



Characterization, Thermal and Antimicrobial Properties of Hybrid Cellulose Nanocomposite Films with *in-Situ* Generated Copper Nanoparticles in *Tamarindus indica* Nut Powder

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Abstract

Hybrid cellulose nanocomposite films (HCNFs) were fabricated using cellulose from cotton linters as the matrix, and varying loadings (5 to 25wt.%) of modified tamarind nut powder (TNP) with in situ generated copper nanoparticles as the reinforcing fillers. These hybrid composite films were characterized by X-ray diffraction (XRD), Fourier transform infrared (FTIR) spectroscopy, thermogravimetric analysis (TGA), and antibacterial tests. The inclusion of modified tamarind nut powder improved the crystallinity of the hybrid nanocomposites by 13%. From the FTIR analysis, it could be ascertained that there was no notable change in the chemical composition of the matrix with the amendment of the filler. Further, the shift in peak intensity with increasing concentration of the filler shows the formation of hydrogen bond between the filler and the matrix. The TGA analysis revealed that the degradation temperature of hybrid nanocomposites increased until HCNFs with 15wt.% modified tamarind nut powder. The hybrid nanocomposites exhibited better antibacterial properties against all the pathogens used in the study. These hybrid nanocomposites were found to have superior antibacterial activity, thermal resistance, and improvement in crystallinity. Thus, these bio-based materials can be used to replace the existing packaging materials in the food packaging and help to reduce the impact of conventional plastics on the environment.

Keywords Hybrid nanocomposite films · Cellulose · Modified tamarind nut powder · Antimicrobial properties

Introduction

In the recent years development of materials based on plastics is happening in almost all industries in order to reduce the weight of the components. However, another debate is

going on to reduce the usage of petroleum-based plastic materials and the use of bio-based polymeric materials. In this case, many countries have formulated stringent norms to reduce plastic pollution, which is growing day by day and causing serious effects on human beings and other living

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organisms both on land and water [1]. Usage of polymer-based food packaging materials has been on a constant rise in the last three decades due to the increase in the production and consumption of processed foods. However, the synthetic polymers used in such applications pollute the ecosystem after their disposal. All the above factors paved the way for fully biodegradable food packaging materials made from the bio-polymers and bio-fillers. Biodegradable polymers can be a feasible solution to these problems since they are produced from renewable sources like cellulose, starch, polysaccharides, proteins etc. Biodegradable polymers used in such applications include polylactic acid (PLA), polypropylene carbonate (PPC), Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), cellulose etc. [2]. Even though biopolymers provide various advantages, they lack in certain functional properties such as mechanical, thermal stability, and they are also slightly expensive. Hence, inexpensive fillers are reinforced to enhance the properties of the ensuing composites.

In the case of bio-fillers, municipal solid wastes such as spent coffee bean powder [3], banana peel powder [4], tamarind nut powder (TNP) have been reported earlier [5, 6]. Preserving the processed food against contamination is inevitable for the packing bags and containers. It has been identified from literature that incorporation of the metal nanoparticles into the leaf extracts from various plants such as Terminalia catappa/cellulose [7], Ocimum sanctum/cellulose [8], Cassia alata/cellulose [9] enhances all the above said properties. In this study, TNP filler modified by the incorporation of copper nanoparticles (CuNPs) was used as reinforcement. This was mainly due to their good antibacterial activity [10].

TNP, an agro-by-product obtained after the extraction of tamarind pulp, has been used in the processing of paper, jute, and textiles. Gelling characteristics of TNP also make them prospective fillers in food packaging and pharmaceutical industries [11]. Cellulose is an organic compound used as matrix due to its high crystallinity and ability to form transparent film [9]. The effect of modified TNP/CuNPs filler wt.% on the dispersion characteristic, thermal, crystalline, and antimicrobial properties of the hybrid cellulose nanocomposite films (HCNFs) was investigated.

Materials and Methods

Materials

The matrix material cotton linter in the form of sheets was supplied by Hubei Chemical Co. Ltd., China. Lithium hydroxide, urea, and ethyl alcohol were supplied by Ganapathi Chemicals, Tamil Nadu, India. $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ was obtained from SD Fine Chemicals, Mumbai, India. The filler

material TNP was purchased from SV Sri Ganga Ayurveda, Hyderabad. The filler was sieved, and then the fillers with particle size less than $40 \mu\text{m}$ were used. This TNP filler was then dried overnight using a hot air oven at 100°C . The pathogens such as *E.coli 1652*, *P.aeruginosa 2453*, *S.aureus 96*, *B.licheniformis 75527* were purchased from IMTECH, Chandigarh, India for conducting the antibacterial test.

Fabrication of Cellulose/Modified TNP Hybrid Nanocomposites

Copper nanoparticles were in-situ generated into the TNP filler by the hydrothermal method [10] to form a modified TNP filler. The cellulose matrix solution was prepared by dissolving the cotton linters into a solution containing 8% of Lithium hydroxide (LiOH) and 15% of Urea at -12°C [3]. Modified TNP filler with varying weight percentages (5–25) was added to the matrix. In order to have a uniform distribution of the fillers in the matrix, the solution was continuously stirred for about 20 min. The resulting solutions were cast in different glass plates and were dipped in ethyl alcohol bath to form hybrid nanocomposite films. The nanocomposites were then rinsed carefully in deionized water for the removal of unwanted materials. Subsequently, the wet nanocomposite films were air-dried and stored in a desiccator before testing.

Characterization of Cellulose/Modified TNP Hybrid Nanocomposites

FTIR Analysis

The FTIR spectrometer (Bruker, Richmond Scientific Ltd, Great Britain) was used in the acquisition of the hybrid cellulose nanocomposite film spectra. A total of 32 scans at a resolution of 4 cm^{-1} and ranging from 4000 to 500 cm^{-1} were used to measure the spectra of the nanocomposite films.

XRD Analysis

Crystalline changes in the HCNFs with various filler wt.% was evaluated using the XRD technique. HCNFs with various modified TNP filler loading were scanned in the 2 Theta range of 10 – 80° at a rate of $4^\circ/\text{min}$. The crystallinity index (CI) was calculated using the Segal empirical method [12] as depicted in Eq. (1):

$$\text{CI} (\%) = \left(\frac{I_{002} - I_{\text{am}}}{I_{002}} \right) * 100 \quad (1)$$

where I_{002} and I_{am} intensity peak of the crystalline and amorphous region, respectively.

Thermogravimetric Analysis

The thermal degradation behavior of the HCNFs was analyzed in the temperature range of between 25 °C and 600 °C using a thermogravimetric analyzer (Mettler Toledo/ TGA/ DSC 3⁺ Series). The heating rate and the nitrogen gas flow rate were selected as 10 °C min⁻¹ and 60 mL min⁻¹, respectively.

Antimicrobial Analysis

Bacterial resistance of the HCNFs was analyzed by the diffusion method [4]. For this, pathogens such as *E.coli* 1652, *P.aeruginosa* 2453, *S.aureus* 96, *B.licheniformis* 75527 were used. The inhibition of the bacteria through the clear zones surrounded on the 5 mm diameter samples were used to assess the bacterial resistance of the HCNFs. The test was carried out with the samples kept in an inoculated medium and incubated at 37 °C for 2 days.

Results and Discussion

FTIR Analysis

Chemical groups and the interaction between the cellulose matrix and the modified TNP were determined from the FTIR analysis. It can be inferred from Fig. 1a that the HCNFs have a similar chemical group and some additional peaks.

It can be found that the CuNPs did not change the chemical composition of the matrix. Therefore, electrostatic forces might have held the CuNPs in the nanocomposite films [13]. FTIR spectra of the cellulose matrix, TNP/CuNPs filler,

and HCNFs with 5wt.% and 25wt.% fillers are presented in Fig. 1b. The intense broad peak between 3000 cm⁻¹ and 3700 cm⁻¹ indicates the O–H stretching and intermolecular hydrogen bonding groups in the cellulose matrix, modified TNP and nanocomposite films [14–18]. The peaks became broader for the HCNFs in the wave number range of 3000–3700 cm⁻¹ with an increase in the filler loading. This might be due to the strong interaction between (a) the OH- and reduced Cu²⁺ ions released from the CuNPs and (b) CuNPs with the TNP filler [6].

The intense peaks between 1630 and 1650 cm⁻¹ as shown in Fig. 1b and Table 1 correspond to the O–H group. The band stretching at 2895 cm⁻¹ was due to the presence of C–H stretching in methyl and methylene groups [19], and the band at 1216 cm⁻¹ corresponds to the –COO of hemi-celluloses [4]. On the other hand, the band at 1018 cm⁻¹ was due to C–O stretching of primary alcohol. The intensity peaks around 894 cm⁻¹, 1157 cm⁻¹, 1313 cm⁻¹, 1369 cm⁻¹, and 1421 cm⁻¹ were ascribed to the C–H rocking vibrations, C–O–C asymmetric valence vibration, C–H₂ rocking vibration, C–H₂ deformation vibration, and in the form of cellulose, respectively, of the carbohydrates [14]. The presence of the lignocellulosic, methyl and methylene groups in the cellulose matrix and hybrid nanocomposites were evident from the presence of characteristic peaks discussed above.

From Fig. 1b, it can be noticed that the intensity of the main peaks of the HCNFs increased with increasing filler content. Besides that, shifting of bands can be seen in the spectra of cellulose matrix and the HCNFs such as 894–896 cm⁻¹, 1014–1018 cm⁻¹, 1157–1155 cm⁻¹, 1421–1419 cm⁻¹, 1643–1645 cm⁻¹, 2160–2164 cm⁻¹, and 2890–2894 cm⁻¹ that the carboxyl and hydroxyl of the TNP filler had influence in the synthesis of the CuNPs. Further, the band located at 3324 cm⁻¹ had shifted to 3340 cm⁻¹,

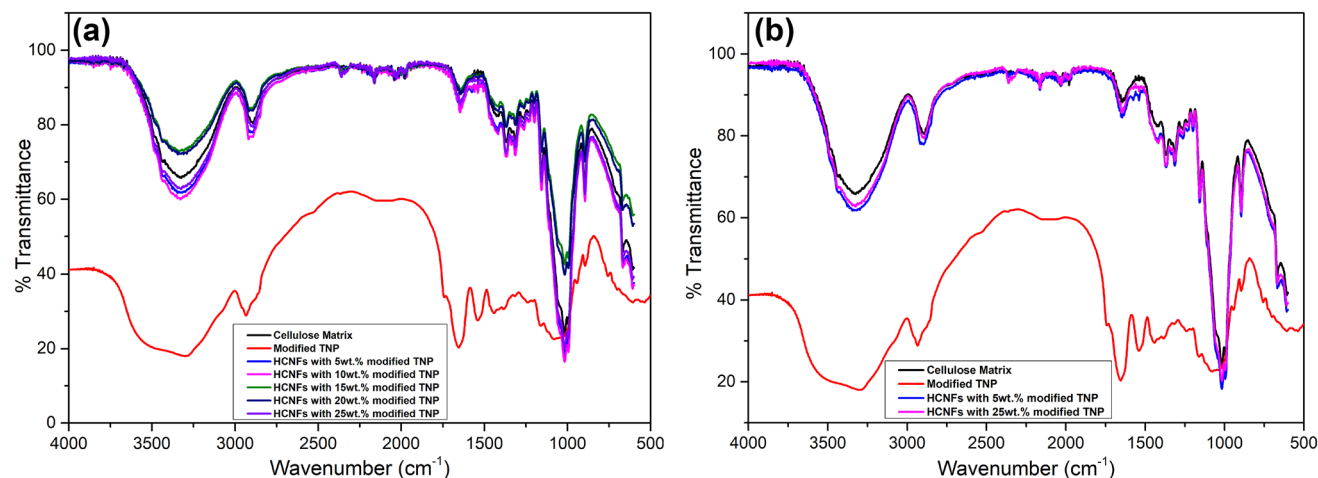


Fig. 1 a. FTIR spectra of cellulose matrix, modified TNP, HCNFs overlapped. b. FTIR spectra of cellulose matrix, modified TNP, HCNFs with 5wt.% and 25wt.% modified TNP fillers

Table 1 Details of peaks observed from the FTIR spectra of the cellulose matrix and HCNFs

Peak assignment	Wavenumber (cm ⁻¹)	Structural polymer	References
Cellulose	894	C–H rocking vibrations	[14, 20, 21]
Cellulose	1157	C–O–C asymmetric valence vibration	[16, 19]
Cellulose	1313	C–H ₂ rocking vibration	[15, 21]
Cellulose	1369	C–H ₂ deformation vibration	[17, 21]
Cellulose	1421	–	[17, 21]
Cellulose and hemicellulose	1018	C–O stretching of primary alcohol	[19]
Hemicellulose	1216	–COO group	[4]
Lignin	1227	C–C plus C–O plus C=O stretch; G condensed >G etherified	[19]
–	665	aromatic substituted groups	[4]
–	2895	C–H stretching in methyl and methylene groups	[19]
–	1650–1630	O–H group	[22]
–	3000–3700	O–H group	[23, 24]

for HCNFs with 25wt.% fillers. Peak intensity had a greater shift with the increasing concentration of the filler. These changes are due to the formation of hydrogen bonding or Van der Waals force between the modified TNP filler and cellulose matrix.

XRD Analysis

XRD analysis was performed, and the results are displayed in Fig. 2a. It can be observed from Fig. 2a that XRD analysis displayed approximate peaks for each sample. Besides that, various ordered crystalline arrangements were caused from both intra- and intermolecular hydrogen bonding that takes place in cellulose hydroxyl groups. The intensity of the characteristic peaks of the filler was well below the magnitude of the cellulose matrix and the HCNFs.

However, when the modified TNP filler was reinforced into cellulose, the CI varied from 61.1% for the cellulose matrix to 74.2% in the case of HCNF with 25wt.% filler loading.

Figure 2b illustrates the XRD spectra of the cellulose matrix, modified TNP filler, HCNFs with 5wt.% fillers, and HCNFs with 25wt.% fillers. In the case of the HCNFs, the intense peaks at 41.3° and 51.1° arising because of the reflections from the (1 1 1) and (2 0 0) planes, respectively. It indicates that the CuNPs were crystalline in nature and spherical in shape [10]. These patterns confirmed the presence of spherical CuNPs in the HCNFs. The observations of intense peaks at 41.3° and 51.1° corresponding to the presence of CuNPs were also reported in various nanocomposites films [6, 25, 26]. For cellulose matrix and HCNFs, intensity peaks observed at 12.05°, 20.1°, and 21.3° is attributed

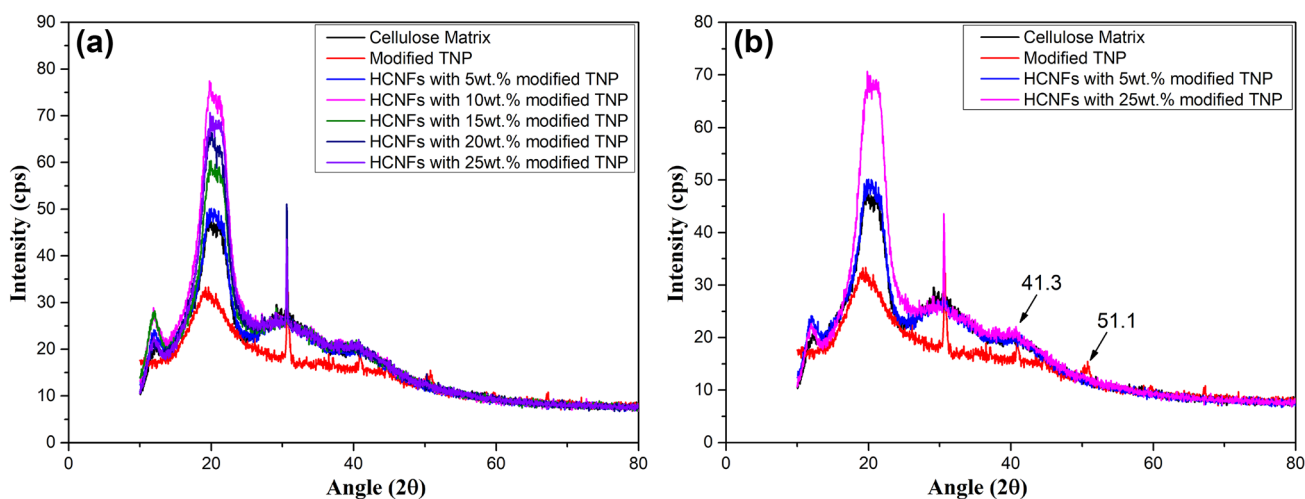


Fig. 2 a. XRD spectra of cellulose matrix modified TNP and HCNFs. b. XRD spectra of cellulose matrix, modified TNP, HCNFs with 5wt.%, and 25wt.% modified TNP fillers

to cellulose II, which arises from the reflections of (1 1 0), (1 1 0), and (2 0 0) planes respectively [18, 27, 28].

Thermogravimetric Analysis (TGA)

TGA curves of pure cellulose matrix and their hybrid nanocomposites are shown in Fig. 3. From the figure, it is observed that all the hybrid composites underwent a two-step degradation process. In the first step, the hybrid composites degraded between the temperature range of 296–303 °C. It could be attributed to the degradation of hemicellulose [29] in the hybrid composites. Yang et al. [30] reported that the decomposition temperature of hemicellulose at 220 °C; however, the decomposition continues until it reaches 315 °C. Then, the final degradation of hybrid nanocomposites occurred between 326–335 °C, which was ascribed to the decomposition of fillers and the cellulose matrix. Nevertheless, after decomposition of hemicellulose, cellulose, etc. some residue remained due to non-degraded CuNPs in the hybrid composites. The final residues for each sample are presented in Table 2.

The thermal behavior of the HCNFs was determined by (a) temperature at which the weight loss of hybrid composites occurred (i.e., 5%, 25%, 50%, and 65%) and (b) based upon the attained final residue (%). Besides, the improvement in the thermal behavior of hybrid composites was witnessed by comparing their degradation temperatures and final residue with the cellulose matrix composites such as cellulose/5wt.% TNP and cellulose/25wt.% TNP (without the infusion of CuNPs) obtained from the first phase of the research [23].

From Table 2, it is essential to point out that the addition of modified TNP in the cellulose matrix, the 5% degradation temperature of all the hybrid composites showed between 101 and 183 °C. Besides, the final residue of hybrid composites ranged between 22 and 24.5%; it could be attributed to the infusion of CuNPs in the TNP. This, however, is not in the case of pure cellulose matrix and non-hybrid composites, where the temperatures were in the range of 80–94 °C. These results confirm the improvement in the thermal stability of the films by incorporating the modified TNP with CuNPs. However, the pure cellulose matrix exhibited higher thermal stability than the

Fig. 3 TGA curves of cellulose matrix and HCNFs

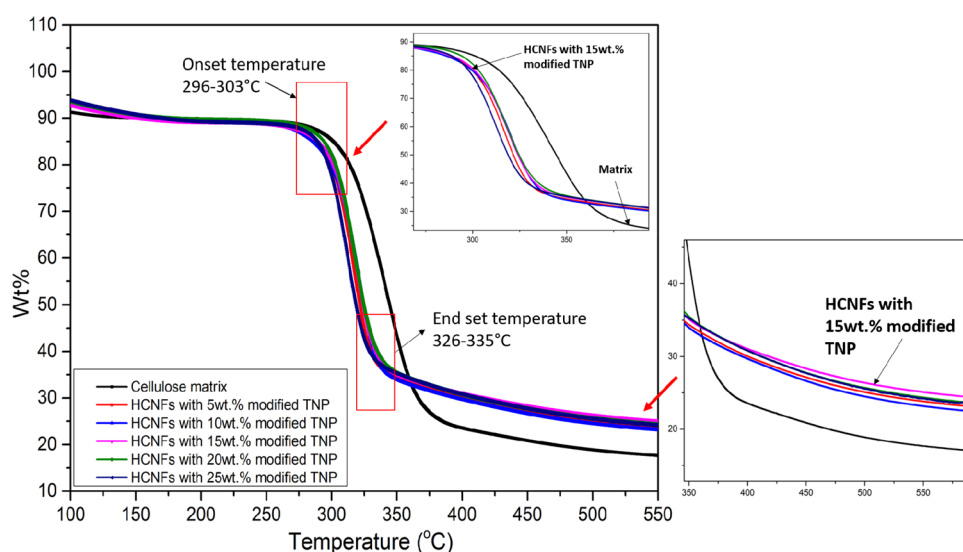


Table 2 Thermal degradation characteristics of the cellulose matrix and HCNFs

Specimens	T °C at 5%	T °C at 25%	T °C at 50%	T °C at 65%	Final residue (%)
Cellulose	93.71	325.13	347.24	363.23	17.61
Cellulose/5wt.%TNP	85.08	313.76	332.71	365.19	18.23
HCNFs with 5wt.% modified TNP	101.45	307.37	322.95	363.82	22.97
HCNFs with 10wt.% modified TNP	120.01	309.49	326.35	366.70	22.30
HCNFs with 15wt.% modified TNP	182.32	311.72	328.37	430.93	24.32
HCNFs with 20wt.% modified TNP	102.6	309.29	325.82	368.03	23.46
HCNFs with 25wt.% modified TNP	105.08	303.92	320.01	367.43	23.42
Cellulose/25wt.%TNP	80.37	312.37	332.94	379.55	18.95

hybrid composites until 357 °C. It was ascribed to the (a) crystalline nature of the cellulose matrix, and (b) the cellulose matrix is relatively possessed higher thermal stability [31]. This result agrees with the poly(propylene) carbonate/tamarind seed polysaccharide/silver nanoparticles [32].

Besides, it is observed that the degradation temperatures were found to be improved in hybrid composites with increasing the dosage of fillers within the cellulose matrix (in Table 2). The enhancement in crystallinity for the hybrid composites also supports the improvement of thermal behavior. However, the increasing trend of degradation temperature of hybrid composites was noticed until 15wt.%. The insignificant decrease of thermal stability for the hybrid composites prepared with 20 and 25wt.% of fillers could be attributed to the possible agglomeration of modified TNP fillers. Based upon the degradation temperatures and final residue, the HCNFs with 15wt.% modified TNP showed higher thermal stability than the others.

Antimicrobial Activity

The use of antibacterial copper is considered in place of other expensive antibacterial agents such as silver, gold, platinum, etc. due to its cheap cost and comparable antibacterial abilities. The antibacterial activity of HCNFs was assessed based on the disc diffusion method, and the results are presented in Fig. 4.

From Fig. 4, it is evident that the infusion of modified TNP in the cellulose matrix exhibited excellent antibacterial activity against all the pathogens used in the study, namely *E. coli*, *P. aeruginosa* (Gram Negative) and *S. aureus*, *B. licheniformis* (Gram Positive). The mechanism of the antibacterial activity is presented in Fig. 5. The reason for the excellent antibacterial activity can be ascribed to the presence of photochemical components present in the TNP and also the influence of CuNPs. It is well known that the Cu^+ ions in the CuNPs prevents cell respiration and punch holes in the bacterial cell membrane and thus destroys the DNA and RNA inside. The reaction with Cu^+ ions forms reactive

Fig. 4 Antibacterial activity of HCNFs with varying concentrations of modified TNP fillers against **a** *E. coli*; **b** *P. aeruginosa*; **c** *B. licheniformis* and **d** *S. aureus*

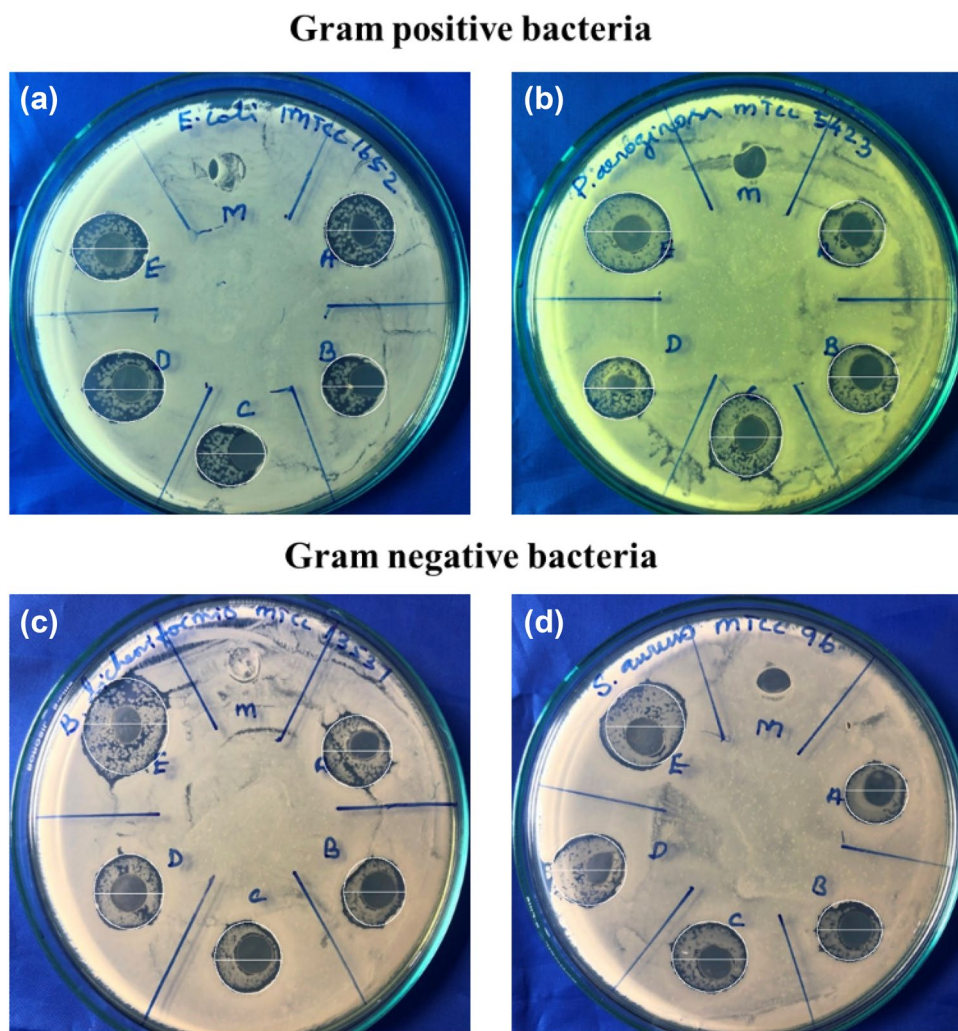
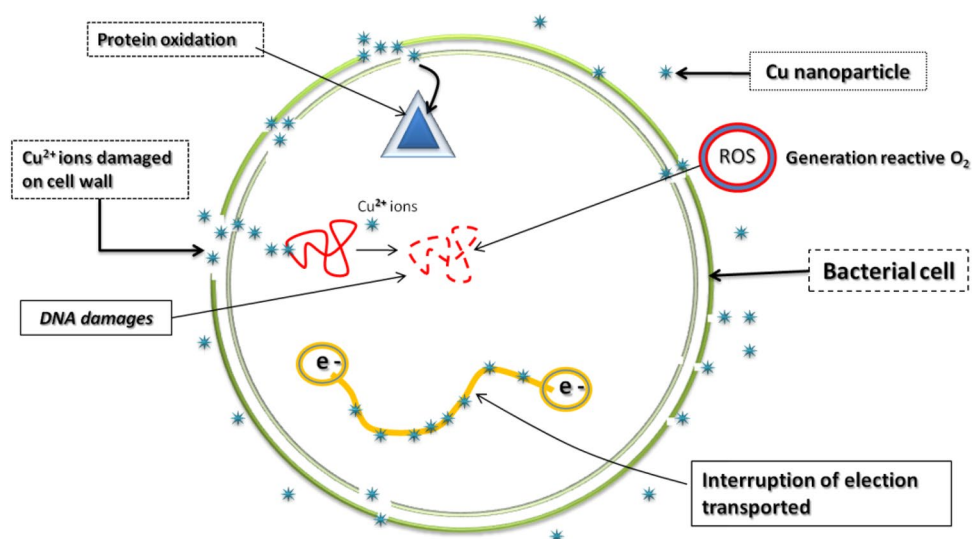


Fig. 5 Mechanism of antibacterial inhibition



oxygen, which also attacks and damages the microorganisms in multiple areas [33]. The inhibition diameters of the different samples subjected in the test to the corresponding bacteria are presented in Table 3. It is also evident from Table 3 that effective antimicrobial activity against all the microbes has been established, and the inhibition diameters of the nanocomposite films were almost equal with minimal deviations in each case. Owing to their excellent antibacterial behavior, these HCNFs can be considered potential replacement in food packaging and biomedical applications.

Conclusions

Hybrid nanocomposites were prepared using cellulose as matrix material and modified tamarind nut power as reinforcing filler by regeneration method. It is evident from the FTIR analysis that the infusion of modified TNP did not change the chemical composition of the cellulose matrix. Modification of TNP with in-situ generated CuNP resulted

in the strong adhesion of reduced Cu²⁺ ions on TNP powder as evident from the increase in the broadness of –OH peaks. The interactions of hydrogen bonding between hydroxyl groups of the cellulose matrix and the filler could also be observed from the FTIR spectra. When the filler loading was increased, the crystallinity of the composites' improved. The relative crystallinity of the cellulose matrix was found to be 61.1%, whereas after the addition of 25 wt.% filler, the crystallinity index increased to 74.2%. The thermal stability of the hybrid nanocomposites at 5% weight loss was found to be better than the matrix. However, the thermal stability marginally reduced with increased filler content. Further, it could be also be seen that the residues increased with increased filler content. The nanocomposite films showed outstanding resistance against the bacteria used in the study. With better thermal and antibacterial properties, these hybrid nanocomposites can be potentially used in food packaging applications.

Table 3 Inhibition zone diameters

Control and modified TNP infused films	Inhibition zone diameters (cm)			
	Gram negative bacteria		Gram positive bacteria	
	<i>E.coli</i>	<i>P.aeruginosa</i>	<i>B. licheniformis</i>	<i>S.aureus</i>
Cellulose matrix (M)	–	–	–	–
HCNFs with 5 wt.% modified TNP (A)	1.020	0.930	1.010	0.900
HCNFs with 10 wt.% modified TNP (B)	0.870	1.010	0.960	0.870
HCNFs with 15 wt.% modified TNP (C)	0.880	1.190	0.970	1.010
HCNFs with 20 wt.% modified TNP (D)	1.000	0.880	0.990	0.990
HCNFs with 25 wt.% modified TNP (E)	1.010	1.130	1.330	1.280

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Data availability The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

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