

Development of Eco-Friendly LLDPE Composite Films with Antimicrobial Functionality Using Citrus Peel Waste

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Abstract: Over a few decades, plastic packaging has been mainly used for the storage of food products. However, a major concern has been the contamination of food due to microbial activities. Various plastic materials or composites have been developed for their use in packaging applications so that the microbial activities can be stopped or diminished. The main aim of the current project is to incorporate antimicrobial activities in the commonly used LLDPE films by utilising antimicrobial activities of naturally occurring biowaste i. e. orange-peel powder. The films were produced by conventional blown film process and are studied for various mechanical properties as well as their antimicrobial activities.

Keywords: Orange peel powder, green composite, active packaging, antimicrobial film, sustainability

1. Introduction

Plastics, as polymeric materials, can be shaped into various forms under heat and pressure owing to their plasticity. These materials are widely produced through industrial processes, with packaging films being one of their primary applications due to their ability to preserve food freshness and shape, ensure product safety, facilitate identification, and aid transportation. Packaging use accounts substantially for the global consumption of plastic materials. [1-4]

Sustainable packaging has increasingly gained importance as consumers become more aware of foodborne illnesses and seek safer food options. This heightened awareness has driven demand for packaging that prevents spoilage and maintains food quality throughout storage and distribution. Food contamination caused by bacteria and fungi leads to degradation of texture, flavour, colour, and nutritional value, rendering food unsafe for consumption. To mitigate these issues, active packaging incorporating antimicrobial properties is being developed to inhibit microbial growth and extend shelf life. [5-7]

A promising natural additive for such applications is orange peel powder, a by-product constituting nearly half the weight of citrus fruits. It contains bioactive compounds such as flavonoids, pectin, terpenoids, and limonene, which exhibit potent antioxidant and antimicrobial activity. When incorporated into polymer matrices like LDPE or LLDPE, orange peel powder enhances mechanical properties, recyclability, and resistance to degradation, while contributing biodegradability to the composite films. Utilizing orange peel as filler supports environmental sustainability by transforming fruit waste into value-added packaging materials. [7-9]

2. Experimental Procedures

2.1 Materials used

The following materials which used in the preparation of LLDPE film in this research work.

S. No.	Materials	Function	Address
1	Linear Low- Density Polyethylene LLDPE (Grade -010F18S)	Matrix	IOCL
2	Orange Peel Powder 40 mesh (425 μ m) t	Filler	Self- Prepared

Linear Low-Density Polyethylene (LLDPE) grade 010F18S, procured from Indian Oil Corporation Limited (IOCL), was used as the base polymer matrix due to its excellent flexibility, toughness, and processability, making it suitable for packaging applications. Orange peel powder, self-prepared and ground to a particle size of 40 mesh (approximately 425 μ m), acted as a natural filler. Rich in bioactive compounds such as flavonoids and limonene, the orange peel powder provides antioxidant and antimicrobial properties, enhancing the functionality and sustainability of the composite material. [10]

2.2 Methodology

For the related Research work five sample were prepared in order to attain the better antimicrobial and mechanical property of the film with different weight composition of orange peel powder as given table below.

S. NO	Samples	LLDPE (kg)	OPP (%)
1	S1	1kg	0
2	S2	1kg	3
3	S3	1kg	4
4	S4	1kg	5
5	S5	1kg	6

During the preparation of film all the granules was dried in the hot air oven at the temperature of 70°C for 24hrs. The preparation of film was done by blown film extrusion. The machine of blown film extruder was from KONARK company from the Gururaj Engineers. The diameter of the screw is 35 mm and the L/D ratio is 40. The advanced technology unique to Gururaj Engineers has been accumulated over many years and results in blown film extrusion more accuracy. [11-13]

Before the preparation of film material were dried in the hot air oven at 70°C for 12hrs. The dried material was fed into the hopper of blown film extrusion melt process. The temperature for melting process was 155-190°C and the speed of the screw was set to 36 rpm. The melted material makes the membrane kind of circle then the air blow to the melted material circle by compressor. [14-15]

2.3 Characterization of orange peel powder /LLDPE film:

The thermal behavior of orange peel powder and composite films which different concentration were investigated by thermogravimetric analysis (TGA 8000) by using Perkin Elmer's PYRIS 1TGA thermogravimetric analyzer. This TGA plots the graph of wt. loss% at different temperatures. Graphs were recorded from ambient to 450°C with a warming rate of 10°C/min in the presence of N₂ (inert atmosphere).

Tensile testing of film: Tensile testing of the fabricated films was carried out using a Instron universal mechanical tester equipped with a 5KN load cell. The tests were conducted at a constant speed of 50 mm/min and maintained at a temperature of 23 ± 2°C. Samples measuring 25mm by 160mm were cut from the extruded film bubble in both the machine direction (MD) and transverse direction (TD) following the ASTM D882-10 standard.

For antimicrobial test, there are many diffusion methods which used such as Disk Diffusion method, agar disk diffusion method, agar well diffusion, agar plug diffusion method etc. The most commonly used method is agar disk diffusion method. In this work the agar disk diffusion method is used for antimicrobial test. the samples were tested against the Staphylococcus (Gram +) bacteria.

3. Results & Discussions

Thermogravimetric analysis (TGA) of orange peel powder (OPP) shows an initial moisture loss at 125°C with about 7.9% weight reduction, followed by major decomposition near 230°C due to esterification and decarboxylation in sample 1. For the LLDPE composites, sample 2 containing 3% OPP exhibited two degradation steps between 370–460°C and 465–560°C, completing degradation at ~570°C. Sample 3 with 14 wt% OPP was stable up to 400°C with total

degradation at 585°C. Sample 4 (5% OPP) showed two degradations at 435–450°C and 455–548°C, decomposing fully at 560°C. Sample 5 (6% OPP) had degradation ranges of 365–445°C and 450–555°C, finishing at 567°C. Initial moisture loss was observed in all samples.

The Tensile strength of LLDPE+OPP composite film of sample code S1 to S5 ranges between 11.2 to 18.75 MPa. The value recorded for neat film is 15.4 MPa. The tensile strength is affected by OPP presence. As the wt% of OPP increased to 2 % the tensile strength also increased but later with increasing % of OPP the tensile strength decreased to 11.2MPa at 2wt% this may be due the poor adherence with LLDPE. As 3 wt% of OPP with 100% LLDPE showed the best mechanical properties i. e. tensile strength showed in figure 2.

Sample prepared by Blown Film Extrusion process with different concentrations were tested for antimicrobial activity by agar disc diffusion method. The best result against gram positive bacteria (*S. Aureus* strain) were obtained by the specimen 5. For *S. Aureus*, Specimen 1, 2, 3, 4 and 5 have the inhibition zone diameters are 6, 10, 14, 17 and 22 mm respectively as shown in fig 2.

4. Conclusion

This research successfully developed composite films by incorporating varying concentrations of orange peel powder (OPP) into a Linear Low-Density Polyethylene (LLDPE) matrix using blown film extrusion. The study confirmed that 3wt% OPP addition yielded the best enhancement in mechanical properties, particularly tensile strength, attributed to improved matrix-filler interaction. Thermal analysis showed the composites exhibited satisfactory thermal stability up to 400–570°C depending on filler content, with typical degradation patterns indicative of the composite nature. Antimicrobial tests demonstrated that films with higher OPP content provided increased inhibition against Gram-positive *Staphylococcus aureus*, underscoring the added functional benefit of the bioactive natural filler.

Overall, these findings indicate that incorporating orange peel powder into LLDPE films not only enhances mechanical strength and thermal stability at optimal filler loadings but also imparts valuable antimicrobial properties, making these composites promising for sustainable packaging applications. Future work could explore optimizing filler dispersion and interface adhesion to further improve mechanical performance and extend application potential. This study contributes to the development of eco-friendly and multifunctional polymer composites combining polymer processability with natural bioactive fillers to meet packaging industry demands without compromising environmental considerations.

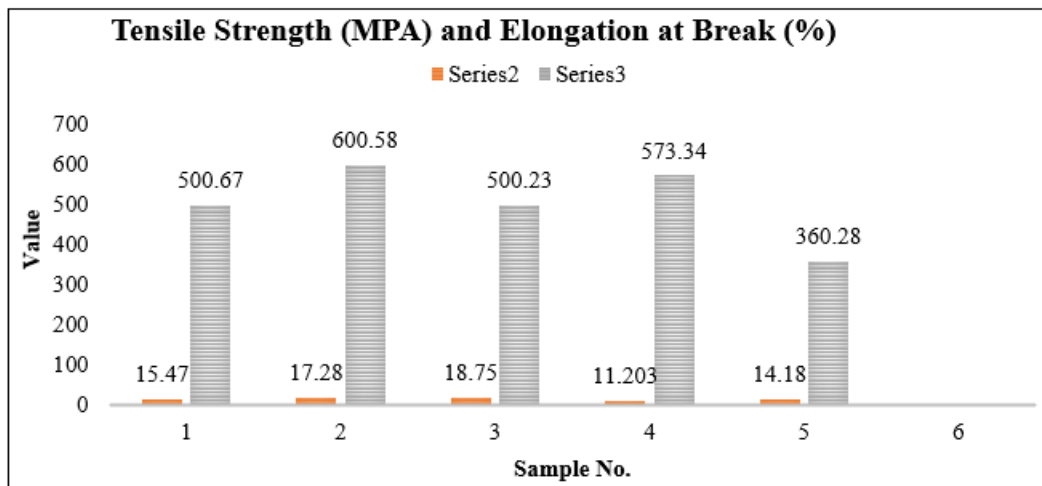


Figure 2: Tensile strength (Mpa) & %Elongation of LLDPE+OPP composite film of sample S1 to S5

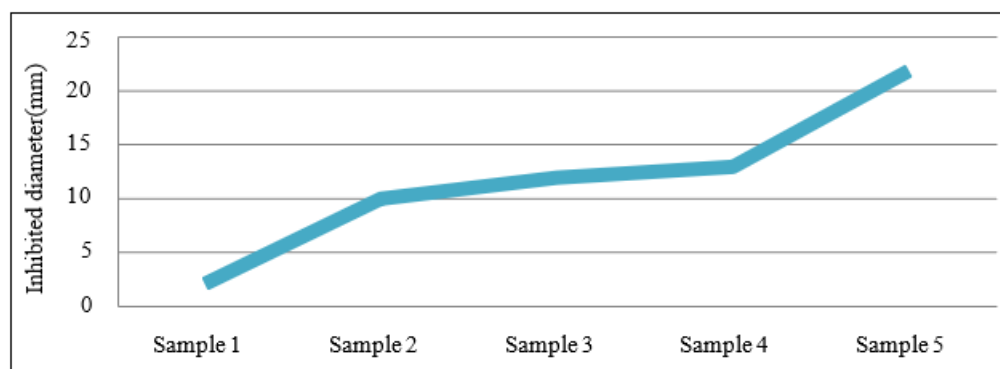


Figure 2: Specimen against S.Aureus Strain

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