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Antimicrobial properties of poly(propylene) carbonate/Ag nanoparticle-modified tamarind seed polysaccharide with composite films

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Abstract

Completely biodegradable poly(propylene)carbonate-based composite films with 10 wt.% of tamarind seed polysaccharide (TSP) as filler were prepared by solution casting method. In these composite films, using TSP as the reducing agent, silver nanoparticles (AgNPs) were in situ generated using 1 to 5 mM aq. AgNO₃ source solutions. The hybrid nanocomposite films were characterized by FTIR spectroscopy, X-ray diffraction, scanning electron microscopy (SEM), polarized optical microscopy (POM), tensile testing, thermogravimetric analysis, and antibacterial studies. From the SEM analysis, it was evident that the particle size of the AgNPs varied between 44 and 86 nm when 1 to 4 mM source solutions were used. But on the other hand, the particle size increased to 406 nm when 5 mM source solution was used indicating agglomeration of the AgNPs. The reinforcement of TSP enhanced the crystallinity of the poly(propylene carbonate) (PPC) matrix. The hybrid nanocomposite films exhibited enhanced tensile and thermal properties when compared with the PPC matrix. Further, the hybrid nanocomposites exhibited excellent antibacterial properties against *Escherichia coli* (*E. coli*), *Pseudomonas aeruginosa* (*P. aeruginosa*), *Bacillus licheniformis* (*B. licheniformis*) and *Staphylococcus aureus* (*S. aureus*) bacteria. These hybrid nanocomposites with excellent tensile and antibacterial properties can be potentially used for food packaging applications.

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Introduction

Today, the impact of the plastic food packaging materials on the environment includes the utilization of resources for packaging production, environmental pollution caused by the process and disposal [1, 2]. On the other hand, there is a huge demand for microwave food, frozen food, and snacks from the consumers. The packaging materials must therefore offer a balanced advantage on both food protection and environmental protection [3]. Accordingly, many works have been reported using different biodegradable polymers such as cellulose [4], starch [5, 6] soy proteins [7], polylactides [8], polyhydroxyalkanoates [9], poly(vinyl alcohol) [10], with reinforcing fillers for packaging applications. Similarly, poly(propylene carbonate) (PPC) is also a biodegradable polymer which is synthesized by reaction of CO₂ and propylene oxide in the presence of catalyst [11]. In PPC matrix, several biofillers have been reinforced for improving its properties. Some of them are spent tea leaf powder [12], spent coffee bean powder [13], egg shell powder [14], wood flour [15], cellulose nanocrystals [16], tamarind nut powder [17] etc.

Polymer composites can potentially be used as food packaging materials and can be categorized into three types based on the purpose of usage such as improved packaging, active packaging, and intelligent packaging [18]. Using fillers with nano-size can be a promising option to improve the functional properties [6] of the composites. In recent years, nano metals and metal oxides of silver, zinc, magnesium etc. [19] were used to improve the properties such as flexibility, gas barrier, and antimicrobial or antioxidant properties [20] of polymer composites. The interaction and the compatability between the filler and the matrix is an important factor to achieve the reinforcing effect of nanocomposites. Mainly the properties of the nanocomposites depend on the size of the filler, its dispersion, and the interfacial bonding [21]. Further, the incorporation of nanofillers with antimicrobial or antioxidant activities can result in an inhibiting effect on the microbial growth which can protect the food from spoiling. Silver nanoparticles are most widely used in laundry detergents, disinfectant sprays, and kitchen utensils to inhibit the growth of microorganisms. They are also effective against Gram-positive and Gram-negative bacteria and have a significant antimicrobial performance against multidrug-resistant microorganisms [22, 23]. When silver nanoparticles are incorporated and immobilized in a polymer film by in situ generation, the particles disperse evenly [24] and retain their nano-size in the films. Such films help in extending the shelf life of the food materials [25]. But there can be potential hazard to the consumer health if there is a migration of the nanoparticles into the food stuff. The antibacterial effect in food packaging applications can be performed without the migration of the silver nanoparticles, with the polymer acting as carriers of silver nanoparticles [26]. To the best of our knowledge, there are no reports on using PPC-based hybrid nanocomposites targeting the food packaging application. Hence, in the present work, it was proposed to in situ generate the silver nanoparticles (AgNPs) in the biodegradable polymer composite films with poly (propylene) carbonate and tamarind seed polysaccharide. The hypothesis is that the addition of silver nanoparticles increases the antibacterial properties, and addition of tamarind seed polysaccharide increases the thermal and mechanical properties, while also reducing the final cost of the material.

In this work, hybrid nanocomposite films were fabricated with two different fillers into PPC matrix, namely, tamarind seed polysaccharide (TSP) and the generated AgNPs using solution casting and in situ generation methods. The effect of in situ generated AgNPs on the functional properties of PPC/TSP matrix system was studied. It was proved that the resulting hybrid nanocomposites with better functional properties could be a possible alternate in the food packaging applications.

Materials and methods

Materials

Poly(propylene) carbonate (PPC) was procured from Tianguan Enterprise Group Company (Henan, China) in the form of pellets. The tamarind seed polysaccharide and dimethyl formamide (DMF) were purchased from the local market, India. Silver nitrate (AgNO₃) was purchased from Sigma-Aldrich (Mumbai, India). Standard and clinically isolated microorganism strains of Gram-negative *Escherichia coli* (*E. coli*) and *Pseudomonas aeruginosa* (*P. aeruginosa*) and Gram-positive *Staphylococcus aureus* (*S. aureus*) and *Bacillus licheniformis* (*B. licheniformis*) were procured from MTCC, IMTECH, Chandigarh, India, and were used for antimicrobial assays.

Preparation of poly(propylene) carbonate/tamarind seed polysaccharide/silver nanoparticle hybrid nanocomposites

The TSP was dried in a hot air oven at 100 °C for 1 h to remove the moisture content. With the aid of a sieve, the powder with particle size less than 20 μ m was separated and mixed with the PPC solution using a mechanical stirrer. Prior to this, the PPC solution was prepared for the required wt% using the same procedure reported earlier [12, 14]. The mixture of PPC/TSP solution was spread over the glass plate to obtain the uniform thickness as per the spacer dimension. Then, the PPC/TSP composite films were formed using simple casting technique. The composite films were then dipped in 1 to 5 mM aq.AgNO₃ source solutions separately to get in situ generated AgNPs in the films to form hybrid nanocomposites. The films with change in color were washed thoroughly with distilled water and then dried. The color remained unchanged despite repeated washings with distilled water indicating the permanent generation of AgNPs in the hybrid nanocomposite films. Then, films were stored in desiccators prior to further testing.

Microscopy

A scanning electron microscope (SEM) (Zeiss O 18) and a polarized optical microscope with a digital image analyzer (Olympus SZX12) were used to investigate the distribution and particle size of the TSP fillers and the AgNPs in the nanocomposite films.

Fourier transform infrared spectroscopy

An IR spectrometer (Bruker Vector 22) was used to record the Fourier transform infrared spectroscopy (FTIR) spectra of the PPC matrix, the filler, and the PPC/TSP/AgNPs hybrid nanocomposites in reflection mode. All the spectra were recorded in the 4000–500 cm⁻¹ region with 32 scans in each case, at a resolution of 4 cm^{-1} .

X-ray diffraction

An X-ray diffractometer (Rigaku Corporation, Japan) was used to record X-ray diffractograms of the PPC/TSP/ AgNPs hybrid nanocomposites in 10-80° range at a scan rate of 4°/min.

Thermogravimetric analysis

The primary and derivative thermograms of the PPC/TSP composite matrix and the PPC/TSP/AgNPs hybrid nanocomposite films were recorded using a thermogravimetric analyzer (Mettler Toledo) in the temperature range of 25–600 °C in nitrogen atmosphere at a heating rate of 10 °C/min.

Tensile testing

The tensile properties of PPC/TSP composite matrix and PPC/ TSP/AgNPs hybrid nanocomposite films were determined as per the ASTM D882 specifications. Film strips were cut into 100 mm \times 10 mm sections and then characterized using INSRON 3369 Universal tensile machine equipped with 5 KN load cell with an initial grip separation and crosshead speeds of 50 mm and 5 mm/min, respectively. Tensile strength, modulus, and elongation at break were calculated. Five identical specimens were used in each case, and the average values are reported.

Antibacterial activity

The antibacterial test conducted for the hybrid nanocomposite films was carried against four bacteria using two Gramnegative namely Escherichia coli (E. coli) and Pseudomonas aeruginosa (P. aeruginosa) and two Gram-positive Staphylococcus aureus (S. aureus) and Bacillus licheniformis (B. licheniformis) by disc method as described elsewhere [27]. The inhibition zones indicating the inhibition of bacteria were photographed, and the zone diameter in each case was measured. All test organisms were inoculated in LB broth (pH 7.0.) for 8 h. The concentration of the suspensions was adjusted to 0.5 (optical density) by using a spectrophotometer. Isolates were seeded on LB agar plates by using sterilized sterile plastic inoculation loop and L rod. One hundred microliters of 10-6 cells was spread over the LB agar plate. Plates were incubated at 37 °C for 48 h. Triplicate plates were maintained for each organism.

Results and discussion

Appearance of the matrix and the PPC/TSP/AgNPs hybrid nanocomposite films

The digital images of the matrix and the PPC/TSP/AgNPs hybrid nanocomposite films are shown in Fig. 1. From Fig. 1, it is evident that both the matrix and the PPC/

Fig. 1 Digital images of PPC/TSP matrix (a) and PPC/TSP/AgNPs hybrid nano composite films made using 1 mM (b), 2 m (c), 3 mM (d), 4 mM (e), and 5 mM (f) aq.AgNO₃ source solutions





Fig. 2 Optical images of PPC/TSP/AgNPs hybrid nanocomposites made using 1 mM (a), 2 m (b), 3 mM (c), 4 mM (d), and 5 mM (e) aq.AgNO₃ source solutions

TSP/AgNPs hybrid nanocomposite films were uniform. As the TSPs have proteins, carbohydrates, crude fibers etc., in order to probe the presence of the crude fibers, the polarized optical micrographs were recorded and are presented in Fig. 2. From Fig. 2, we can observe, in addition to the carbohydrate particles, the nanocomposite films had some fibers [28] which could not be observed in the digital images (Fig. 1). The presence of the fibers in the fillers of the nanocomposites is expected to improve their tensile properties. Further, from the digital images of the composite films, it can be understood that, upon increasing the filler concentration, the color of the films darkened, but they



Fig. 3 SEM images of PPC/TSP/AgNPs hybrid nanocomposite films with in situ generated AgNPs on the surface of the TSPs made using 1 mM (a), 2 mM (b), 3 mM (c), 4 mM (d), and 5 mM (e) aq.AgNO₃ source solutions



Fig. 4 Histograms of the particle size distribution of in situ generated AgNPs in the PPC/TSP/AgNPs hybrid nanocomposite films made using 1 mM (a), 2 mM (b), 3 mM (c), 4 mM (d), and 5 mM (e) aq.AgNO₃ source solutions

have sufficiently enough transparency. Hence, they can also be utilized for wrapping applications.

SEM analysis

The SEM images of the PPC/TSP/AgNPs hybrid nanocomposite films are presented in Fig. 3. From Fig. 3, it can be seen that the nanocomposite films had spherical AgNPs on the surface of the TSP filler. It can also be observed that with the increase in the concentration of the source solution, the resulting AgNPs were found to agglomerate at some places. The particle size in each case was measured using SmartTiff program, and the histograms indicating the particle size distribution are presented in Fig. 4.



Fig. 5 FTIR spectra of PPC/TSP (Matrix) and PPC/TSP/AgNPs hybrid nanocomposite films made using 1 to 5 mM aq.AgNO₃ source solutions (**a**) and PPC/TSP and PPC/TSP/AgNPs composite film made using 5 mM aq.AgNO₃ source solution (**b**)

From Fig. 4, it is evident that the average particle size when the 1 to 4 mM source solutions were used varied between 44 and 86 nm. However, when 5 mM source solution was used, the average size of the AgNPs was found to be around 406 nm indicating agglomeration of the particles. It is understandable as for the highest concentrated source solution, many number of AgNPs were generated leading to their agglomeration. It is worthy to note here that the agglomeration of the AgNPs did not affect the tensile strength and modulus of the hybrid nanocomposites. It can be seen that the tensile strength and modulus had an increasing trend even at the highest source solution concentration.

Fourier transform infrared spectroscopy analysis

The FTIR spectra of the matrix and the composite films were recorded and presented in Fig. 5a. From Fig. 5a, it is evident that the spectra of both the matrix and the hybrid nanocomposite films were similar. For clarity, the spectra of the matrix and the PPC/TSP/AgNPs composite film made using 5 mM aq.AgNO₃ source solution are presented separately in Fig. 5b. From Fig. 5b, it is evident that the matrix and the hybrid nanocomposite film had similar functional groups. The broad and shallow absorption band at 3500 cm^{-1} corresponds to the stretching vibrations of the OH groups of TSP. The bands at 2995 and 1730 cm⁻¹ correspond to the C-H and C=O stretching vibrations. Further, the peaks at 1460 and 1237 cm⁻¹ were attributed to the -C-H bending and -CO vibrations of carbonate. The C-C of methyl group resulted a peak at 941 cm⁻¹ while the deformation of the methylene group at 780 cm^{-1} . The sharp band at 1066 cm⁻¹ was ascribed to the C-O-C vibrations [17]. Hence, the in situ generated AgNPs in the TSP filler did not alter the structure of the hybrid nanocomposite films.



Fig. 6 XRD pattern of a pure PPC, TSP filler and matrix (PPC/TSP composite) and PPC/TSP/AgNPs hybrid nanocomposite films made using 1 mM (a), 2 mM (b), 3 mM (c), 4 mM (d), and 5 mM (e) aq.AgNO₃ source solutions

and 5 mM	I aq.AgNO ₃ source solutions		
Table 1	The FWHM (B _{size}) and crystallite size (Dp) of PPC/TSP/AgNPs composites with in situ generated AgNPs using T	mM, 2 mM, 3 mM,	4 mM,

Peak	1 mM AgNPs		2 mM AgNPs		3 mM AgNPs		4 mM AgNPs		5 mM AgNPs	
2θ (°)	FWHM B _{size} (°)	Dp (nm)								
38.37	0.62742	14.01	0.87441	10.05	0.56338	15.61	0.7402	11.88	0.5723	15.36
44.57	0.52076	17.23	0.54729	16.4	0.56342	15.93	0.6329	14.18	0.5629	15.94
65.01	0.54107	18.2	0.5444	18.09	0.52497	18.76	0.4983	19.76	0.5597	17.59
78.07	0.59124	18.08	0.59549	17.95	0.49158	21.75	0.5446	19.63	0.6081	17.58
Dp Average (nm) 16.8		16.88		15.62		18.01		16.36		16.62

X-ray diffraction analysis

The PPC, TSP filler, and PPC/TSP composites and PPC/ TSP/AgNPs hybrid nanocomposites were characterized using X-ray diffraction (XRD) to comprehend the structural behavior. Figure 6a shows the XRD spectra of PPC, TSP filler, and PPC/TSP composites. The appearance of a broad peak indicates the amorphous nature of PPC. Similar observation was also reported earlier [29, 30]. Further, the presence of sharp intensified peak at $2\theta = 20^{\circ}$ was attributed to the crystalline nature of TSP filler with cellulose-II structure. From Fig. 6a, it is clearly evident that the addition of TSP filler in the matrix influenced the intensity of the peak in the case of PPC/TSP composites [17]. Figure 6b shows the XRD spectra for PPC/TSP/AgNPs hybrid nanocomposites with in situ generated AgNPs made using 1 to 5 mM aq.AgNO₃ source solutions. Strong peaks appeared at $2\theta = 38.37^{\circ}$, 44.57° , 65.01° , and 78.07° corresponding to the reflections from (111), (200), (220), and (311) planes of AgNPs in the hybrid nanocomposite films. Such observations were also made for AgNPs in similar systems [31-33]. Besides, the peak at 32.28° might be due to the organic compounds in the TSP filler [34]. A

similar result was reported earlier, where they identified the crystalline peaks at 32.28° [34, 35].

Further, the crystallite size of the AgNPs was determined using Debye–Scherrer's equation as reported earlier [36]:

Crystallite size $D_A = K \lambda/(B \cos \theta)$

where D_A —average crystallite size (nm); K—Scherrer constant. K varies from 0.68 to 2.08. K = 0.94 for spherical crystallites with cubic symmetry; λ —X-ray wavelength, Cu K α radiation (1.54178 Å); **B**—FWHM (Full width at half maximum) of XRD peak; θ —XRD one half of 2θ peak position. The average crystallite size values for PPC/TSP composites with in situ generated AgNPs using 1 to 5 mM aq.AgNO₃ source solutions are presented in Table 1. The calculated crystallite sizes are in good agreement with the reported values published by earlier workers for AgNPs [31-33]. The calculated average size of the particle is 16.8 nm, 15.6 nm, 18.0 nm, 16.3 nm, and 16.2 nm for the PPC/TSP/ AgNPs hybrid nanocomposites with in situ generated AgNPs using 1 mM, 2 mM, 3 mM, 4 mM, and 5 mM aq.AgNO₃ source solutions respectively.



Fig. 7 Primary thermograms (a) and derivative thermograms (b) of matrix and PPC/TSP/AgNPs hybrid nanocomposite films made using 1 to 5 mM aq.AgNO₃ source solutions



Fig. 8 Tensile strength (a), tensile modulus (b), and %elongation at break (c) of the matrix and the PPC/TSP/AgNPs hybrid nanocomposite films made using 1 to 5 mM aq.AgNO₃ source solutions

Thermogravimetric analysis

In order to probe the thermal stability of the PPC/TSP/AgNPs hybrid nanocomposites, the thermogravimetric analysis was performed and the results are presented in Fig. 7. It can be noticed that there is no inflection peak found in the region of 30–100 °C, indicating the hydrophobic nature of PPC. From Fig. 7, it is evident that the thermal stability of the composites increased with increase in filler content. However, till a temperature of 350 °C, the thermal stability of the composites was lower than the matrix but after that, a reverse trend was observed. This could be attributed to the polyphenols present in the TSP filler [17] and also the nanoparticles could have influenced such an effect. Further, the thermal stability of the composites in the TSP which include cellulose and carbohydrates.

Tensile properties

In order to probe the effect of in situ generated AgNPs on the tensile properties of the PPC/TSP/AgNPs hybrid nanocomposite films, the tensile test was carried out. The histograms representing the tensile strength, tensile modulus, and the % elongation at break of the matrix and the PPC/TSP/AgNPs hybrid nanocomposite films are presented in Fig. 8. The tensile properties of the matrix (PPC/TSP composite) used in the present work were compared with those of same grade PPC used in the earlier work of one of our groups [12]. The tensile strength, modulus, and elongation at break of PPC were reported to be 10.5 MPa, 417 MPa, and 821%, respectively. From Fig. 8, it can be observed that the tensile strength and modulus of PPC/TSP composite used in the present work increased by 16 and 5% while the elongation at break decreased by 138% over the PPC. This may be due to the presence of microfibers and polyphenols in the TSP. Further, from Fig. 8, it is evident that the tensile strength and the modulus of the hybrid nanocomposite films were higher than those of the matrix and increased with increasing concentrations of the source solutions. However, the % elongation at break exhibited a reverse trend. The possible reasons for such a trend could be the uniform dispersion of the TSP fillers with in situ generated rigid AgNPs in the PPC/TSP/AgNPs hybrid nanocomposites. Further, the effective stress transfer among



[After 24 hours]



[After 36 hours]



[After 48 hours]

Fig. 9 Antibacterial inhibition zones of PPC matrix and PPC/TSP/ AgNPs hybrid nanocomposite films made using 1 mM (1); 2 mM (2); 3 mM (3); 4 mM (4), and 5 mM (5) aq.AgNO₃ source solutions against

the components may be another reason for the improvement in the tensile properties of the hybrid nanocomposite films under study [10, 37]. Similar observation was also made in the case of PPC/TNP composites [17] and cellulose/TNP composites [38].

Antibacterial activity

It is an established fact that the AgNPs exhibit good antibacterial activity. In order to probe the antibacterial activity of the PPC/TSP/AgNPs hybrid nanocomposite films, the antibacterial test by disc method was carried out against both Gram-negative and Gram-positive *Escherichia coli* MTCC 1652 (**a**), *Pseudomonas aeruginosa MTCC 2453* (**b**), *Bacillus licheniformis MTCC 73537* (**c**), and *Staphylococcus aureus MTCC 96* bacteria [after 24 h, 36 h, and 48 h]

bacteria. The zones of clearance indicating the inhibition of bacteria were photographed after 24 h, 36 h, and 48 h, and the respective images photographed are presented in Fig. 9. Results were analyzed on the basis of diameter of inhibition zones of nanocomposite films with varying concentration of AgNPs under different bacterial medium. From Fig. 9, it is evident that though the matrix did not exhibit any antibacterial activity, the nanocomposite films showed good antibacterial activity against both Gram-negative (*Escherichia coli* and *Pseudomonas aeruginosa*) and Gram-positive (*Bacillus licheniformis* and *Staphylococcus aureus*) bacteria after 24 h, 36 h, and 48 h of the test. Further, in all the cases, the diameters of the clear zones increased with increasing concentration of the source solutions.

From the result, a slight increase in clear zone diameter was observed for the particular bacteria in each case of Gramnegative (P. aeruginosa) and Gram-positive (S. aureus) at 36 and 48 h of inhibition. It could happen because of highly sensitive nature and chemical structure of P. aeruginosa than that of E. coli bacteria though both belong to Gram negatives. A similar observation was also found for S. aureus in Grampositive case because of a compound nature. However, it was observed that the clear zone formation was found to vary due to various factors such as type of bacterial medium, concentration, cell wall mechanism etc. Some of the previous reports discussed about the restriction of antibiotic behavior of bacteria through various mechanisms have been considered to support the aforesaid statements. Accordingly, some microorganisms are capable of changing the antibiotic's chemical structure. This mechanism is applicable in bacteria which produce β -lactamases, and these enzymes cleave the β -lactam ring of the penicillin, or other β -lactams, which leaves the antibiotic obsolete [39]. Secondly, some bacteria have an increased number of efflux pumps in their cellular membrane. These efflux pumps can rapidly expel the antibiotic that enters the cell, keeping the internal antibiotic concentration to a minimum [40]. Lastly, bacteria also have the ability to modify chemically or genetically the target site of the antibiotic, and hence, once the target is modified, the antibiotic cannot bind and resistance is acquired [41]. Further, increasing concentration of AgNO₃ could lead to the increasing surface area of nanoparticles which in turn can have interactions with larger microorganisms and thus alter the microbial metabolism [42]. It is well known that AgNPs are toxic to microorganisms, and hence, the AgNPs embedded TSP filler in the nanocomposite films are expected to lysis the cell wall of the bacteria and restrict the further growth of bacteria [42, 43]. After the formation of antimicrobial activity in the AgNP compound, the retention of clear zone was not found to occur even after 48 h of inhibition which could confirm the effectiveness of AgNPs present in the medium. It ensures the stability of the AgNP compound in the medium which can happen due to the existence of Ag nanoparticle after the oxidation process.

Conclusions

The AgNPs were in situ generated in the PPC/TSP composite films to form hybrid nanocomposites. The average particle size of the AgNPs was found to be 44 to 86 nm when 1 to 4 mM source solutions were used. On the other hand, when 5 mM source solution was used, there was an increase in the particle size to 406 nm which evidence the formation of particle agglomeration. The infusion of TSP in the PPC matrix increased the crystallinity. The FTIR spectra were almost similar for both the matrix and composites indicating no changes in the structure of the composites. The XRD diffractograms proved that the crystallinity of the composites increased with the addition of fillers. The hybrid nano composite films exhibited better thermal stability compared to the matrix. This may be attributed to the rigid polyphenols and polysaccharides present in the TSP filler and the presence of AgNPs. The tensile strength and modulus of the composites increased with increase in the filler content, while a reverse trend was observed in case of % elongation at break. This behavior of the nanocomposite films may be due to the in situ generation of rigid AgNPs in them which increased the tensile stress and modulus while decreasing the % elongation at break. Further, the films exhibited excellent antibacterial activity against both Gram-positive and Gram-negative bacteria. These hybrid nanocomposite films with excellent tensile and antibacterial properties can be potentially used in active food packaging applications.

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