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Enhancing Antimicrobial Properties of LLDPE/Orange Peel Powder/Titanium Dioxide Composite Films for Food Packaging Applications

Malika Singh[1]; Rajneesh Singh[2]; Irfan Ahmed Mir[1]

[1]Research Scholar, Central Institute of Petrochemical Engineering & Technology, Lucknow.
[2]Research Scholar, Maulana Azad National Institute of Technology, Bhopal
[1]Senior Technical Officer, Central Institute of Petrochemical Engineering & Technology, Lucknow.

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Abstract: Food-borne microbial outbreaks have become a serious issue worldwide, pushing researchers to find better ways to stop microbes from spoiling food while keeping it fresh and safe. One promising approach is antimicrobial packaging, which actively fights bacteria and fungi, unlike regular packaging, which only physically protects food. Linear low-density polyethene (LLDPE) is a common plastic used for packaging because of its strength and flexibility. This study focused on improving LLDPE films by adding natural and safe antimicrobial agents. Orange peel powder, a waste product rich in natural compounds like flavonoids and essential oils, was used for its natural ability to fight microbes. Using orange peel powder also helps reduce waste and supports eco-friendly goals. To boost the antimicrobial effect, titanium dioxide (TiO2) nanoparticles were added. TiO2 can kill bacteria by producing reactive molecules under light. When combined, orange peel powder and TiO2 work together to more effectively inhibit microbes. The mixture was melted and shaped into films, which retained their strength and allowed air and moisture control ideal for packaging. Tests showed these films could reduce microbial growth on food, keeping it safer for longer. This method offers a sustainable, efficient way to protect food by using natural waste and modern nanotechnology.

Keywords: *Antimicrobial Packaging, Food Safety, Microbial Inhibition, Food Preservation, Sustainable Packaging, Nanofillers.*

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I. INTRODUCTION

Plastics, as polymeric substances, can be formed into various shapes when exposed to heat and pressure due to their malleable nature. Among their many uses, packaging films represent a significant application, playing a crucial role in maintaining food freshness, protecting products, aiding identification, and simplifying transportation. Packaging accounts for a large portion of global plastic usage.

With growing consumer awareness of foodborne diseases, the demand for sustainable packaging solutions that maintain food quality and prevent spoilage has increased. Contamination from bacteria and fungi can alter food's texture, taste, color, and nutritional value, making it unsafe for consumption. To combat these problems, active packaging materials that include antimicrobial agents are being developed to slow microbial growth and extend the shelf life.

A natural additive gaining attention is orange peel powder, a by-product making up nearly half of the fruit's weight. Rich in bioactive compounds such as flavonoids, pectin, terpenoids, and limonene, it exhibits both antioxidant and antimicrobial effects. When incorporated into polymer matrices like low-density polyethylene (LDPE) or linear low-density polyethylene (LLDPE), orange peel powder boosts mechanical strength, enhances recyclability, improves resistance to degradation, and promotes biodegradability of the composite films. Additionally, the inclusion of titanium dioxide nanoparticles in these composites further improves their antimicrobial properties and UV resistance, thereby increasing durability and shelf-life effectiveness. Utilizing orange peel powder alongside titanium dioxide nanoparticles offers an eco-friendly solution by converting citrus waste into valuable packaging materials that meet safety and sustainability standards.

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II. EXPERIMENTAL PROCEDURES

A. Materials:

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

Table 1 Preparation of LLDPE Film Work

Sr. No.	Materials	Function	Address
1	LLDPE (010F18A)	Matrix	IOCL
2	Orange Peel Powder	Filler	SELF PREPARED
3	Titanium dioxide Nanoparticles (50-80 nm)	Antimicrobial agent	NANORESEARCH LAB, Jameshdpur, India

LLDPE 010F18A from IOCL is a linear low-density polyethene known for good processability and strong mechanical properties. It has a melt flow index of 0.9 g/10 min and a density of about 0.918 g/cm³. Its tensile strength is around 13 MPa, with elongation at break up to 850%, making it flexible and tough. This grade is suitable for films like lamination, stretch cling, and drip laterals and meets food safety standards. When combined with self-prepared orange peel powder as a natural filler and titanium dioxide nanoparticles (50-80 nm) from Nano Research Lab, it forms an eco-friendly antimicrobial film with enhanced microbial protection and durability. This composite supports sustainable packaging with improved safety and quality.

B. Methodology:

➢ *Preparation of Orange Peel Powder (OPP)*

Purchased oranges were washed and sundried for 14 days in order to remove moisture. Now the pre dried material was blended into a fine powder, then passed through the sieve mesh 69 to collect the fine powder of orange peel, Fig1 almost the same particle size.

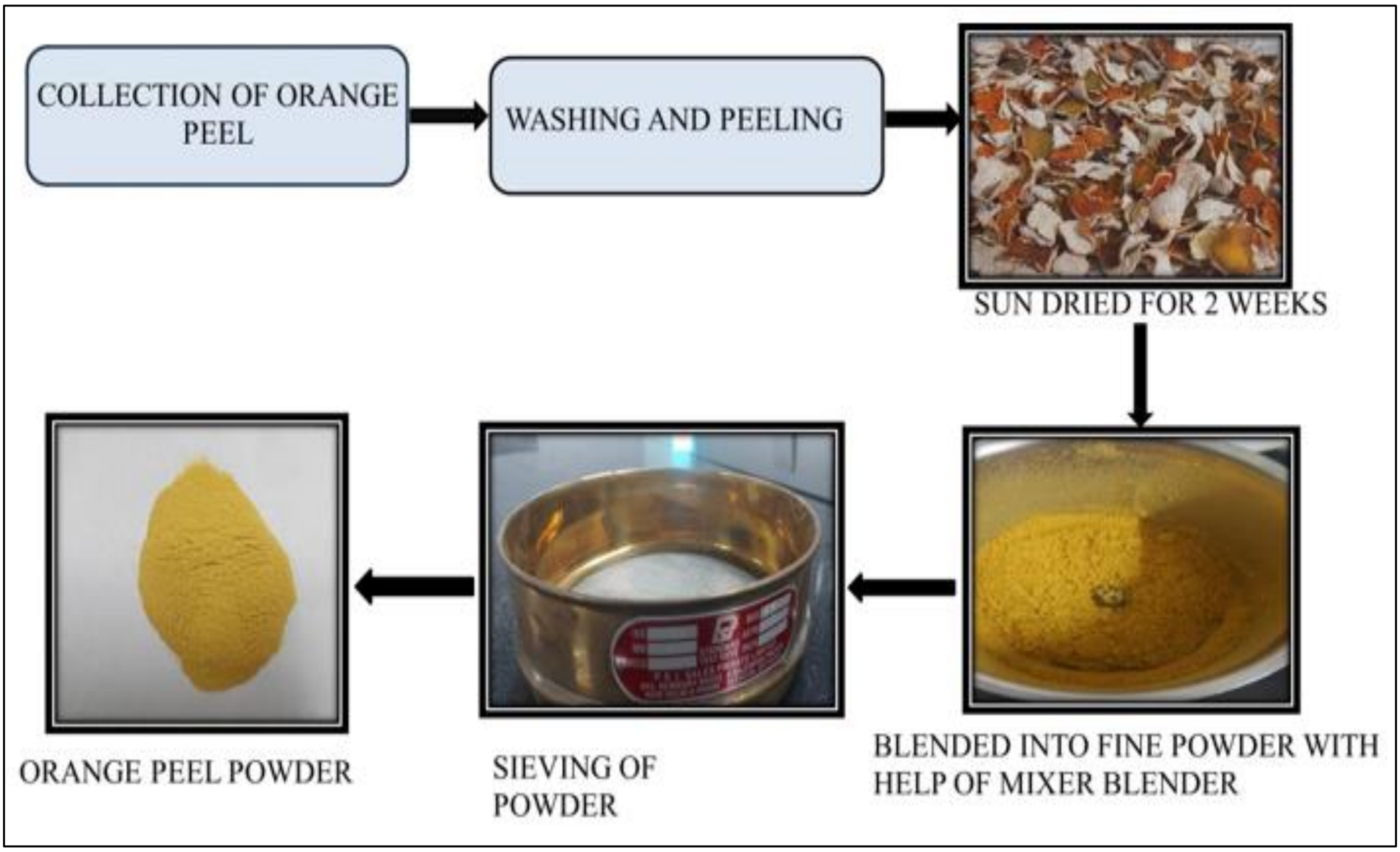

Fig 1 Orange Peel Powder Preparation

➢ *Compounding of the Composite Film*

1% (w/w) of OPP was taken as ideal, and further titanium dioxide nanoparticles in different ratios with LLDPE were mixed, and then the composite film was made of (OPP+TiO_2+ LLDPE), and mixing of material was again done with the help of a twin screw extruder, and film was manufactured by the blown film extrusion process. Finally, tests such as mechanical, optical and antimicrobial test was done to evaluate the properties of the film for the end-use application.

- *Formulation of Composite Film*

Formulation of film shown in Table 2.

Table 2 Formulation of Composite Film

Sample	LLDPE (kg)	OPP(%)	TiO_2 (%)
S1	1kg	1	0.1
S2	1kg	1	0.2
S3	1kg	1	0.3
S4	1kg	1	0.4

- *Preparation of Film from Blown Film Extrusion*

For the preparation of film, the mixture of orange peel and titanium dioxide nanoparticles in LLDPE is subjected to preheating at 80°C for 4 hours and the formulation of the mixture is discussed earlier in table fig 2. Film of four sample with their code name, S1, S2,S3 and S4 is prepared in the blown film. During this process the temperature is maintained in machine for positive flow in different zone of the barrel. The prepared granule with the help twin screw extruder, is fed into the hopper of the blown film extrusion machine. Different temperatures are maintained from one zone to another zone inside the barrel which is discussed in Table fig2. The film prepared for the food packaging application is shown in table 3.

Table 3 Blown Film Extrusion Temperature Zone for Final Film

Feed Zone	Compression Zone		Metering Zone	Die Temperature	
Zone - 1	Zone -2	Zone -3	Zone – 3	Zone -4	Zone – 5
145°C	155°C	165°C	175°C	180°C	195°C

III. TESTING & CHARACTERIZATION OF COMPOSITE FILM

- *Tensile Testing*

Tensile test was performed on the Universal Testing Machine (UTM – Make/type Instron 3360 series, Capacity KN), as indicated by the ASTM-D 882 on testing speed of 500 mm/min. four samples of each sort of film were tried, and their mean value was determined. Every sample was cut into a rectangular shape with dimensions of (15*2)cm.

- *Agar Disc Diffusion Test for Antimicrobial Activity*

Samples prepared by the Blown Film Extrusion process with different concentrations were tested for antimicrobial activity by the agar disc diffusion method. The formulations that were tested are listed in Figure 2. The results of the different concentrations are shown in Figure 2. The samples were tested against E.Coli (Gram –) bacteria, where the circle in Figure 4 shows the inhibition zone.

- *Scanning Electron Microscopy of Composite Film*

The Scanning Electron Microscope from JEOL Ltd., in Tokyo, Japan, were used for detailed surface and structural analysis of composite films. Testing involves preparing the film sample, often coating it with a thin conductive layer to prevent charging. The SEM then scans the surface with an electron beam, producing high-resolution images of the film's morphology, filler dispersion, and nanoparticle distribution. This helps assess the uniformity and interaction of components like orange peel powder and TiO_2 nanoparticles.

- *Optical Properties of Composite Film*

The spectrophotometer used for measuring haze and transmittance in films and plastics typically features an integrating sphere design following ASTM D1003 standards. It measures how much light passes through (transmittance) and the degree of light scattering (haze) caused by the sample. The instrument provides accurate, reproducible results over the visible wavelength range (400-700 nm) .

IV. RESULTS & DISCUSSIONS

The tensile strength of samples S1 to S4 ranged from 14.18 MPa to 23.24 MPa, indicating the influence of OPP content and TiO_2 nanoparticle addition on mechanical properties. Sample S4, containing 0.3 wt% TiO_2 along with 1 wt% OPP and 100 wt% LLDPE, showed the highest tensile strength of 23.24 MPa. This suggests that the addition of TiO_2 nanoparticles enhances the strength of the composite film. Meanwhile, intermediate samples demonstrated tensile strengths between 14.18 MPa and 15.31 MPa, reflecting variations in OPP content and adhesion with LLDPE. The data indicate that tensile strength improves with the optimal amount of OPP (around 1 wt%) but decreases at higher concentrations, likely due to reduced interfacial bonding as shown in the figure 2.

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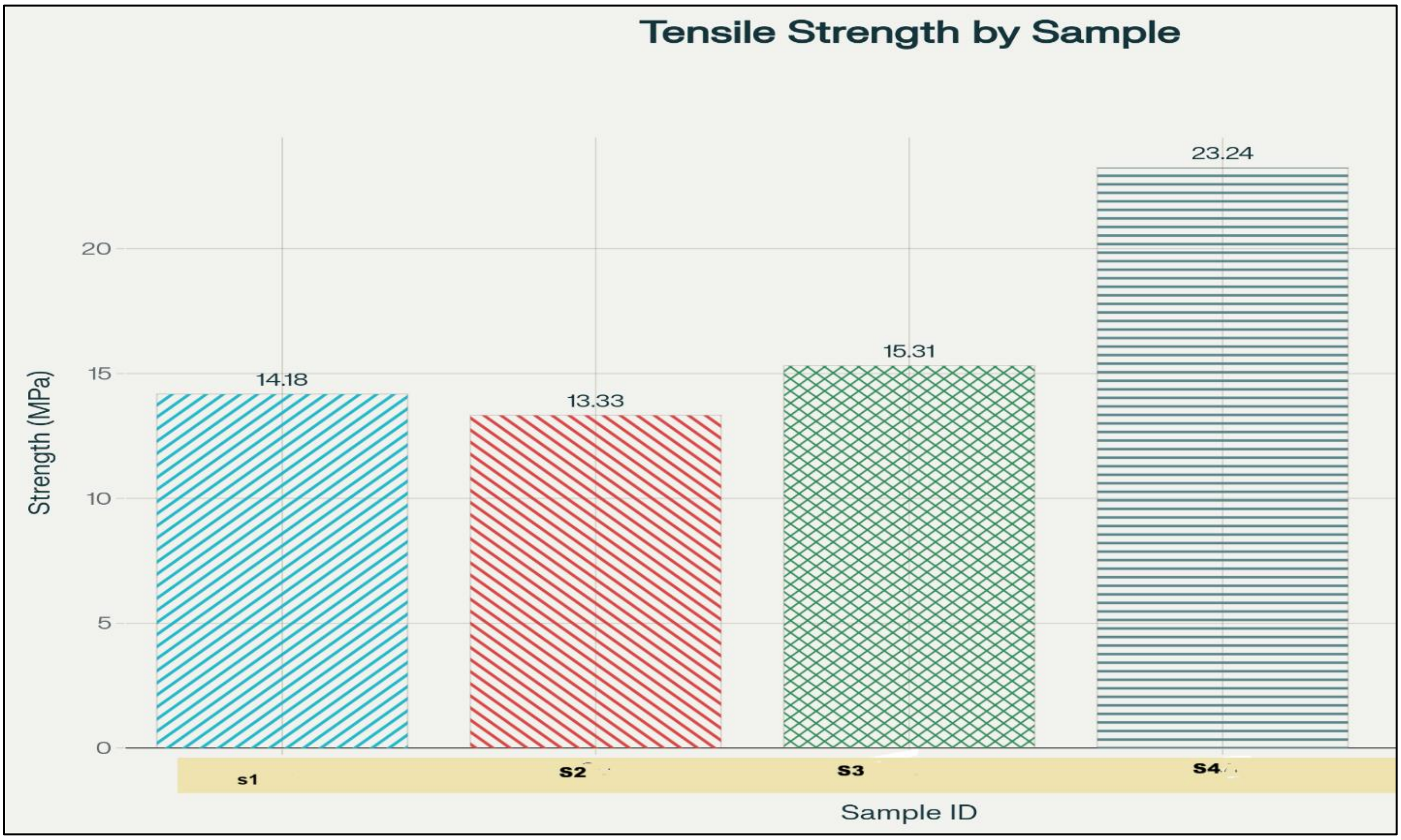

Fig 2 Tensile Strength of Composite Films ((MPa)

➢ *Antimicrobial Testing of the Composite Films*

For this work, the antimicrobial test which we were used is Agar Disc Diffusion Test. The results of antimicrobial activity (which is for gram-negative bacteria) of all the specimen are shown in figure 3. The ciruclar part in the figure shows the inhibited zones against microbe.

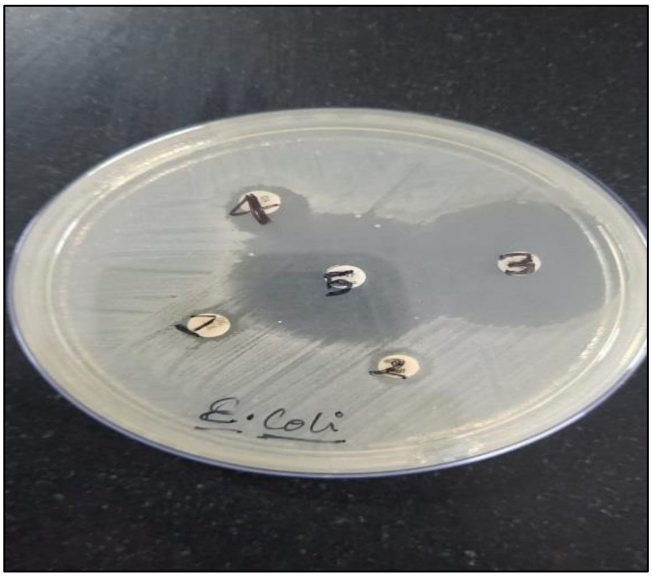

Fig 3 Antimicrobial Activity of All the Specimen Against Gram Negative Bacteria

The best result against gram negative bacteria (E. Coli strain) were obtained by the sample 2 which have a formulation of both orange peel powder and titanium dioxide nanoparticle in LLDPE material. For E. coli, Specimen 1,2,3,4,5 where speciemn 1is virgin lldpe film and specimen2 is sample 1,specimen 3 is sample 2 ,specimen 4 is sample 3 and specimen 5 is sample 4 have the inhibit zone diameters are 7,10,34,14and 33 mm respectively.

➢ *Scanning Electron Microscope*

SEM micrographs for film show slightly rough surface characteristic of titanium dioxide NPs and orange peel powder immobilized LLDPE film. From the micrographs it is also evident that the film looks slightly rough due to the presence of OPP , minute amount of micropores are available on the composite films shown in figure 4 to 7.

Fig 4 SEM Image of 0.1% TiO2NPs

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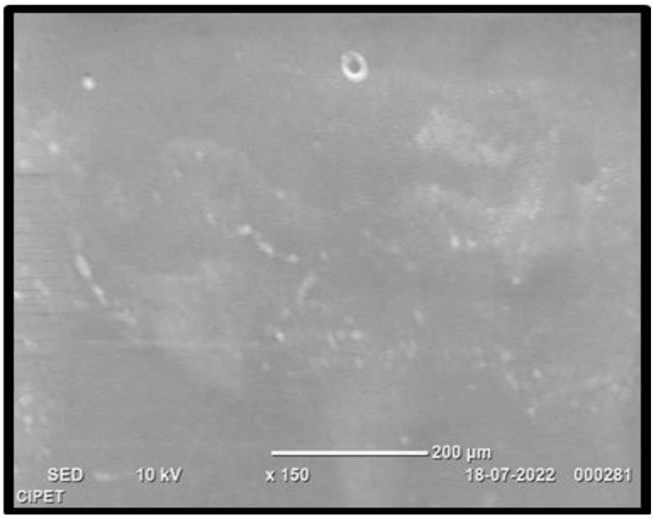

Fig 5 SEM Image of 0.2%TiO2NPs

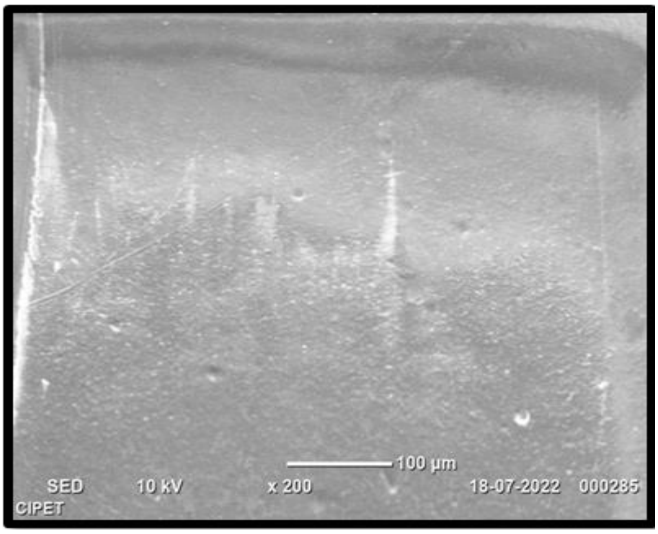

Fig 6 SEM image of 0.3% TiO2NPs

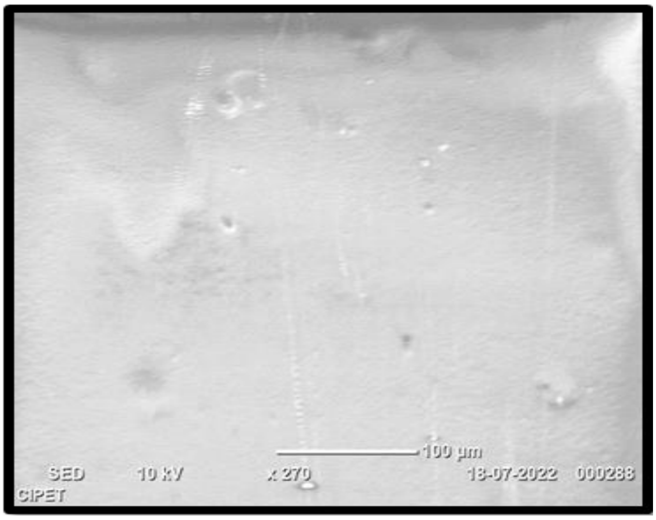

Fig 7 SEM Image of 0.4%TiO2NPs

➢ *Optical Properties of Composite Film*

The optical behaviours of samples S1 through S4 were carefully examined with respect to haze and transmittance. Sample S1 recorded a haze value of 31.4% and transmittance of 88.2%, indicating a balance of light scattering and clarity. Increasing the concentration of OPP and TiO_2 nanoparticles in samples S2 and S3 raised the haze notably to 44.4% and 46.3%, while transmittance dipped slightly to 88.7% and 87.5%, respectively. These changes suggest that added fillers promote more scattering, thereby reducing clarity to some extent. Sample S4 demonstrated a reduced haze at 35.3% and improved transmittance at 89%, implying a better dispersion or interaction of components that mitigated light diffusion. Overall, these trends reflect the delicate trade-off between the composite's composition and its optical transparency, important for tailoring films in packaging to optimize both protection and visibility.

V. CONCLUSION

The present study confirms that the incorporation of orange peel powder (OPP) and titanium dioxide (TiO_2) nanoparticles into a linear low-density polyethylene (LLDPE) matrix leads to notable improvements in mechanical, antimicrobial, and optical characteristics. The composite containing 0.3 wt% TiO_2 and 1 wt% OPP exhibited the highest tensile strength of 23.24 MPa, demonstrating enhanced filler–matrix interaction. Agar disc diffusion results indicated that this formulation also provided the strongest inhibition against E. coli, highlighting its potential for antimicrobial packaging applications. SEM analysis revealed a moderately rough morphology with fine micropores resulting from the uniform distribution of OPP and TiO_2 within the polymer. Optical evaluation further showed that controlled filler loading maintained high transmittance and acceptable haze levels. These findings suggest that optimizing the composition of OPP–TiO_2–LLDPE composites can yield multifunctional films suitable for sustainable and active food packaging

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