Archives of Materials Science and Engineering

Volume 79
Issue 1
May 2016
Pages 12-18

International Scientific Journal
published monthly by the
World Academy of Materials
and Manufacturing Engineering

Bio-composites based on polypropylene filled with waste of camellia sinensis

N. Kucukdogan [a,*], S. Ozturk [b], M. Sutcu [b]

[a] Graduate School of Natural and Applied Science, Material Science and Engineering Department, Izmir Katip Celebi University, Izmir, Turkey

[b] Material Science and Engineering Department, Izmir Katip Celebi University, Izmir, Turkey

* Corresponding e-mail address: kucukdogan.nilay@gmail.com

ABSTRACT

Purpose: This study aims to examine the properties of polypropylene (PP) composites filled with household waste tea (camellia sinensis) (HWT) to development of their mechanical properties and also to gain the economic value of HWT.

Design/methodology/approach: HWT was grinded using a disk mill in the finely powder form. Prepared finely tea powders were characterized compositional and morphologically by using Fourier transform infrared spectroscopy, X-Ray diffractometer and scanning electron microscope. PP composites were prepared at different proportions of HWT ranging from 0 wt% to 40 wt% with two particle sizes (<30 μm and <60 μm) by high-volume energy mixer so as to evaluate the effect of the HWT filler on mechanical properties.

Findings: The mechanical properties of composites were evaluated through modulus of elasticity, ultimate tensile strength and flexural strength in terms of filler particle size and filler contents. The results showed that the modulus of elasticity of composites increased as a function of additional HWT particles and the highest value was observed which was containing 40 wt% filler contents.

Research limitations/implications: Clearly, the maximum flexural and tensile strength of HWT particles filled composites were observed for 10 wt% HWT loading into PP composite increased when compared to other filler contents (20-40 wt%).

Practical implications: For potential applications in the plastics industry, such as outdoor deck floors, cladding and siding, indoor furniture etc., it is desirable to form polymer composite systems by combining phases with polymer, so as to reduce expenses of materials, to have strength and, at the same time to be biodegradable.

Originality/value: Despite HWT was chemically untreated, the mechanical properties of composites increased it would appear that it can be a renewable alternative material for manufacturing of PP matrix composites.

Keywords: Thermoplastic resin; Natural reinforcement; Mechanical properties

Reference to this paper should be given in the following way:

N. Kucukdogan, S. Ozturk, M. Sutcu, Bio-composites based on polypropylene filled with waste of camellia sinensis, Archives of Materials Science and Engineering 79/1 (2016) 12-18.

PROPERTIES

1. Introduction

Recently, development of commercially feasible eco-friendly products based on natural sources for both fillers and matrices for many applications is a steady rise [1-5]. The new regulations with environmental constraints on the recycling of polymer composites have forced manufacturer to progress new materials from renewable sources [2]. Therefore, this effort contains new approaches to manufacture natural fiber filled polymer composites with better mechanical and the other intended properties [1].

Both commercial and agricultural wastes are constituted important problem for the environment. For this reason, the proper utilization or recycling of wastes has been recognized worldwide not only as developing sustainability in industry but also for solving environmental problems [3].

All over the world, in the context of the sustainable development approach; waste management strategies aims to transform from not a treat the environmental and human health to the economic input. Japan commonly used thermal methods for disposal of solid waste due to the limited habitable space. In Japan, produced 50 million tons of household solids are disposal by burning methods in approximately 1300 thermal burning plants [4]. It is seen in the literature survey, solid wastes used in production of composites are by-products from agricultural wastes [4,5]. Other waste material like HWT can be used as filler in production of composites.

Many ligno-cellulosic materials, such as jute, cotton, ramie and coir are utilized as reinforcements or fillers in thermoplastic composites, especially including the automotive and construction industries. Ligno-cellulosic based fillers or reinforcements have important advantages in polymer matrices in relation to other inorganic or synthetic materials. The most significant advantages of these materials are: (i) easy recyclability, (ii) biodegradability, (iii) low energy requirements for processing, (iv) low density, (v) natural appearance, (vi) easy color processing and (vii) low cost. The low cost is the most effective factor for the materials selection. The cost of natural fiber filled polymer composites has a quietly low value. Moreover this cost can be reducible with used commercial or agricultural waste in polymer composite [3-16]. Camellia sinensis is a non-woody plant and grow in north-eastern of Turkey, where it has been used in the preparation of a traditional tea drink. Turkey is a significant producer of tea which is an annual production of dry-tea approximately 100.000 tones [17] hence commercially viable. Apparently, the interpretation of financial of HWT into any products, such as wood-polymer composites, becomes a strategic approach for developing polymer composite sectors and also it will increase the economic value of camellia sinensis plant.

In this study, HWT powders were used as a filler material for a PP matrix. The objectives of this study were (1) to research composition and morphology of the HWT (2) to evaluate the mechanical properties of waste filled PP composites without any coupling agents or additives.

2. Material and method

2.1. Raw materials

A matrix of PP (MH-418) supplied by Petroleum Chemical Industry (Turkey, Izmir) with density 0.905 g/cm^3 (at 23°C) and melt flow rate 4.5 g/10 min (2.16 kg at 190°C) was used in experimentally.

The camellia sinensis by-product used as filler materials collected from a tea-café in Izmir city (Turkey). The filler materials are subjected to some pre-processing for use in composite manufacturing. Firstly, HWT particles were dried at 70±2°C in drying oven (Binder ED53) for 24 hours. Secondly, HWT particles were milled by vibratory disc mill (Retsch RS200) for 2 minutes. And finally, milled particles were sieved via sieve shaker (Retsch AS200) to obtain particle size under 30 μm and 60 μm which were used in experimentation.

2.2. Preparation of composites

HWT particles were mixed according to Table 1 in distinct concentrations of particles/PP-matrix.

Table 1.
Composition of the studied formulations

Components, weight%	Composite types				
	A	B[a,b]	C[a,b]	D[a,b]	E[a,b]
HWT	0	10	20	30	40
PP	100	90	80	70	60

[a] HWT particles under 30 μm
[b] HWT particles under 60 μm

The composites were produced by high-volume energy mixer and, they were molded using a heated-frigorific hydraulic press. In this study, coupling agents or additives were not used in the manufacturing process of the PP-composites. The mechanical tests were performed according to ASTM D790 and D3039 standards [20,21].

2.3. Characterization of HWT particles

X-Ray diffraction (XRD) analysis

The X-ray diffraction analysis of HWT particles were carried out with Bruker D2 Phaser, X-ray diffractometer system with Ni filtered Cu-Kα radiation (λ=1.54 Å) generated at 30 kV and 10 mA. The diffracted X-ray beam was collected by scanning the detector between 2θ=5° and 50°. The step size and scan time were 0.014° and 6.78s, respectively.

Fourier transform infrared spectroscopy (FTIR) analyses

Fourier transform infrared spectroscopy-Attenuated Total Reflectance (ATR) analyses were carried out with Thermo-Scientific Nicolet IS5 Spectrometer. IR spectra were recorded in the spectral range of 400-4000 cm^{-1} with a 2 cm^{-1} resolution and 25 scans.

Scanning electron microscopy (SEM) analysis

The scanning electron microscopy analysis of HWT particles and HWT/PP composites samples were performed by using SEM (FEI QUANTA 250).

2.4. Mechanical properties of composites

The tensile and three point bending tests of neat PP and HWT/PP composites were carried out using Shimadzu Autograph AG-IS series universal testing machine at crosshead speeds of 5 mm/min according to ASTM 3039 standard [20] and 1 mm/min by following ASTM D790 standard [21], respectively.

3. Results and discussion

3.1. Characterization of HWT particles

XRD analysis

It can be seen that the major phase in HWT particle is cellulose I (see Figure 1). The characteristic crystalline form cellulose I-α shows peak at 14.95°, 16.28° and, 22.11° also the characteristic crystalline form cellulose I-β shows peak at 14.05°, 16.28° and, 21.52° [14,23,24]. Crystallinity index value of HWT was found as 24.1%.

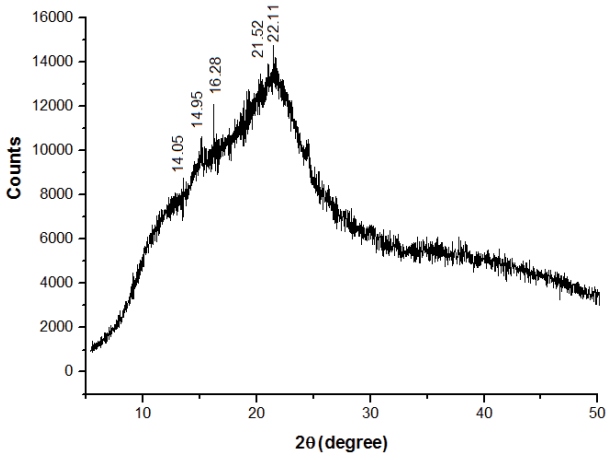

Fig. 1. XRD patterns of HWT

FT-IR analysis

The functional groups present in HWT particle was analyzed through FTIR analysis (see Figure 2). The major constitutes of ligno-cellulosic materials are cellulose, hemicellulose and lignin, meanwhile the minor compositions include pectin, proteins and water-soluble components [22].

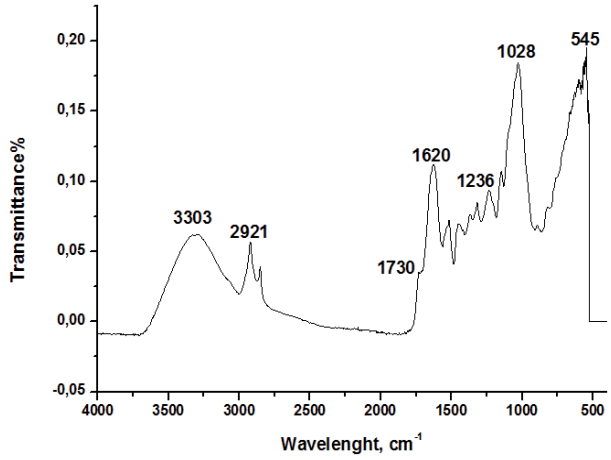

Fig. 2. FTIR spectrum of HWT

The most significant absorbance bands assigned to crystalline cellulose I [24,25]. 3175-3350 cm^{-1} regions are assigned to hydrogen bonded (O-H) stretching, absorbed water 1620 cm^{-1}, also bands assigned to different lignin groups, such as 1730, 1518, 1368 and, 1236 cm^{-1}. The main bands assigned to asymmetric methoxyl C-H stretching at 2921 cm^{-1}. The C-OH stretching vibration of the cellulose backbone was located at 1028 cm^{-1} [26,27].

SEM analysis

The scanning electron micrographs of HWT particles sieved under 30 µm and 60 µm are indicated in Figure 3a and 3b, respectively. The morphology of the particles has irregular shape like fibrous and spherical.

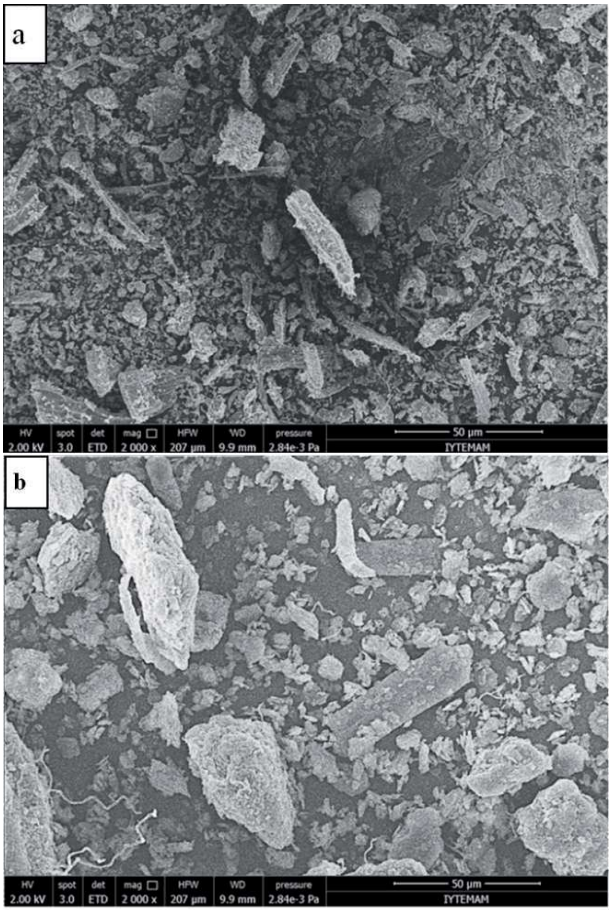

Fig. 3. SEM micrographs of HWT: a) <30 µm, b) <60 µm

3.2. Mechanical properties of composites

The mechanical properties of composites are determined in terms of tensile strength, flexural strength and modulus of elasticity. The tensile strength values of both neat PP and HWT/PP composites at different filler content with two different particle sizes are shown in Figure 4. It can be seen in Figure, the tensile strength of neat PP (A), $B^{a,b}$, $C^{a,b}$, $D^{a,b}$ and $E^{a,b}$ composites were obtained to be approximately 26.42, 27.07, 21.68, 19.47, and 16.69 N/mm^2 with particle size under 60 µm, and 26.42, 28.31, 27.78, 21.41 and 18.74 N/mm^2 with particle size under 30 µm, respectively.

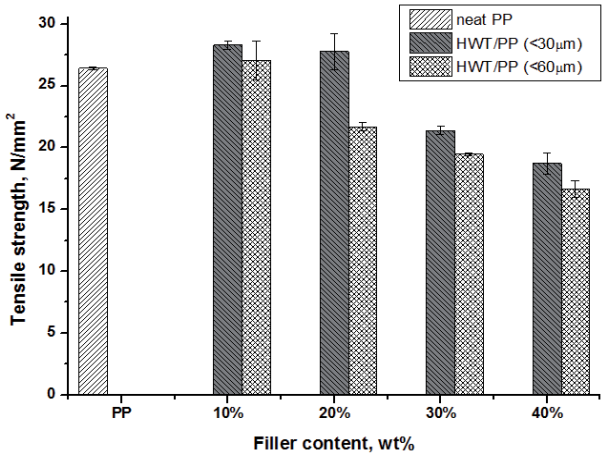

Fig. 4. Effect of filler content and particle size on tensile strength of PP composites filled with HWT

It can be seen that among produced composites, B type composites with two particle sizes were presented the highest improvement in terms of tensile strength (as seen from Figure 4).

The micrographs of the tensile fractured surface for $B^{a,b}$ composites with HWT particle size under 30 µm and 60 µm are shown in Figure 5 (a) and (b), respectively. The HWT particles homogenous dispersed in composites are observed from these fractured surfaces.

The flexural strength values of both neat PP and HWT/PP composites at different filler content with two different particle sizes are shown in Figure 6. The flexural strength values of $B^{a,b}$, $C^{a,b}$, $D^{a,b}$ and $E^{a,b}$ composites were obtained to be approximately 44.19, 54.97, 45.89, 40.89 and 37.42 N/mm^2 with particles size under 60 µm, and 44.91, 49.18, 44.86, 41.28 and 35.1 N/mm^2 with particles size under 30 µm, respectively. It should be noted that among these composites with two particle sizes, 10%HWT/PP ($B^{a,b}$) composites exhibited the best improvement in flexural strength. For higher filler loadings (20-40 wt%) presented a sudden decrease in flexural strength. However, the flexural strength of the composite with addition of 20 wt% HWT is still same with that of the neat PP.

When investigating the tensile and flexural strengths in terms of the size of the fillers, the tensile strength values of B^a, C^a, D^a, E^a composites are higher than B^b, C^b, D^b, E^b composites. Similar results are also appeared in the literature that the tensile strength of the polymer composites increases with the decrease of the particle size of fillers [28]. However, opposite to tensile strength results, flexural strength results of composites reduced with the decrease of the particle size. In tensile test, it may be

explained that the stress transferred between filler and polymer matrix cannot be support in this regard the loaded material play like filler than as reinforcement. The length and shape of particles influence the mechanical properties of polymer composites [7]. In flexural test, HWT particles with irregular shape and fibrous structure behave like reinforcement and contribute to the flexural strength.

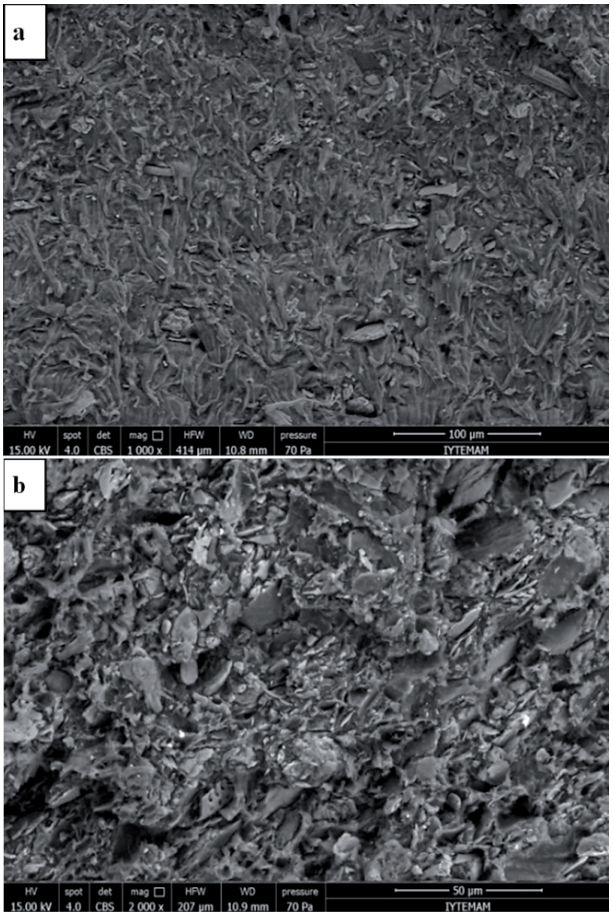

Fig. 5. SEM micrographs of the tensile fractured surface for $B^{a,b}$ composites with HWT particle size under: a) 30 µm, and b) 60 µm

Generally, the modulus of elasticity of composites increased as a function of increased HWT fillers. It can be seen that the modulus of elasticity steadily increases with increasing the filler contents. The effect of two different particle size (undersize: 30 and 60 µm) distributions on the modulus of elasticity of composites is shown in Figure 7. For higher filler contents of HWT (30-40wt%), the modulus of elasticity is not almost dependent of particle size.

The presence of fillers with a non-uniform shape does not the stress transferred between filler and the polymer matrix [6]. In this regard, the fillers act more than reinforcement and literature emphasized that shape, length and dimension of fibers or powders influence the mechanical properties of polymer composites. In this study, the shapes of HWT have been irregular shape, which could be influenced in the relationship filler to reinforcement [6,7].

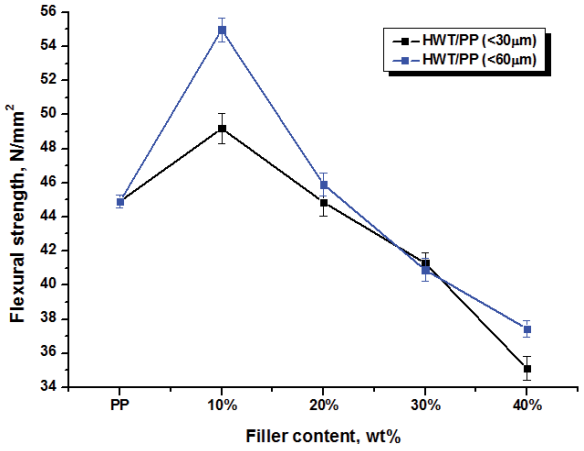

Fig. 6. Effect of filler content and particle size on flexural strength of PP composites filled with HWT

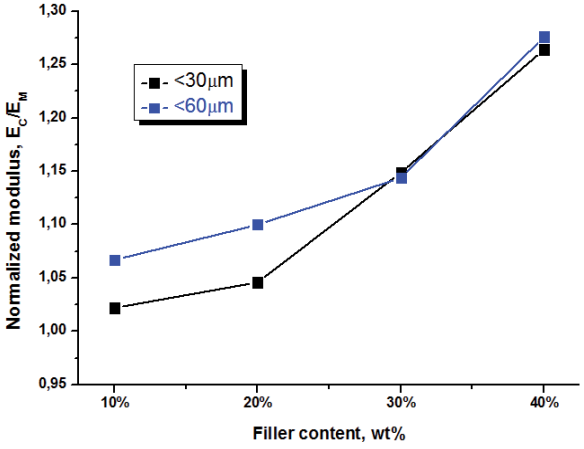

Fig. 7. The modulus of elasticity HWT filled PP composites as a function of filler content and particle size (E_c: modulus of composite, E_M: modulus of matrix)

4. Conclusions

To gain the economic value of HWT, as a household wastes, production of HWT filled PP composites were performed by using high-volume energy mixer.

Tensile and flexural strength of PP increased up to HWT loading of 10 wt%, then started to decrease. When HWT of 10 wt% were used in HWT/PP composites, tensile and flexural strength of neat PP increased 7.15% and 24.4%, respectively.

Modulus of elasticity values, which is strongly dependent on filler content, were increased by increasing of HWT content in composites with two particle sizes and these results are compatible with the literature [28]. Also, the modulus of composites, especially C and D, are not affected significantly by particle size of HWT.

10 wt% HWT/PP composites for both particle sizes were presented the highest improvement in terms of tensile strength and flexural strength. Furthermore, it is observed that the modulus of elasticity of composites increase with increasing filler content.

Considering environmental and economic aspects, it can be said that HWT could be used as filler material up to 10% by weight without coupling agent in production of bio-composites.

References

[1] K. Satyanarayana, G. Arizaga, F. Wypych, Biodegradable Composites Based on Lignocellulosic fibres-An overview, Progress in Polymer Science 34 (2009) 982-1021.

[2] H. Essabir, E. Hilali, A. Elgharad, H. El Minor, A. Imad, A. Elamraoui, O. Al Gaoudi, Mechanical and thermal properties of bio-composites based on polypropylene reinforced with Nut-shells of Argan particles, Materials and Design 49 (2013) 442-448.

[3] F. Al-Oqla, S.M. Sapuan, Natural fiber reinforced polymer composites in industrial applications: feasibility of date palm fibers for sustainable automotive industry, Journal of Cleaner Production 66 (2014) 347-354.

[4] European Commission, Reference Document on the Best Available Techniques for Waste Incineration, Integrated Pollution Prevention and Control, Brussel, 2006.

[5] F. Saltabas, Y. Soysal, V.S. Yıldız, Thermal Disposal Methods of Household Solid Waste and Applicability in Istanbul, Scientific National Symposium, Symposium of Solid Waste Management in Turkey, TURKAY'2009, Istanbul-Turkey, 2009.

[6] W.L.E. Magalhaes, S.A. Pianaro, C.J.F. Granado, K.G. Satyanarayana, Preparation and characterization of polypropylene/heart of peach palm sheath composite, Journal of Applied Polymer Science 127/2 (2013) 1285-1294.

[7] B.D. Mattos, A.L. Miss, P.H. Cademartori, E.A. Lima, W.L. Magalhaes, D.A. Gatto, Properties of Polypropylene Composites Filled With a Mixture of Household Waste of Mate-Tea and Wood Particles, Construction and Building Materials 61 (2014) 60-68.

[8] L. Yusriah, S.M. Sapuan, E. S. Zainudin, M. Mariatti, M. Jawaid, Thermo-physical, thermal degradation, and flexural properties of betel nut husk fiber reinforced vinyl ester composites, Polymer Composites 37 (2015) 2008-2017.

[9] J. Zhang, H. Zhang, J. Zhang, Evaluation of liquid ammonia treatment on surface characteristics of hemp fiber, Cellulose 21/1 (2014) 569-579.

[10] F. Mizi, D. Dai, B. Huang, Fourier transform infrared spectroscopy for natural fibres, Fourier Transform–Materials Analysis. Intech (2012) 45-68.

[11] Y. Zhang, C. Yin, Y. Zhang, H. Wu, Synthesis and characterization of cellulose carbamate from wood pulp, assisted by supercritical carbon dioxide, BioResources 8/1 (2013) 1398-1408.

[12] N. Reddy, Y. Yang, Structure and properties of high quality natural cellulose fibers from cornstalks, Polymer 46/15 (2005) 5494-5500.

[13] L. Segal, J. Greely, A.E. Martin, L.M. Conrad, An empirical method for estimating the degree of crystallinity of native cellulose using X-ray diffractometer, Textile Research Journal 29/10 (1959) 786-794.

[14] H. Matsumura, J. Sugiyama, W.G. Glasser, Cellulosic nanocomposites. I. Thermally deformable cellulose hexanoates from heterogeneous reaction, Journal of Applied Polymer Science 78 (2000) 2242-2253.

[15] W.L.E. Magalhaes, S.A. Pianaro, C.J.F. Granado, K.G. Satyanarayana, Preparation and characterization of polypropylene/heart of peach palm sheath composite, Journal of Applied Polymer Science 127/2 (2013) 1285-1294.

[16] A. Ashori, A. Nourbakhsh, Preparation and characterization of polypropylene/wood flour/nanocla y composites, European Journal of Wood and Wood Products 69/4 (2011) 663-666.

[17] Caykur Tea Operating Data, www.tarim.gov.tr/sgb/Belgeler/ SagMenuVeriler/CAYKUR.pdf, Access date: 01.03.2015.

[18] U.C. Yildiz, S. Yildiz, E.D. Gezer, Mechanical properties and decay resistance of wood-polymer composites prepared from fast growing species in Turkey, Bioresource Technology 96/9 (2005) 1003-1011.

[19] S. Panthapulakkal, M. Sain, Agro-residue reinforced high-density polyethylene composites: fiber characterization and analysis of composite properties, Composites Part A: Applied Science and Manufacturing 38/6 (2007) 1445-1454.
[20] ASTM D3039, Standard test method for tensile properties of polymer matrix composite materials, ASTM International, West Conshohocken, PA, 2014, www.astm.org.
[21] ASTM D790, Standard Test methods for flexural properties of unreinforced and reinforced plastics and electrical insulating materials, ASTM International, West Conshohocken, PA, 2010, www.astm.org.
[22] C.M. Popescu, M.C. Popescu, G. Singurel, C. Vasile, D.S. Argyropoulos, S. Willfor, Spectral characterization of eucalyptus wood. Applied spectroscopy 61/11 (2007) 1168-1177.
[23] Z.T. Liu, Y.N. Yang, L.L. Zhang, Z.W. Liu, H.P. Xiong, Study on the cationic modification and dyeing of ramie fiber, Cellulose 14/4 (2007) 337-345.
[24] A.C. Kılınc, M. Atagur, O. Ozdemir, I. Sen, N. Kucukdogan, K. Sever, O. Seydibeyoglu, M. Sarikanat, Y. Seki, Manufacturing and characterization of vine stem reinforced high density polyethylene composites, Composites Part B: Engineering 91 (2016) 267-274.
[25] F. Carrillo, X. Colom, J.J. Sunol, J. Saurina, Structural FTIR analysis and thermal characterisation of lyocell and viscose-type fibres, European Polymer Journal 40/9 (2004) 2229-2234.
[26] S.Y. Oh, D.I. Yoo, Y. Shin, G. Seo, FTIR analysis of cellulose treated with sodium hydroxide and carbon dioxide, Carbohydrate Research 340/3 (2005) 417-428.
[27] M.A.S. Spinace, C.S. Lambert, K.K.G. Fermoselli, M.A. De Paoli, Characterization of lignocellulosic curaua fibres, Carbohydrate Polymers 77/1 (2009) 47-53.
[28] S.Y. Fu, X.Q. Feng, B. Lauke, Y.W. Mai, Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical properties of particulate polymer composites, Composites Part B: Engineering 39/6 (2008) 933-961.