

# Co-Extruded Wood-Plastic Composites with Talc-Filled Shells: Morphology, Mechanical, and Thermal Expansion Performance

Runzhou Huang,<sup>a</sup> Birm-June Kim,<sup>b</sup> Sunyoung Lee,<sup>c,\*</sup> Zhang Yang,<sup>a</sup> and Qinglin Wu<sup>a,d,\*</sup>

The effect of unmodified talc particles on the mechanical and thermal expansion performance of talc-filled high density polyethylene (HDPE) and co-extruded wood plastic composite (WPC) with talc-filled shells was studied. The use of talc in HDPE helped enhance its tensile, bending, and dynamic modulus, but lowered its tensile and impact strength. The selected models for composite modulus and tensile strength fit the data well after adjusting the model parameters. Talc-filled HDPE had lower linear coefficient of thermal expansion (LCTE) values in comparison with the neat HDPE values, and the LCTE reduction rate increased after the talc loading levels increased above the 30 wt%. Extruding a relatively thick, less-stiff HDPE shell with a large LCTE value over a stiff and thermally stable WPC core decreased overall composite modulus and increased the LCTE values. The composite modulus and strength increased and LCTE values decreased with increase of the talc loading levels in the shell. The impact strength of co-extruded WPC was greatly enhanced with unfilled- and filled HDPE shells.

*Keywords:* Co-extrusion; Core-shell structure; Mechanical property; Talc; Thermal expansion; WPC

*Contact information:* a: College of Wood Science and Technology, Nanjing Forestry University, Nanjing, 210037, China; b: Department of Forest Products & Biotechnology, Kookmin University, Seoul 136-702, Korea; c: Korea Forest Research Institute, Seoul 130-712, Korea; d: School of Renewable Natural Resources, Louisiana State University Agricultural Center, Baton Rouge, LA 70803, USA;

\* Corresponding author: wuqing@lsu.edu and nararawood@forest.go.kr.

## INTRODUCTION

As a class of new-generation green structural materials, co-extruded wood plastic composites (WPCs) with a core-shell structure have been recently developed to enhance performance characteristics of WPC (e.g., Stark and Matuana 2007; Yao and Wu 2010; Kim *et al.* 2012). The shell layer, made of thermoplastics unfilled or filled with minerals and other additives, plays a critical role in modifying the overall composite properties (e.g., Yao and Wu 2010). It was demonstrated that a pure high density polyethylene (HDPE) or a pure polypropylene (PP) shell over a WPC core reduced moisture uptake compared with non-coextruded WPC. However, the addition of a pure plastic shell with a relatively low modulus and large thermal expansion over a WPC core negatively affected overall composite modulus and thermal stability (Jin and Matuana 2010; Yao and Wu 2010). Effort has been made to develop stabilized shell layers by using carbon nanotubes (Jin and Matuana 2010) and by using combined wood and mineral fillers (Yao and Wu 2010; Kim

*et al.* 2012) in the shell layer. Huang *et al.* (2012) demonstrated the influence of varying shell moduli and thermal expansion coefficients of glass-fiber filled HDPE shells on the overall thermal expansion of co-extruded WPCs using a finite element model. Further development of filled materials as more cost-effective shell layers for co-extruded WPC is still necessary.

Talc,  $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$  with an average density of  $2.75\text{g/cm}^3$ , is one of the most commonly used fillers in plastic-based composites (Karrad *et al.* 1998; Huda *et al.* 2007). The use of talc can have a positive influence on modulus and strength but may decrease toughness and ductility of filled plastics (Weon and Sue 2006). In wood and natural fiber plastic composites, talc is used to reduce material cost and to improve stiffness and durability (Noel and Clark 2005; Klyosov 2007). It is shown that talc (up to 30 wt%) can have a positive effect on modulus, strength, processing efficiency, creep, and elastic recovery performance of WPCs (Noel and Clark 2005). Lee *et al.* (2008) studied thermal and mechanical properties of wood flour (WF) and talc-filled poly(lactic) acid (PLA) composites. The addition of WF and WF-talc mixture into neat PLA resulted in some decreases in the glass transition and crystallization temperatures of the composites. The addition of talc in combination with a silane agent to PLA-WF composites improved the tensile modulus. The tensile strength of the composites decreased slightly with the addition of talc, but it was considerably improved with the use of 1 wt% silane. Hybrid bio-composites from talc, WF, and polyhydroxybutyrate-co-valerate (PHBV) were analyzed by Singh *et al.* (2010). Synergistic improvement in the mechanical properties of PHBV-WF composites were obtained with the additional reinforcement of micro-sized talc. The hybrid green composites showed up to 200% increase in the Young's and flexural moduli with the dual reinforcement of 20 wt% talc and 20 wt% wood fiber in the PHBV matrix. A 36% decrease of linear coefficient of thermal expansion (LCTE), and an improvement in heat deflection temperature of the hybrid composite were also observed.

The use of talc in the shell layer of co-extruded WPC is very common to help lower material cost and improve composite processability (Klyosov 2007). However, very little published data, especially on LCTE values, has been found directly dealing with the effect of talc-filled plastic shells on overall properties of co-extruded WPCs. The objective of the study described in this paper was to investigate the effect of talc content on morphology, mechanical performance, and thermal expansion of the talc-filled HDPE and co-extruded WPC with talc-filled HDPE shells.

## EXPERIMENTAL

### Raw Material

Raw materials for the study included talc, wood flour, high density polyethylene (HDPE), maleic anhydride grafted polyethylene (MAPE), and lubricant. Talc was from Fiber Glast Development Corp. (Brookville, OH, USA) and had an oil absorption rate of 30 g/100 g talc, a density of  $2.8\text{g/cm}^3$ , and a particle size with 99.7% material passing through a 325-mesh screen. Twenty mesh pine WF was from American Wood Fibers Inc. (Schofield, WI, USA). HDPE was grade AD60 with an MFI at  $190\text{ }^\circ\text{C}$  and 2.16 kg weight of 0.7g/10 min, and a density of  $0.96\text{g/cm}^3$  (ExxonMobil Chemical Co., Houston, TX,

USA). MAPE was grade Epolene™ G2608 with an MFI at 190 °C and 2.16 kg weight of 6~10 g/10 min,  $M_w$  of 65,000 g/mol, and an acid number of 8 mgKOH/g from Eastman Chemical Co. (Kingsport, TN, USA). Lubricant was grade TPW 306 from Struktol Co. (Stow, OH, USA).

## Experimental Design

The test design included two separate sets of experiments. The objective of the first set was to investigate the effect of talc loading level on talc-filled HDPE composites (*i.e.*, shell layers for co-extruded WPC) through injection molding, consisting of six blends with talc loading levels of 0, 10, 20, 30, 40, and 50 wt% of the total composite weight. The second set was designed to study properties of co-extruded WPC with talc-filled shells at 0, 5, 10, 15, 25, 35, and 50 wt% of talc loading level in the shell layers and a fixed WPC core. The shell formulation for co-extruded WPCs also included the use of 2 wt% colorant based on the total shell material weight.

## Composite Manufacturing

### (1) Talc filled HDPE

Melt compounding was performed using a Leistritz Micro-27 co-rotating parallel twin-screw extruder (Leistritz Corporation, Allendale, NJ, USA) with a screw speed of 60 rpm. The temperature profile of barrels ranged from 150 to 175 °C. The extrudates were air-cooled and then pelletized into granules. The granules were injection-molded into standard mechanical test specimens using a Batenfeld Plus 35 injection molding machine (Batenfeld, NJ). The injection temperatures were 190 and 180 °C for HDPE-talc composites and neat HDPE, respectively. All specimens were then conditioned for 72 h at a temperature of 23±2 °C and a relative humidity of 50±5% for later characterization.

### (2) Co-extruded WPC

The core material (HDPE: WF: Lubricant: MAPE: talc=33:55:5:2:5 wt%) was a HDPE-based blend with 20-mesh pine flour, which was compounded prior to the co-extrusion runs using the Leistritz Micro-27 extruder. The co-extruded composites were manufactured with a pilot-scale co-extruded system (Yao and Wu 2010). This system consists of the Leistritz Micro-27 twin-screw extruder for the core and a Brabender 32 mm conical twin-screw extruder (Brabender Instruments Inc., South Hackensack, NJ, USA) for the shell. A specially-designed die to produce a profile with a target cross-section area of 13 x 50 mm was used. A vacuum sizer was used to maintain the targeted size. The co-extruded profiles passed through a 2 m water bath with water spraying using a downstream puller. Manufacturing temperatures for core were controlled at 155 (feeder), 160, 165, 170, 170, 170, 160, 150, 140, 130, and 155 °C (die). Manufacturing temperatures for shells varied from 150 to 165 °C in the variation of different shell formulations.

## Characterization

### (1) Talc-filled HDPE

Flexural testing was done with specimens of 80x13x3-mm in a three-point bending mode using an Instron model 5582 testing machine (Instron Inc., Norwood, MA, USA) following the ASTM D790 standard. A crosshead speed of 5 mm/min and a span length of

50 mm were used for all tests. Tensile testing was done with Type-I dumbbell-shape tensile specimens having a dimension of 165x13x3 mm using the same Instron machine according to the ASTM D638 standard. A crosshead speed of 5 mm/min and an extensometer with a gage length of 25 mm for strain measurement were used for the test. Notched Izod impact strength was determined from specimens of 63.5x12.7x3-mm in size using a Tinius Olsen Mode 1892 impact tester (Tinius Olsen, Horsham, PA, USA) according to the ASTM D256 standard. A notch angle of 45° and a “V”-type notch depth of about 2.5 mm were used for the test. Five specimens were taken for each test, and average data along with corresponding standard deviations were reported.

The storage modulus, loss modulus, and  $\tan \delta$  of neat HDPE resin and its composites were evaluated using a TA DMA Q800 (TA Instruments Inc., New Castle, DW, USA) with samples of 59.8 (length) x 12.7 (width) x 3.2 (thickness) mm. The specimens were loaded at room temperature and cooled to -40 °C. They were then heated from -40 to 120 °C with a heating rate of 5 °C/min. A dual cantilever mode was used to test all specimens at a fixed frequency of 1 Hz.

Thermal expansion samples were machined with a miniature table saw along the long direction of injection molded samples, having a final dimension of 12.7 (length) x 5.1 (width) x 1.6 (thickness)-mm. Special attention was paid to ensure that the sample ends were parallel. The test was done with a TA Q400 Thermomechanical Analyzer, TMA. Each sample was placed on a quartz base, and an extension quartz probe was then placed on the top end of the sample along the length direction. A loading of 5-g force was applied to the probe to ensure the proper contact of the probe and the sample. The change in the length of the sample with temperature was measured using a linear variable differential transducer (LVDT) with a sensitivity of  $\pm 0.02 \mu\text{m}$ . The length and temperature data were recorded and analyzed with TA's Universal Analysis software. All tests were done with a complete cooling and heating cycle: 60 °C to -30 °C and -30 °C to 60 °C. The heating and cooling rates were kept constant at 5 °C /min. The LCTE ( $\alpha_L$ , 1/ °C) is calculated as,

$$\alpha_L = (1/L) dL/dT \quad (1)$$

where  $L$  is the linear dimension of the test sample and  $dL/dT$  is the rate of change in the linear dimension per unit temperature.

The morphologies of selected composite samples were analyzed with a Hitachi S-3600N VP Scanning Electron Microscope (SEM) (Hitachi Ltd., Tokyo, Japan). The fractured surfaces of selected test samples were coated with Pt to improve the surface conductivity before observation and observed at an acceleration voltage of 15 kV.

## (2) Co-extruded WPC

The specimens having a typical dimension of 390 (length) x 50.4 (width) x 12.5 (thickness) mm were measured under a four-point bending mode using the Instron model 5582 testing machine (Instron, Norwood, MA, USA) following the ASTM 6272 standard. A crosshead speed of 8.7 mm/min and a span length of 250 mm were used for the test. Izod impact strength was measured using the Tinius Olsen Model 1892 impact tester. Samples with a 3-mm thickness along the extrusion direction were obtained by cross-cutting the extruded profiles. Ten replicate samples from each group were tested without

notching in order to protect the integrity of the shell. The impact force on the test samples was perpendicular to the extrusion direction.

The LCTE value of each co-extruded specimen was measured parallel to the extrusion direction over a temperature range from 25 to 60 °C and 60 to -13 °C. The specimens had a long dimension of  $76\pm 9$  mm along the extrusion direction. They were conditioned at 60 °C in an oven and -13 °C in a freezer from their initial equilibrium temperature of 25 °C prior to size measurements with a Mitutoyo digimatic indicator of  $\pm 0.01$  mm accuracy (Mitutoyo Co., Kanagawa, Japan). Five specimens were used for each group. The LCTE for each sample was calculated based on size changes before and after conditioning.

### Data Analysis

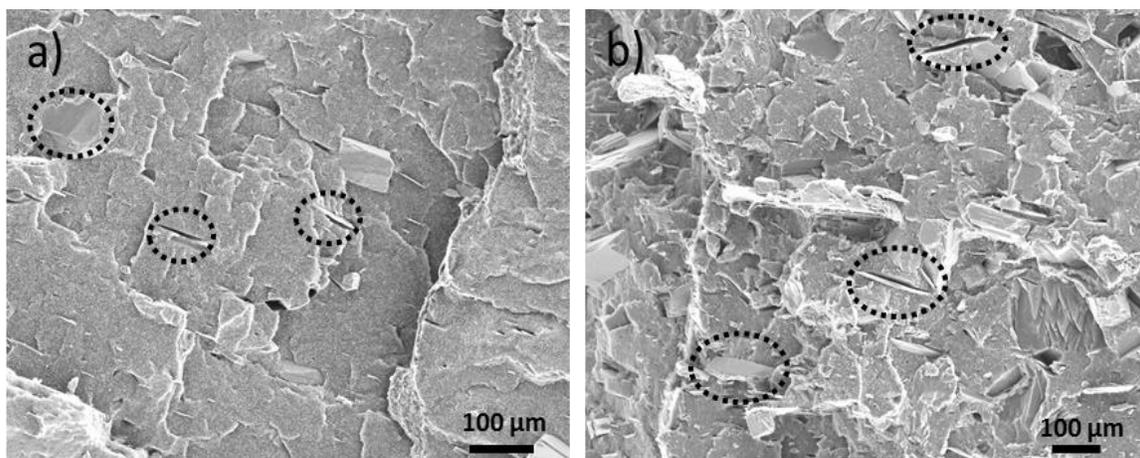
Duncan's multiple range tests for pairwise comparison were used to test the effect of various treatments using Statistical Analysis Software SPSS 10.0. Statistical ranking at the 5% significance level was provided among the treatments for each property.

## RESULTS AND DISCUSSION

### Properties of Talc-filled HDPE

#### (1) Morphology

Typical SEM micrographs for talc-filled HDPE composites are shown in Fig. 1 (a: 10 wt% and b: 30 wt% talc). Plate-type talc particles (Fig. 1) can be clearly seen from the fractured surfaces with both in-plane and out-of-plane orientations. Some line-type voids showed talc plate particles being pulled-out during the fracturing process. The talc particles appeared to be loosely mixed in the plastic matrix with little bonding to the plastic matrix since the talc particles were not surface modified. There were more aggregations of the talc particles in composites at the 30 wt% loading level, as indicated by the rougher surface (Fig. 1b) compared with composites with the 10 wt% talc loading (smoother fracture surface shown in Fig. 1a).



**Fig. 1.** SEM micrographs of talc-filled HDPE composites; (a) 10 wt% talc and (b) 30 wt% talc

## (2) Tensile properties

Tensile properties of neat HDPE and its composites with different talc loading levels are summarized in Table 1 and plotted in Fig. 2. Tensile modulus increased with the increase of the filler loading levels. For the neat HDPE, the tensile modulus was 0.86 GPa. The modulus increased to 5.86 GPa at the 50% talc loading level. The enhanced composite modulus from filler systems was due to dispersed stiff talc filler in the plastic matrix, as shown in the SEM micrographs (Fig. 1). Figure 2 shows significantly different behaviors of tensile strength between HDPE and talc-filled HDPE composites. There was a slightly decreasing trend of tensile strength for talc-filled composite. The strength decreased to 21.78 MPa from the neat HDPE strength of 23.8 MPa when it was loaded with 50 wt% talc. The decreasing trend of talc-filled composites was probably due to the stress concentration formed around the talc particles in the stressed composites, leading to the debonding between the filler and the matrix at the interface. It is expected that surface modification of talc particle using chemical agents such as silane can significantly enhance talc-matrix bonding and composite strength (Lee *et al.* 2008).

**Table 1.** Summary of Mechanical Properties of Neat HDPE and Talc-Filled HDPE Composites

Talc content (wt%) <sup>a</sup>	Strength			Modulus	
	Tensile (MPa) <sup>bc</sup>	Flexural (MPa)	Impact (kJ/m <sup>2</sup> )	Tensile (GPa)	Flexural (GPa)
0	23.80(1.40)D	21.8(1.0)A	28.57(2.0)C	0.86(0.09)A	0.85(0.06)A
10	23.10(0.42)BC	24.9(1.3)B	7.97(0.58)C	1.92(0.16)B	1.20(0.10) B
20	22.78(0.13)B	25.3(0.5)B	6.03(0.13)B	2.09(0.21)C	1.50(0.01)BC
30	22.54(0.57)AB	27.3(0.5)C	5.54(0.46)B	2.60(0.39)D	1.80(0.10)D
40	22.22(0.56)AB	29.0(0.8)D	4.60(0.39)A	3.24(0.17)E	2.35(0.08)E
50	21.78(0.73)A	32.8(0.2)E	3.87(0.10)A	5.86(0.34)E	3.20(0.08)F

<sup>a</sup> The filler content was based on the total composite weight.

<sup>b</sup> Mean values with the same capital letter for each property are not significantly different at the 5% significance level.

<sup>c</sup> Numbers in the parenthesis are standard deviation based on five specimens.

Several theoretical models can be used to predict tensile properties of fiber composites in terms of the properties of the constituent materials (Nielson and Landel 1993). The Halpi-Pagano micromechanical model (Tsai and Pagano 1968), which has been utilized to predict modulus of the composites with randomly oriented short fibers (Pilla *et al.* 2008; Johnson *et al.* 2008), is given as below,

$$E_C^{\text{ran}} = \frac{3}{8} E_L + \frac{5}{8} E_T \quad (2)$$

where  $E_C$  is Young's modulus of random fiber composites, and  $E_L$  and  $E_T$  are longitudinal and transverse Young's modulus, respectively, of corresponding uniaxially oriented discontinuous fiber composites. This equation is an averaging procedure for estimating elastic moduli of quasi-isotropic laminates.  $E_L$  and  $E_T$  can be estimated from the following Halpin-Tsai equations (Fornes and Paul 2003; Sperling 2006):

$$E_L = E_m \left[ \frac{1 + (2l/d)\eta_L\phi_f}{1 - \eta_L\phi_f} \right] \text{ and } E_T = E_m \left[ \frac{1 + 2\eta_T\phi_f}{1 - \eta_T\phi_f} \right] \quad (3)$$

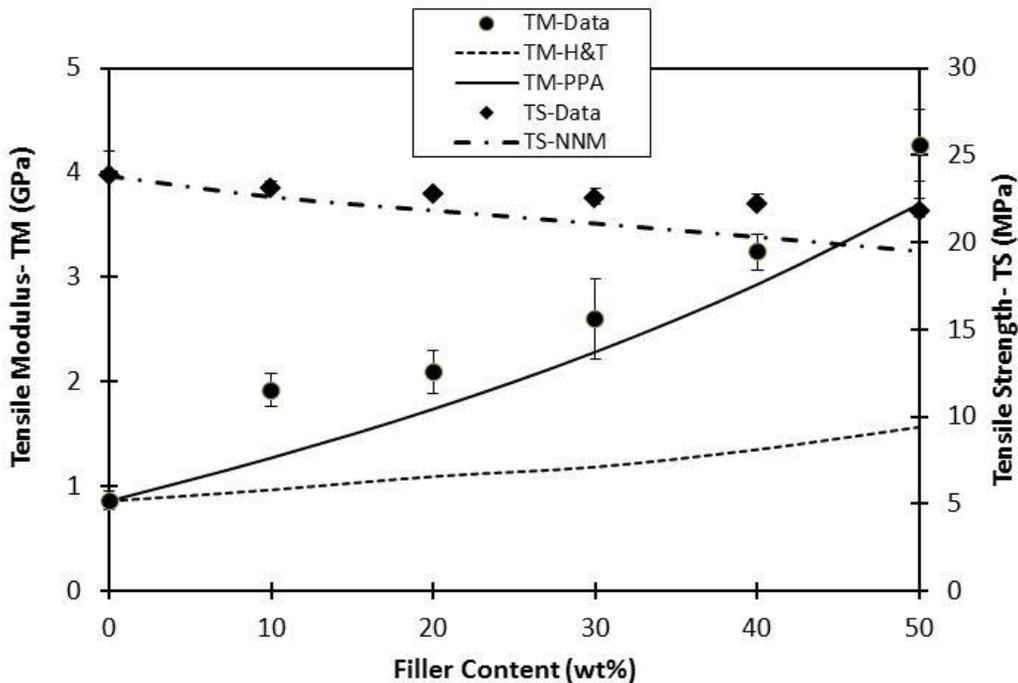
Among which the constant  $\eta_L$  and  $\eta_T$  are defined as,

$$\eta_L = \frac{(E_f/E_m) - 1}{(E_f/E_m) + (2l/d)} \text{ and } \eta_T = \frac{(E_f/E_m) - 1}{(E_f/E_m) + 2} \quad (4)$$

where  $E_f$  and  $E_m$  are Young's moduli of the fiber and polymeric matrix, respectively;  $\phi_f$  and  $l/d$  is volume fraction and aspect ratio of the fiber in the composites, respectively. The elastic modulus of short fiber composite can also be predicted based on paper physics approach - PPA (Cox 1952; Jayaraman and Kortschot 1996; Fu and Lauke 1998):

$$E_c^{PPA} = \chi_1\chi_2 E_f\phi_f + E_m(1 - \phi_f) \quad (5)$$

where  $\chi_1$  and  $\chi_2$  are, respectively, the fiber length and orientation factors for the composite elastic modulus. When the  $\chi_1 * \chi_2 = 1.0$ , Equation 5 becomes the rule of mixture law.



**Fig. 2.** Tensile properties of talc-filled HDPE composites. Line showing predicted values with various models. TM- tensile modulus, TS-tensile strength, H&T-Halpin and Tsai model, PPA-paper physics approach, and NNM-Nicolais and Nicodemo model

To assess predictability of the tensile modulus of talc-reinforced HDPE blends, experimental results were compared with calculated data from the above models (*i.e.*, Equations 2 and 5). The properties of the constituent materials of the composite used in the model prediction are  $E_f=12$  GPa,  $E_m=0.86$  GPa, density = 2.8 g/cm<sup>3</sup>, and  $l/d = 5$  for talc (Ma *et al.* 2010; Stixrude 2002).

Figure 2 shows comparisons of predicted results with the experimental data obtained from tensile tests as a function of filler loading levels. As shown, the theoretical curves predicted by Equation 2 are far below the experimental data points for HDPE-talc systems. The behavior could be attributed to several reasons. First, the model assumes perfect filler dispersion, while the presence of filler agglomeration may lead to an underestimate of filler volume. Second, particle orientation may strongly affect the calculated Young's modulus, while the model assumes a random fiber orientation. SEM micrographs (Fig. 1) show significant alignment of the filler. Equation 5 based on paper physics approach, PPA, was also used to calculate the tensile modulus. The calculated values based on a value of the lumped parameter,  $\chi_1*\chi_2$ , of 1.0 for talc (*i.e.*, rule of mixture law) are also plotted in Fig. 2. The predicted values were in a fairly good agreement with the experimental data.

To predict tensile strength, Nicolais and Nicodemo (1974) proposed a model that gives an equation of tensile stress as a function of filler fraction assuming no adhesion between filler and matrix,

$$\sigma_c = \sigma_m (1 - a \phi_f^b) \quad (6)$$

where  $\sigma_c$  and  $\sigma_m$  are composite and matrix tensile strength, respectively, and  $a$  and  $b$  are material constants. The “ $a$ ” value is assumed to be a function of aspect ratio ( $l/d$ ) of the filler, which decreases with increasing aspect ratio. For talc-filled composites, the tensile strength data fit very well with the proposed model assuming the values of  $a = 0.45$  and  $b = 2/3$  (Fig. 2).

### (3) Flexural properties

Flexural properties of the talc-filled composites are shown in Table 1. Flexural modulus for the composites exhibited an increasing trend with increased filler content. The filler had an important influence on flexural properties. Compared with neat HDPE, the talc improved flexural properties up to 50% and 276% in strength and modulus, respectively. The neat HDPE had a flexural modulus of 0.85 GPa. The modulus increased to 3.2 GPa for the talc-HDPE composite at the 50% talc level. Flexural strength for talc-filled composites shows an increase from 24.9 MPa (neat HDPE resin) to 32.8 MPa at the 50% talc loading level. Talc particle alignment as shown in the SEM micrographs (Fig. 1) might play an important role in determining the composite flexural strength.

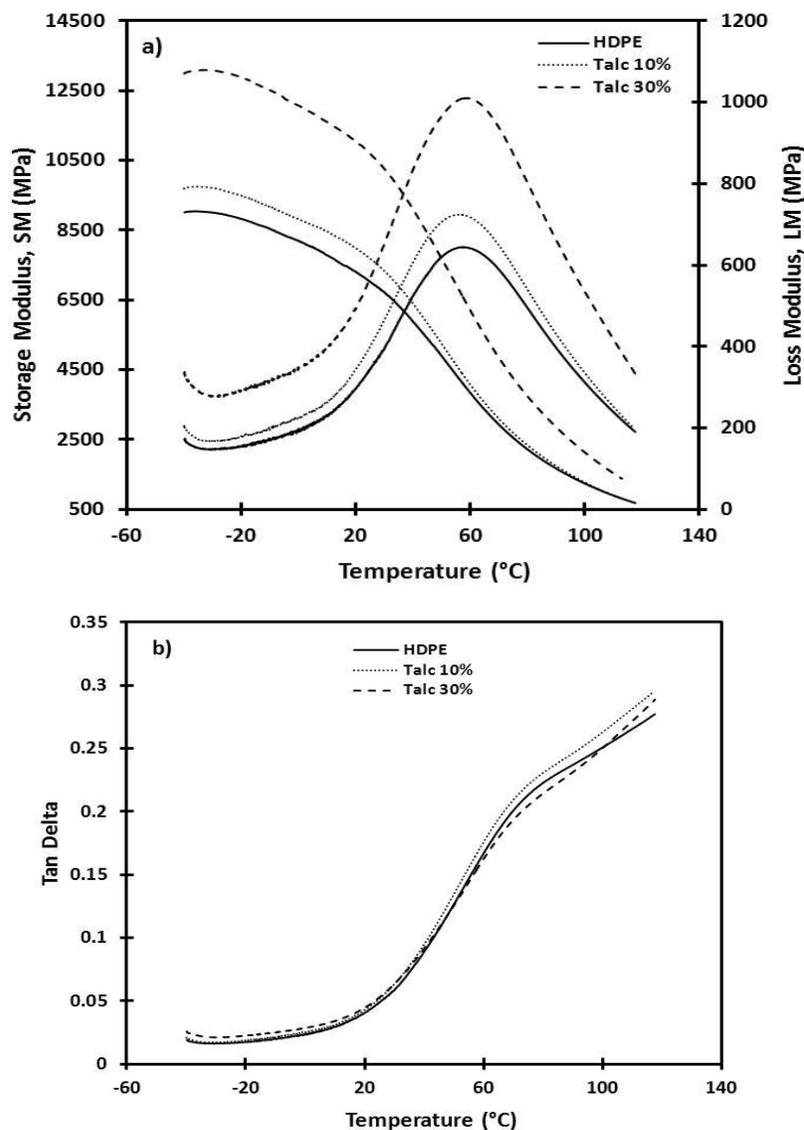
### (4) Impact strength

Table 1 lists the notched Izod impact strength data of HDPE-talc composites at various talc loading levels. The neat HDPE resin had an impact strength of 28.57 kJ/m<sup>2</sup>. The strength decreased significantly when the talc was added to the system. At the 10% talc loading level, the impact strength for HDPE-talc composites decreased to 6.03 kJ/m<sup>2</sup>

(Table 1). The impact strength further decreased as talc content increased. The reduced impact strength at higher loading levels of talc could be due to some degree of clustering of talc particles in the test specimens, which might lead to localized failure under impact force.

##### (5) Dynamic mechanical properties

Storage Modulus ( $E'$ ) – The effect of temperature on the storage modulus of HDPE-talc composites having 0, 10, and 30 wt% talc loading levels is shown in Fig. 3a. A general trend of increase of the storage modulus with increased filler content in the composites was observed.  $E'$  is more associated with the molecular elastic response of the composites, indicating the stiffness of the material. The increase of  $E'$  with increased filler



**Fig. 3.** Effects of temperature level on storage modulus and loss modulus (a) and damping factor (b) of talc filled HDPE composites

content was due to mechanical limitation posed by increasing filler concentration embedded in the viscoelastic matrix. The value of  $E'$  decreased with increasing temperature and converged to a narrow range at higher temperatures. The reduction of  $E'$  with increasing temperature was due to the softening of the matrix and initiation of the relaxation process (Huda *et al.* 2005).

**Loss Modulus ( $E''$ )** – The loss modulus is a measure of the absorbed energy due to the relaxation and is associated with viscous response or the damping effect of the material. Figure 3a shows the effect of varying filler contents and temperature on loss modulus of the composites.  $E''$  increased with the increased filler concentration and had a peak in the transition region around 50 °C. This relaxation peak is known as an indication of  $\alpha$ -relaxation of HDPE (Matuana *et al.* 1998), which was related to a complex multi-relaxation process associated with the molecular motion of the PE crystalline region (Danyadi *et al.* 2007; Mohanty and Nayak 2006). The  $E''$  at this relaxation temperature was markedly increased with the increase of talc loading level. The presence of fillers in the plastic resin reduced the flexibility of the material by introducing constraints on the segmental mobility of the polymer molecules. The  $\alpha$ -relaxation peaks of talc-filled composites were almost unchanged.

**Tan  $\delta$**  – The damping factor,  $\tan \delta$ , is a ratio of the loss modulus to the storage modulus. The parameter is independent of the material's stiffness and is widely used to study viscoelastic response of the materials. Figure 3b shows  $\tan \delta$  curves of neat HDPE and talc-filled HDPE composites. For the composite systems,  $\tan \delta$  curves had less distinctive  $\alpha$ -relaxation process compared with the  $E''$  data. The damping curves for talc-filled composites at 10 and 30 wt% talc levels almost overlapped the curve of neat HDPE. This indicates that the damping effect was reduced with the increased filler content in the matrix. This result could be interpreted for the reinforced composites that had the poor interfacial bonding between filler and HDPE matrix and tended to dissipate more viscous energy. Unmodified talc composites showed less or no interfacial bonding, leading to little change in the damping effect.

#### (6) Thermal Expansion Properties

Typical dimensional change data in relation with temperature for the composite system are shown in Fig. 4 for the neat HDPE resin and talc filled systems. The sample dimension increased as the temperature increased, and decreased as temperature decreased. The LCTE value is represented by the slope of the linear portion of the curve. Neat plastic samples had a noticeably larger dimensional change than that from filled composites for a given temperature change. Residual deformation is seen for the neat HDPE at the end of the heating cycle. The residual deformations for talc-composite systems were all similar to that of the neat plastic.

The measured LCTE values are plotted in Fig. 5 as a function of talc content levels. The cooling and heating cycles led to similar LCTE values at each given filler content level. For talc filled composites, talc content increase up to the 30 wt% level did not cause large LCTE value reduction. Thus, the use of unmodified talc at these loading levels was not effective in controlling thermal expansion behavior of the filled plastics. The LCTE reduction rate increased greatly with the use of talc after the 30 wt% filling level. At the 50 wt% talc level, the LCTE values were, respectively,  $85.4 \times 10^{-6} / ^\circ\text{C}$  and

$79.7 \times 10^{-6} / ^\circ\text{C}$  from the cooling and heating cycles. These values are still higher than the reported LCTE values of well-made commercial WPC filled with wood flour and other fillers. Surface modification of the talc particles for enhancing their bonding to the matrix may help improve the thermal expansion behaviors of filled composites (e.g., Singha *et al.* 2010; Huang *et al.* 2012).

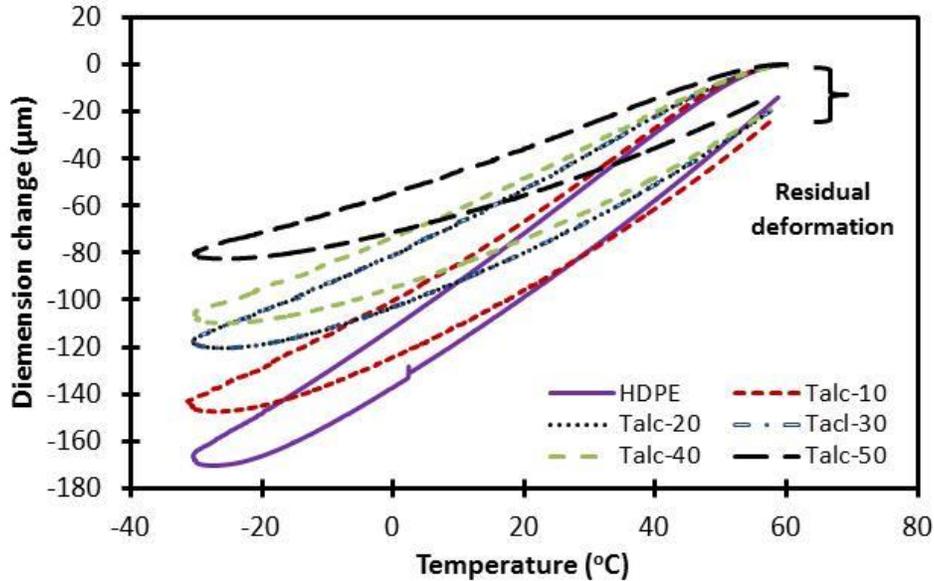


Fig. 4. Typical dimension change-temperature history for talc filled HDPE composite systems

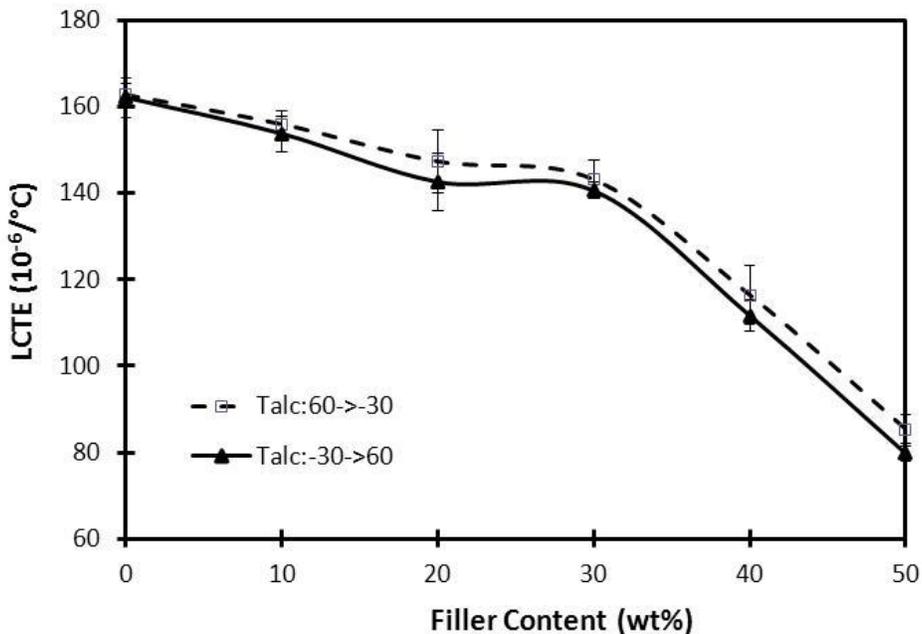
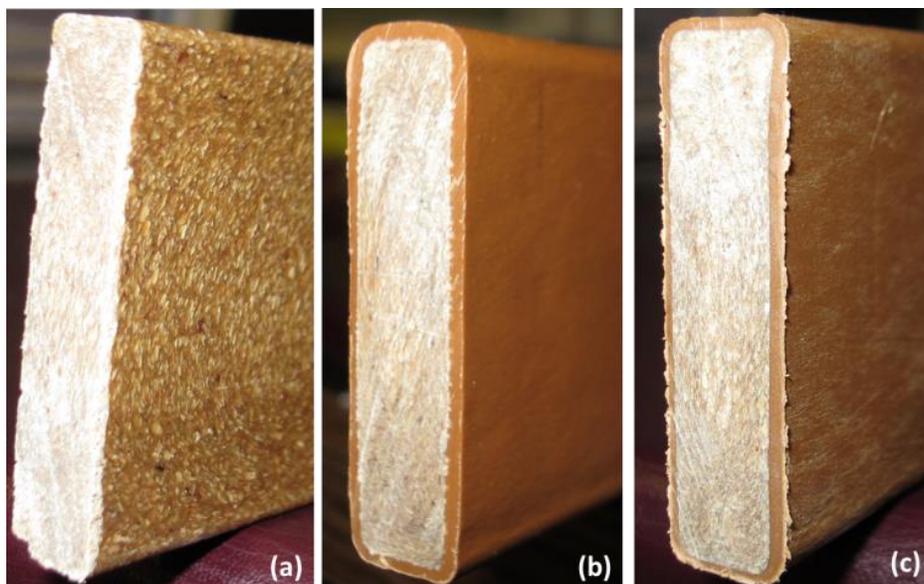


Fig. 5. Measured LCTE values as a function of filler content level for talc filled composites

## Co-extruded WPC with Talc Filled HDPE Shells

### (1) Composite structure

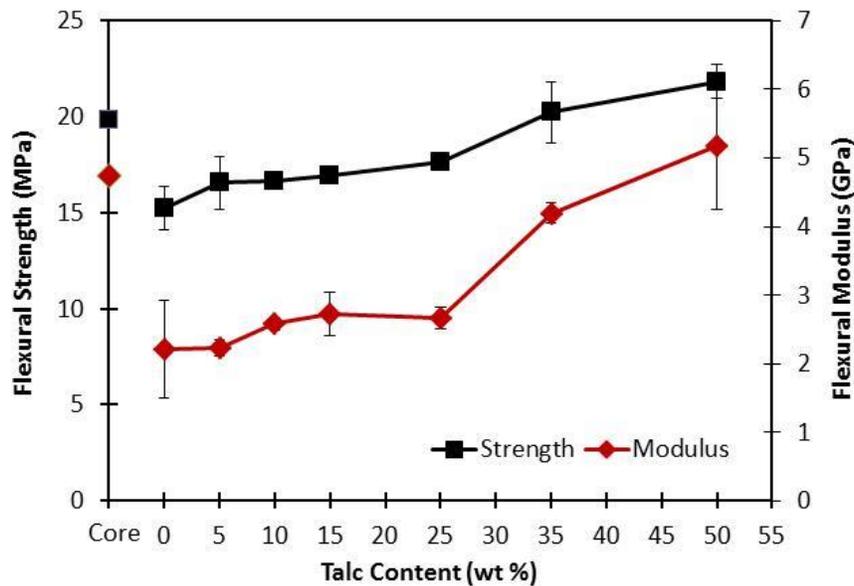
Figure 6 shows photographs of extruded core material (a) and co-extruded core-shell WPC with 10 wt% (b) and 50 wt% of talc in the shell layer (c). The shell thickness averaged at 1.2 mm, which represents a fairly thick shell over a 10-mm thick WPC core material. At higher talc loading levels (*e.g.*, 50 wt%) and a constant colorant loading rate (*i.e.*, 2 wt%), composite surfaces became lighter (Fig. 6b vs. Fig. 6c).



**Fig. 6.** Extruded core material (a), and co-extruded core-shell WPCs with 10 wt% (b) and 50 wt% talc in shell (c)

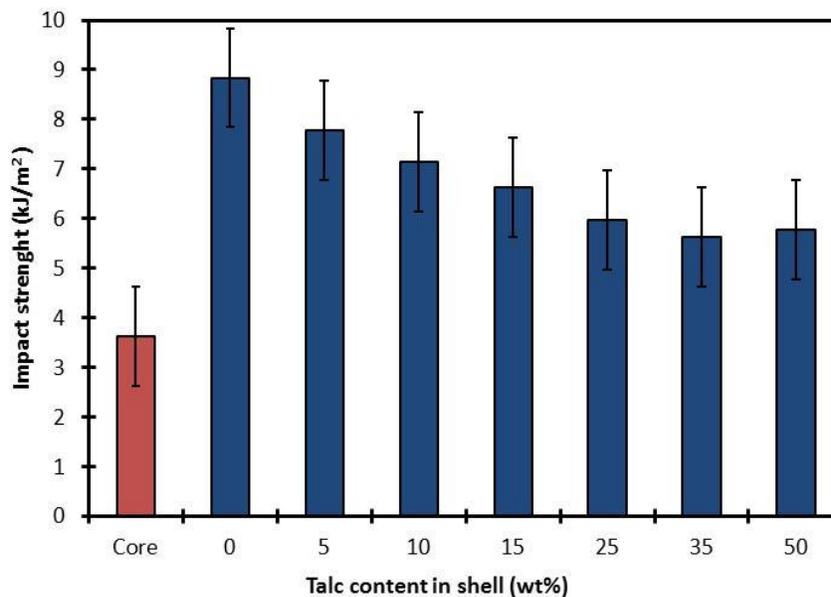
### (2) Flexural and impact properties

The effect of seven different talc loadings (*i.e.*, 0, 5, 10, 15, 25, 35, and 50 wt%) in the shell layer on the four-point flexural properties of co-extruded composites is plotted in Fig. 7 in comparison with the corresponding core-only material properties. The extruded core-only materials had a flexural modulus of 4.74 GPa. The modulus decreased to 2.21 GPa for co-extruded composites with a pure HDPE shell (*i.e.*, 0 wt% talc loading). Thus, coating a relatively thick and less-stiff HDPE shell over a stiff-core material reduced overall composite modulus (Yao and Wu 2010; Kim *et al.* 2102). The modulus for the co-extruded composite system increased with increase of the talc loading levels in the shell. When the talc content in the shell reached the 35% and 50% levels, the flexural modulus increased to 4.17 and 5.19 GPa, respectively. The flexural strength decreased to 15.23 MPa from the core-only strength of 19.81 MPa when composite had a core-shell structure with a pure HDPE shell. With increased talc loading level in the shell layer, the composite strength gradually increased and reached 21.84 MPa at the 50% talc loading level. The strengthened shells at higher talc loading levels (Table 1) helped to enhance overall composite strength properties.



**Fig. 7.** Flexural strength and modulus from four point bending tests of co-extruded WPC with talc filled shells in comparison with the core-only properties

Measured impact strengths of the extruded core and coextruded core-shell WPCs are shown in Fig. 8. The impact strength for co-extruded WPCs noticeably increased in comparison with the corresponding core value. Thus, the shell layer showed an obvious protection effect for the more brittle core layer. The shell layer contained more HDPE, which tended to be tougher and more flexible (Yao and Wu 2010). The shell layer absorbed more impact energy and prevented crack propagation during fracturing process.

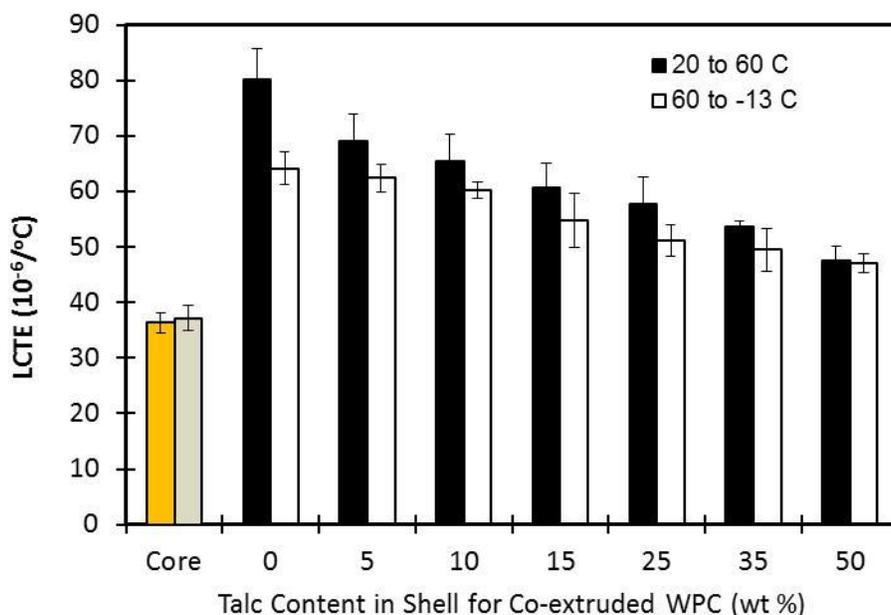


**Fig. 8.** Measured impact strength of co-extruded WPC with talc filled shells in comparison with the core-only property

As talc loading increased in the shell layers, which caused shells to be more brittle, the overall composite impact strength decreased. The trend followed impact strength behavior shown in Table 1 for talc-filled HDPE composites.

### (3) Thermal Expansion Properties

Figure 9 shows measured LCTE values from a heating cycle (20 to 60 °C) and a cooling cycle (60 to -13 °C) for the core-only control samples and for co-extruded WPCs in the variation of shell talc loading levels. The extruded core-only material had the LCTE values of  $36.3 \times 10^{-6}/^{\circ}\text{C}$  and  $37.2 \times 10^{-6}/^{\circ}\text{C}$  for 20 to 60 °C and 60 to -13 °C, respectively. The LCTE values increased to  $80.08 \times 10^{-6}/^{\circ}\text{C}$  and  $64.11 \times 10^{-6}/^{\circ}\text{C}$  for co-extruded composites with a pure HDPE shell (i.e., 0wt% talc loading) with heating and cooling cycles, respectively. Thus, coating a relatively thick HDPE shell with a large LCTE value over a stiff and thermally stable WPC core material increased the overall composite LCTE value. The LCTE values for the co-extruded composite system decreased with increase of the talc loading levels in the shell.



**Fig. 9.** LCTE values of co-extruded WPC with talc-filled shells in comparison of the core-only properties

When the talc content in the shell reached the 50 %wt level, the composite LCTE values decreased to  $47.59 \times 10^{-6}/^{\circ}\text{C}$  and  $47.09 \times 10^{-6}/^{\circ}\text{C}$ , respectively, for the two thermal cycles. These values are still noticeably higher than the corresponding core-only LCTE values. As shown in Fig. 5, the shell-only LCTE values at the 50% talc loading level were  $85.4 \times 10^{-6}/^{\circ}\text{C}$  from the cooling (60 to -30 °C) and  $79.7 \times 10^{-6}/^{\circ}\text{C}$  from the heating cycle (-30 to 60 °C). The larger shell LCTE values even at higher talc-loading levels (compared with the core-only data) led to overall composite LCTE increases. The negative effect of shells

on composite LCTE can be reduced by using thinner shells (Huang *et al.* 2012) and by using a more thermally stable shell material with surface modified talc particles.

It should be pointed out that a relatively stiff and thermally stable WPC core was used in manufacturing the co-extruded WPCs in this study. It is expected the talc-filled shells can help improve both strength and thermal expansion properties of co-extruded WPC when a relatively weak and less-thermally stable core is used (Kim 2012). The use of talc can also help lower the material cost and enhance composite processability (*i.e.*, increased shell stability during the extrusion process).

## CONCLUSIONS

1. The use of unmodified talc particles in high-density polyethylene (HDPE) helped enhance its tensile, bending, and dynamic moduli, but lowered its tensile and impact strengths.
2. The selected paper-physics model for composite modulus and Nicolais and Nicodemo model for tensile strength fit the data well after adjusting the model parameters for talc-filled HDPE composites.
3. Talc-filled HDPE had a reduced linear coefficient of thermal expansion (LCTE) values compared with that of neat HDPE. The LCTE reduction rate increased after the talc loading level increased to above the 30 wt%.
4. Coating a relatively thick, less-stiff HDPE shell with a large LCTE value over a stiff and thermally stable WPC core material decreased the overall composite modulus and increased the composite LCTE value. The composite modulus and strength increased and LCTE values decreased with increase of the talc loading levels in the shell.
5. The overall composite impact strength was greatly increased with the use of shell layers, whether or not the shells were filled with talc particles.

## ACKNOWLEDGMENTS

The authors would like to thank the Louisiana Board of Regents (Contract Number: LEQSF-EPS (2011)-OPT-IN-04), Korea Forest Research Institute, and Nanjing Forestry University, Nanjing, China for financial support.

## REFERENCES CITED

- Cox, H. L. (1952). "The elasticity and strength of paper and other fibrous materials," *Br. J. Appl. Phys.* 3, 72-79.
- Danyadi, L., Renner, K., Moczo, J., and Pukanszky, B. (2007). "Wood flour filled polypropylene composites: Interfacial adhesion and micromechanical deformations," *Polym. Eng. Sci.* 47(8), 1246-1255.

- Fornes, T. D., and Paul, D. R. (2003). "Modeling properties of nylon 6/clay nanocomposites using composite theories," *Polymer* 44(17), 4993-5013.
- Fu, S. Y., and Lauke, B. (1998). "The elastic modulus of misaligned short fiber reinforced polymers," *Composite Sci. Technol.* 58, 389-400.
- Huda, M. S., Drzal, L. T., Mohanty, A. K., and Misra, M. (2007). "The effect of silane treated- and untreated-talc on the mechanical and physico-mechanical properties of poly(lactic acid)/newspaper fibers/talc hybrid composites," *Composites: Part B*, 38, 367-379.
- Huda, M. S., Mohanty, A. K., Drzal, L. T., Misra, M., and Schut, E. (2005). "Green composites from recycled cellulose and poly(lactic acid): Physicomechanical and morphological properties evaluation," *J. Mater. Sci.* 40(16), 4221-4229.
- Huang, R., Xiong, W., Xu, X., and Wu, Q. (2012). "Thermal expansion behavior of co-extruded wood plastic composites with glass-fiber reinforced shells," *BioResources* 7(4), 5514-5526.
- Huang, R., Zhang, Y., Xu, X., Zhou, D., and Wu, Q. (2012). "Effect of hybrid mineral and bamboo fillers on thermal expansion behavior of bamboo fiber and recycled polypropylene-polyethylene composites," *BioResources* 7(4), 4563-4574.
- Jayaraman, K., and Kortschot, M. T. (1996). "Correction to the Fukuda-Kawata Young's modulus theory and the Fukuda-Chou strength theory for short fibre-reinforced composite materials," *J. Mater. Sci.* 31, 2059-2064.
- Jin, S., and Matuana, L. M. (2010). "Wood/plastic composites co-extruded with multi-walled carbon nanotube-filled rigid poly(vinyl chloride) cap layer," *Polym. Inter.* 59, 648-657.
- Johnson, R. K., Zink-Sharp, A., Rennekar, S. H., and Glasser, W. G. (2008). "Mechanical properties of wet-laid lyocell and hybrid fiber-reinforced composites with polypropylene," *Composite Part A*. 39(3), 470-477.
- Karrad, S., Lopez Cuesta, J. M. and Crespy, A. (1998). "Influence of a fine talc on the properties of composites with high density polyethylene and polyethylene/polystyrene blends," *J. Mater. Sci.* 33, 453-461.
- Kim, B. J., F. Yao, G. Han, Q. Wang, and Wu, Q. (2012). "Mechanical and physical properties of core-shell structured wood plastic composites: Effect of shells with hybrid mineral and wood fillers," *Composite Part B*. 45, 1040-1048.
- Kim, B. J. (2012). "The effect of inorganic fillers on the properties of wood plastic composites," *PhD Dissertation*, School of Renewable Natural Resources, Louisiana State University, 125 pp.
- Klyosov, A. A. (2007). *Wood-Plastic Composites*, John Wiley & Sons, Inc., Hoboken, New Jersey, 698 pp.
- Lee, S. Y., Kang, I. A., Doh, G. H., Yoon, H. G., Park, B. D., and Wu, Q. (2008). "Thermal and mechanical properties of wood flour/talc filled polylactic acid (PLA) composites: Effect of filler content and coupling treatment," *J. Thermoplastic. Composite Mater.* 21, 209-223.
- Ma, O., Tibbenham, P. C., Lai, X., Glogovsky, T., and Su, X. (2009). "Micromechanical modeling of the mechanical behavior of thermoplastic olefin," *Polymer Eng. Sci.* 50(3), 536-542.
- Matuana, L. M., Balatinecz, J. J., and Park, C. B. (1998). "Effect of the surface properties

- on the adhesion between PVC and wood veneer,” *Polymer Eng. Sci.* 38(5),765-773.
- Mohanty, S., and Nayak, S. K. (2006). “Interfacial, dynamic mechanical, and thermal fiber reinforced behavior of MAPE treated sisal fiber reinforced HDPE composites,” *J. Appl. Polym. Sci.* 102, 3306-3315.
- Nicolais, L., and Nicodemo, L. (1974). “The effect of particles shape on tensile properties of glassy thermoplastic composites,” *Inter. J. Polym. Mater.* 4, 229-243.
- Nielson, L.E, and Landel, R.F. (1993). *Mechanical Properties of Polymers and Composites*, Marcel Dekker, New York, NY.
- Noel, O., and Clark, R. (2005). “Recent advances in the use of talc in wood-plastic composites,” *The 8th International Conference on Wood Fiber-plastic Composites*, Madison, Wisconsin, USA.
- Pilla, S., Gong, S., O’Neil, E., Rowell, R. M., and Krzysik, A. M. (2008). “Polylactide-pine wood flour composites,” *Polym. Eng. Sci.* 48(3), 578-587.
- Singha, S., Mohanty, A. K., and Misra, M. (2010). “Hybrid bio-composite from talc, wood fiber and bioplastic: Fabrication and characterization,” *Composites Part A* 41(2), 304-312.
- Sperling, L. H. (2006). *Introduction to Physical Polymer Science*, John Wiley & Sons Inc., Hoboken, New Jersey, pp.702-705.
- Stark, N. M., and Matuana, L. M. (2007). “Coating WPCs using co-extruded to improve durability,” *Conference for Coating Wood and Wood Composites: Designing for Durability*, Seattle, WA. pp. 1-12.
- Stixrude, L. (2002). “Talc under tension and compression: Spinodal instability, elasticity, and structure,” *J. Geophysical Research* 107(B12), 23-27.
- Tsai, S. W., and Pagano, N. J. (1968). “Invariant properties of composite materials,” *Composite Materials Workshop*, S.W. Tsai, J. C. Halpin, and N. J. Pagano (eds). Technomic Publishing Co. Inc., Stamford, CT, USA, pp.233-253.
- Weon, J. I., and Sue, H. J. (2006). “Mechanical properties of talc and CaCO<sub>3</sub>-reinforced high-crystallinity polypropylene composites,” *J. Mater. Sci.* 41, 2291-2300.
- Yao, F., and Wu, Q. (2010). “Co-extruded polyethylene and wood-flour composite: Effect of shell thickness, wood loading, and core quality,” *J. Appl. Polym. Sci.* 118, 3594-3601.

Article submitted: February 2, 2013; Peer review completed: March 6, 2013; Revised version received and accepted: March 18, 2013; Published: March 19, 2013