Preparation and Characterization of Hydroxyethyl Cellulose/Nanolignin Composite Films

Mustafa Zor,[a,d,]* Hikmet Yazici,[a,]* Ferhat Şen,[a] Erdal Eroğlu,[b] Zeki Candan,[c,d]
Denis Rodrigue,[e] and Xiaodong (Alice) Wang [f]

Hydroxyethyl cellulose/nanolignin composite films were prepared and characterized. The composite films were produced *via* casting of synthesized nanolignin added to hydroxyethyl cellulose at different concentrations (2.5%, 5%, 10%, and 20% by mass). A control film without nanolignin was also prepared for comparison. The thermal properties of the composite films were examined by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC), while the mechanical properties were determined by tensile testing and the surface properties were determined by water contact angle measurements. In addition, the morphologies of the samples were examined by scanning electron microscopy (SEM). It was observed that with the addition of nano lignin, the glass transition temperature of the composite films increased from 109 °C to 262 °C; the elongation at break increased from 19% to 51%; and the contact angles increased from 53 °C to 73 °C. The results showed that the presence of nanolignin produced materials being more flexible and more hydrophobic with higher glass transition temperatures.

DOI: 10.15376/biores.19.4.7354-7365

Keywords: Hydroxyethyl cellulose; Nanolignin; Composites; Films

Contact information: a: Zonguldak Bulent Ecevit University, Department of Nanotechnology Engineering, 67100, Zonguldak, Türkiye; b: Manisa Celal Bayar University, Department of Bioengineering, 45140, Manisa, Türkiye; c: İstanbul University-Cerrahpasa, Department of Forest Industrial Engineering, 34473, İstanbul, Türkiye; d: Biomaterials and Nanotechnology Research Group & BioNanoTeam, 34473, İstanbul, Türkiye; e: Laval University, Department of Chemical Engineering, Quebec, Canada, G1V 0A6; f: Laval University, Department of Wood and Forest Sciences, Quebec, Canada, G1V 0A6;
** Corresponding authors: mustafa.zor@beun.edu.tr; hikmet.yazici@beun.edu.tr*

INTRODUCTION

Green materials, known as environmentally friendly materials, highly contribute to developing better biocomposites, both in terms of recycling/composting possibilities while meeting the increasing demands to eliminate non-renewable resources. The most important advantages of these materials are their availability, low costs, biodegradability, and easy processing. In contrast, they have some disadvantages such as low hardness, stability, and moisture resistance. Nevertheless, biocomposite films are now seen as high-value-added engineering materials useful in several industrial applications such as in UV protective packaging materials, sweeteners to obtain nutritious and antioxidant-rich food ingredients in sugar-based cuisines, and hydrophilic films with high water vapor permeability food and non-food edible biocomposite films (Gennadios *et al.* 1994; Noshirvani *et al.* 2017; Achachlouei and Zahedi 2018; Lee *et al.* 2018; Balasubramanian *et al.* 2019; Rajasekharan *et al.* 2018; Candan *et al.* 2022).

To use environmentally friendly materials (green marketing) more effectively in this connection between people and the environment, alternatives have been developed that will contribute to the more sustainable growth of society, especially in the 21st century (Nogueira 2020). Accordingly, the research on environmentally friendly materials and their production has also gained momentum. Currently, the most practical way to develop an environmentally friendly material is to use natural/biosourced biopolymers. Cellulose and cellulose derivatives, which are the most abundant biopolymers in nature, are the main options for this purpose (Liu *et al.* 2021).

Hydroxyethyl cellulose (HEC) is a derivative of cellulose that is easily soluble in cold water. It has attracted the scientists' attention because films can be easily formed in an economical and practical way. The HEC is also used as a processing aid due to its important properties such as binding, suspending, thickening, dispersing, and stabilizing effects (Abdel-Halim 2014). The HEC is used in a wide range of areas including cement, paints, textiles, and paper products (Kugge *et al.* 2004; Dal-Bó *et al.* 2011; Gorgieva and Kokol 2011; Patural *et al.* 2011). The HEC is generally obtained from cellulose by a nucleophilic ring opening reaction of ethylene oxide over hydroxyl anions on the cellulose structure (Şen *et al.* 2020).

In contrast, lignin is a highly condensed polymer based on a group of compounds containing methoxyl (OCH_3), carbonyl (CO), and hydroxyl (OH) aromatic groups, with similar characteristics and chemical properties (Abreu *et al.* 1999). Over the past 20 years, several industrial organizations have benefited from the high value-added cutting-edge technologies of nanoscience. Because wood and wood-based biomaterials are sustainable, renewable, and recyclable, the number of nanotechnological studies has increased, leading to a great potential to reduce the demand for petroleum-based resources (Kocaturk *et al.* 2023). Recently, the focus has been on obtaining and improving the functionality of nanomaterials (such as nanocellulose and nanolignin) from lignocellulosic materials (Stojanovska *et al.* 2018; Stojanovska *et al.* 2019; Durmaz *et al.* 2023; Dalkılıç *et al.* 2024). For example, lignin-based nanoparticles have been used as functional additives in biobased matrices instead of lignins as additives in biobased polymeric materials on a macro scale. Liao *et al.* (2020) showed that the structure and chemical properties of lignin depend on the method used for their extraction and their source. Various methods have been discussed, and a wide range of application areas have been presented, such as wood, polymer composite and pharmaceutical-anti-corrosion industries.

According to the literature, limited studies investigated composite films based on HEC functionalized with lignins. For example, Rukmanikrishnan *et al.* (2020) examined the potential use of composite biopolymers prepared from gellan gum, HEC, and lignin for food packaging and biomedical applications. They reported that the addition of lignin (10 wt%) improved the thermal, mechanical, and hydrophobic properties of the composite films. In addition, the radical scavenging behavior of the materials was shown to be very effective in terms of antioxidant and non-cytotoxic activities.

Chen *et al.* (2023) prepared hydrophobic, magnetic hydroxyethyl cellulose-lignin composite aerogels. They reported that n-dodecyl mercaptan (NDM) effectively increased the hydrophobicity and magnetic susceptibility of the aerogels showing very high adsorption capacity for various oils. Yang *et al.* (2021) prepared polyvinyl alcohol (PVA) nanocomposite films containing different amounts (1, 2, and 3 wt%) of lignin nanoparticles using two crosslinkers (glutaraldehyde and citric acid). They reported that both crosslinkers improved the thermal, mechanical, and wettability properties of nanocomposite films. Sohni *et al.* (2019) produced lignin nanoparticles from palm kernel shell (biomass residue) and a novel lignin nanoparticles-based chitosan nanocomposites for the removal of methylene blue dye contaminant from aqueous solutions. According to the adsorption studies, various factors, such as pH, contact time, dye concentration, and temperature, were shown to have an improving effect on the removal of contaminants from aqueous solutions.

In this study, it was aimed to prepare composite films containing hydroxyethyl cellulose and nanolignin in different proportions and to investigate the changes in the properties of the composite films caused by nanolignin. Hydroxyethyl cellulose composite films containing nanolignin (0%, 2.5%, 5%, 10%, and 20% by mass) were prepared to produce films *via* casting. The samples were characterized in terms of morphological, mechanical, surface, and thermal properties.

EXPERIMENTAL

Materials

Hydroxyethyl cellulose (Mv ~90 000 g/mol), NaOH (97%, 40g/mol), and HNO_3 (70%, 63 g/mol) were purchased from Sigma Aldrich, St. Louis, MO, USA). Low sulfonated lignin Indulin AT (MeadWestVaco Corporation, Richmond, VA, USA) was used as the lignin source.

Preparation of Nanolignin

The method proposed by Frangville *et al.* (2012), was followed to synthesize the nanolignin used in this study with some minor modifications. To start, 20 g of Indulin AT was dissolved in 100 mL distilled water (d-water), and then the pH was adjusted to 11.44 with 1 M NaOH. After stable pH, the final volume of Indulin AT (7.7%) solution was made to 240 mL by adding deionized water. From this stock solution, the pH of 0.05% Indulin AT solution (10 mL) was adjusted to 1.97 with HNO_3. The lignin nanoparticles were finally obtained by centrifuging the colloidal solution at 4000 rpm for 5 min.

Preparation of the Composite films

To prepare the HEC/nanolignin (HECNL) composite films, the amounts of HEC and nanolignin (Table 1) were weighed and placed in a beaker. Then, 50 mL of deionized water was added to the beaker and mixed with a magnetic stirrer until HEC was completely dissolved. Each beaker was then placed in an ultrasonic bath (Alex Ultrasonic Cleaner Machine, Istanbul, Türkiye) for 30 min at room temperature to homogeneously distribute the nanolignin. Then, the mixture was poured into petri dishes and left to dry in an oven at 40 °C for 48 h. The composite films were finally obtained by peeling from the petri dish and coded according to their nanolignin content. For example, HEC5NL contains 5% of nanolignin by mass of HEC.

Table 1. Composition of the HEC/nanolignin Composite Films

Sample	Hydroxyethyl Cellulose (g)	Nanolignin (g)
HEC	3	0
HEC2.5NL	3	0.075
HEC5NL	3	0.15
HEC10NL	3	0.3
HEC20NL	3	0.6

Characterization

Morphological properties

SEM images were taken from the cut surfaces of the samples at different magnifications using a model MAIA3 XMU from Tescan (Brno, Czech Republic). Before each analysis, the samples were coated with platinum (Quorum Q150T ES, Quorum Technologies, East Sussex, United Kingdom).

Thermal properties

Thermogravimetric analysis (TGA) was carried out under a nitrogen atmosphere with a heating rate of 10 °C/min between 30 and 750 °C using a TG/DTA model Hitachi STA 7300 (Tokyo, Japan). Differential scanning calorimetry (DSC) was performed under a nitrogen atmosphere with a heating rate of 10 °C/min between 50 and 350 °C using a DSC 8000 from Perkin Elmer (Hopkinton, MA, USA). Thermal analyses were carried out at the "Thermal Analysis Laboratory" and "Nanotechnology Laboratory" located in İstanbul University.

Mechanical properties

The mechanical properties were determined by tensile testing using a Zwick Roell Tester (Ulm, Germany) at room temperature. The composite films were cut into dimensions of 60 mm x 10 mm x 1 mm. The tensile speed was set to 100 mm/min and 5 repetitions were performed to obtain an average with standard deviation.

Wettability

Hydrophobicity was evaluated by water contact angle measurements. The contact angle of 2 μL of d-water on the film surface was measured using a goniometer (DSA-100S, Advance Drop Shape Analyzer, Krüss, Hamburg, Germany). The measurements were taken within 5 s after placing the drops and 5 samples were used to report the data.

RESULTS AND DISCUSSION

Morphological Properties of the Composite Films

The morphology of the samples was examined by scanning electron microscopy. Figure 1 shows that the composite films had particles stacked on top of each other. However, in films containing nanolignin, mostly nanolignin particles (almost spherical) were clustered together. From these micrographs, it can be concluded that the nanolignin particles had some level of agglomeration, which increased with increasing nanolignin content, leading to a less homogeneous distribution of particles and overall structure.

Thermal Properties of the Composite Films

Thermogravimetric analysis was performed to determine the thermal properties of the composite films. Typical thermograms are presented in Fig. 2, while the data extracted from the curves are reported in Table 2. The thermal stability, determined as the temperature to achieve 10% mass loss (T_{10}%), increases with increasing nanolignin content. The initial mass loss is due to bound water. However, it is thought that higher nanolignin content leads to retention of more bound water, making it more difficult to remove from the films.

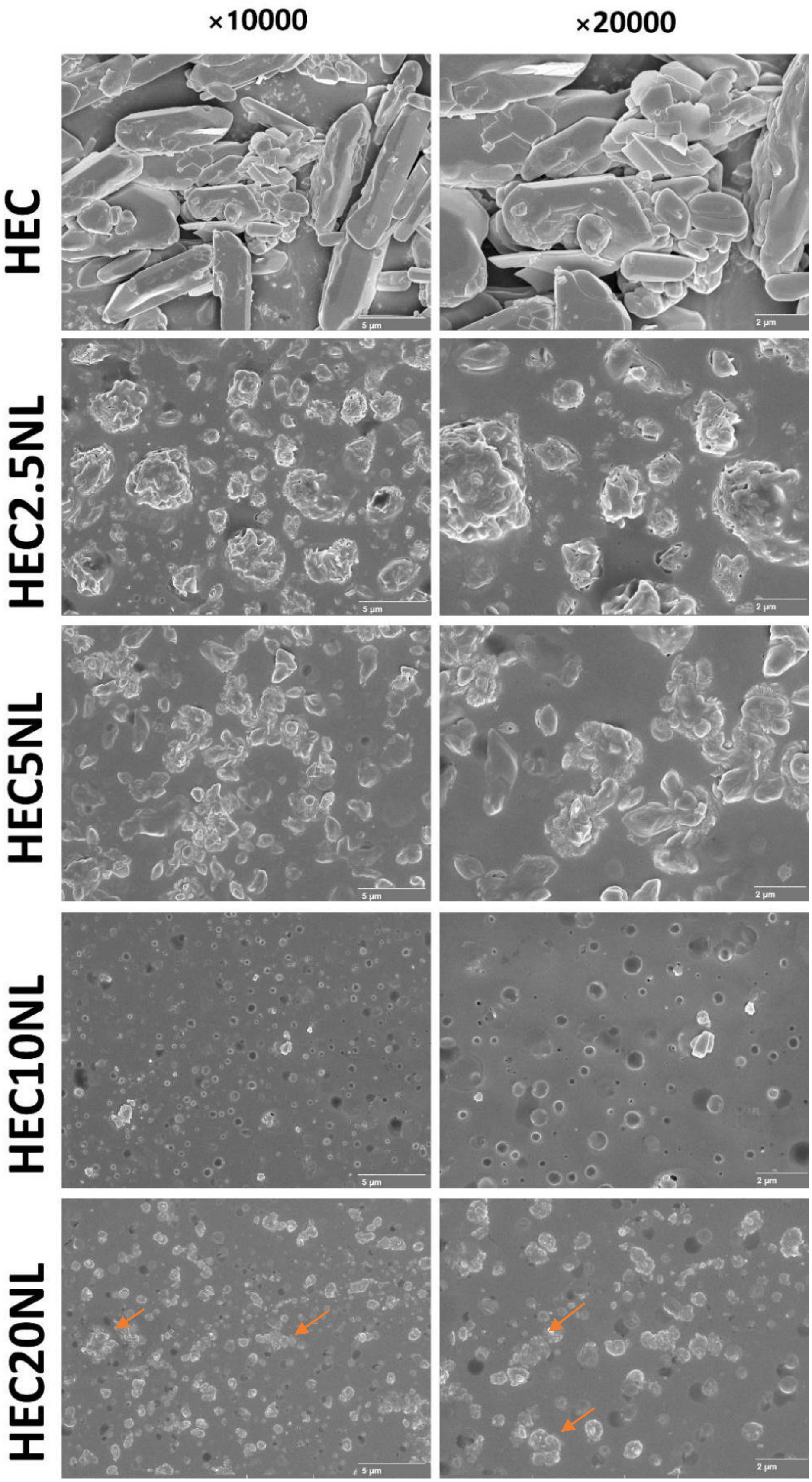

Fig. 1. SEM images of the composite films at different magnifications

The maximum mass loss temperatures (DTG peak) of the composite films were around 300 °C, and a slight decrease was observed with increasing nanolignin content. This is due to the random and complex chemical structure of lignins. While regular anhydroglucoside units are present in the structure of hydroxyethyl cellulose, several different chemical groups are present in the lignin's structure. These groups can decompose at low temperatures, leading to lower thermal stability (easier degradation) of the composite films. Zhang *et al.* (2023) also reported that the thermal degradation temperature decreased as the nanolignin content increased in pectin-nanolignin composite films. Finally, the composite films showed a decrease in char yield with increasing nanolignin concentration. The char yield is a crucial factor for combustion resistance as under combustion, the char left by the material prevents the flame from going deeper into the material and delays the combustion reactions.

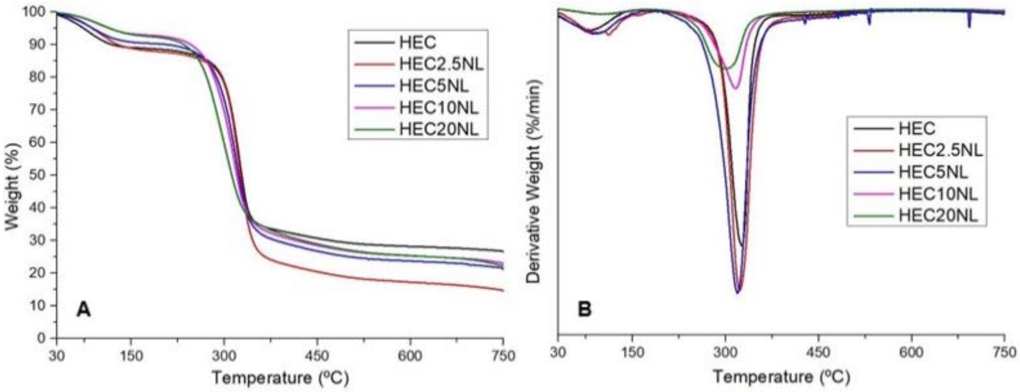

Fig. 2. Thermograms of the composite films: TGA (A) and DTG (B)

Table 2. Thermal Properties of the Composite Films

Sample	$T_{10\%}$ (°C)	DTG peak (°C)	Char Yield (%)	T_g (°C)
HEC	120	327	26.6	Not observed – 109 (Yang *et al.* 2021)
HEC2.5NL	129	322	14.6	146
HEC5NL	210	319	21.3	Not observed
HEC10NL	246	316	23.1	241
HEC20NL	235	302	22.2	262

Differential scanning calorimetry analysis was performed to determine the thermal transitions of the composite films. Figure 3 shows that the thermal transitions are not always clear to determine. It is known that amorphous samples do not show thermal transitions in a narrow temperature range and cannot be easily detected by DSC. However, when the curves are carefully examined, endothermic transitions related to the glass transition temperature (T_g) can be detected. Although the T_g of the film obtained from neat HEC could not be determined in this study, it was determined to be 109 °C in a previous study (Şen and Kahraman 2018). The results reported in Table 2 show that the T_g increased with increasing nanolignin content in the composite films. This is due to a decrease in the free volume of the composite films with increasing nanolignin content. As the free volume decreases, the mobility of the polymer chains is more restricted and T_g increases (Şen and Kahraman 2014).

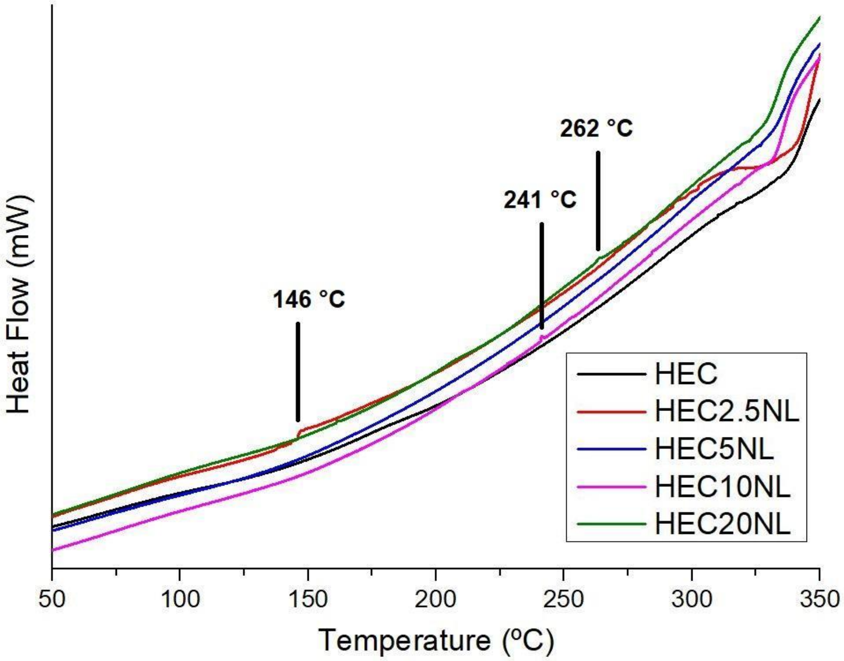

Fig. 3. DSC curves of the composite films

Mechanical Properties of the Composite Films

The mechanical properties of the samples were determined by tensile tests, and the results are presented in Table 3. The tensile strength decreases while the elongation at break increases with increasing nanolignin content. For example, the sample with 5% nanolignin (HEC5NL) has around 50% elongation at break, while the neat polymer (HEC) has only 19%. As a result, the materials are more flexible with nanolignin incorporation. However, a decrease of up to 44% was observed in the elongation at break of the composite sample containing 20% nanolignin. This decrease is probably related to nanolignin agglomeration at higher concentration. As a result, the optimum nanolignin content is 10% based on the results of Table 3. Similar trends were reported in the literature. For example, Jiang *et al.* reported that compared to pure natural rubber, a gradual decrease in tensile strength and tear strength was observed with lignin addition in the composites due to poor interfacial compatibility between the polar lignin and non-polar natural rubber (Jiang *et al.* 2013). Finally, the tensile modulus also decreases with increasing nanolignin content, indicating that the nanolignin did not reinforce the HEC matrix and mainly disrupted the polymer network and interactions, leading to lower rigidity of the composite films.

Table 3. Mechanical Properties of the Composite Films

Sample	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)
HEC	25.9 (± 0.9)	5253 (± 51)	19.4 (± 1.6)
HEC2.5NL	14.1 (± 0.8)	1358 (± 87)	40.9 (± 2.2)
HEC5NL	9.9 (± 0.4)	763 (± 31)	51.1 (± 2.3)
HEC10NL	5.1 (± 0.1)	390 (± 8)	51.5 (± 2.6)
HEC20NL	4.1 (± 0.2)	365 (± 9)	44.2 (± 0.9)

Values in parentheses are standard deviations

Wettability of the Composite Films

Contact angle measurements were performed to determine the surface hydrophobicity of the composite films. The average values for the contact angles increased from 53.2º (HEC) to 73.2º (HEC20NL), as reported in Fig. 4.

This increasing trend in surface hydrophobicity with nanolignin content can be attributed to both the hydrophobic characteristics of lignin and increasing surface roughness. Firstly, the hydrophobic characteristics of lignin directly contribute to higher film hydrophobicity (Yu *et al.* 2023). Secondly, it has been reported in the literature that the advancing contact angle of a water drop on a hydrophobic material increases with increasing surface roughness (Şen and Kahraman 2014). A study reported that the surface roughness, and thus the hydrophobicity of cotton fabrics, was significantly improved by adding lignin/metal nanoparticles (Liu *et al.* 2023). According to the authors' SEM analysis (Fig. 1), nanolignin particles were clearly visible on the surface of the composite films and would contribute to the hydrophobicity of films. Therefore, nanolignin addition significantly increased the water contact angle leading to more hydrophobic films.

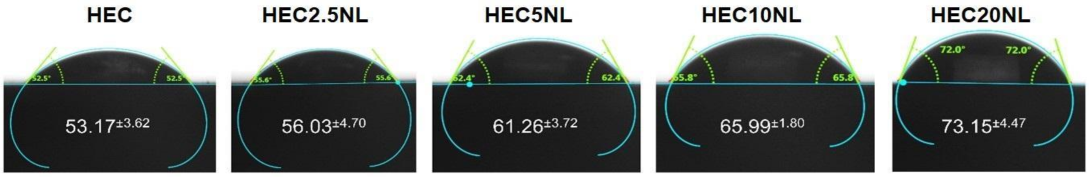

Fig. 4. Water contact angle for the composite films. All the values are expressed as an average of 10 measurements ± one standard deviation.

CONCLUSIONS

In this study, nanolignin was incorporated into hydroxyethyl cellulose (HEC) to produce biocomposite films *via* casting. From the samples produced, a wide range of characterization was performed including morphological, mechanical, surface, and thermal properties.

1. The thermogravimetric analysis (TGA) results showed that nanolignin significantly increased the initial mass loss temperatures and partially decreased the main decomposition temperatures. Additionally, it was determined that the char yield of the composite films was between 14% and 20%.
2. According to differential scanning calorimetry (DSC) results, nanolignin addition increased the glass transition temperature from 109 ºC (neat HEC) to 262 ºC (20% lignin). From the mechanical testing, the presence of nanolignin increased the flexibility of the composite films but decreased their tensile strength.
3. The optimum nanolignin content was determined to be 10% because higher concentrations decreased both the flexibility and tensile strength of the composite films. Water contact angle measurements showed that the contact angle of the composite films increased from 53º to 73º with nanolignin addition.
4. From the scanning electron micrograph (SEM) images, it was clear that some nanolignin particles were agglomerated, but the agglomerated structures had a homogeneous distribution within the samples.

5. As a result, materials being more flexible and more hydrophobic with higher glass transition temperatures were obtained with the incorporation of nanolignin. Based on the results obtained, it is believed that lignin and lignin-based nanocomposites have good opportunities for commercial use, and can be of interest in several industrial areas, especially for biopolymer-based formulations, such as pulp and paper, plastic, and agriculture, as they have a potential to contribute to a greener future.

ACKNOWLEDGMENTS

The authors would like to thank the Turkish Academy of Sciences & TÜBA for their financial support and the Biomaterials and Nanotechnology Research Group BioNanoTeam for their valuable contributions during the work.

REFERENCES CITED

Abdel-Halim, E. S. (2014). “Chemical modification of cellulose extracted from sugarcane bagasse: Preparation of hydroxyethyl cellulose,” *Arabian Journal of Chemistry* 7(3), 362-371. DOI: 10.1016/j.arabjc.2013.05.006

Abreu, H. S., Nascimento, A. M, and Maria, M. A. (1999). “Lignin structure and wood properties,” *Wood and Fiber Science* 31(4), 426-433.

Achachlouei, B. F., and Zahedi, Y. (2018). “Fabrication and characterization of CMC-based nanocomposites reinforced with sodium montmorillonite and TiO_2 nanomaterials,” *Carbohydrate Polymers* 199, 415-425. DOI: 10.1016/j.carbpol.2018.07.031

Balasubramanian, R., Kim, S. S, Lee, J., and Lee, J. (2019). “Effect of TiO_2 on highly elastic, stretchable UV protective nanocomposite films formed by using a combination of kcarrageenan, xanthan gum and gellan gum,” *International Journal of Biological Macromolecules* 123, 1020-1027. DOI: 10.1016/j.ijbiomac.2018.11.151

Candan, Z., Tozluoglu, A., Gonultas, O., Yildirim, M., Fidan, H., Alma, M. H., and Salan, T. (2022). “Nanocellulose: Sustainable biomaterial for developing novel adhesives and composites,” *Industrial Applications of Nanocellulose and Its Nanocomposites. Elsevier*, UK, pp. 49-137. DOI:10.1016/B978-0-323-89909-3.00015-8

Chen, S., Shao, Q., Hu, L., Tan, Z., and Zheng, D. (2023). “Hydrophobic and magnetic fabrication of hydroxyethyl cellulose-lignin aerogel through ultrasound enhancement for efficient oil/water separation,” *Journal of Water Process Engineering* 52, article ID 103503. DOI: 10.1016/j.jwpe.2023.103503

Dal-Bó, A. G., Laus, R., Felippe, A. C., Zanette, D., and Minatti, E. (2011). “Association of anionic surfactant mixed micelles with hydrophobically modified ethyl(hydroxyethyl)cellulose,” *Colloids and Surfaces A: Physicochemical Engineering Aspects* 380(1-3), 100-106. DOI: 10.1016/j.colsurfa.2011.02.028

Dalkilic, B., Durmaz, E., Oncul, B., and Candan, Z. (2024). “Nanosensors based on lignocellulosic materials,” *BioResources* 19(4), 6970-6974. DOI:10.15376/biores.19.4.6970-6974

Durmaz, E., Sertkaya, S., Yilmaz, H., Olgun, C., Ozcelik, O., Tozluoglu, A., and Candan, Z. (2023). “Lignocellulosic bionanomaterials for biosensor applications,” *Micromachines* 14(7), article 1450. DOI:10.3390/mi14071450

Frangville, C., Rutkevicius, M., Richter, A., Velev, O. D., Stoyanov, S. D., and Paunov, V. N. (2012). “Fabrication of environmentally biodegradable lignin nanoparticles,” *Chemphyschem* 13(18), 4235-4243. DOI: 10.1002/cphc.201200537

Gennadios, A., Weller, C. L., and Gooding, C. H. (1994). “Measurement errors in water vapor permeability of highly permeable, hydrophilic edible films,” *Journal of Food Engineering* 21(4), 395-409. DOI: 10.1016/0260-8774(94)90062-0

Gorgieva, S., and Kokol, V. (2011). “Synthesis and application of new temperature-responsive hydrogels based on carboxymethyl and hydroxyethyl cellulose derivatives for the functional finishing of cotton knitwear,” *Carbohydrate Polymers* 85(3), 664-673. DOI: 10.1016/j.carbpol.2011.03.037

Jiang, C., He, H., Jiang, H., Ma, L., and Jia, D. M. (2013). “Nano-lignin filled natural rubber composites: Preparation and characterization,” *eXPRESS Polymer Letters* 7(5), 480-493. DOI: 10.3144/expresspolymlett.2013.44

Kocaturk, E., Salan, T., Ozcelik, O., Alma, M. H., and Candan, Z. (2023). “Recent advances in lignin based biofuel production and utilization,” *Energies* 16(3). DOI: 10.3390/en16083382

Kugge, C., Craig, V. S. J., and Daicic, J. (2004). “A scanning electron microscope study of the surface structure of mineral pigments, latices and thickeners used for paper coating on non-absorbent substrates,” *Colloids and Surfaces A: Physicochemical Engineering Aspects* 238(1-3), 1-11. DOI: 10.1016/j.colsurfa.2004.02.029

Lee, J. S., Ramalingam, S., Jo, I. G., Kwon, Y. S., Bahuguna, A., Oh, Y. S., Kwon, O. J, and Kim, M. (2018). “Comparative study of the physicochemical, nutritional, and antioxidant properties of some commercial refined and non-centrifugal sugars,” *Food Research International* 109, 614-625. DOI: 10.1016/j.foodres.2018.04.047

Liao, J. J., Abd Latif, N. H., Trache, D., Brosse, N., and Hussin, M. H. (2020). “Current advancement on the isolation, characterization and application of lignin,” *International Journal of Biological Macromolecules* 162, 985-1024. DOI: 10.1016/j.ijbiomac.2020.06.168

Liu, X., Chen, X., Bian, H., Ni, S., Li, Z., Liu, N., Qin, M., and Zhang, F (2023). “Highly hydrophobic cotton fabric by *in-situ* co-deposition of lignin/metal particles for oil/water separation,” *Industrial Crops and Products* 204(Part B), Article ID 117393. DOI: 10.1016/j.indcrop.2023.117393

Liu, Y., Ahmed, S., Sameen, D. E., Wang, Y., Lu, R., Dai, J., Li, S., and Qin, W. (2021). “A review of cellulose and its derivatives in biopolymer-based for food packaging application,” *Trends in Food Science & Technology* 112, 532-546. DOI: 10.1016/j.tifs.2021.04.016

Nogueira, S. (2020). “The importance of a green marketing strategy in brand communication—M. Coutinho multi-brand car dealer case in Northern Portugal,” in: *Proceedings of the Third EBOR Conference*, Rome, Italy, pp. 351-373.

Noshirvani, N., Ghanbarzadeh, B., Mokarram, R. R., and Hashemi, M. (2017). “Novel active packaging based on carboxymethyl cellulose-chitosan-ZnO NPs nanocomposite for increasing the shelf life of bread,” *Food & Packaging Shelf Life* 11, 106-114. DOI: 10.1016/j.fpsl.2017.01.010

Patural, L., Marchal, P., Govin, A., Grosseau, P., Ruot, B., and Devès, O. (2011). "Cellulose ethers influence on water retention and consistency in cement-based mortars," *Cement and Concrete Research* 41(1), 46-55. DOI: 10.1016/j.cemconres.2010.09.004

Rajasekharan, S. K., Byun, J., and Lee, J. (2018). "Inhibitory effects of deoxynivalenol on pathogenesis of *Candida albicans*," *Journal of Applied Microbiology* 125(5), 1266-1275. DOI: 10.1111/jam.14032

Şen, F., and Kahraman, M. V. (2014). "Hybrid dual-curable cyanate ester/boron phosphate composites *via* sequential thiol-ene photopolymerization and thermal polymerization," *Progress in Organic Coatings* 77(6), 1053-1062. DOI: 10.1016/j.porgcoat.2014.03.004

Şen, F., and Kahraman, M. V. (2018). "Preparation and characterization of hybrid cationic hydroxyethyl cellulose/sodium alginate polyelectrolyte antimicrobial films," *Polymers for Advanced Technology* 29(7), 1895-1901. DOI: 10.1002/pat.4298

Şen, F., Uzunsoy, İ., and Kahraman, M. V. (2020). "Hydroxyethyl cellulose-based indicator labels to track freshness of anchovy (*Engraulis encrasicolus*)," *Color Research and Application* 45(5), 962-967. DOI: 10.1002/col.22517

Sohni, S., Hashim, R., Nidaullah, H., Lamaming, J., and Sulaiman, O. (2019). "Chitosan/nano-lignin based composite as a new sorbent for enhanced removal of dye pollution from aqueous solutions," *International Journal of Biological Macromolecules* 132, 1304-1317. DOI: 10.1016/j.ijbiomac.2019.03.151

Stojanovska, E., Kurtulus, M., Abdelgawad, A., Candan, Z., and Kilic, A. (2018). "Developing lignin-based bio-nanofibers by centrifugal spinning technique," *International Journal of Biological Macromolecules* 113, 98-105. DOI: 10.1016/j.ijbiomac.2018.02.047

Stojanovska, E., Pampal, E. S., Kilic, A., Quddus, M., and Candan, Z. (2019). "Developing and characterization of lignin-based fibrous nanocarbon electrodes for energy storage devices," *Composites Part B: Engineering* 158, 239-249. DOI: 10.1016/j.compositesb.2018.09.072

Rukmanikrishnan, B., Ramalingam, S., Rajasekharan, S. K., Lee, J., and Lee, J. (2020). "Binary and ternary sustainable composites of gellan gum, hydroxyethyl cellulose and lignin for food packaging applications: Biocompatibility, antioxidant activity, UV and water barrier properties," *International Journal of Biological Macromolecules* 153, 55-62. DOI: 10.1016/j.ijbiomac.2020.03.016

Yang, W., Ding, H., Qi, G., Li, C., Xu, P., Zheng, T., Zhu, X., Kenny, J. M., Puglia, D., and Ma, P. (2021). "Highly transparent PVA/nanolignin composite films with excellent UV shielding, antibacterial and antioxidant performance," *Reactive and Functional Polymers* 162, 1304-1317. DOI: 10.1016/j.reactfunctpolym.2021.104873

Yu, S., Wang, M., Xie, Y., Qian, W., Bai, Y., and Feng, Q. (2023). "Lignin self-assembly and auto-adhesion for hydrophobic cellulose/lignin composite film fabrication," *International Journal of Biological Macromolecules* 233, article ID 123598. DOI: 10.1016/j.ijbiomac.2023.123598

Zhang, S., Cheng, X., Fu, Q., Li, Y., Wu, P., Qiao, Y., Yan, J., Si, L., Waterhouse, G. I. N., Li, H., *et al*. (2023). “Pectin-nanolignin composite films with water resistance, UV resistance, and antibacterial activity,” *Food Hydrocolloids* 143, article ID 108783. DOI: 10.1016/j.foodhyd.2023.108783

Article submitted: June 24, 2024: Peer review completed: July 31, 2024; Revised version received: August 8, 2024; Accepted: August 9, 2024; Published: August 21, 2024. DOI: 10.15376/biores.19.4.7354-7365