weight fraction with its maximum value occurring at 40 wt% fiber fraction for the PP-only matrix and at 50 wt% for the MAPP added matrix. Clearly, the MAPP additives significantly improved the flexural strength of the APCs. The maximum flexural strength of the APC was approximately two times that of PP.

### 3.2. Temperature effects

APCs with 30 wt% and 50 wt% woodfiber content were further tested at room temperature, 4 °C, and 40 °C. Table 2 shows the tensile strength of the APC-30s and APC-50s

with and without MAPP additives. The tensile strength at 40 °C showed approximately 10% reduction compared to the tensile strength at room temperature. However, the tensile strength at 4 °C was approximately 20–40% higher than those tested at room temperature. The temperature dependence on the tensile properties was not a function of the woodfiber content. Table 2 also shows the flexural strength of the APCs at 40 °C and at 4 °C. A considerable reduction (approximately 50%) in flexural strength was observed at the elevated (40 °C) temperature compared to those at room temperature. In contrast, the flexural strength at the near frozen (4 °C) temperature decreased (approximately 10–30%) except for the APC-50 without MAPP additives, which showed approximately 10% increase.

### 3.3. Environmental effects

APCs with 30 wt% and 50 wt% woodfiber content after 7000 h conditionings that mimic typical domestic house application were further tested under quasi-static uniaxial tension at room temperature, 4 °C, and 40 °C. Fig. 3 shows the tensile strength of the APCs of 30 wt% aspen fiber contents (APC-30s) and APCs of 50 wt% aspen fiber contents (APC-50s) with and without MAPP additives after exposed to 40 °C/82% RH, 4 °C/82% RH, 40 °C/30% RH, and 4 °C/82% RH for 7000 h respectively. First, comparing to the corresponding tensile strengths at room temperature, 4 °C, and 40 °C shown in Table 2, the APCs demonstrated reductions in tensile strength of approximate 20% when tested at room temperature and approximate 30% when tested at 4 °C and 40 °C. APC-50s showed higher reduction in tension strengths than APCs-30s. Second, the reduction in tensile strengths of APCs' conditioned in higher humidity, 82% RH, is higher than that of the APCs conditioned in the lower humidity, 30% RH. When conditioned at the same humidity level, the APCs displayed similar tensile strength reductions while conditioned at 4 °C and 40 °C. Some large statistical variation in tensile strengths were shown in Fig. 3, which suggests more samples should be tested for better understanding of the properties of APCs for future study.

### 3.4. Microstructure

The influence of the MAPP coupling agent on the mechanical properties was widely observed in mechanical testing. Figs. 4 and 5 show SEM micrographs of the frac-

tured surface of the APCs. Without MAPP, fiber/matrix debonding and fiber pullout were observed, especially at high aspen fiber content (see Fig. 5a), resulting in lower APC mechanical properties. With MAPP, fiber breakage and matrix fractures were observed (see Figs. 4b and 5b). Therefore, the MAPP coupling agent helped to enhance the interfacial bonding between the aspen fiber and the polypropylene matrix.

## 4. Micromechanics models

A micromechanical model was developed to capture the elastic moduli of the composite based on the properties and the interfacial bonding of the composites' constituents. The typical fiber distribution in the WPCs was demonstrated by Gamstedt et al. [21] for an unbleached kraftcooked wood fiber reinforced vinyl ester using a scanning electron microscope. Using image analysis, the distribution of woodfiber was characterized as a Gaussian distribution as shown in Fig. 6. Here the angle  $\theta$  represents the fiber orientation's offset from the extrusion direction.

A composite with unidirectional oriented short fibers as reinforcements formed a transversely isotropic composite. The longitudinal, transverse, and shear moduli were modeled using the Halpin–Tsai semi-empirical model [19] with the empirical parameter  $\xi_i$  obtained from model calibration which is written as the following:

$$E_1 = E_m \left[ \frac{1 + \xi_1 \eta_1 v_f}{1 - \eta_1 v_f} \right] \left( \psi \eta_1 = \left[ \frac{E_{f1}}{E_m} \right] \left( -1 \right) \left[ \frac{E_{f1}}{E_m} \right] \left( + \xi_1 \right)^{-1}, \psi \xi_1 = \frac{2L}{d} \right), \quad (1a)$$

$$E_2 = E_m \left[ \frac{1 + \xi_2 \eta_2 v_f}{1 - \eta_2 v_f} \right] \left( \psi \eta_2 = \left[ \frac{E_{f2}}{E_m} \right] \left( -1 \right) \left[ \frac{E_{f2}}{E_m} \right] \left( + \xi_2 \right)^{-1}, \psi \xi_2 = 2, \right) \quad (1b)$$

$$G_{12} = G_m \left[ \frac{1 + \xi_3 \eta_3 v_f}{1 - \eta_3 v_f} \right] \left( \psi \eta_3 = \left[ \frac{G_f}{G_m} \right] \left( -1 \right) \left[ \frac{G_f}{G_m} \right] \left( + \xi_3 \right)^{-1}, \psi \xi_3 = 1, \right) \quad (1c)$$

where  $E_1$ ,  $E_2$ ,  $G_{12}$  are the longitudinal, transverse and shear moduli of the composite, respectively;  $E_{f1}$  and  $E_{f2}$  are the longitudinal and transverse modulus of the fiber;  $E_m$  is the modulus of the matrix;  $v_f$  is the volume fraction of the fiber; and  $\xi_i$ ,  $i = 1, 2, 3$ , represents suggested values based on experiments by Halpin [19].  $L$  is the average length of the aspen fiber and  $d$  is the averaged diameter of the aspen fiber in the APCs. Poisson's ratio of the

Table 2

Tensile and flexural strength of 30 wt% and 50 wt% aspen fiber–polypropylene composites with and without maleated anhydride polypropylene at room temperature, 40 °C, and 4 °C, where the variation of the strength is  $V(T_1, T_2) = [V_{11} - V_{12}] / V_{12}$

Materials	Tensile strengths (MPa) @					Flexural strengths (MPa) @				
	25 °C	40 °C	4 °C	SDV (40 °C, RT)	SDV (4 °C, RT)	25 °C	40 °C	4 °C	SDV (40 °C, RT)	SDV (4 °C, RT)
APC-30-00	31.10	26.99	54.59	−0.13	0.76	49.93	29.89	39.77	−0.40	−0.20
APC-30-02	45.44	37.37	54.59	−0.18	0.20	61.55	35.05	52.14	−0.43	−0.15
APC-50-00	30.69	26.69	79.60	−0.13	1.59	51.4	29.39	56.84	−0.43	0.11
APC-50-02	51.76	44.87	79.93	−0.13	0.54	77.21	38.71	55.35	−0.50	−0.28

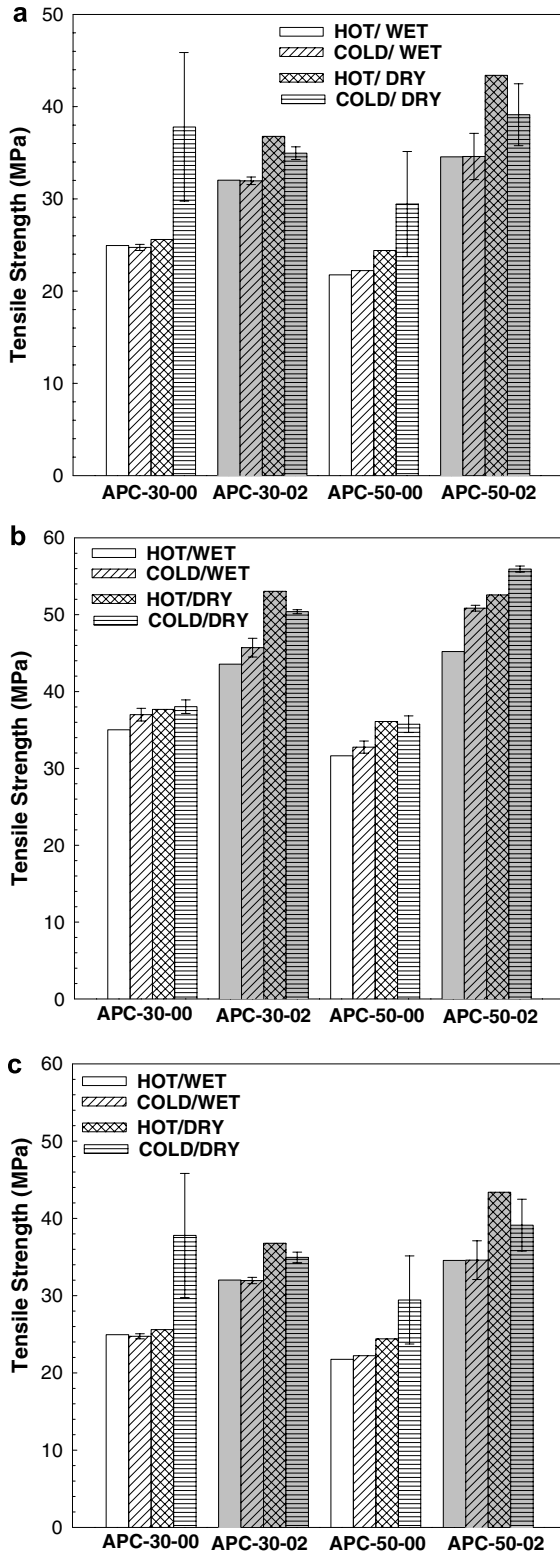


Fig. 3. Tensile strengths of APC-30s and APC-50s, at (a) room temperature, (b) elevated temperature (4 °C) and (c) near frozen temperature (−40 °C), respectively, after 7000 h conditioning at: (1) hot/wet at 40 °C and 82% RH, (2) cold/wet at 4 °C and 82% RH, (3) hot/dry at 40 °C and 30% relative humidity (RH), and (4) cold/dry at 4 °C and 30% RH.

composite was calculated using a simple rule of mixtures. Using the transformation relations for the elastic con-

stants, we can obtain the properties of the composite at directions  $\theta$  from the fiber orientation angle [22]. If  $\theta$  is the angle between the fiber orientation and the axis- $x$  in the global Cartesian coordinate ( $x, y, z$ ), we have

$$\frac{1}{E_x} = \frac{c^4}{E_1} + \left[ \frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1} \right] s^2 c^2 + \frac{s^4}{E_2}, \quad (2a)$$

$$\frac{1}{E_y} = \frac{s^4}{E_1} + \left[ \frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1} \right] s^2 c^2 + \frac{c^4}{E_2}, \quad (2b)$$

$$\frac{1}{G_{xy}} = 2 \left[ \frac{2}{E_1} + \frac{2}{E_2} + \frac{4\nu_{12}}{E_1} - \frac{1}{G_{12}} \right] s^2 c^2 + \frac{1}{G_{12}} (c^4 + s^4), \quad (2c)$$

$$\nu_{xy} = E_x \left\{ \frac{\nu_{12}}{E_1} (c^4 + s^4) - \left[ \frac{1}{E_1} + \frac{1}{E_2} - \frac{1}{G_{12}} \right] s^2 c^2 \right\}, \quad (2d)$$

where  $c = \cos(\theta)$ ,  $s = \sin(\theta)$ ,  $E_x$ ,  $E_y$ ,  $G_{xy}$  are the longitudinal, transverse, and shear moduli respectively, and  $\nu_{xy}$  is Poisson's ratio of the composite. To determine the mechanical properties of a composite that account for an arbitrary fiber distribution  $c(\theta)$ , the modified Nielsen–Chen equation [23] using the statistical distribution function was employed, *i.e.*

$$\tilde{E}_x = \frac{\int_0^{\pi/2} [E_x(\theta)] c(\theta) d\theta}{\int_0^{\pi/2} c(\theta) d\theta}, \quad (3)$$

where  $\tilde{E}_x$  is the averaged modulus. Since the distribution function is symmetric on  $\{-\pi, \pi\}$ , the integration is simply limited at  $\{0, \pi/2\}$ . This equation can also be used to calculate the transverse modulus, shear modulus, and Poisson's ratio of the composite.

## 5. Model prediction for APCs

The mechanical properties of aspen fiber are as follows,  $E_f = 10.9$  GPa,  $E_t = 1.7$  GPa,  $G_{ft} = 2.7$  GPa,  $\nu_{ft} = 0.45$  and of PP are  $E_m = 2.8$  GPa and  $\nu_m = 0.37$ . We also assumed that the fiber orientation obeyed a Gaussian distribution,  $G(\theta) = A e^{-\frac{\theta^2}{2\sigma^2}}$  with the standard deviation of the fiber orientation distribution as  $\sigma = 0.62$ , as measured by Gamstedt [22], where  $\theta$  is the fiber orientation angle against the extrusion direction. As such, the mechanical properties of the APCs were calculated and shown in Table 3. The fiber weight percentage in the composite was calculated using  $w_f\% = \frac{\text{pvf} \cdot d_f}{\text{pvf} \cdot d_f + (1 - \text{pvf}) \cdot d_m}$ , and  $d_f$  and  $d_m$  are the density of the aspen fiber and PP, respectively. Fig. 7 shows that the predicted elastic moduli of APCs demonstrated almost a linear increase as the fiber content increased. The Halpin–Tsai model along with the Nielsen–Chen statistical treatment predicted the tensile modulus along the extrusion direction that correlated fairly well with the experimental results as shown in Fig. 7. It should be noted that the predicted modulus along the extrusion direction was slightly lower than the experimental results, which arose because the Halpin–Tsai model was a conservative estimate of the elastic modulus.

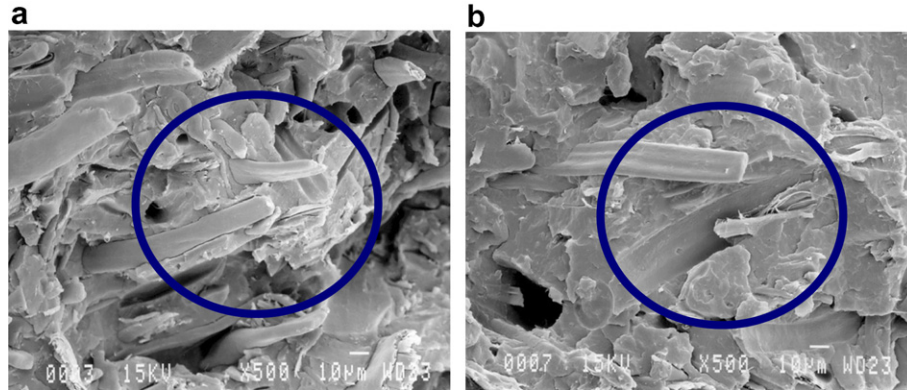


Fig. 4. SEM micrographs showing the cross section of fractured 30 wt% aspen fiber–polypropylene composites (a) without maleated anhydride polypropylene and (b) with 2 wt% maleated anhydride polypropylene.

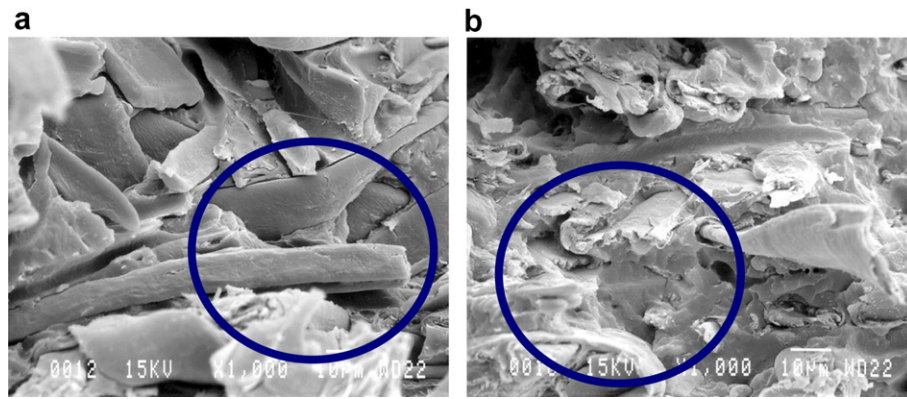


Fig. 5. SEM micrographs showing the cross section of fractured 30 wt% aspen fiber–polypropylene composites (a) without maleated anhydride polypropylene and (b) with 2 wt% maleated anhydride polypropylene.

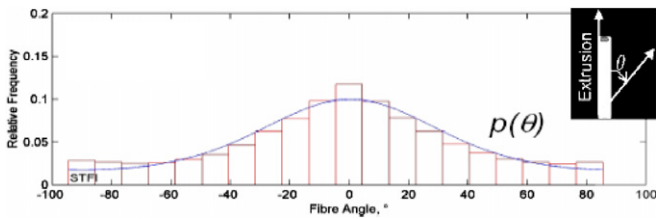


Fig. 6. The fiber orientation distribution,  $\theta$  is the fiber orientation angle with respect to the extrusion direction, and  $p(\theta)$  is the distribution function of the angle  $\theta$ .

6. Conclusions

The mechanical properties of aspen fiber–polypropylene composites (APCs) were studied with the aspen fiber weight percentages of 0, 30, 40, 50, and 60, and with 2 wt% of maleated anhydride polypropylene (MAPP) by replacing the corresponding polypropylene (PP). The tensile moduli, flexural moduli, and flexural strength of the APCs at room, elevated, and near frozen temperatures were represented as a function of aspen fiber content in

Table 3

The elastic moduli of aspen fiber–polypropylene composites predicted using Halpin–Tsai (H–T) model with the fiber volume fraction (vol%) varying from 0% to 70%

Fiber (vol%)	Fiber (wt%)	E1-APC (GPa)	E2-APC (GPa)	G-APC (GPa)	$\nu$ -APC
0	0.00	2.08	2.08	0.52	0.329
10	4.55	2.43	2.11	0.83	0.321
20	9.68	2.76	2.14	0.80	0.325
30	15.52	3.09	2.17	0.41	0.336
40	22.22	3.43	2.19	0.84	0.352
50	30.00	3.76	2.22	1.14	0.37
60	39.13	4.10	2.25	1.52	0.391
70	50.00	4.45	2.27	1.56	0.414

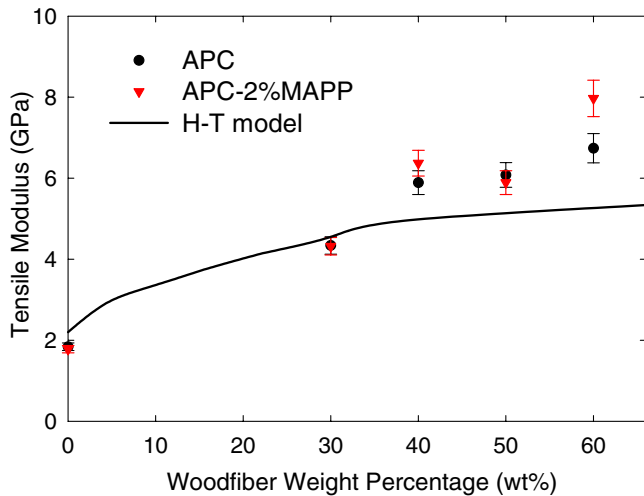


Fig. 7. Tensile moduli of aspen fiber–polypropylene composites (APC) along the extrusion direction obtained experimentally and predicted using Halpin–Tsai (H–T) model.

the composites. Aspen fiber–polypropylene composites were found to exhibit increased mechanical properties over the non-reinforced polymer under tensile and flexural loads. In general, the APC mechanical properties decreased as the temperature increased. The MAPP as an additive to the polypropylene matrix was found to enhance the tensile and flexural properties and also to improve the microscopic interfacial bonding between the fibers and matrix. A micromechanics formulation based on the Halpin–Tsai model was developed to study the moduli of APCs. The model predictions of the elastic moduli of the APCs of various woodfiber volume fractions correlated very well with the experimental results.

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