Rheological and Mechanical Properties of Ultra-fine Cellulose-Filled Thermoplastic Epoxy Composites

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Thermoplastic epoxy resin (TPER)-based composites containing different amounts of ultra-fine cellulose (UFC) were prepared *via* melt compounding and injection molding. The effect of UFC loading on the mechanical properties and dynamic rheological behavior of the UFC-filled TPER composites was analyzed. The UFC-filled composites displayed higher complex viscosities than those of the neat TPER composites, especially at low frequencies. The elastic modulus of the 20 wt.% UFC-filled composite was up to 6- and 2-fold higher than that of TPER at 0.1 and 100 Hz, respectively. The loss factor decreased over the entire frequency range with the incorporation of UFC. The tensile modulus of elasticity (TMOE) of neat TPER was 3.13 GPa, and it increased as a function of UFC loading. The neat TPER exhibited the lowest flexural strength (108.1 MPa), and the flexural strength increased by 14% with the incorporation of 20 wt.% UFC. The results of the TMOE and the flexural modulus of elasticity (FMOE) were in agreement with rheological data on complex viscosity, elastic modulus, and viscous modulus. Ultra-fine cellulose-filled TPER composites may provide special capabilities for automotive applications and may also meet requirements for end-of-life vehicle (ELV) directives.

Keywords: Thermoplastic epoxy (TPER); Melt compounding; Rheology; Mechanical properties; Ultra-fine cellulose (UFC); Van Gurp-Palmen plot

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INTRODUCTION

To meet the progressively increasing efficiency requirements by year 2025, corporate average fuel economy (CAFE) regulations (54.5 miles/gallon), emissions standards (163 gCO_2/mile), and end-of-life vehicle directives require automakers to find innovative solutions. These technologies include weight reduction (lightweighting) and advanced engine and powertrain innovations. In addition, battery electric vehicles (BEVs) and plug-in hybrid electric vehicles (PHEVs) are encouraged (Bastani *et al.* 2012; ACC 2014). Plastic and polymer composites can provide light-weight solutions for body interior and exterior, power train, and lighting applications, and the automotive industry is looking for cost-effective and environmentally friendly ways to produce polymer composites for future vehicles (ACC 2014). Natural fibers, in particular cellulose micro- and nano-fibers, have received considerable attention in automotive applications because of their promising properties, such as non-abrasiveness for compounding or processing tools, renewability, biodegradability, low density, world-wide availability, and effectiveness for end-of-life vehicle (ELV) directives (Kalia *et al.* 2011). Thus, cellulose micro- and nano-fiber-reinforced composites may greatly accelerate the development of light-weight, sustainable, more fuel-efficient, and cost-effective vehicles.

Although recycling of plastic and polymer composites is receiving considerable attention in the automotive industry, especially in Europe, the amount of plastic recovered and recycled from end-of-life vehicles has not met the quota of the Green Vehicle Disposal statute (Miller *et al*. 2014; Verespej 2015). Unlike thermosetting polymers, thermoplastics offer a short cycle time (faster), more flexible manufacturing, higher toughness, and the potential for improved recyclability (Halliwell 2006; Petriccione 2011). Recycling issues surrounding thermosets contribute to the extensive use of thermoplastic and thermoplastic composites in the automotive industry. In addition, materials containing natural fiber-reinforced composites (NFCs) have significant advantages in terms of recycling because they do not create residues upon incineration (Halliwell 2006).

Thermoplastic epoxy resin (TPER), a new class of amorphous thermoplastic resin, is an excellent candidate for use in the matrix phase of composite materials for automotive applications. This is due to its unique chemistry (great compatibility with both synthetic and natural fibers), high mechanical properties, low processing temperature, and high forming and recycling efficiencies (Chmielewski and Kaffenberger 2008; Taniguchi *et al.* 2009; Petriccione *et al.* 2014). Chmielewski and Kaffenberger (2008) studied the mechanical and morphological properties of chopped glass and natural fiber-filled TPER composites and found that these new composite materials could be used in a variety of applications in the automotive industry, based on their properties and appearance. Petriccione *et al*. (2014) investigated the viscosity change in TPER composites using the incorporation of multi-walled carbon nanotubes (MWCNT), showing that MWCNT considerably reduced the viscosity of the TPER system. Taniguchi *et al*. (2009) studied the dynamic tensile property of carbon fiber-filled TPER composites. The results revealed that there was no linear relationship between the tensile strength and the strain rate of carbon fiber-filled composites. However, there are few publications addressing TPER-based composites and their characterization. The objective of this study was to investigate the effect of ultra-fine cellulose (UFC) on the mechanical and rheological properties of TPER-based composites. The development of UFC-filled thermoplastic composites is expected to provide light weight and high strength and modulus materials and also to bring additional functionalities such as renewability and effectiveness for ELV directives for automotive applications. Mechanical properties were determined using tensile, flexural, and Izod impact tests. The complex viscosity, storage modulus, loss modulus, loss factor (loss modulus/storage modulus), and phase angle of neat TPER- and TPER-based composites were also compared.

EXPERIMENTAL

Materials

Thermoplastic epoxy resin was supplied by L&L Products (Romeo, MI, USA) with a density of 1.20 g/cm^3 and a melt flow index of 5 g/10 min (190 °C/2.16 kg). Ultra-fine cellulose 100 was obtained from J. Rettenmaier & Söhne (Rosenberg, Germany), with a specific density of 1.4 g/cm^3 and an average particle size of 8 µm. The epoxy resin structure and properties influenced by the various chemical groups and the cellulose polymer chain structure are illustrated in Fig. 1.

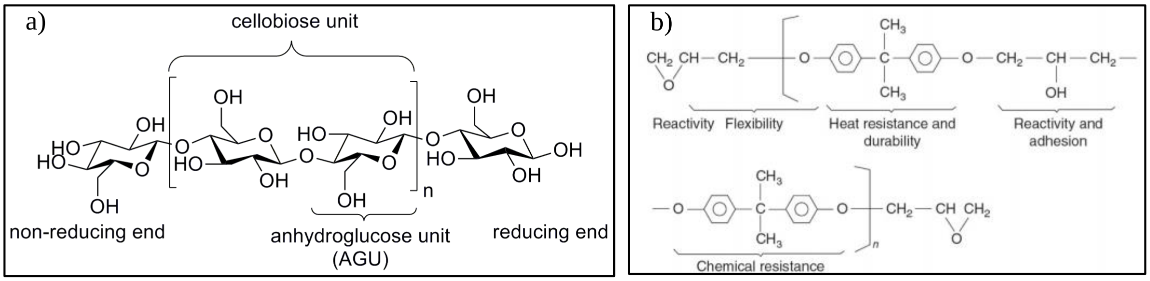

Fig. 1. The cellulose polymer chain structure (a) and typically epoxy resin structure (b) (Petriccione 2011; Olsson and Westman 2013)

Methods

The dried UFC and TPER were extruded at 60 rpm with the temperature of the heating zone from hopper to the die set to 180, 185, 190, 195, 200, and 200 °C, with a melt pressure at the die ranging from 15 to 40 bars, depending on the formulation through a twin screw extruder (TSE20/40D, C. W. Brabender Instruments Inc., South Hackensack, NJ, USA). Thermoplastic epoxy resin and cellulose were mixed using a Speed Mixer® (Flack Tek, Landrum, SC, USA) for 3 min at 1500 rpm to improve the dispersion of cellulose in the TPER matrix prior to melt compounding. For the fabrication of cellulose-filled TPER composites, TPER was mixed with cellulose, specified at 2.5, 5.0, 10, and 20 wt.% of the TPER matrix. Upon completion of melt mixing, the extrudate was cooled in an air-cooling system (2200 Series End Drive Conveyors, Dorner Manufacturing Corp., Hartland, WI, USA) and pelletized using a laboratory scale pelletizer (C. W. Brabender Instruments Inc., South Hackensack, NJ, USA). The samples obtained from the pelletizer were dried and injection-molded in a Mini-jector (Model 50, Amtek Company Inc., Arnold, MD, USA) prior to testing. Injection molding pressure, temperature, and cooling time were set to 2500 psi, 200 °C, and 10 sec, respectively.

The testing standards ASTM D 638-10 (2010), ASTM D 790-10 (2010), and ASTM D 256-10 (2010) were followed for the tensile, flexural, and Izod impact tests, respectively. The tensile and flexural behavior of neat TPER and cellulose-filled composites were measured (5966, Instron Corp., Norwood, MA, USA), with a 10 kN load cell and a 0.27 kN load cell, respectively. All the tension tests were conducted at a rate of 5 mm/min, and an extensometer was employed to determine elongation of the samples. Flexural tests were run at a test speed of 1.27 mm/min. The notches for Izod impact test were added using a NotchVIS machine manufactured by Ceast and the specimens were tested on a Resil 50 B impact test machine, manufactured by Ceast. At least six specimens were tested for each composition for tensile, flexural and impact properties and the results are presented as an average for tested samples. The details of mechanical testing were described in Kiziltas *et al*. (2014a) and Aydemir *et al*. (2015).

The rheological behavior of the neat TPER and cellulose-filled composites were analyzed using a dynamic oscillatory rheometer (Malvern Instruments, Worcestershire, UK) in the melt state, with a stress-controlled, Bohlin Gemini rheometer (Malvern Instruments, Worcestershire, UK) equipped with a 25-mm diameter parallel plate geometry. Composite samples were directly loaded and melted between plates, and the rheological tests were performed at 200 °C with a gap distance of 2.0 mm, under a nitrogen atmosphere, as detailed in Kiziltas *et al*. (2014b).

RESULTS AND DISCUSSION

The complex viscosities (η^*) for TPER and UFC-filled composites as a function of frequency (ω) at 200 °C are shown in Fig. 2a. The variation in η^*, with cellulose loading at 0.1, 1.0, 10, and 100 Hz, is presented in Fig. 2b. It was observed that η^* decreased with increasing ω for neat TPER and UFC-filled composites, indicating a non-Newtonian behavior and pseudoplastic characteristics over the entire ω range studied (Wang *et al*. 2008; Kim 2009; Barick and Tripathy 2011). This behavior can be explained as being due to the random orientation of the rigid molecular chains upon the application of shear force (Kim 2009). The UFC-filled composites exhibited higher η^* than the neat TPER, especially at low frequencies. This may be due to an interconnectivity or network-like structure created by the UFC in the TPER composites because of the strong interfacial interactions between the UFC-UFC or UFC-TPER matrix (Barick and Tripathy 2011). The UFC-filled composites also exhibited strong shear thinning behavior, and the enhanced effect of UFC-filled composites with increasing UFC content was more apparent at lower frequencies (less than 1 Hz) (Fig. 2a). For example, the η^* at 0.1 and 100 Hz for 20 wt.% UFC-filled composites were 3.9- and 1.7-fold higher than that of the neat TPER composites, respectively. Strong shear thinning behavior was also reported for other cellulose-filled polymer systems (Shumigin *et al*. 2011; Bitinis *et al*. 2013; Kiziltas *et al*. 2014b). The UFC-filled composites exhibited a positive relationship between the shear thinning behavior and mechanical properties, such that higher cellulose content led to an increase in the shear thinning behavior. Therefore, comparatively higher additions of UFC enhanced the reinforcement of the composites.

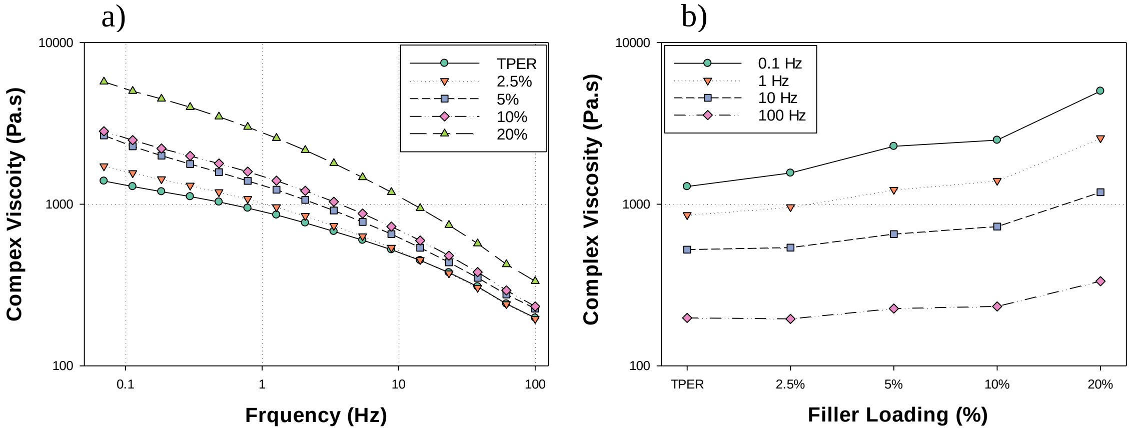

Fig. 2. a) Complex viscosity plotted against the ω at different UFC loadings, and b) the complex viscosity *vs*. UFC loadings at various frequencies for neat TPER and UFC-filled composites at 200 °C

The storage modulus (G') and loss modulus (G'') of the TPER and UFC-filled composites at 200 °C as a function of ω are shown in Fig. 3. It was apparent that cellulose exhibited a distinct effect on the G' and G'' of the TPER matrix, especially at the loading level of 2.5 wt.%. At lower frequencies, the values of G' and G'' of UFC-filled TPER composites dramatically increased with increasing UFC content when compared with the TPER matrix because of the intrinsic rigidity of UFC. Storage modulus data often serves as a measure of molecular rigidity; in this case, the addition of UFC increased the rigidity of composites, especially at high filler loadings. The G' of the 20 wt.% UFC-filled composite was up to 6- and 2-fold higher than that of the TPER composite at 0.1 and 100 Hz, respectively. The *G*” of the 20 wt.% UFC-filled composite was also up to 4- and 1.5-fold higher than that of the TPER composite at 0.1 and 100 Hz, respectively. This improvement in *G*’ and *G*” can be explained by the restriction of deformation with UFC in polymer melts (Shumigin *et al.* 2011). Even though a positive shift occurred in the *G*’ and *G*” values at all frequencies, and the low frequency levels (terminal) behavior of the materials remained unchanged as the addition of UFC increased, which can be attributed to a lack of a three-dimensional elastic structure in the material (Wang *et al.* 2008). The slopes of *G*’ and *G*” decreased minimally with increasing UFC content in the TPER matrix (Fig. 3a and b), and the decrease can be explained by the fact that UFC-UFC or UFC-TPER interactions can induce interconnected or network-like structures, leading to the pseudo-solid-like behavior (Wang *et al*. 2008; Kim 2009). Pseudo solid-like behavior was also reported for other cellulose-filled polymer systems (Shumigin *et al*. 2011; Kiziltas *et al*. 2014b; Kiziltas *et al*. 2016a; Kiziltas *et al*. 2016b).

The loss factor, tan *δ* (*G*”/*G*’), was plotted against the *ω* for neat TPER and UFC-filled composites at 200 °C (Fig. 3c). The loss factor decreased over the entire *ω* range through the incorporation of UFC, which mainly resulted from interactions between the UFC and TPER matrix that improved the elastic properties of the composites. This reduction was more noticeable at low *ω* (less than 0.5 Hz), representing solid-like structures; the loss factor difference between UFC-filled composites and neat TPER lessened at higher *ω* (liquid-like) may be because of the change in microstructure and formation of an elastic network-like structure (Wang *et al*. 2008; Barick and Tripathy 2011). The phase angle (*δ*) results *versus* the complex modulus (*G**) are shown in the Van Gurp-Palmen plot, which identifies the rheological percolation for the UFC-filled composites (Fig. 3d) (Nandi *et al*. 2013; Paleo *et al*. 2015; Piontech *et al*. 2015). Figure 3d shows that the polymer chains were entirely relaxed at approximately 75° for *δ* in the low *G** region, and a continuous reduction in *δ* occurred with increasing *G** for neat TPER and UFC-filled composites. The value of *δ* in the *G** low region for TPER was 88.1, which indicated the dominant response of viscous flow. The deviation from 75° in Fig. 3d shows an elastic response of UFC-filled composites, owing to the creation of a percolated structure in the melt sample (Paleo *et al*. 2015). The UFC-filled composites of 2.5, 5.0, and 10 wt.% were highly elastic, and the 20 wt.% UFC-filled composite was slightly elastic. In contrast, the neat TPER composites were predominantly viscous in the terminal region. Similar results were also reported for high viscosity (hv-PP)-expanded graphite (EG) composites, high density polyethylene (HDPE)-fumed silica composites, and polypropylene-carbon nanofiber (CNF-PS-XT grade) composites (Nandi *et al*. 2013; Paleo *et al*. 2015; Piontech *et al*. 2015).

The tensile properties of UFC-filled TPER composites at various UFC loadings are shown in Fig. 4. The incorporation of UFC improved the tensile properties of UFC-filled TPER composites because of the strong interfacial adhesion between TPER and UFC (high polarity of TPER and its chemical compatibility with UFC), and the greater efficiency in load transfer from TPER to UFC, thus improving the tensile properties upon higher UFC content (20 wt.%) (Chmielewski and Kaffenberger 2008).

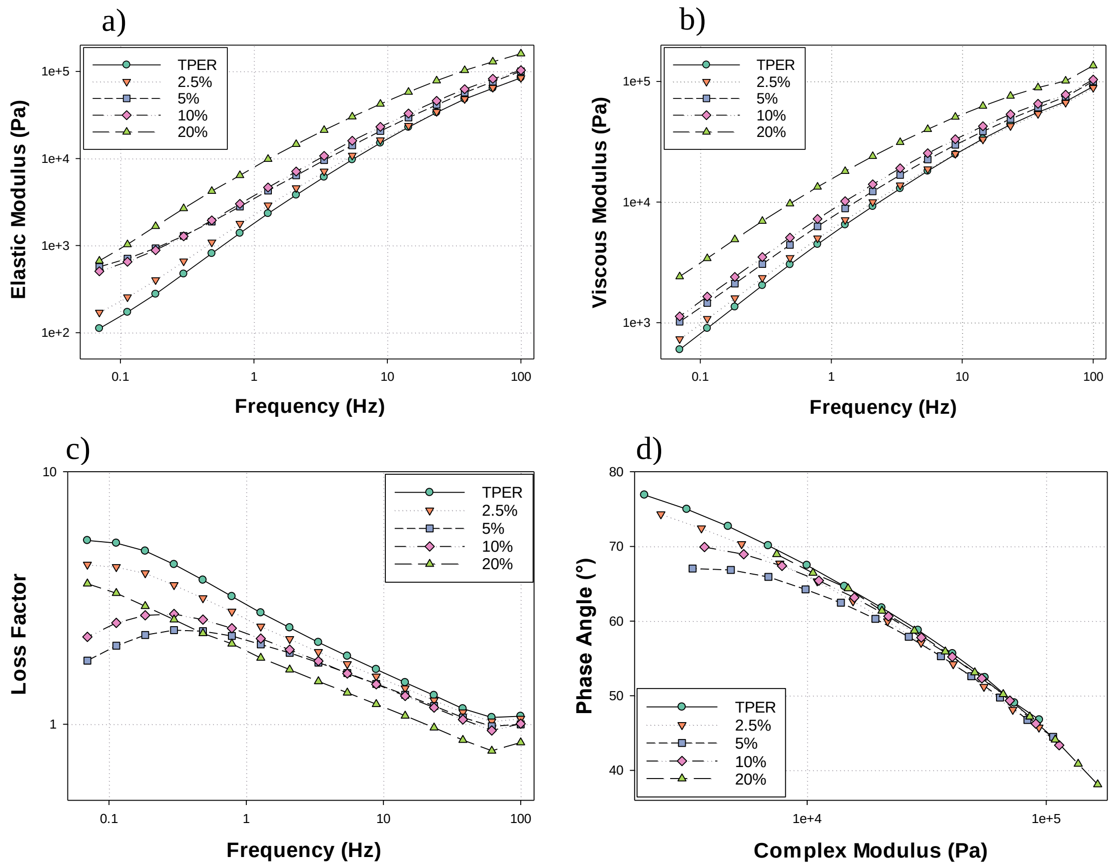

Fig. 3. Dynamic frequency sweep of a) G', b) G", c) the loss factor, tan δ, and d) Van Gurp-Palmen plot for the neat TPER and UFC-filled TPER composites, at a test strain of 1.0% and a temperature of 200 °C

The tensile strength of neat TPER was 62.1 MPa, and the cellulose-filled composite exhibited greater tensile strength, reaching 69.4 MPa at 20 wt.% UFC. In other words, the 20 wt.% UFC-filled composite had a tensile strength increase that was 12% greater than the neat TPER composite. The tensile modulus of elasticity (TMOE) of the neat TPER composite was 3.13 GPa and increased with UFC loading.

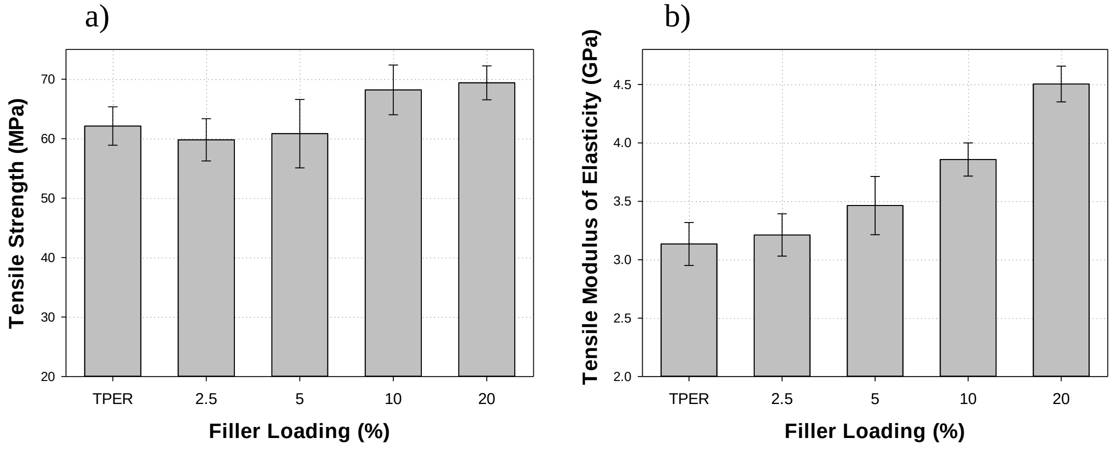

Fig. 4. Effect of filler loading on the a) tensile strength and b) TMOE of UFC-filled TPER composites

The TMOE reached 4.50 GPa with the incorporation of 20 wt.% UFC because of the restricted mobility and deformability of the TPER chain with increasing UFC content. As a result, TMOE increased by 44% with 20 wt.% UFC. An improvement in the tensile strength and TMOE were also reported for the MWCNT/TPER, short glass fiber/TPER, wood flour and micro-scale cellulose/TPER, and wood flour and micro-scale cellulose/TPER-HDPE blended composites (Chmielewski and Kaffenberger 2008).

The influence of UFC on the flexural properties of cellulose-filled TPER composites is shown in Fig. 5. Similar to tensile strength, the addition of a low filler loading (2.5 and 5.0 wt.%) showed no relative variation in the flexural strength of the composites, but an increased UFC loading level above 10 wt.% slightly improved the flexural strength compared with neat TPER. The neat TPER showed the lowest flexural strength (108.1 MPa), yet the flexural strength increased by 14% with the incorporation of 20 wt.% UFC. This result was attributed to better adhesion and stress transfer from the TPER matrix to the UFC. Filler content and uniformity of filler dispersion in the matrix greatly affects composite stiffness, and this effect can be observed as a linear increase in the composite modulus (Tawakkal *et al.* 2012). As with the tensile modulus, the flexural modulus of elasticity (FMOE) increased linearly with the incorporation of UFC. The FMOE of neat TPER was 2.99 GPa, which increased by 40% with the addition of 20 wt.% UFC (4.19 GPa). A similar observation was also reported by Chmielewski and Kaffenberger (2008) for TPER/MWCNT, TPER/wood flour, TPER/micro scale cellulose, TPER/HDPE blend/wood flour, TPER/HDPE blend/micro-scale cellulose, and TPER/short glass fiber composites. The interfacial interactions between UFC and TPER, and the uniform dispersion of UFC in the TPER matrix, could be the reasons for improved tensile and flexural properties in UFC-filled TPER composites (Kim 2009).

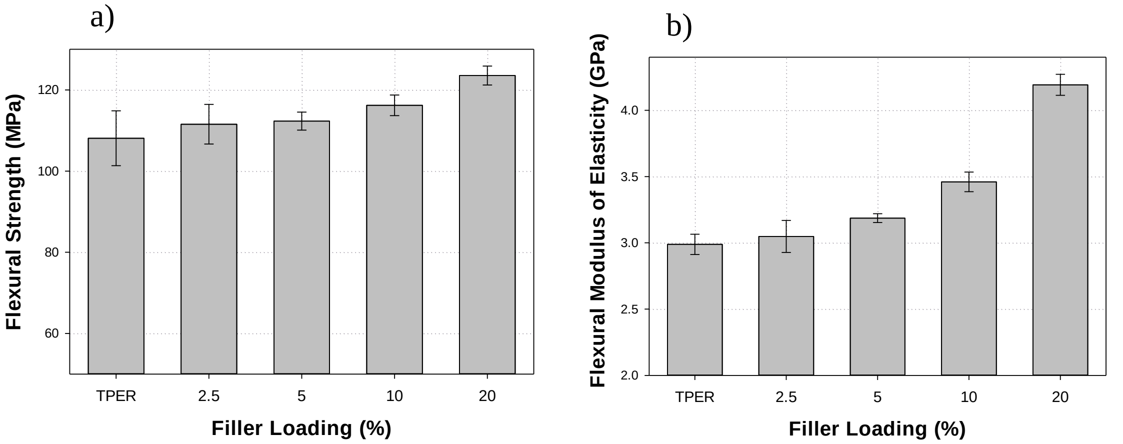

Fig. 5. Effect of filler loading on the a) flexural strength and b) flexural modulus of elasticity of UFC-filled TPER composites

To gain a better understanding of interactions and interface between TPER and UFC, and improvement on tensile and flexural properties in UFC-filled composites, techniques such as microscopic observation using scanning electron microscopy, FT-IR nano-indentation, dynamic mechanical thermal analysis, and microbond testing methods could be employed. The results of TMOE and FMOE are in accordance with rheological data on complex viscosity, elastic modulus, and viscous modulus (Aydemir *et al.* 2015). Higher filler loading resulted in higher TMOE, FMOE, complex viscosity, elastic modulus, and viscous modulus of the cellulose-filled TPER composites because of the restriction in the mobility of chain segments in the presence of cellulose (Shumigin *et al*. 2011).

In general, the filler loading level, the type of impact testing (notched or unnotched), and the surface functionalization of fillers can affect the impact strength of composites (Sanadi *et al*. 1997). The effect of various UFC contents on the notched Izod impact strength of UFC-filled TPER composites is presented in Fig. 6. The impact strength of neat TPER was 45.4 J/m, which decreased by 31% with the incorporation of 20 wt.% UFC. This result indicated that the addition of the UFC into the TPER matrix generated in stress concentration regions that cause higher crack propagation and crack initiation (Huda *et al*. 2006). Impact modifiers or coupling agents that can provide flexible and strong interfacial bonding in the composite may improve the impact strength of UFC-filled TPER composites (Sanadi *et al*. 1997).

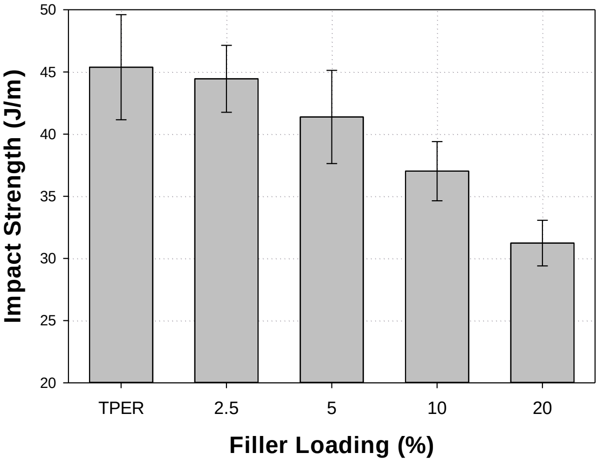

Fig. 6. Effect of filler loading on the impact strength of UFC-filled TPER composites

CONCLUSIONS

1. The η^* of % UFC-filled composites was higher than the neat TPER.
2. The values of G' and G'' of UFC-filled TPER composites notably increased with UFC content and ω compared to the TPER matrix, particularly at low frequencies.
3. The tan δ decreased over the entire ω range by the incorporation of UFC.
4. It is believed that an enhanced interfacial adhesion between UFC and TPER, and the uniform dispersion of UFC in the TPER matrix, might have contributed to substantial improvements in the tensile and flexural properties of the UFC-filled composites.
5. The results of the TMOE and the FMOE were in accordance with rheological data.
6. The Izod impact strength of neat TPER was decreased by the incorporation of UFC.
7. Higher mechanical properties, good surface appearance (smooth looking reflective surface with no distortion light highlights), and easy processability make these UFC-filled TPER composites a potential candidate for structural and semi-structural applications.

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