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SYNTHESIS AND PROPERTIES OF DEGRADABLE BIOFILMS FROM RAW BANANA POWDER TO GROW NANOCOMPOSITESIN THE FOOD PACKAGING APPLICATIONS

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Abstract

Silver nanoparticles were developed using aqueous extract of Citrus limetta leafas a reducing as well as capping agents, and Silver Nitrate as a precursor. Raw banana powder was used to develop starch-based nanocomposites films containing glycerol as plasticizers incorporated with silver nanoparticlesat different concentrations via solution casting method. Themain objectives of this study were to improve the properties such as mechanical, thermal and physical properties of starch nanocomposite films incorporated with silver nanoparticles containing glycerol as plasticizer. The physicochemical properties of the film were characterized.In general, a high concentration of silver nanoparticles in the film could increase the tensile strength,reduce the water vapor permeability and water solubility and also antibacterial activity.

Keywords:Citrus limetta leaf,nanocomposites,packaging,silver nanoparticles,Thermal property.

Introduction

Synthetic Plastics have been employed in discrete due to their physicochemical furnishings and economic growth(Ballesteros-Martinez*et al*.,2020). Environment and the exhaustion of natural assets caused by non-biodegradable petroleum-based plastics have led to great interest in developing environmentally benign natural polymers and biodegradable films for application in food packaging and other purposes(Liu *et al*., 2016).Renewable and degradable biopolymer are the most essential alternative for producing green raw materials in the forthcoming(Orsuwan*et al*., 2017). Nature has provided various natural biopolymers such as polysaccharides (starch, cellulose and chitosan) and proteins such as soy protein, wheat protein, casein and gelatine(Fahad *et al*., 2017).

Green bananas have a large amount of starch during its unripe stage, which consists of around 20-25 % found in the pulp of the fruits(Coelho *et al*., 2020).The banana starch might be able to use as substitution of other starches such as potato, corn and wheat(Nimsung*et al*., 2007).Starch-based biofilms show excellent optical properties like transparent,colorless and barrrier properties like oxygen and carbon-dioxide permeability (Cheng et al., 2019; Yildirim-Yalcin et al., 2019; Saberi et al., 2018).Several starches from different sources, either alone or in combination with other biopolymers,have been assessed as biodegradable packaging agents or as edible coatings to extend the shelf life of fresh produce(Guo *et al*.,2019; Abdelghaffar, 2021; Valodkar *et al*., 2012; Roy and Rhim, 2019).

To produce efficient edible films, the properties of films must be optimized for industrial applications (Jost*et al*., 2014). The type and content of plasticizer are critical factors affecting the properties of films.plasticizers are generally small molecules such as glycerol and sorbitol that intersperse and intercalate among and between polymer chains, disrupting hydrogen bonding and spreading the chains apart, which not only increases flexibility, but also water vapor and gas permeabilities(Vieira *et al*., 2011).Among the plasticizers, glycerol has often been used as a plasticizer for starch films due to its compatibility with amylose (Farhan *et al*., 2017) which promotes better mechanical properties by interfering with amylose packing, thereby decrease the intermolecular forces between the starch molecules(Nordin*et al*., 2020).

In this present work silver nanoparticles were prepared simple green method by using fresh Citrus limetta leaf extract. There are no reports of silver nanoparticles which combined with raw banana powderto develop nanocomposites at different concentrations. This work aims to evaluate the mechanical,thermal and physical properties of starch nanocomposite films incorporated with silver nanoparticles containing glycerol as plasticizer.These prepared nanocomposites biofilm may act as convenientfood packaging applications.

2. MATERIALS AND METHODS

2.1 Synthesis of Silver nanoparticles

Silver nanoparticles using leaf extract Citrus limetta (CL-Ag) nanoparticles were synthesized by adding the corresponding leaves extracts to 0.1 m of the AgNO3 solution with a volume ratio of 1:10.The mixture were stirred on magnetic stirrer at room temperature for 1 hour and left for 48 hours. These set up were covered with Aluminium foil to avoid photo activation of silver nitrate. Monitoring of black precipitate indicates the formation of silver nanoparticles. These nanoparticles were allowed to centrifuged 7,000 rpm for 10 min, filtered, washed with de-ionised water for several times followed by ethanol and dried at 90^0c in an oven for 2 hours.

2.2 Preparation of Starch based biofilm and their Nano composites

Starch based nano composite synthesized by solution casting method. In this method, first raw banana powder (3g) dispersed in de-ionized water (30ml), which was gelatinized by stirring and heated up to 75^0c.A clear viscous solution was obtained. To this solution, silver nanoparticles with different % weight (0.02) has been dispersed. Then the solutions were casted into the petri dish coated with petroleum jelly the system was kept aside for two days at room temperature, Nanocomposite biofilms were obtained.

2.3 Evaluation of mechanical properties

Tensile strength of the prepared novel starch-based biofilm and its nanocomposites were evaluated by Universal Testing Machine at across speed of 100 mm for minute using rectangle shaped samples (10 x 4 cm) punched out from starch-based biofilm and its nanocomposites as for ASTM D6100(Gadhave*et al*., 2018). The gauge length was set at 3cm in each test. The Young's modulus, tensile strength and elongation at break were evaluated using standard methods.

2.4 Determination of Water absorption percentage (WA %)

Each starch-based biofilm was put in 5ml of distilled water for 24 hrs. After 24 hrs, the excess solvent on the outside of starch-based biofilm was detached by using filter paper. Then it was weighed and the solvent absorption percentage was calculated using the following equation, were W_1 = Weight of the starch-based composites, W_2 = Weight of the starch-based composites after absorption of the solvent.

2.5 Soil burial degradation test

The novel starch-based biofilm and its nanocomposites (5 x 2 cm) were buried in the soil at a base of 25 cm from the ground surface for 60 days, inoculated with compost having the capacity to grip and degrade the starch-based nanocomposites. At fixed time, the samples were removed, washed with water in order to ensure the stop of the degradation, dried out at 30^{o}C weighted and stored in dark(Gabriel*et al*., 2021).

2.6 Evaluation of performance under bacterial exposure

Solid media ager-plate tests were conducted to evaluate the biodegradation of starch-based biofilm and its nanocomposites by specific strains of microorganisms for 48 hours.

3. Characterization studies

Scanning Electron Microscopy is used to determine particle size and surface morphology of the starch-based nanocomposites and silver nanocomposites. The thermal stability of the nanocomposite biofilm was evaluated using TGA and the derivative thermogravimetric curve was expressed as the mass variation as a function of temperature (Shapii*et al*.2022).

4. Result and discussion

4.1 Mechanical properties

The data of mechanical properties such as young’s modulus, tensile strength and elongation at break of newly prepared starch-based biofilm and its nanocomposites are given in Table 1. It is observed that the 0.02 concentration ofsilver nanocomposites possesses greater mechanical properties than starch-based biofilm due to the presence of smaller size nanoparticles.The mechanical property shows that the 0.02 concentration ofsilver nanocomposites possess higher young’s modulus and tensile strength than the starch-based biofilm. Nanosized particles, size decreases, these nanoparticles shared the tensile stress to improve the tensile strength and young’s modulus significantly.

Table 1. Mechanical properties of starch and their nano composite

Polyesters and their composites	Tensile strength (MPa)	Elongation at break (%)	Young’s modulus (MPa)
starch	14.2	95.70	11.8
0.02 Ag	14.6	87.25	11.1

4.2 Thermal properties

In this study, the TGA and DTA thermograms of starch-based biofilm and its 0.02g of silver nanocomposites are given in Fig 1. Thermogravimetric analysis reveals that all the composites have higher starting degradation temperature and weight losses. The decomposition of composites shows three stages. The third stage of decomposition is about 99.28% and 99.71% in the temperature range 350^0C to 500^0C due to the complete decomposition of thesilver nanocomposites and starch-based biofilm. Thermal decomposition of the starch-based biofilmpossesses higher degradation percentage than the silver nanocomposites. The DTA thermogram of silver nanocomposites and starch-based biofilm shows an endothermic peak at 250^0C to 300^0C, it is due to the degradation of crosslinking starch bio composites fragments.

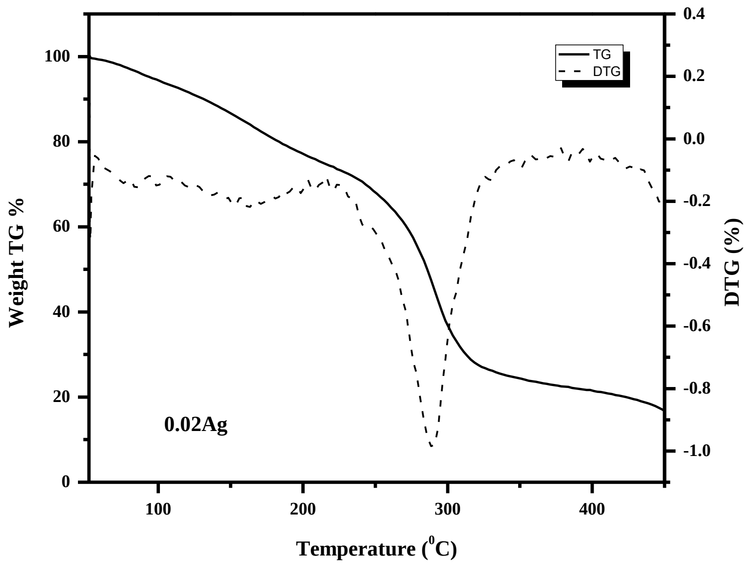

Figure 1: Thermal analysis of 0.02 g concentration of silver nanocomposites

4.3 Physical and chemical properties of starch-based nanocomposites

The result of the water solubility, water vapor permeability, water absorption and moisture content of the newly prepared starch-based biofilm and their silver nanocomposites are shown in Table.2. The water solubility, moisture content, and water absorption of different concentration (0.02Ag) of starch-based nanocomposite decreased significantly compared to the neat starch-based biofilm. These decreasing properties probably due to the addition of silver nanoparticles having hydrophobic character, which reduced the hydrophilicity of starch-based biofilm. Comparing different concentration of biofilm 0.02Ag have highest water solubility, water vapor permeability, water absorption than starch-based nanocomposite. When increasing concentration of nanoparticles incorporated with biofilm the hydrophilicity character reduces.

Table 2. Physico chemical properties of starch-based biofilm and their nanocomposites

Starch based nanocomposites	Moisture content	Solubility in water	Absorption of water	Water Vapor permeability
starch	19.68	29.37	37.5	3.56
0.02 Ag	19.12	18.77	33.3	3.24

The water vapor permeability of the starch-based biofilm decreased when silver nanoparticles incorporated with starch-based biofilm. The decrease in the water vapor permeability of composite films was attributed to the distribution of silver nanoparticles as a discontinuous phase in the film matrix, which prevented the diffusion of water vapor resulting in an increase in the tortuous path (Taha*et al*., 2022).

4.4 Solvent absorptivity

The solvent absorptivity percentage of the newly prepared starch-based biofilm and their corresponding 0.02 Ag nanocomposites are given in Table 3. The solvent absorptivity is carried out in different solvents such as Ethylene glycol, Methanol, Glycerol, ethyl methyl ketone (EMK), DMA(dimethyl acetamide) Acetone, Chloroform, Xylene. This indicates that the newly prepared starch-based biofilm and their corresponding 0.02 Ag nanocomposites are hydrophobic in nature. In the present study, the newly preparedstarch-based biofilm and their corresponding 0.02 Ag nanocomposites, the solvent absorptivity percentage is observed in the order Raw banana powder starch>0.02Ag. starch-based biofilm has higher solvent absorptivity percentage in comparison with that of0.02 Ag nanocomposites.

Table 3. Solvent absorptivity percentage of starch and their nano composite

Starch and their composites	Solvent absorptivity (%)							
	Ethylene glycol	Methanol	Glycerol	EMK	DMA	Acetone	Chloroform	Xylene
(RB)Starch	27.11	16.66	10.2	16.42	54.02	20.46	37.77	7.40
0.02Ag	22.30	11.76	18.9	15.40	48.43	19.28	17.85	6.73

4.5 Soil burial test

Soil burial test was used to detect the environmental resistance of the 0.02 concentration of silver nanocomposites and starch-basednanocomposites. The weight loss percentage of newly prepared 0.02g of silver nanocomposites and starch-based compositesin soil burial test is represented in Table 4.

Table 4. Weight loss % of starch-based nanocomposites under soil burial test

Starch based nanocomposites	Weight loss%
Raw banana starch	89.38
0.02 Ag nanocomposites	85.59

As the percentage of nanoparticle content increases the degradation rate decreases. The weight loss percentage of newly prepared 0.02g of silver nanocomposites and starch-based nanocomposites are 89.38% and 85.59% It concluded that the starch-based nanocomposites possess higher degradation percentage than the 0.02g silver nanocomposites.The starch-based composites have higher degradation rate due to the rate of increased starch content.

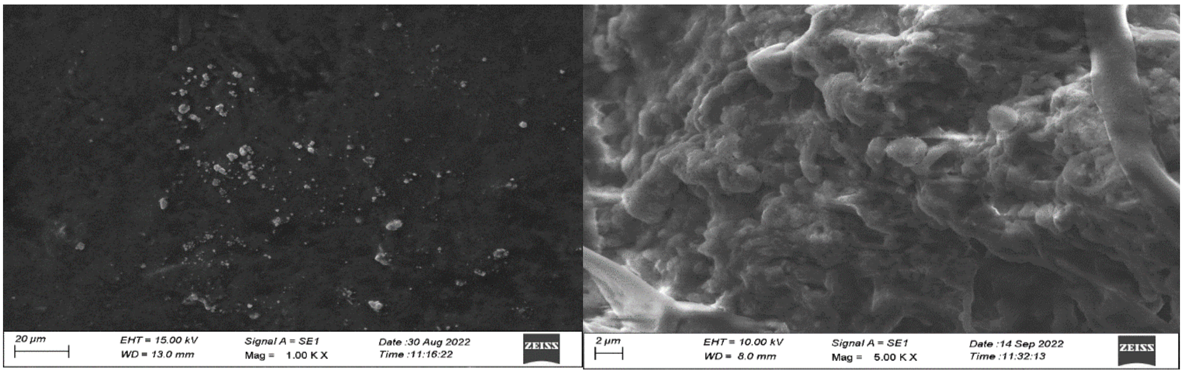

Figure 2:Before and after soil burial 0.02 g concentration of silvernanocomposites

4.6Antifungal activity

The antimicrobial activity of the newly prepared starch -based biofilm and their 0.02Ag silver nanocomposite films was tested against bacterial strains, *Serratia marcens Gram (-), Bacillus cereus Gram(+)*,are shown in Figure 3. Raw banana powder itself showed minimum inhibition activity against gram positive and gram negative bacterial strains. Starch based biofilms exhibited significant antibacterial activity of 10 mm against *Bacillus cereus* and 11mm against *Serraciamarcens.*The starch-based 0.02Ag silver nanocomposite films exhibited no inhibition against *Bacillus cereus* and 9 mm inhibition showed against *Serraciamarcens.*Antibacterial activity is mainly due to differences in the cell wall structure of the test microorganisms (Corre*et al*., 2010). Starch- based biofilm exhibited significant antibacterial activity compared to and 0.02 Ag nanocomposite. Edible food packaging material which contains great antibacterial activity can be used as food packaging applications in industry.

Figure 3: Antibacterial activity of starch-based biofilm and 0.02 Ag silver nanocomposites

Conclusion

The green synthesis of silver nanoparticles has been achieved using the bio reducing agent *Citrus limetta* leaf extract. Some of the starch-based materials which are incorporated with nanomaterials to enhance the thermal and mechanical properties of biofilm have been essential in developing feasible replacements for petroleum-based plastics.Silver nanoparticles played an important role in maintaining film properties such as water solubility, water vapor permeability,mechanical, and antimicrobial properties. Starch based biofilm and their silver nanocomposite possesses greater mechanical properties and lower water solubility, moisture content, and water absorption. The results suggested that the 0.02 Ag starch-based bio nanocomposites can be used as an active food packaging material compared to plain starch-based biofilm.

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