



Nanocellulose-Polypropylene Nanocomposites Enhanced With Coupling Agent

Ece Yakkan¹, Tuğçe Uysalman¹, Metehan Atagür¹, Kutlay Sever², M. Özgür Seydibeyoğlu^{1,*}

¹Department of Materials Science and Engineering, Izmir Katip Celebi University

²Department of Mechanical Engineering, Izmir Katip Celebi University

Abstract

As the growth of the nanocellulose is evident, this important material was used to reinforce polypropylene matrix with a coupling agent. Polypropylene (PP) - cellulose nanofibril (CNF) and Fusabond hybrid composites were prepared using twin screw extrusion technique. The commercial coupling agent was used to improve mechanical properties of PP/ CNF composites. The main challenges were to obtain well-dispersed CNFs in the polymer matrix and to establish compatible linkages between the CNFs and PP. The various loadings of coupling agent were examined to improve the interfacial adhesion between PP and CNFs. FTIR characterization of the composites were performed to confirm the interfacial adhesion of polypropylene and cellulose nanofibrils. The effect of coupling agent on the mechanical and morphological properties of polypropylene/ cellulose nanofibrils hybrid composites was studied by tensile testing, dynamic mechanical analysis, and SEM images, respectively. The best mechanical properties of the composite were 19.99 MPa (tensile strength), 1.067 GPa (Young's Modulus), which represented about 87% improvement, respectively, compared to that of pure polypropylene (14.45MPa, 0.570 GPa). Fracture morphology examination indicated good dispersion of CNFs in the PP matrix was achieved in the case of loading coupling agent (0.1 wt %). TGA results show that thermal stability of PP/CNF composites did not change much but slightly increased in the treated composites compare to that of the untreated PP/CNF composite.

Keywords: Polypropylene, Nanocellulose, Nanocomposites.

Uyumlaştırıcı Kimyasalla Güçlendirilmiş Nanoselüloz-Polipropilen Nanokompozitleri

Öz

Nanoselüloza olan talep giderek artmasıyla, bu önemli materyal uyumlaştırma kimyasallarıyla polipropilen matrikslerin güçlendirilmesinde kullanılmaktadır. Polipropilen (PP)-selüloz nanofibril (CNF) ve Fusabond Hibrit kompozitler çift vidalı ekstruder kullanılarak hazırlanmıştır. Ticari uyumlaştırıcı kimyasal PP/CNF kompozitlerinin mekanik özelliklerini iyileştirmek için kullanılmıştır. Buradaki esas zorluk, PP ve CNFs arasında uyumlu bağları oluşturmak ve polimer matriks içerisinde CNFs iyi bir dağılımını elde etmektir. Çeşitli oranlarda uyumlaştırma kimyasalı PP ve CNFs arasında yüzeyler arası bağlanmayı iyileştirmek için incelendi. Kompozitlerin FTIR karakterizasyonu polipropilen ve selüloz nanofibrilin yüzeyler arası yapışmasını belirlemek için gerçekleştirildi. Polipropilen/selüloz nanofibril kompozitlerinin mekanik ve morfolojik özellikleri üzerinde uyumlaştırıcı kimyasalın etkisi sırasıyla çekme testi, dinamik mekanik analiz ve SEM resimleriyle çalışıldı. En Kompozitlerin en iyi mekanik özellikleri, saf polipropilen (14.45 MPa, 0.570 GPa) ile karşılaştırıldığında 19.99 MPa (çekme direnci) ve yaklaşık %87 iyileşme gösteren 1.067 GPa (Young's modülü) idi. Kırılma morfoloji incelemesi PP matriks içerisinde uyumlaştırıcı kimyasal ilavesi (0.1 wt%) durumunda CNFs'nin iyi dağılımı sağlandı. TGA sonuçları PP/CNF kompozitlerinin termal kararlılığını değiştirmedini gösterdi, buna karşın muamelesiz PP/CNF Kompozitleriyle karşılaştırıldığında muamele edilmiş kompozitlerde hafif artış kaydedildi.

Anahtar Kelimeler: Polipropilen, Nanoselüloz, Nanokompozitler.

*Sorumlu Yazar (Corresponding Author):

M. Özgür Seydibeyoğlu (Dr.); Izmir Katip Celebi University, Department of Materials Science and Eng., 35620, Cigli, Izmir, Turkey, T: +90 232 3293535-3773, M: +90 532 5475217, E-mail: seydibey@gmail.com. ORCID No:

Geliş (Received) : 27.08.2018

Kabul (Accepted) : 11.09.2018

Basım (Published) : 15.12.2018

1. Introduction

With increasing environmental concerns, ecofriendly materials have gained significant importance due to the need to preserve our environment (Zini and Scandola, 2011; Asokan et al., 2012). Material selection plays a critical role not only for human life but also for engineering design of a successful sustainable product. Increasing demands for special materials led to the conception of composites, since valuable properties of different types of materials can be combined. Nowadays, engineering materials at the atomic and molecular levels are creating a revolution in the field of materials and processing (Jancar et al., 2010; Fermeglia et al., 2013). The discovery of new nano scaled materials such as nanoclays (Chan et al., 2011), carbon nanomaterials (Andrews and Weisenberger, 2004), and nanocellulose (Azeredo et al., 2010) significantly enhances mechanical and physical properties of polymers.

Polymeric materials and composites are attractive for a variety of applications due to many features including low weight, low cost, ease of processing and shaping, and corrosion resistance. Polymeric composite materials are consist of matrix phase and reinforcements such as commercial and traditional fibers (Ashori 2008). Reinforcing phase is loaded typically to improve chemical and/or physical properties of polymers. Inorganic fibers (glass and carbon fibers) and aromatic organic fibers (Aramid) use as traditional fibers and natural fibers use as ecofriendly types of fibers in the polymer matrices (Ashori, 2008; Xie et al., 2010).

Ecofriendly types of fibers such as kenaf, hemp, flax, jute, and sisal have a pioneering role in composite technology and large number of applications in automotive, constructions, marine, electronic, packaging, and aerospace (Zini and Scandola, 2011; Nair et al., 2014; Bledzki et al., 2006; Koronis et al., 2013; Holbery and Houston, 2006).

Among these natural fibers, cellulose fibers, especially nanocellulose, are attracting much attention. Cellulose is a polymer which can be found in the nature abundantly and can be obtained from many various plants and living organisms (Nakagaito et al., 2005, Bledzki and Gassan, 1999). Cellulose is light and environment friendly and also biodegradable. It has low cost, high specific strength and modulus (Malainine et al., 2005). In recent years scientists have succeeded in producing nano-size cellulose in different morphologies such as whiskers and fibrils, which can reinforce the polymer matrices enormously. Nakagaito et al. (2009), Suryanegara et al. (2009) and Iwatake et al. (2008) studied microfibrillated cellulose (MFC)-PLA nanocomposites. It was shown that MFC can reinforce PLA 40% (Iwatake et al., 2008) and MFC acts as nucleating agent and increases the crystallinity, accompanied with a tensile modulus increase of 42% (Iwatake et al., 2008). Seydibeyoğlu and Oksman (2008) successfully reinforced the polyurethane matrix with micron- and nano-sized cellulose. They showed the importance of the nano-size reinforcement by tensile testing and dynamic mechanical testing coupled with significant thermal stability. Nakagaito and Yano (2008) prepared micro- and nanocomposites of cellulose fibers and phenol formaldehyde. They modified the surface by NaOH treatment, which resulted in superior performance of the nano-size fibers and a further improvement in the toughness and percentage elongation.

All the advantages of cellulose and natural fibers in composite materials listed earlier, there are also some drawbacks. The major constraint for cellulose composites is their hydrophilicity. Most polymers are hydrophobic and this incompatibility creates adhesion problems at the interface of the composites, resulting in poor properties (Mohanty et al., 2002). However, there are many challenges to be solved for cellulose composites. There have been studies on overcoming the incompatibility of biocomposites using different coupling agents (Mulinari et al., 2009), such as silane (Bengtsson and Oksman, 2006), isocyanates (Raj et al., 1989), and titanate coupling agents (Spoljaric et al., 2009).

In this study, nanocellulose–polypropylene composites were manufactured via twin screw extrusion technique with and without coupling agent. Composite materials were characterized in terms of their mechanical, thermal and spectroscopic properties. In the last part of the study, the final composition was injection molded into an automotive part showing the importance of the study for industrial uses of this nanocellulose reinforced polypropylene nanocomposites.

2. Material ve Methods

Materials

Heterophasic polypropylene copolymer Moplen 2000 HEXP (medium high fluidity, $T_m=165^\circ\text{C}$, $\rho=0.9\text{ g/cm}^3$, Melt flow rate ($230^\circ\text{C}/2.16\text{ kg}$)= $16\text{g}/10\text{ min}$) was provided by Lyondell Basell Polymers. Cellulose nanofiber

(CNF) with a diameter of 0.02 microns was provided as slurries of approximately 4% solids by Process Development Center at The University of Maine. FUSE® E265 (FUSE) was purchased from DuPont Packaging & Industrial Polymers. It has a melt flow rate (190 °C/2.16kg): 12 g/10 min.

Processing

Lab Tech Twin-Screw Extruder (screw diameter: 20mm, L/D ratio of 32:1) was used to produce PP/CNF composites. First, nanocellulose was oven-dried at 35-37 °C for 24 hours. Second it was put into laboratory mixer for 8 minutes to minimize the water content. Then, PP/CNF composite containing 2 wt % nanocellulose was manufactured. In this process, the extrusion temperature profile was 190 °C, 200 °C, 210 °C, 210 °C, 210 °C, and 220 °C from the feeding zone to the die and the extruder rotation speed was 190 rpm. Furthermore, PP/CNF composite containing 2 wt % nanocellulose treated with FUSE. In this treatment three different percentage were studied (0.1wt%, 0.5wt%, 1wt %). The extruded composites were pelletized and dried at room temperature for a week. Then, these composite pellets were pressed by Lab-Tech hydraulic hot press under the pressure of 39 bar to manufacture of composite plates. Moreover, composite pellets were mixed by gelimat mixer for 15 second prior to pressing. Finally, the samples prepared in this study are coded and sample codes are shown in Table 1.

Table 1. The code of samples.

Samples	Sample Codes
PP	E0
Untreated PP/CNF composite	E1
PP/CNF composite treated with 0.1wt% Fusabond	E2
PP/CNF composite treated with 0.5wt% Fusabond	E3
PP/CNF composite treated with 1wt% Fusabond	E4

The final composition was compounded in large scale and the compounded nanocomposites were injection molded to get an automotive part. The injection molding was done at 180-210°C with heating barrel and mold temperature with a pressure of 48 bar.

Methods

Mechanical Testing

The mechanical properties such as the tensile strength, the value of elongation at break, and the value of Young's Modulus of composite samples and neat PP were characterized by tensile testing. The test samples were prepared according to the ASTM D638 standard. Then, the mechanical properties of samples were tested by SHIMADZU AGS-X tensile testing machine with a 5kN load cell and testing speed of 50mm/min.

Dynamic Mechanical Analysis

Dynamic mechanical analysis gives information about the modulus values with respect to changes in the temperature during testing. TA Instrument Q800 machine was used to determine the modulus value of composite samples and neat PP. The tested materials have dimensions of 17.5×12.2×2.3 mm. Moreover, the tested materials were heated from 30 °C to 120 °C with a heating rate 3 °C/min. The applied frequency was 1 Hz.

Thermal Gravimetric Analysis

PerkinElmer Simultaneous Thermal Analyzer (STA 8000) was used to measure thermal properties of composite materials. Samples were heated from 10 °C to 700 °C at a heating rate of 10° C /min. under nitrogen flow.

Fourier Transform Infrared Spectroscopy

Thermo Scientific TM FTIR spectrometer in Attenuated Total Reflection Infrared (ATR-IR) mode was used to obtain the spectra of composites. Attenuated total reflection infrared (ATR-IR) spectra of the materials were collected at a resolution of 4 cm⁻¹ and a number of 16 scans per sample. Spectral outputs were recorded in absorbance mode as a function of wave number.

Contact Angle Measurement

The surface wettability of PP/CNF composites was evaluated by contact angle measurement, using a KSV Attension Theta Lite Optical Tensio meter, combined with a high-speed camera. Water droplets were deposited directly at the top surfaces of the material, and the water contact angles were measured. Three measurements were performed per sample and averaged.

Scanning Electron Microscopy

The morphologies of the composite samples were studied by SEM using a VEGA3 TC TESCAN scanning electron microscope. SEM images were taken at an accelerating voltage of 30 kV at various magnifications. The samples were coated with gold.

3. Results and Discussions

Fourier Transform Infrared Spectroscopy

The FTIR technique was used to study the main functional groups present in composites. The most distinct spectral peaks were published about cellulosic fibers. The absorption band around $3329\text{--}3334\text{ cm}^{-1}$ was attributed to stretching vibrations of hydroxyl groups in cellulose molecules (Garside and Wyeth, 2003). The OH groups attributed to adsorbed water, aliphatic primary and secondary alcohols found in cellulose (Rosli et al., 2013). Near the OH stretching vibrations around $2892\text{--}2898\text{ cm}^{-1}$ was attributed to C-H stretching vibrations (Zain et al., 2014). The bands in the region around $1428\text{--}1315\text{ cm}^{-1}$ were assigned to C-H in plane deformation of CH₂ groups (Keshk et al., 2015). The absorption band observed around 898 cm^{-1} corresponded to β -1, 4 glycosidic linkages in and this peak is the most significant absorbance band which corresponds to glycosidic C-H deformation, with a ring vibration contribution on O-H bending (Keshk et al., 2015; Murigi et al., 2014).

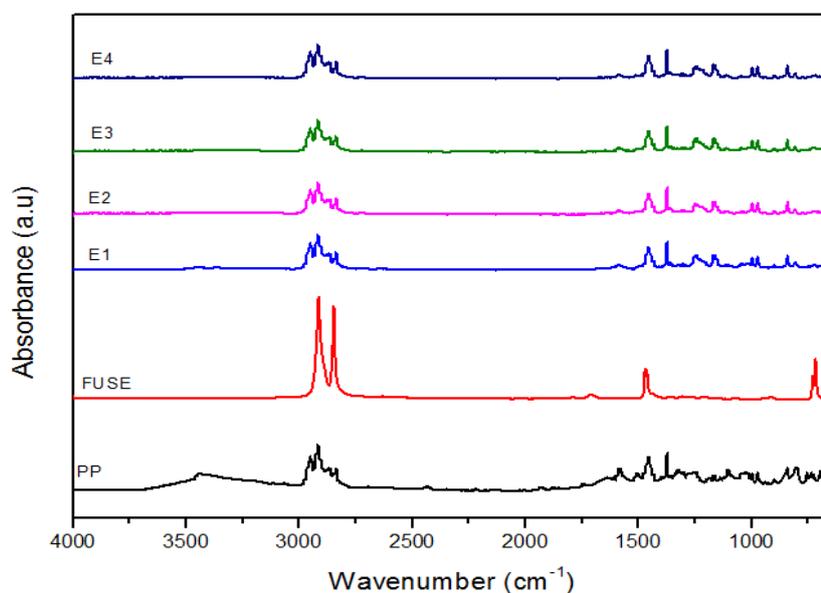


Figure 1. FTIR spectra of neat PP, coupling agent, and treated & untreated PP/CNF composites.

The FTIR spectroscopic analysis of neat PP, coupling agent, and PP/CNF composites are shown in Figure 1. The peaks at 3335 cm^{-1} corresponding to the molecular vibrations of the PP structure disappeared with addition of nanocellulose. Zain et al. (2014) stated that there is O-H stretching vibration and O-H bending of the adsorbed water, respectively. This was attributed to cellulose are inter molecularly bonded to PP chain. Furthermore, the peaks at around 3000 cm^{-1} corresponding to the molecular vibrations of the coupling agent overlapped the characteristic peaks of PP structure in the composite form. The major difference between the different spectrums was observed at around 1500 cm^{-1} and 1000 cm^{-1} after loading of nanocellulose. The band around 1057 cm^{-1} corresponds to C-O-C skeletal vibration and the band around 1060 cm^{-1} that represents C-O and C-H stretching vibration, confirms the structure of cellulose. Another band observed around 898 cm^{-1} . This peak is the most significant absorbance band, which corresponds to glycosidic C-H deformation, with a ring vibration contribution on O-H bending (Keshk et al., 2015). These features are characteristic linkage between the anhydro glucose units in cellulose (Rosli et al., 2013). These bands existence is an evidence of interfacial adhesion of polypropylene and nanocellulose.

Mechanical Testing

The reinforcement effect of coupling agent on the mechanical properties of nanocellulose–polypropylene

composites was evaluated as a function of the coupling agent content. The tensile strength, elongation at break and the Young's modulus of PP/CNF composite were characterized and the mechanical properties of composites are shown in Table 2.

Table 2. Mechanical properties of neat PP and PP/CNF composites.

Sample	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation at break (%)
E0	14.45 ± 0.5	0.570 ± 0.047	14 ± 5.9
E1	14.56 ± 0.15	0.736 ± 0.055	15.56 ± 3.6
E2	19.99 ± 1.8	1.067 ± 0.073	15.99 ± 6.8
E3	14.62 ± 0.43	0.720 ± 0.038	17.17 ± 5.05
E4	15.09 ± 0.39	0.730 ± 0.033	18.85 ± 2.2

In the present study, small amount of nanocellulose (2wt %,) was loaded in the polypropylene matrix polymer without coupling agent to perform the effect of this small loading. Then, coupling agent was loaded to improve the mechanical properties and interfacial adhesion of polypropylene and nanocellulose. The interfacial interaction of the nanocellulose and the polypropylene was important in order to increase the mechanical properties. According to the spectroscopic analysis, cellulose are inter molecularly bonded to PP chain in all cases. However, efficiency of loading is very important factor for mechanical properties. The results show that the loading of 0.1wt% FUSE increased the strength of composite. The increase in the strength values was 38 % at 0.1wt% FUSE loading compared to neat PP. Moreover, the results showed an 87% increase in the Young's Modulus of PP/CNF composite containing 0.1wt % FUSE compared to that of neat polypropylene. Electron microscopy also confirms that the tensile testing results. The cellulose nanofibrils were dispersed more effectively in composite containing 0.1wt % FUSE than other composite samples. This is consistent with the data obtained by Krishnan et al. (2015) and Hietala et al. (2013). According to their results, mechanical properties improved with effectively loading of cellulose fibers.

Dynamic Mechanical Analysis

The DMA results of pure polypropylene and polypropylene-nanocellulose composites are shown in Table 3 and Figure 2.

Table 3. Storage modulus (MPa) of neat PP and PP/CNF composites.

Sample	30 (°C)	60 (°C)	90 (°C)	120 (°C)
E0	1156.64	789.86	444.18	275.52
E1	1033.56	727.79	395.62	225.23
E2	1148.07	801.44	435.19	251.89
E3	979.64	686.62	372.91	212.91
E4	863.33	625.53	345.55	196.14

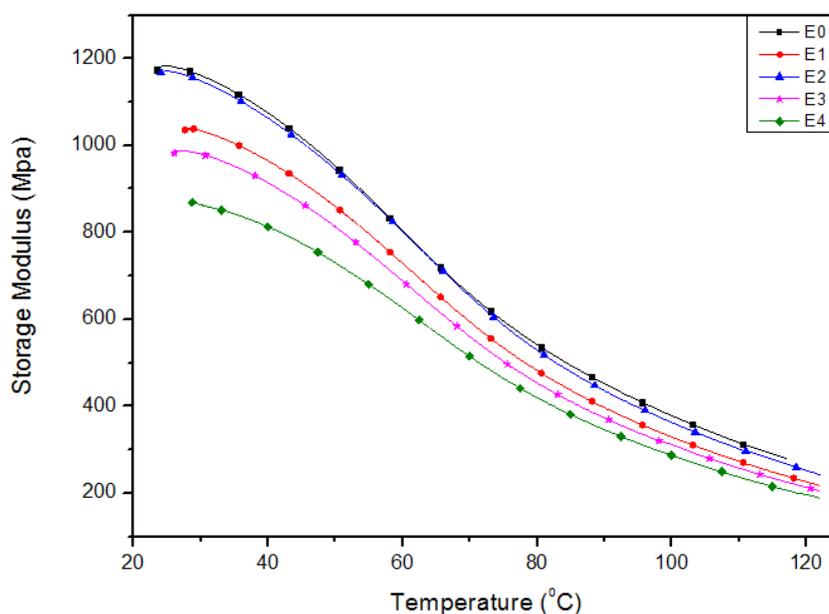


Figure 2. DMA curves of PP, NC and FUSE treated samples.

Dynamic mechanical analysis gives information about the modulus values with respect to changes in the temperature during testing. PP/CNF composites were tested in a temperature scale between the 30 °C-120 °C. The modulus value of neat PP decreased at all temperatures compared to that of composite samples. However, the modulus values of the composite containing 0.1 wt % FUSE exhibited the better results than other composite samples. The high strength of composite containing 0.1wt % FUSE was confirmed with the tensile testing as well. Moreover, the value of storage modulus decreased with increased FUSE content in the range of 0.1-1 wt%. This result is consistent with contact angle measurements because increasing amount of FUSE decreased samples hydrophobicity and homogeneity. Therefore, the values of storage modulus decreased with increasing amount of FUSE content.

Thermal Gravimetric Analysis

Thermogravimetric analysis was conducted a nitrogen atmosphere to investigate the thermal stability of composite materials. Nanocellulose, polypropylene, and nanocellulose–polypropylene composite prepared without coupling agent exhibits single degradation step is shown in Figure 3.

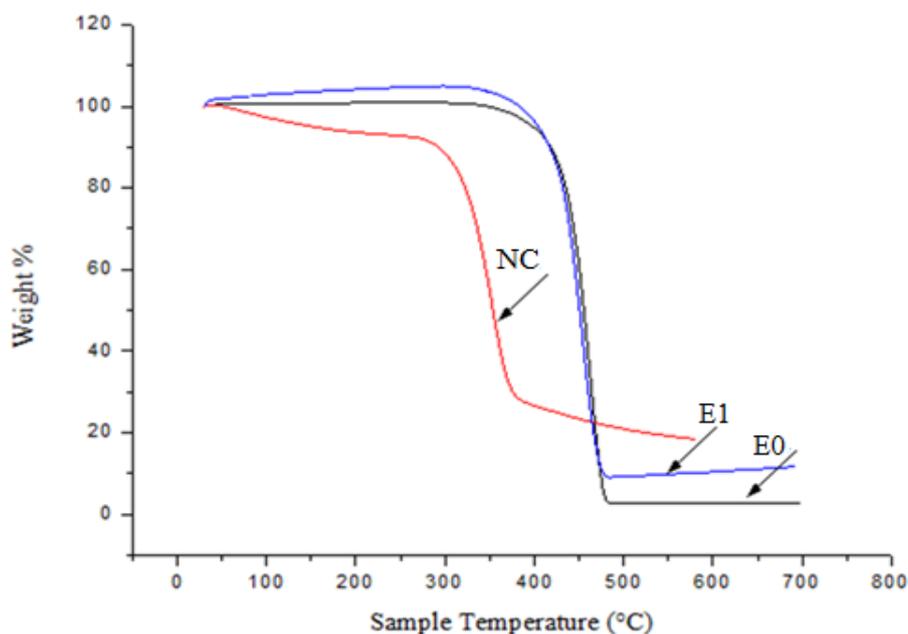


Figure 3. TGA curves of a) neat NC b) neat PP c) untreated NC/PP composite.

This was determined from weight changes in TGA curves. Compare to the PP/CNF composites, small weight loss occurred in pure cellulose between 50-150°C that is attributed to the removal of absorbed water in cellulose (Reixach et al., 2015; Yuan and Lee 2013). Then, pure nanocellulose sample indicated a more pronounced degradation process at around 280°C to 370°C. This is the consistent with data obtained by Lee and Luner (1972). Moreover, the TGA of neat PP showed a single degradation step, starting at 348°C. This is the consistent with data obtained by Reixach et al. (2015). The TGA of treated composites also followed the similar degradation patterns and some important properties are shown in the Table 4.

Table 4. Onset temperature, degraded weight, end temperature and maximum degradation temperature of the specimens.

Specimens	T _{onset} (°C)	Degraded Weight (%)	T _{end} (°C)	Charred Residues at 600°C (%)
E0	443.1	98	472.8	2
E1	424.16	89	477.3	11
E2	440.18	99	480.1	1
E3	442.20	98	479.8	2
E4	447.27	99	487	1

Thermogram of composite treated with 0.1 wt % FUSE exhibits single stage decomposition starting from 402 to 485°C. The region between 200-400°C in composite treated with 0.1 wt % FUSE is almost linear and accounts

for meager 3% weight loss due to moisture. Rate of decomposition is very fast between the region 430° - 485° C resulting in 91% weight loss. It can be ascribed to dehydration, depolymerization and pyrolysis at higher temperature. It stabilizes and tremendously slows down in the temperature range 300°-410°C, accounting for only 5% weight loss. The final degradation temperature is recorded at 480°C leaving behind 1% residue. In case of composite treated with 0.5wt % FUSE initial decomposition temperature is observed at 330°C and degradation completes at 485°C leaving 2% residue. Decomposition curve shows that degradation completes at 390°-500° C region, resulting 96 %weight loss. Furthermore, composite treated with 1wt % FUSE exhibits single stage decomposition starting from 300 to 487°C. This degradation indicated the pyrolysis of cellulose and PP because it was mainly happened at 315– 400°C (Bryant, 1968). Rate of decomposition is very fast between the region 380° - 487° C resulting in 97% weight loss. The final degradation temperature is recorded at 487° C leaving behind 1% residue. On the other hand, onset temperature of neat PP slightly decreased with addition of 2wt % nanocellulose because cellulose has the hydrophilic nature. However, onset temperature and thermal stability of PP/CNF composite slightly increased with addition of coupling agent. The TGA analysis results show that the thermal stability of PP/CNF composites did not change much but addition of coupling agent was improved the composite thermal stability and homogeneity.

Contact Angle Measurement

More specifically, a contact angle less than 90° indicates that wetting of the surface is favorable, and the fluid will spread over a large area on the surface; while contact angles greater than 90° generally means that wetting of the surface is unfavorable so the fluid will minimize its contact with the surface and form a compact liquid droplet (Yuan and Lee, 2013). Wettability is an important property to adhesion of cellulosic materials (Lee and Luner, 1972). Wetting quality of this types of materials are influenced by many factors including macroscopic characteristics (e.g., porosity, surface roughness, wood surface polarity, pH value, moisture content, grain orientation, and extractives), surface quality (e.g., virgin, aging, and contamination), processing temperature, and properties of adhesives (e.g., acidity, rheology, and viscosity) (Yuan and Lee, 2013; Bryant, 1968). Therefore, the wettability of composites was studied by contact angle measurements and results of measurements are shown in Table 5. The results show that contact angle values of composites were measured the range from 112° to 98°. Compared to the pure PP, hydrophobicity of composites increased. Composite treated with 0.1wt% FUSE indicated the highest CA value was 112°. This is consistent with the data obtained by Matuana et al (28). Furthermore, CA values and homogeneity of composites decreased with increasing amount of FUSE. They are shown in Figure 4 and Table 5.

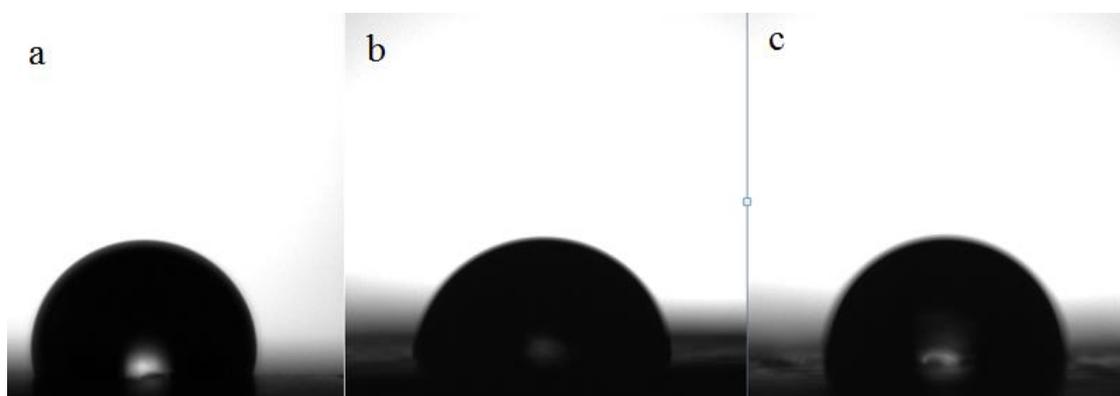


Figure 4. Contact angle images of a) PP/CNF composite treated with 0.1wt% FUSE b) PP/CNF composite treated with 0.5wt% FUSE c) PP/CNF composite treated with 1wt% FUSE

Table 5. Contact angle measurement results.

Sample	CA left [°]	CA right [°]	CA mean [°]
E1	121.92	121.90	121.91
E2	112.15	112.15	112.15
E3	99.92	99.90	99.91
E4	98.20	95.94	97.07

Cellulose fibers became observable and the homogeneity of the composite decreased in the case of increasing FUSE content. The existence of cellulose fibers on the composite surface led to decrease CA value, because cellulose has the hydrophilic nature. Therefore, CA value of FUSE treated composites decreased while increasing amount of FUSE.

Scanning Electron Microscopy

SEM was carried out to determine the dispersion of cellulose fiber in the matrix, adhesion between fiber and matrix and to detect the presence of any defect. SEM images of neat nanocellulose and PP/CNF composites are shown in Figure 5a-5d.

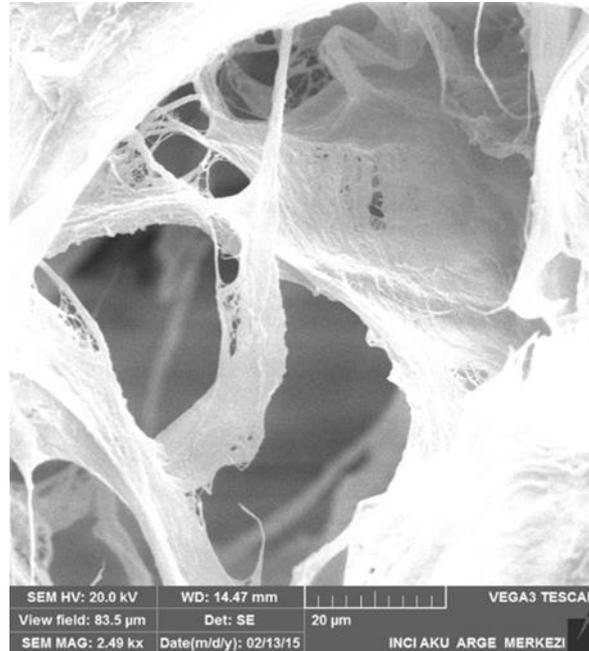


Figure 5a. SEM images of neat NC

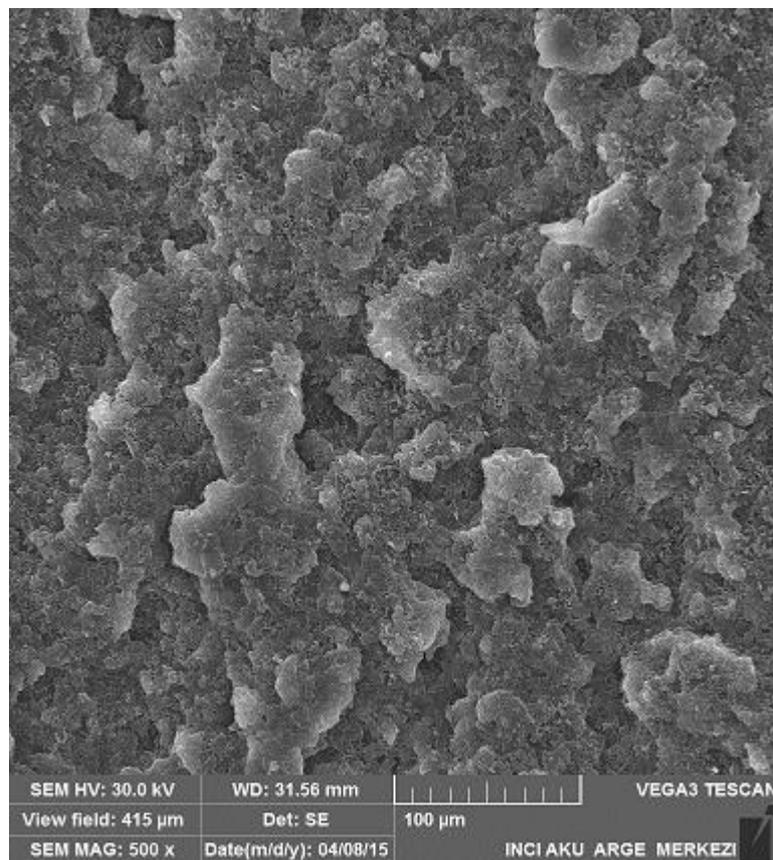


Figure 5b. SEM images of PP/CNF composite treated with 0.1 wt% FUSE

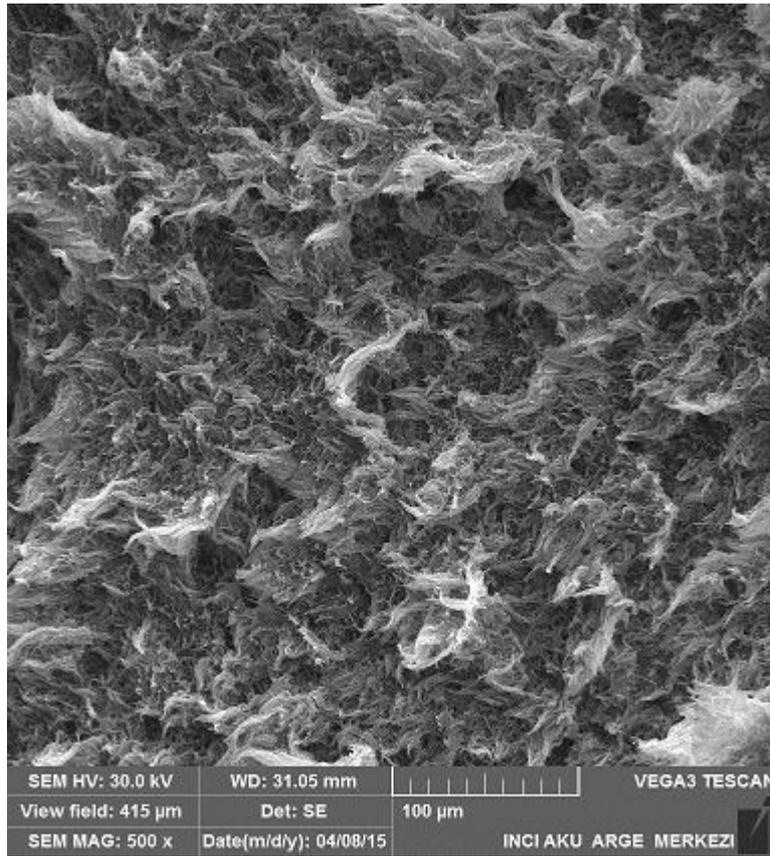


Figure 5c. SEM images of PP/CNF composite treated with 0.5wt% FUSE

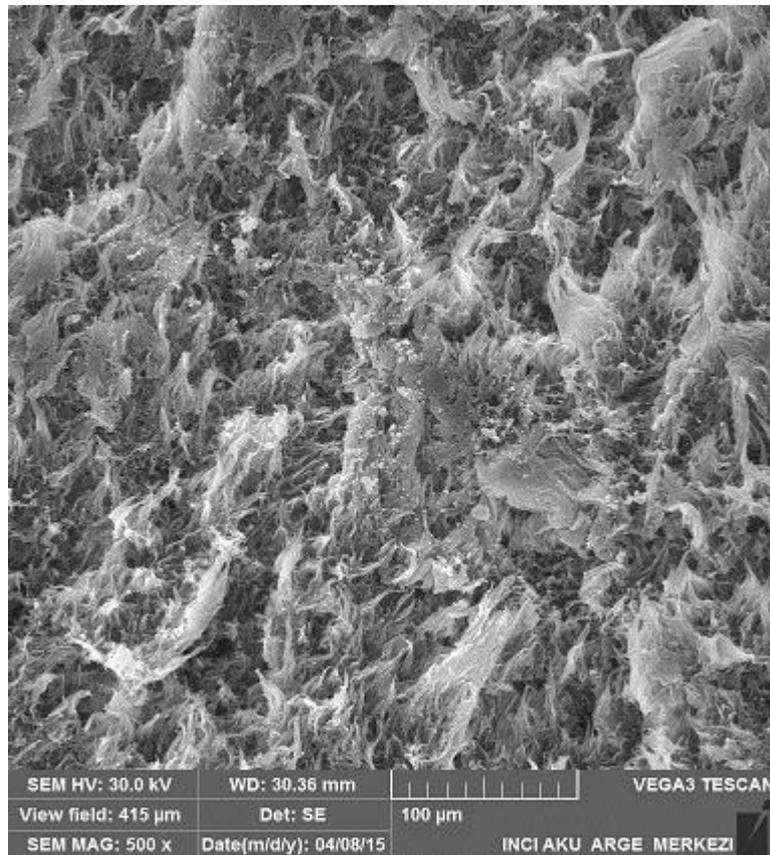


Figure 5d. SEM images of PP/CNF composite treated with 1wt% FUSE

SEM images of fracture surfaces of composites clearly indicated the differences. The sample reinforced with 0.1wt% FUSE exhibited the better dispersion from the other samples. In this treatment, adhesion between cellulose fiber and polypropylene indicated the better result because cellulose fibers were buried more effectively in the polypropylene matrix from the others sample. On the other hand, the homogeneity of composites treated with FUSE decreased with increasing amount of FUSE.

Injection Molded Part

In the final part of the study, the optimized formulation was shaped into an automotive part via injection molding. The study was carried jointly with Farplas Auto Spare Parts Production Company and one of the most important part of this study was to shape it into a final commercial product. There are numerous studies on nanocellulose based nanocomposites in the literature but it is very rare to see a final product that can be used in a final application area. This will also help to produce sustainable auto parts with improved properties. The final product is shown in Figure 6.



Figure 6. The final product.

The product was smooth and the shrinkage of the injection molded part was minimized using certain injection molding parameters. This product will be used by Farplas.

4. Conclusion

Nanocellulose–polypropylene composites were manufactured via twin screw extrusion technique with and without coupling agent. The matrix polymer (PP) and the prepared composites mechanical properties, thermal properties and interfacial adhesion were analyzed and the results showed that thermal stability and mechanical properties of composites were improved by adding 0.1wt% FUSE. This sample tensile strength observed around 20 MPa while PP tensile strength value was around 14MPa. Compared to the neat PP, the Young's (elastic) modulus of the 0.1wt% FUSE containing PP nanocomposites was higher than 87% and the tensile strength was higher than 38 %.

Acknowledgement

2013-STZ-061 SanTez project is greatly acknowledged for the study. 2016-ÖNP-MÜM-002 project is also highly important for this study and it is also acknowledged.

Kaynaklar

- Zini, E. and M. Scandola, Green composites: An overview. *Polymer composites*, 2011. 32(12): p. 1905-1915.
- Asokan, P., M. Firdoous, and W. Sonal, Properties And Potential Of Bio Fibres, Bio Binders, And Bio Composites. *Rev. Adv. Mater. Sci*, 2012. 30: p. 254-261.

- Jancar, J., et al., Current issues in research on structure–property relationships in polymer nanocomposites. *Polymer*, 2010. 51(15): p. 3321-3343.
- Fermeglia, M., P. Posocco, and S. Pricl, Nano tools for macro problems: multiscale molecular modeling of nanostructured polymer systems. *Composite Interfaces*, 2013. 20(6): p. 379-394.
- Chan, M.-I., et al., Mechanism of reinforcement in a nanoclay/polymer composite. *Composites Part B: Engineering*, 2011. 42(6): p. 1708-1712.
- Andrews, R. and M.C. Weisenberger, Carbon nanotube polymer composites. *Current Opinion in Solid State and Materials Science*, 2004. 8(1): p. 31-37.
- Azeredo, H., et al., Nanocellulose reinforced chitosan composite films as affected by nanofiller loading and plasticizer content. *Journal of Food Science*, 2010. 75(1): p. N1-N7.
- Ashori, A., Wood–plastic composites as promising green-composites for automotive industries! *Bioresource Technology*, 2008. 99(11): p. 4661-4667.
- Xie, Y., et al., Silane coupling agents used for natural fiber/polymer composites: A review. *Composites Part A: Applied Science and Manufacturing*, 2010. 41(7): p. 806-819.
- Nair, S.S., et al., High performance green barriers based on nanocellulose. *Sustainable Chemical Processes*, 2014. 2(1): p. 23.
- Bledzki, A.K., O. Faruk, and V.E. Sperber, Cars from Bio-Fibres. *Macromolecular Materials and Engineering*, 2006. 291(5): p. 449-457.
- Koronis, G., A. Silva, and M. Fontul, Green composites: a review of adequate materials for automotive applications. *Composites Part B: Engineering*, 2013. 44(1): p. 120-127.
- Holbery, J. and D. Houston, Natural-fiber-reinforced polymer composites in automotive applications. *Jom*, 2006. 58(11): p. 80-86.
- Nakagaito, A., S. Iwamoto, and H. Yano, Bacterial cellulose: the ultimate nano-scalar cellulose morphology for the production of high-strength composites. *Applied Physics A*, 2005. 80(1): p. 93-97.
- Bledzki, A. and J. Gassan, Composites reinforced with cellulose based fibres. *Progress in polymer science*, 1999. 24(2): p. 221-274.
- Malainine, M.E., M. Mahrouz, and A. Dufresne, Thermoplastic nanocomposites based on cellulose microfibrils from *Opuntia ficus-indica* parenchyma cell. *Composites Science and Technology*, 2005. 65(10): p. 1520-1526.
- Nakagaito, A.N., et al., Production of microfibrillated cellulose (MFC)-reinforced polylactic acid (PLA) nanocomposites from sheets obtained by a papermaking-like process. *Composites Science and Technology*, 2009. 69(7): p. 1293-1297.
- Suryanegara, L., A.N. Nakagaito, and H. Yano, The effect of crystallization of PLA on the thermal and mechanical properties of microfibrillated cellulose-reinforced PLA composites. *Composites Science and Technology*, 2009. 69(7): p. 1187-1192.
- Iwatake, A., M. Nogi, and H. Yano, Cellulose nanofiber-reinforced polylactic acid. *Composites Science and Technology*, 2008. 68(9): p. 2103-2106.
- Seydibeyoğlu, M.Ö. and K. Oksman, Novel nanocomposites based on polyurethane and micro fibrillated cellulose. *Composites Science and Technology*, 2008. 68(3): p. 908-914.
- Nakagaito, A.N. and H. Yano, Toughness enhancement of cellulose nanocomposites by alkali treatment of the reinforcing cellulose nanofibers. *Cellulose*, 2008. 15(2): p. 323-331.
- Mohanty, A., L. Drzal, and M. Misra, Engineered natural fiber reinforced polypropylene composites: influence of surface modifications and novel powder impregnation processing. *Journal of adhesion science and technology*, 2002. 16(8): p. 999-1015.
- Mulinari, D.R., et al., Preparation and properties of HDPE/sugarcane bagasse cellulose composites obtained for thermokinetic mixer. *Carbohydrate Polymers*, 2009. 75(2): p. 317-321.
- Bengtsson, M. and K. Oksman, The use of silane technology in crosslinking polyethylene/wood flour composites. *Composites Part A: applied science and manufacturing*, 2006. 37(5): p. 752-765.
- Raj, R., et al., Use of wood fibers in thermoplastics. VII. The effect of coupling agents in polyethylene–wood fiber composites. *Journal of applied polymer science*, 1989. 37(4): p. 1089-1103.
- Spoljaric, S., A. Genovese, and R.A. Shanks, Polypropylene–microcrystalline cellulose composites with enhanced compatibility and properties. *Composites Part A: Applied Science and Manufacturing*, 2009. 40(6): p. 791-799.
- Garside, P. and P. Wyeth, Identification of Cellulosic Fibres by FTIR Spectroscopy-Thread and Single Fibre Analysis by Attenuated Total Reflectance. *Studies in Conservation*, 2003. 48(4): p. 269-275.
- Rosli, N.A., I. Ahmad, and I. Abdullah, Isolation and characterization of cellulose nanocrystals from *Agave angustifolia* fibre. *BioResources*, 2013. 8(2): p. 1893-1908.
- Zain, N., S. Yusop, and I. Ahmad, Preparation and Characterization of Cellulose and Nanocellulose From Pomelo (*Citrus grandis*) Albedo. *J Nutr Food Sci*, 2014. 5(334): p. 2.

- Keshk, S.M., M.S. Hamdy, and I.H. Badr, Physicochemical Characterization of Mercerized Cellulose/TiO₂ Nano-Composite. *American Journal of Polymer Science*, 2015. 5(1): p. 24-29.
- Murigi, M. K., Madivoli, E. S., Mathenyu, M. M., Kareru, P. G., Gachanja, A. N., Njenga, P. K., ... & Mercy, G. (2014). Comparison of physicochemical characteristics of microcrystalline cellulose from four abundant kenyan biomasses. *J Poly Text Eng*, 1(2), 53-63.
- Krishnan, A., C. Jose, and K. George, Sisal nanofibril reinforced polypropylene/polystyrene blends: Morphology, mechanical, dynamic mechanical and water transmission studies. *Industrial Crops and Products*, 2015. 71: p. 173-184.
- Hietala, M., A.P. Mathew, and K. Oksman, Bionanocomposites of thermoplastic starch and cellulose nanofibers manufactured using twin-screw extrusion. *European Polymer Journal*, 2013. 49(4): p. 950-956.
- Reixach, R., et al., Orange Wood Fiber Reinforced Polypropylene Composites: Thermal Properties. *BioResources*, 2015. 10(2): p. 2156-2166.
- Yuan, Y. and T.R. Lee, Contact angle and wetting properties, in *Surface science techniques*. 2013, Springer. p. 3-34.
- Lee, S. and P. Luner, Wetting and interfacial properties of lignin. *Tappi*, 1972. 55(1): p. 116-&.
- Bryant, B., Interaction of wood surface and adhesive variables. *Forest Prod. J*, 1968. 18(6): p. 57-62.