International Journal of Engineering
Journal Homepage: www.ije.ir

Experimental Study on Mechanical, Thermal and Antibacterial Properties of Hybrid Nanocomposites of PLA/CNF/Ag

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PAPER INFO

Paper history:
Received 23 September 2020
Received in revised form 15 October 2020
Accepted 30 October 2020

Keywords:
Antibacterial Properties
Cellulose Nano-fiber
Poly Lactic Acid
Silver Nanoparticles
Thermal Properties

ABSTRACT

The main purpose of this study is preparation of the nanocomposite samples with synergistic properties containing the mechanical, thermal and antibacterial properties. For this purpose, the combination of cellulose nanofiber (CNF) and Ag (silver) nanoparticles were incorporated into polylactic acid (PLA) matrix by solution casting method. The CNF in constant content of 1 wt.% and Ag nanoparticles in the content of 1, 3, and 5 wt.% were incorporated into the PLA matrix. The structure and morphology of the nanocomposite samples was characterized by FE-SEM, and mechanical, antibacterial, and thermal properties of the nanocomposites were evaluated by tensile, agar disk-diffusion, and DSC tests, respectively. FE-SEM images showed the uniform dispersion of the nanoparticles within the polymer matrix. The simultaneous addition of two nanoparticles significantly raised the mechanical properties such as tensile strength and tensile modulus by 40 and 9%, respectively. However, CNF had no considerable effect on the thermal and antibacterial properties of the PLA matrix. Unlike CNF, Ag nanoparticles significantly improved the antibacterial properties of the nanocomposites against *staphylococcus aureus* and *Escherichia coli* bacteria, and enhanced the thermal stability of the PLA matrix. Ag nanoparticles improved the degree of crystallinity of PLA from 10.5% to 17.9%, and T_m from 147.8 to 153.6 °C. By incorporating 5wt.% Ag nanoparticles, the inhibition duameter increased from 20 mm to 39 mm for *staphylococcus aureus*.

***doi**: 10.5829/ije.2021.34.02b.23*

NOMENCLATURE

X_c	degree of crystallinity	ΔH_m^c	The heat of diffusion
ΔH_m	The melting enthalpy		

1. INTRODUCTION

Consuming the syntactic polymers manufactured by petrochemical industries cusses environmental damage seriously [1,2]. A considerable amount of plastic material (approx. 45%) is used for disposable packaging [3]. The use of compatible and biodegradable polymers (natural polymers) for consuming in various industries, especially for food packing, could reduce these environmental problems significantly [4]. Poly (lactic acid) (PLA) as a biodegradable polyester is a favorite environmental-friendly polymer that is used widely in the packing industry [5]. PLA has some advantages such as abundant sources of PLA, good resistance to ultraviolet, ease of processability, low toxicity, and acceptable hydrophobicity [6-8]. Notwithstanding these advantages, some defects such as poor heat resistance, high brittleness, poor disinfection properties, and low barrier properties limit the application of PLA [9,10].

The nanotechnology is a new technology which could improve the properties of various materials. Hamedi et al. [11] improved the mechanical properties of asphalt by adding nano silica. Saeid et al. [12] improved the performance of warm mix asphalt by incorporating nano

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Please cite this article as: K. Sheikh, H. Shahrajabian, Experimental Study on Mechanical, Thermal and Antibacterial Properties of Hybrid Nanocomposites of PLA/CNF/Ag, International Journal of Engineering, Transactions B: Applications Vol. 34, No. 02, (2021) 500-507

bentonite and fatty Arbocel. A useful method that is proposed by researchers to improve overcome these defects is incorporating the nano-particles into the PLA matrix to improve the properties such as mechanical properties (tensile strength, tensile modulus, etc.), barrier properties, antibacterial properties, and thermal properties [13]. Among the various nanofillers introduced into the PLA matrix, nano-cellulose has received much attention from researchers due to high availability, low price, high mechanical properties, and biodegradability [14,15]. Trifol et al. [16] compared the effect of cellulose nanofibers (CNF) and clay nanoparticles on the mechanical properties of the PLA matrix film. They observed a slight improvement in the mechanical properties of PLA/CNF film compared to PLA/clay film. Rezaei et al. [17] improved tensile strength and tensile modulus of PLA matrix film up to 32 and 19% by incorporating CNFs, respectively. Jonoobi et al. [18] increased tensile strength and tensile modulus of PLA matrix from 58 to 71 MPa, and 2.9 to 3.6 GPa, respectively, by incorporated CNF content of 1 to 5 wt.%. Arrieta et al. [19] showed that introduce the CNFs into PLA matrix composite increased the transition glass temperature (Tg), and concluded that CNFs restrict the mobility of the polymer chains. Frone et al. [20] increased the degree of crystallinity of the PLA matrix from 39% to 47% by adding the CNFs in the content of 2.5 wt.%.

As mentioned above, one of the major applications of PLA is the food packing industry for meat products, fruits, vegetables, and beverage bottles [21,22]; therefore, the antibacterial property is an essential characteristic for PLA. Among the nano-particles, silver (Ag), with its strong antibacterial properties, can kill the various microorganisms such as *E. coli*, *Staphylococcus aureus*, and *Neisseria gonorrhoeae* [23,24]. Other important properties of Ag include acceptable antiviral effects [25], antiparasitic activities [26], and antifungal properties [27]. Manikandan et al. [28] coated nanoporous Sodium Alginate (SA)/ Poly Vinyl Alcohol (PVA) composite scaffold with Ag nanoparticles for antibacterial activity. Maroti et al. [29] investigated the effect of Ag nanoparticles on thermal and antibacterial properties of PLA for use in 3D print of biomedical devices. Their results showed that PLA/Ag nanocomposites are suitable for biomedical devices. Gan et al. [30] showed that the combined effect of Ag and carbon nanotube (CNT) could improve the antibacterial properties, thermal stability and, tensile strength of the PLA matrix. Fan et al. [31] concluded that the migration of Ag nanoparticles in PLA/Ag nanocomposites increased by Ag content. Growing the migration time made the film rougher, and increased the crystallization temperature of the nanocomposite films.

It is also found that the combination of two nanofillers into the polymer matrix could merge the properties of the both of nanofillers to matrix [27]. Therefore, the co-incorporation of CNFs and Ag nanoparticles into PLA matrix is potential to manufacture the nanocomposite samples with synergistic properties containing mechanical, thermal and antibacterial properties. Since no research has been done to investigate the co-incorporation of CNF and Ag nanoparticles into PLA matrix, in this study, PLA matrix nanocomposites reinforced by CNFs and Ag nanoparticles were prepared by the solution casting method. CNFs in the content of 1 wt.% were added into PLA matrix to improve mechanical properties, and Ag nanoparticles in the content of 1, 3, and 5 wt.% were added to improve anti-bacterial properties of the PLA matrix. The dispersion of nano-fillers in the PLA matrix was investigated by field emission scanning electron microscopy (FE-SEM). Tensile, antibacterial, and differential scanning calorimetry (DSC) tests were done to evaluate mechanical, antibacterial, and thermal properties of the nanocomposite samples.

2. MATERIALS AND METHODS

2. 1. Materials Bio-flex®F 6510 commercial grade of poly(lactic acid) (PLA) with molecular weight (Mw) of 197000 g/mol, the density of 1.25 g/cm^3, and melting temperature point of 160 °C was supplied from Fkur GmbH company. Cellulose nanofiber (CNF) gel (3 wt.%) was prepared from Nano-Novin-Polymer Co. (Iran). Ag nanoparticles with the density of 10.5 g/cm^3, an average diameter of 20 nm, and purity of 99.9% were purchased from Nanosany Co. (Iran). The analytical grade of chloroform (as a solvent), methanol, acetone, and ethanol were purchased from Arman Sina Co. (Iran).

2. 2. Preparation The PLA films and nanocomposite films are prepared by the solution casting method. For the preparation of PLA film, 5 g of PLA was added into 100 ml chloroform, and was stirred by a mechanical a stirrer at rotary speed of 2000 rpm for 6 h, at ambient temperature. The prepared solution was poured in the mold, and was dried at ambient temperature for 24 h to remove the solvent. To remove the solvent completely, the samples were dried in a vacuum oven for 7 days at a temperature of 45 °C. In order to add CNF into PLA solution, the CNF gel in a certain amount was suspended in solvents of methanol, ethanol, acetone, and chloroform respectively and finally centrifuged at a rotary speed of 8000 rpm for 10 min. It should be noted that for each of the mentioned solvents, this procedure was repeated five times. Finally, the CNF content for all the nanocomposite samples was the same in the content of 1 wt.% (because in some literature this content was optimum content [32]). The CNF in the content of 1 wt.% and Ag nanoparticles in content of 1, 3, and 5 wt.% were added into PLA solution, followed by stirring by mechanical stirrer at 2000 rpm for 2 h. For better dispersion of nano-fillers into PLA, the solution was homogenized by the ultrasonic homogenizer model of UP400St (manufactured in Hielscher Co., Germany) for 45 min (Three 15 min intervals). Drying the nanocomposite films was exactly like drying the PLA films. The composition of the samples has been shown in Table 1.

2. 3. Characterization Field emission-scanning electron microscopy (FE-SEM) was used to investigate the morphology of the samples by the MIRA3 TESCAN (manufactured by Tescan company in the Czech Republic). Fourier transform infrared spectra (FTIR) was conducted using the Tensor 27 Bruker instrument with a resolution of 4 cm^{-1}. The mechanical properties of the nanocomposite films such as tensile strength, tensile modulus, and elongation were measured by tensile machine test of H25KS manufactured by Hounsfield Co. at ambient temperature with the speed of 20 mm/min. For each sample, 5 tests was done on the film samples in a dimension of 160 mm×20mm×0.1 mm. The thermal analysis of the samples was evaluated by differential scanning calorimetry (DSC) test using a SANAF DSC analyzer. The samples heated under a nitrogen atmosphere from 20 °C to 200 °C, and cooled to 40 °C with a melting and cooling rate of 10 °C/min. The antibacterial test was investigated by the agar disk-diffusion method. The standard strains of *staphylococcus aureus* (ATCC: 25923) and *E. coli* (ATCC: 25922) as the target bacteria were selected to evaluate the antibacterial properties of the film samples. The disk samples in diameter of 20 mm were cut from nanocomposite films. Both target bacteria were cultivated in the culture media of the blood agar (BA) and tryptic soy broth (TSB). The bacteria suspension of 0.5 MakFarland (1.5×108 CFU/ml) was sprayed over the surface of culture dishes. Then the disk samples were located on the bacterial agar plate, and the dishes were incubated at 37 °C for 24 h. The antibacterial properties of the composite samples were determined by measuring the diameter of the inhibition growth zone by the caliper.

TABLE 1. The composition of the samples

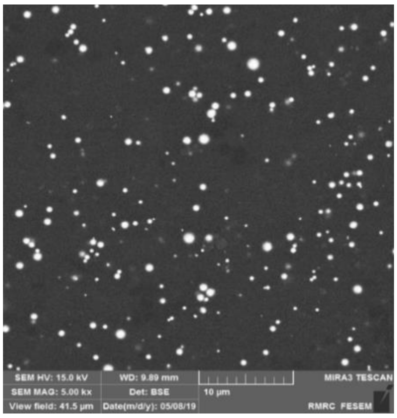
Sample	CNF (wt.%)	Ag (wt.%)	PLA (wt.%)
PLA	-	-	100
PCNF	1	-	99
PCNAg1	1	1	98
PCNAg3	1	3	96
PCNAg5	1	5	94

3. RESULTS

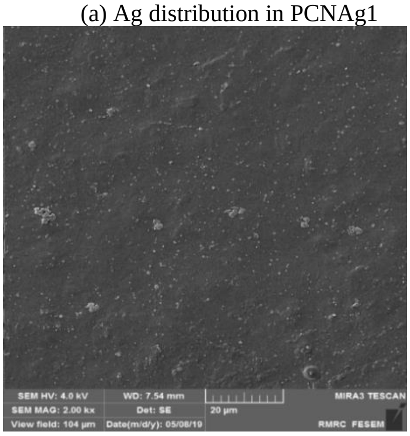
3. 1. Morphology The morphology of the samples was investigated by FE-SEM. Figure 1 shows FE-SEM images of the fracture surface of PCNF, PCNAg1and PCNAg3 nanocomposites. The distribution of the nanoparticles (cellulose and Ag) in PCNAg1 and PCNAg3 has been shown in Figures 1(a), and 1(b), respectively. One of the most important factors affecting the properties of the polymer matrix nanocomposites is the distribution of the nanofillers in the polymer matrix. Uniform dispersion of nanoparticles within the polymer matrix can improve the mechanical and physical properties of the matrix, but on the contrary, the agglomeration of the nanoparticles reduces the properties. Figure 1(a) shows the uniform dispersion of Ag nanoparticles within the PLA matrix in the PCNAg1 sample (the bright points are the Ag nanoparticles). The dispersion of nanoparticles (Ag and cellulose) within the polymer matrix in the PCNAg3 sample has been shown in Figure 1(b). As shown in Figure 1(b), the nanoparticles were almost uniformly dispersed within the matrix. In PCNAg3, several agglomerates with a dimension lower than 5 μm can be seen. By increasing the Ag nanoparticle content in PCNAg5, the agglomeration sites increased, and the surface is more rough compared to PCNAg3, as shown in Figure 1(c). Figures 1(d), and 1(e) illustrate the distribution of nanoparticles in nanoscale. As shown in Figure 1, the nanoparticles the particles are well distributed on a nanoscale.

3. 2. ATR-FTIR The interaction between nanofillers (Ag and cellulose nanoparticles) can be observed by ATR-FTIR measurement. FTIR spectra of neat PLA and PLA/CNF/Ag nanocomposite samples are shown in Figure 2. It can be seen that the neat PLA and nanocomposite samples have the sharp peaks at 1000-1200 cm^{-1} and 1750 cm^{-1}, belonging to the stretching vibration of C-O-C and C=O of PLA. A sharp peak at 1749 cm^{-1} (crystalline sensitive band) is observed, which is due to the C=O ester group. The other peaks were positioned at 1452 cm^{-1} and 1363 cm^{-1} due to the bending vibration of CH_3 (alkyl group). It is also observed from the FTIR spectrum that nanocomposite samples have a small peak at 1253 cm^{-1} (C-O-C), a stretching vibration in the crystalline phase, while in pure PLA, this peak is negligible. This indicates that the nanofillers have an effect on the characteristic band intensities.

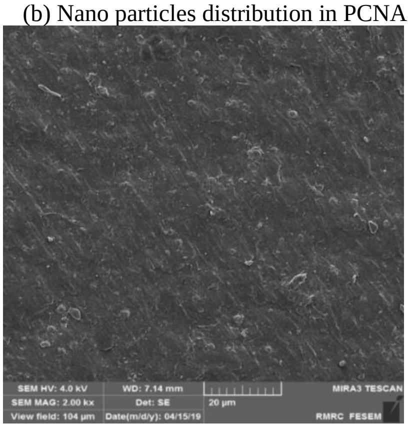
3. 3. Mechanical Properties The impact of nanoparticles (cellulose and Ag) on the mechanical properties of the pure PLA and nanocomposite samples has been investigated using a tensile test. Figure 3 shows the stress-strain curve of PLA, PLA/CNF, and



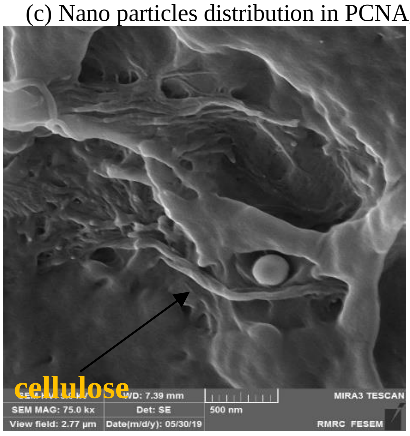
(a) Ag distribution in PCNAg1



(b) Nano particles distribution in PCNAg3



(c) Nano particles distribution in PCNAg5



(d) High magnification image of PCNF

(e) High magnification image of PCNAg1

Figure 1. FE-SEM images of the samples

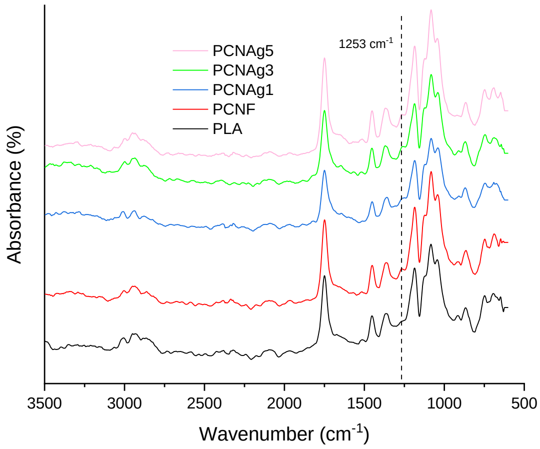


Figure 2. ATR- FTIR spectra of PLA/CNF/Ag nanocomposites

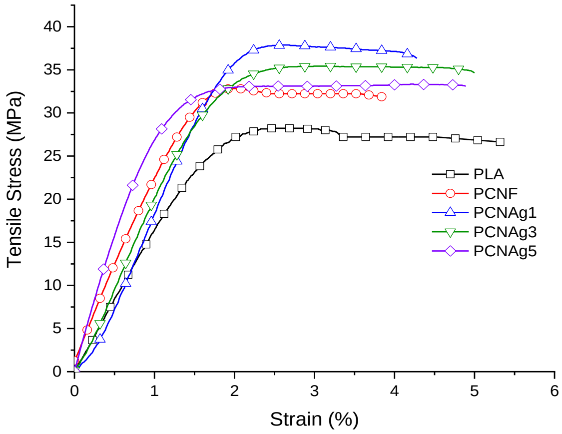


Figure 3. Stress-strain curve for PLA, PLA/CNF, PLA/CNF/Ag nanocomposite films.

PLA/CNF/Ag nanocomposites. Based on the results presented in Table 2, the neat PLA has a tensile strength of 27.1 MPa with elongation (strain at break point) of ~ 6%, and tensile modulus (Young modulus) of 1856 MPa. The results show that adding the CNF and Ag nanoparticles improve the tensile properties of the PLA matrix, indicating a strong interaction between nano-fillers and PLA matrix. The film containing 1 wt% of CNF (PCNF), the tensile strength, Young's modulus increased by 20 and 21%, respectively, and elongation decreased by 34%. The Ag nanoparticles in the content of 1, 3, and 5 wt.% was incorporated into PLA/CNF nanocomposites to investigate the effect of Ag nanoparticles on the tensile properties of nanocomposite films. The results indicate that the adding Ag nanoparticles decreased Young's modulus of PLA/CNF films from 2259 to 2010 MPa, but increased the tensile strength from 32.5 to 38 MPa. The simultaneous increase of tensile strength and elongation after adding the Ag nanoparticles can be due to very small size of nanoparticles that is close to polymer chains and these nanoparticles act as a cross between chains. By increasing the nanoparticle content from 1 to 5 wt.% the Young's modulus increased from 2010 to 2593 MPa, and tensile strength decreased from 38 to 33.2 MPa. A decrease in tensile strength by Ag nanoparticles content can be due to nanoparticles aggregation in higher-level loading of nanoparticles. The agglomeration sites of the Ag nanoparticles can acts as a holes in the polymer matrix and therefore, has negative effect on mechanical and thermal properties of PLA. On the other hand, agglomeration decrease the adhesion between nanoparticles and the polymer matrix.

3. 4. Thermal Properties

The thermal behavior of pure PLA and PLA/CNF/Ag nanocomposite films was investigated by DSC analysis. The melting and cooling curves of the samples has been shown in Figures 4 and 5, respectively, with the results such as glass transition temperature (T_g), melting point temperature (T_m), crystallization temperature (T_c), and degree of crystallinity (χ) shown in Table 3. The degree of crystallinity was calculated through Equation (1):

$$X_c(\%) = \frac{\Delta H_m}{\Delta H_m^c} \times 100 \qquad (1)$$

TABLE 2. Tensile properties of PLA, PLA/CNF, PLA/CNF/Ag nanocomposite films

Sample	Young's modulus (MPa)	Tensile strength (MPa)	Elongation (%)
PLA	1856±210	27.1±2.3	5.81±0.31
PCNF	2259±340	32.5±4.4	3.84±0.22
PCNAg1	2010±150	38.0±3.7	4.28±0.33
PCNAg3	2119±240	35.5±5.2	5.01±0.43
PCNAg5	2593±310	33.2±3.7	4.75±0.55

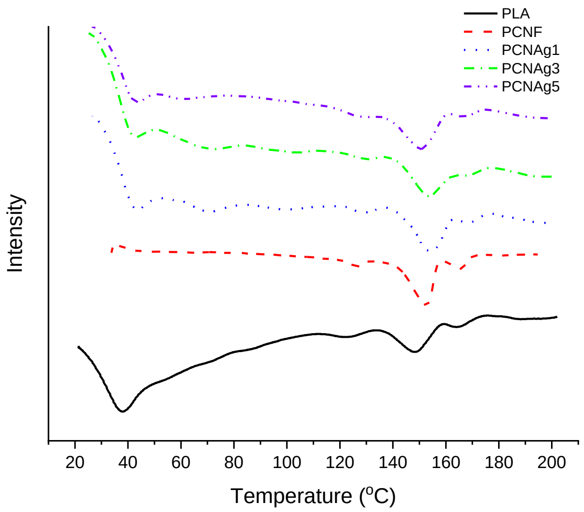


Figure 4. DSC melting scans for the samples

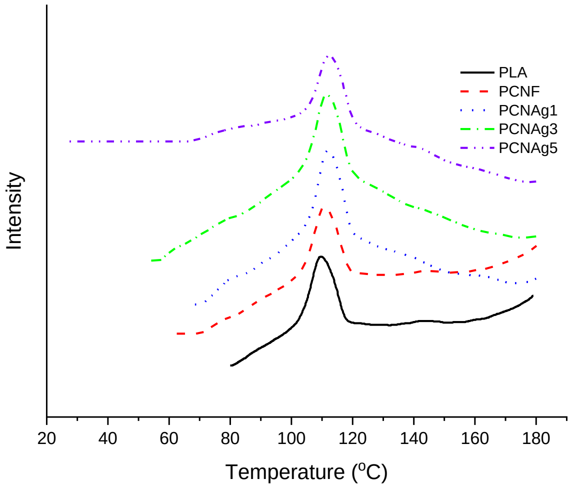


Figure 5. DSC cooling scans for the samples.

TABLE 3. The DSC data for the film samples

Sample	T_g (°C)	T_m (°C)	T_c (°C)	ΔH_m (Jg^{-1})	χ (%)
PLA	48.2	147.8	111	8	10.5
PCNF	47.9	149.7	112.1	7.61	9.98
PCNFAg1	52.5	153.6	112.8	11.1	15
PCNFAg3	51.1	153.2	112.9	12.6	16.5
PCNFAg5	51	150.9	112.7	13.7	17.9

where ΔH_m^c with value of 93 J/g is the heat of diffusion for 100% crystallite PLA.

The data of the DSC test were presented in Table 3 shows that incorporating the CNF into the PLA matrix does not aeffect on T_g, T_m, T_c, and χ of the PLA matrix. The addition of Ag nanoparticles in the content of 1 wt.% increased Tg of PLA matrix from 48.2 to 52.5 °C. The T_g

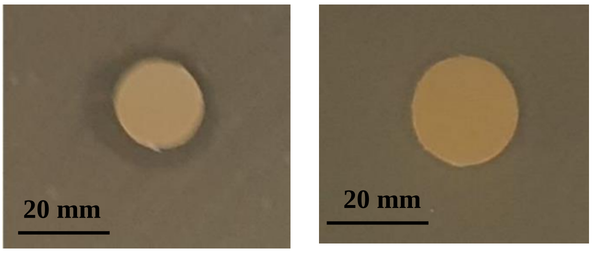
has related to the mobility of the polymer chains of the amorphous phase in the PLA matrix. The increase of T_g with the introducing the Ag nanoparticles shows that Ag nanoparticles have been able to hinder effectively mobility of the molecular chains. Enhancement of T_g by incorporating the Ag nanoparticles could be due to the strong interactions between nanoparticles and PLA matrix [33]. The increase in T_g by using the nanoparticles in the PLA matrix was reported in the other literature. Adding the hydroxyapatite and GO nanoparticles increases the T_g of PLA [34, 35]. The further increase of Ag content has no considerable effect on T_g. The melting point temperature of the PCNAg1 containing 1 wt.% of Ag is 153.6 °C, which is 5.8 °C higher than pure PLA. The amount of T_m depends on the thickness of lamellae, so that the T_m increases by the lamellae thickness. Therefore, it can be concluded that introducing the Ag nanoparticles has increased the lamellae thickness in the crystalline phase. This increase in the melting point temperature of PLA has been observed in other researches [35]. By increasing the Ag content up to 3 wt.%, the considerable change in T_m was not observed, however by further increase of Ag nanoparticles to 5 wt.%, T_m decreased to 150.9 °C, probably due to nanoparticles agglomeration. The results presented in Tabl 3 show that the degree of crystallinity (χ) for pure PLA and PCNF is 10.5 and 9.98%, respectively, indicating that CNF is almost ineffective on χ. Unlike the PCNF sample, the χ for the PCNAg1, PCNAg3, and PCNAg5 increased up to 15, 16.5, and 17.9%, respectively. This increase in χ of the PLA matrix indicates that the Ag nanoparticles are useful to provide more nucleation sites. Frone et al. [20] observed that 2.5 wt.% of nano-cellulose increase the degree of crystallinity of PLA from 39 to 47%. Mukherjee et al. [36] improved the degree of crystallinity of PLA by 4% by adding 2.5 wt.% of microcrystal cellulose.

3. 5. Anti-bacterial Properties

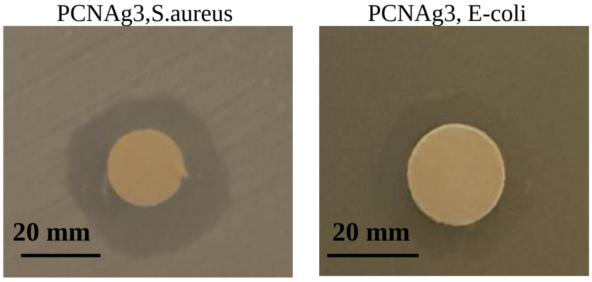
The antibacterial properties of PLA and PLA/CNF/Ag nanocomposite films against the positive strain of staphylococcus aureus and negative strain of *E. coli* were investigated by agar disk diffusion method. For this purpose, the round samples with 20 mm in diameter were separated from the films, and then placed on the Mueller Hinton Agar (MHA) plate on which negative and positive strains have been cultivated. Figure 6 shows the agar plates of *E. coli* and *staphylococcus aureus* treated with the prepared nanocomposites for 24 h. The samples of pure PLA, PCNF, and PCNAg1 are not shown in Figure 6, because, as anticipated, they did not have any antibacterial activity within the tested time (in the PCNAg1 sample, Ag nanoparticles content was insufficient). When Ag nanoparticles in the content of 3 and 5 wt.% were added into PLA, the PLA/CNF/Ag nanocomposite samples showed significant antibacterial performances. The diameter of the inhibition growth zone around the samples is a suitable criterion to estimate antibacterial properties of the nanocomposite samples. Table 4 presents the diameter of inhibition growth zone of nanocomposites. It could be clearly seen in Figure 6 and Table 4 that by increasing the Ag nanoparticles content from 3 to 5 wt.% the inhibition growth diameter increased from 23 to 34 mm for *E. coli*, and from 27 to 39 mm for *Staphylococcus aureus*, indicating a promising antibacterial activity of the nanocomposite samples. By comparing the results presented in Table 4 for two different strains, it can be concluded that Ag nanoparticles have better prevented the growth of staphylococcus aureus strains. The Ag nanoparticles may accumulate in the membrance of bacteria cytoplasmic to increase the permeability of the bacteria considerably, and as a result the cells were killed [37].

4. CONCLUSION

In this study, PLA/CNF/Ag nanocomposite films with constant content of CNF (1 wt.%), and different content of Ag nanoparticles were prepared by solution casting method. The mechanical test showed that the simultaneous addition of CNF and Ag nanoparticles



PCNAg3,S.aureus PCNAg3, E-coli



PCNAg5,S. aureus PCNAg5, E-coli

Figure 6. Antibacterial properties of PLA/CNF/Ag composites

TABLE 2. The diameter of inhibition growth zone of the samples

Sample Bacteria	PCNAg3	PCNAg5
E-coli	23 mm	34 mm
Staphylococcus aureus	27 mm	39mm

could improve the mechanical properties of nanocomposite samples compared with neat PLA samples. The tensile strength and tensile modulus increased by 40 and 39% for PCNAg1 and PCNAg5, respectively. CNF had no considerable effect on the thermal properties of the PLA matrix, but incorporating Ag nanoparticles increased T_g, T_m, and degree of crystallinity of the PLA matrix. T_m and T_g increased from 147.8 to 153.6 °C, and 48.2 to 52.5 °C, respectively. The degree of crystallinity of the PLA was increased from 10.5 to 17.9% by incorporating 5 wt.% Ag nanoparticles. Moreover, the addition of Ag nanoparticles in the content of 3 and 5 wt.% promoted the antibacterial properties of the nanocomposite samples against *staphylococcus aureus* and *E. coli*. When 5 wt.% Ag nanoparticles was added into PLA the inhibition diameter increased from 20 to 34 mm for *E. coli* and from 20 to 39 mm for *staphylococcus aureus*.

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Persian Abstract

چکیده

در این پژوهش نانوکامپوزیت پلیمر با زمینه پلی لاکتیک اسید و تقویت کننده‌های نانوذرات نقره و نانوالیاف سلولوز به روش ریخته‌گری حلال تهیه شد. مقدار نانوسلولوز ثابت و به میزان ۱ درصد وزنی در نظر گرفته شد و مقدار نانوذرات نقره به میزان ۱، ۳ و ۵ درصد وزنی به زمینه پلیمری اضافه شدند. مورفولوژی سطح شکست نمونه‌ها توسط FE-SEM آنالیز شده و خواص مکانیکی، حرارتی و آنتی باکتریال نمونه‌ها به ترتیب توسط آزمون کشش، آزمون کالریمتری روبشی تفاضلی و نفوذ دیسک آگار ارزیابی شدند. تصاویر FE-SEM نشان داد که نانوذرات بصورت نسبتاً همگن و یکنواخت در زمینه پلیمری پراکنده شده‌اند. اضافه کردن همزمان دو نانوذره به زمینه پلیمری باعث افزایش خواص مکانیکی از قبیل استحکام کششی و مدول کششی به مقدار به ترتیب ۴۰٪ و ۹٪ شد، اما نانو الیاف سلولوز تاثیر قابل توجهی بر خواص حرارتی پلی لاکتیک اسید نداشت. برخلاف نانوالیاف سلولوز، نانوذرات نقره پایداری حرارتی زمینه پلیمری را بهبود داده و بطور قابل توجهی خواص آنتی باکتریال پلی لاکتیک اسید را در برابر سویه‌های ای کویل و استافیلوکوس افزایش داد.