

# ZnO/LDPE Nano-composites: Effects of Particle Size and Electrical Resistivity on Mechanical, Electrical, and Thermal Properties: A Review

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## Abstract

Nano-composite materials have been found to have potential in various industrial applications due to their desired novel characteristics. This review explores the influences of zinc oxide (ZnO) particle size and electrical resistivity on several features of low-density polyethylene. (LDPE)/ZnO composites by synthesizing and analyzing findings found in the literature. This work includes a comprehensive analysis of research outcomes, methodologies, and trends related to the incorporation of different ZnO particle sizes into the LDPE matrices. The investigation focuses on the mechanical strength, electrical conductivity, and thermal behavior of the composites. The work systematically analyses and discusses the nanocomposites synthesis and structure on the corresponding mechanical, electrical, and thermal performance of the composites. By providing a comprehensive overview of the existing literature, this work aims to consolidate the current understanding of how ZnO particle size and electrical resistivity influence the multifaceted properties of LDPE/ZnO composites to enhance their implementation in more sustainable industrial applications.

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**Keywords:** Nanocomposites; Particle size; Electrical resistivity; Polymeric composites; Zinc oxide.

## 1. Introduction

The potential of polyethylene (PE) based composites in various industrial applications has encouraged investigating their performance in a wide range of reinforcement conditions. Composite materials are increasingly supplanting traditional materials owing to their superior properties. In many cases, a single component crafted from composites can effectively replace numerous metal parts. The reinforcement of LDPE with nanoparticles has also developed various desired properties for diverse industrial needs. Exploring the high significance of LDPE nanocomposites across various industrial applications has recognized their beneficial attributes, including flexibility, transparency, and chemical resistance [1-5]. The flexibility and advantageous characteristics of LDPE have made it a proper choice of material for packaging, agriculture, and many other fields [3-8]. The integration of zinc oxide ZnO nanoparticles aims to augment the already favorable properties of LDPE, with a specific emphasis on improvements in mechanical, electrical, and thermal

characteristics [9]. This seeks to broaden the range of potential applications for LDPE in response to evolving industrial needs. Emphasizing the potential enhancement of polymer properties through nanoparticle integration, with zinc oxide ZnO serving as a key reinforcing agent. However, addressing challenges in LDPE's mechanical, electrical, and thermal aspects have been found [10]. Analyzing the size impact of ZnO particles and electrical resistivity on the properties of the composite materials is one of the most important issues regarding developing a better understanding of the overall performance [11-16]. Moreover, formulating specific research questions can establish a proper framework outlining objectives to bridge existing knowledge gaps regarding LDPE/ZnO nanocomposites [17-20].

Consequently, this work aims to contribute developing a better understanding of the effect of the reinforcement structure of LDPE/ZnO nanocomposites, specifically the ZnO particle size and their electrical resistivity on the various mechanical, thermal, and electrical performances of the composites. Developing such a proper scientific understanding of the relationship between the reinforcing

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nanoparticles characteristics and the overall performance of such nano-composites was achieved by comprehensively discussing and synthesizing a range of findings and insights obtained by critical researches in the field. Moreover, the authors seek to provide a consolidated and up-to-date overview of how ZnO particle size and electrical resistivity collectively impact the multifaceted properties of LDPE/ZnO nano-composites via reviewing the methodologies utilized to fabricate such composites were provided insight into the subsequent manuscript sections, significantly advancing better understanding of LDPE/ZnO nano composites with potential practical implications in the materials field and polymer engineering.

## 2. LDPE/ZnO nano composites Performance

The performance of the LDPE/ZnO nanocomposites was investigated by several researchers to clarify the effect of several reinforcement parameters. F. Ö. Özmihiç and D. Balköse [21] have conducted research clarifying how some properties of LDPE/ZnO composites are affected by the particle size and electrical resistivity of zinc oxide (ZnO). The nanocomposites were formulated using a rheometer (Rheomixer R600/610) operating at 50 rpm, maintained at 160 °C for duration of 20 minutes. Then the resulting blends were pressed using a force of 6800 kg at 150 °C for 10 minutes with a Carver hot press to obtain sheets with dimensions of 15 cm × 15 cm × 0.1 cm. Micrometric (mZnO), submicronic (sZnO), and nano metric (nZnO) powders with resistivity properties of  $1.5 \times 10^6$ ,  $1.5 \times 10^9$ , and  $1.7 \times 10^8$  were used to prepare composites with 5-20% volume fraction of filler. The X-ray diffractometer (Philips, Cu K $\alpha$  radiation) was employed to ascertain the crystalline structures of both ZnO powders and composites. The electrical resistivity of the composites was assessed in accordance with ASTM D257 standards. This involved the utilization of ZnO pellets, crafted with a diameter of 2.5 cm and a thickness of 2 mm, prepared under a pressure of 10 MPa. Electrical resistivity was measured using a Keithley 6517A electrometer with voltage alternated between 250 and 150 V, and the current was measured. An average value from 8 readings was taken as the resistivity value at the end of the test. The thermal conductivity of the samples was determined through the hot-wire method employing a KEM QTM-500 thermal conductometer.

The composites' morphology was studied by examining fracture surfaces obtained post-rupture following immersion in liquid nitrogen, utilizing a scanning electron microscope (Philips XL-30S FEG). Tensile tests were conducted at room temperature using a Shimadzu AG-I 25 kN tensile testing machine with a crosshead speed of 50 mm/min, adhering to ASTM standard 638.

### 2.1. ZnO Particles Characterization

The X-ray diffraction patterns of mZnO, sZnO, and nZnO displayed uniformity, confirming that all three variants exhibit the characteristic of typical ZnO structure. The SEM micrographs, on the other hand, explain the shapes and size distributions of ZnO particles. The particles of mZnO exhibited rod, sphere, and tripod shapes, highlighting their polycrystalline nature. The sZnO particles

were rod-shaped, whereas the nZnO particles were nearly spherical. SEM images show that the mZnO particles had a higher aspect ratio (4.35) compared to the nZnO (1.96) and sZnO (2.29) particles. In addition, the resistivity of LDPE decreased from  $2.3 \times 10^{16} \Omega \text{ cm}$  to  $1.4 \times 10^{10} \Omega \text{ cm}$  with the incorporation of mZnO. The surface roughness of the pellets was also assessed. The water contact angle of ZnO pellets ranged between 38 and 47, indicating that all powders were hydrophilic (while LDPE is hydrophobic). Nevertheless, nZnO exhibited the lowest contact angle (38), indicating greater hydrophilicity compared to mZnO (47) and sZnO (45). The introduction of ZnO had an impact on the crystallinity of the LDPE matrix.

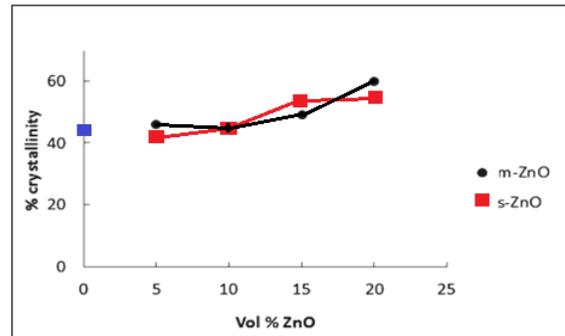


Figure 1. X-ray crystallinity versus volume % of ZnO in the composites [21]

### 2.2. Mechanical Properties of the Composites

The incorporation of ZnO powder had a significant effect on the mechanical properties of the composites, causing a decrease in the Young's modulus and an increase in the elongation at break [22-24].

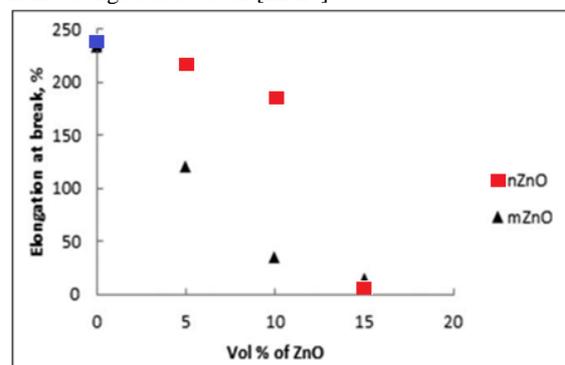


Figure 2. Elongation at break of the composites with volume % of ZnO [21]

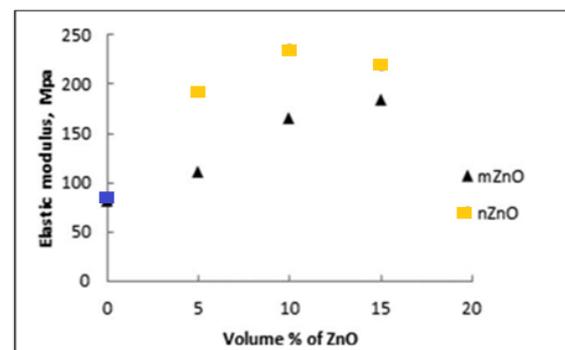


Figure 3. Change of the elastic modulus of the composites with volume % of ZnO [21]

### 2.3. Electrical Resistivity of Composites

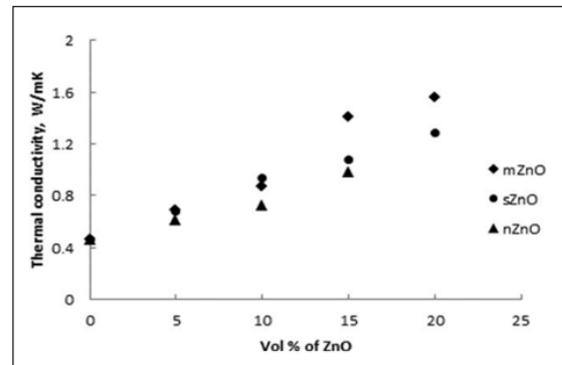
The nanocomposite's electrical conductivity was found to be influenced by several factors and a homogeneous distribution of charges was noted in the composites. The introduction of ZnO resulted in an overall increase in conductivity [25]. The composite's electrical conductivity is influenced by both the inherent electrical conductivity of the filler and the aspect ratio of the reinforcement.

### 2.4. Thermal Conductivity of the Composites

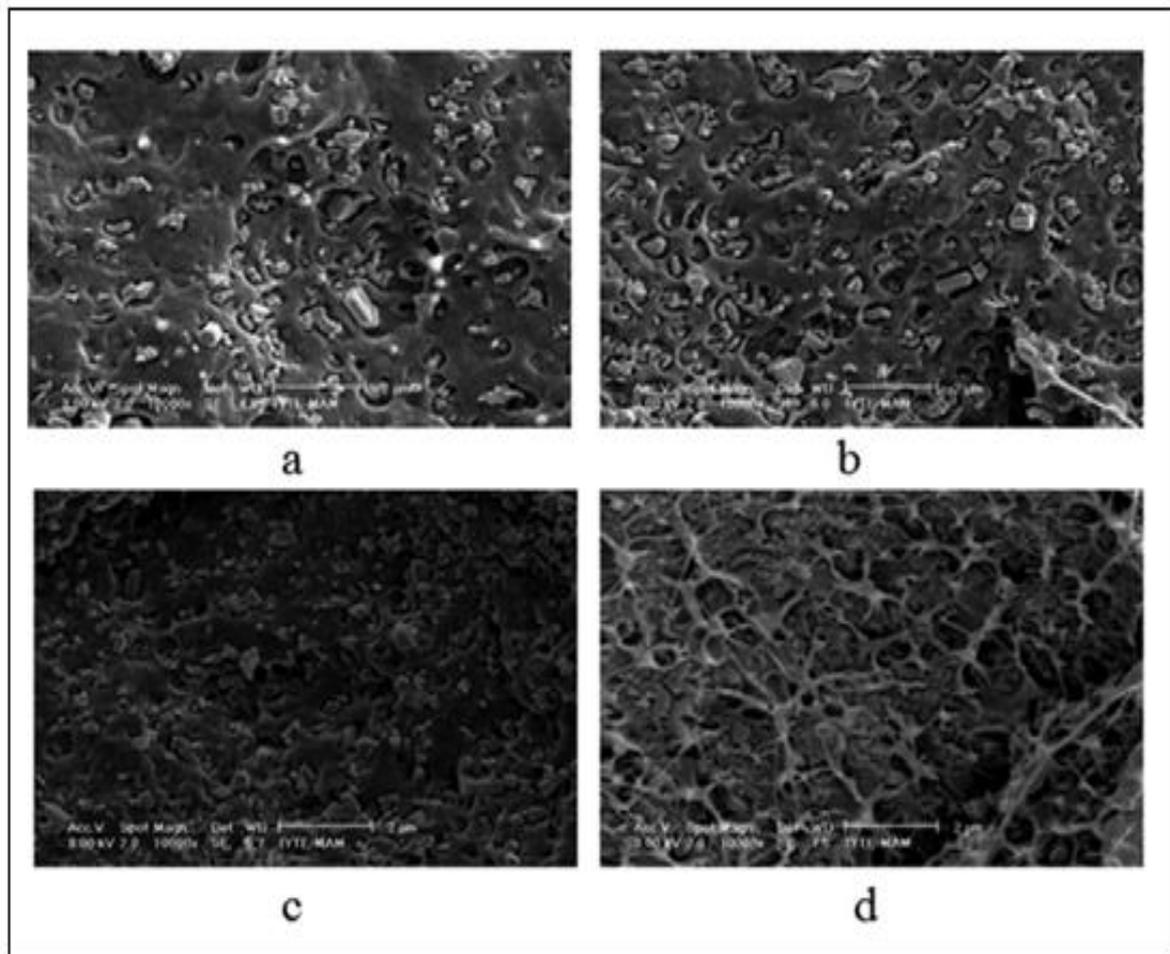
The incorporation of ZnO led to an escalation in the composite's thermal conductivity, reaching 2.5 to 3 times that of the pristine polymer. These composites hold potential for applications involving electrostatic dissipation and thermal dissipation, owing to their low electrical resistivity and heightened thermal conductivity.

Moreover, Yujia Cheng et al. [26] have investigated the effect of different size ZnO particle doping on the dielectric properties of LDPE composites. The ZnO particles with diameters of 30 nm and 1  $\mu\text{m}$  were incorporated as inorganic fillers. The ZnO nano particles after surface

treatment achieved uniform dispersion into the matrix. The Nano-, micro-, and micro-/nano-ZnO/LDPE were prepared by melt mixing. Melt mixing was employed to prepare nano-, micro-, and micro-/nano-ZnO/LDPE blends. The composites exhibited greater crystallinity compared to pure LDPE. The effective enhancement of polymer conductivity characteristics resulted from the micro- and nano-sized ZnO particle doping.



**Figure 5.** Change of the thermal conductivity with volume % of ZnO in composites [21]



**Figure 4.** sZnO-LLDPE composites SEM micrographs for (a) 5 volume %, (b) 10 volume % (c) 15 volume %, and (d) 20 volume % sZnO [21]

## 2.5. Electrical Conductivity

Based on the conductivity properties analysis, doping micro- and nano-sized ZnO particles into LDPE revealed a noticeable impact on interface traps, leading to decreased carrier mobility. The current density across all composites remained lower than that of pure LDPE, with the micro-/nanocomposite exhibiting an intermediate value between micro-composite and nanocomposite. At varying temperatures, the current density of LDPE and its composites demonstrated a linear increase with temperature. Notably, LDPE exhibited the highest current density. Nano-ZnO particles, being polar materials, introduced a new polarization type when doped into LDPE, as evidenced by the dielectric frequency spectrum test. The dielectric constant of nano-ZnO/LDPE surpassed that of pure LDPE, while the dielectric loss of all composites exceeded that of pure LDPE [26].

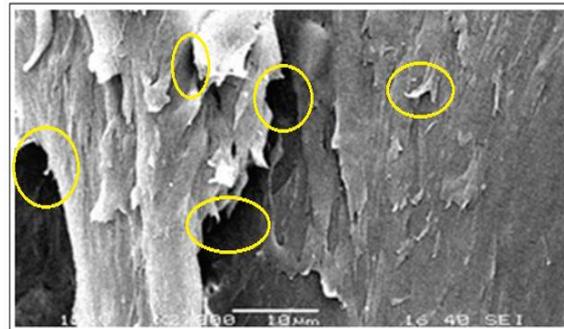
## 2.6. Differential Scanning Calorimetry (DSC)

The results of the DSC test and the influence of ZnO particle doping of different sizes on the crystalline properties of the composites are depicted in Figure 6. LDPE demonstrates a melting temperature of roughly 110°C. According to the results presented in Figure 6, it's evident that the introduction of micro- and nano-sized ZnO particles did not significantly alter the melting peak temperatures of micro- and nanocomposites compared to LDPE; however, micro-composites exhibited a slightly higher melting peak temperature. Additionally, the width of the exothermic crystallization peak ( $\Delta T_c$ ) in micro- and nanocomposites was narrower than in pure LDPE, indicating an enhanced crystallization rate due to micro- and nano-ZnO doping. Although all composites showed improved crystallinity, the extent varied. Specifically, the order of crystallinity among different samples was as follows: 30 nm > 30 nm/1  $\mu$ m > 1  $\mu$ m > LDPE. The crystallinity of micro-/nanocomposites fell between that of micro-composites and nanocomposites [26] (multiple peaks are observed in the graph).

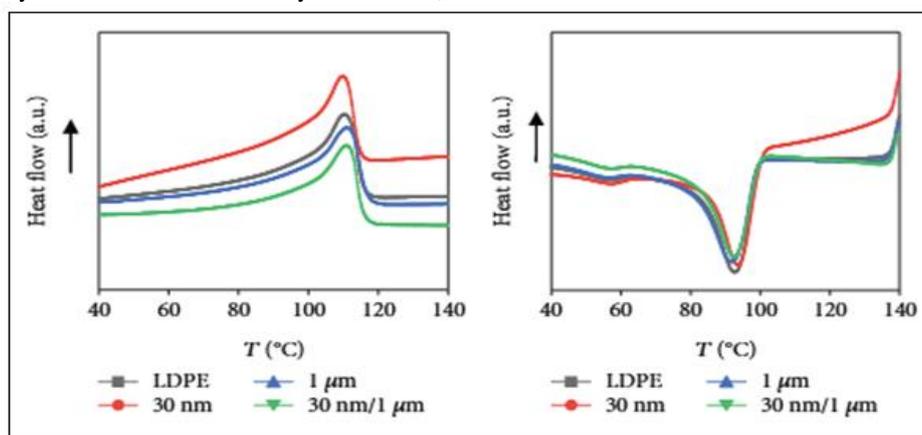
Based on the DSC results, the doping of ZnO particles into LDPE acted as a heterogeneous nucleation agent, enhancing the crystallization rate of the composites. Consequently, the crystallinity of the composites surpassed that of pure LDPE. This increase in crystallinity was accompanied by a rise in the fraction of crystalline areas,

while the amorphous regions exhibited a more circuitous structure. Notably, among the composites, the crystallinity of nano-ZnO/LDPE exhibited the highest enhancement, being 16.1% greater than that of pure LDPE [26]. Furthermore, Halim Hamid Redhwi et al. [27] have studied the effect of ZnO on LDPE properties. LDPE/ZnO nanocomposites were prepared to investigate the impact of the nano-filler on various properties, including mechanical, thermal, and microscopic properties.

A Theyson TSK twin-screw extruder was employed to disperse the nano-filler within LDPE at a 5% weight fraction of the filler, operating at temperatures ranging from 190 to 205°C. Specimen preparation was carried out using an ENGEL injection molding machine at a temperature of 215°C and a pressure of 5000 psi. Tensile testing was conducted following ASTM D638 standards, employing a model 3367 tensile testing machine. Microstructural characterization was performed using a JEOL JSM-6064LV scanning electron microscope (SEM). According to the FTIR spectrum, the incorporation of ZnO results in only a slight change in the peaks compared to the spectrum of virgin LDPE. However, upon comparing the FTIR spectrum of LDPE/ZnO nanocomposites with the reference LDPE sample, an absorption band at 2364  $\text{cm}^{-1}$  attributed to  $\text{CO}_2$  becomes apparent. Based on the DSC analysis, the glass transition temperature is not affected by the addition of the filler (i.e., the DSC of virgin LDPE and LDPE/ZnO appear to be identical). The assessment of reinforcement effectiveness is not solely linked to charge-matrix interactions but also to the dispersion of the filler into the polymer. This was verified by the following SEM image [27].

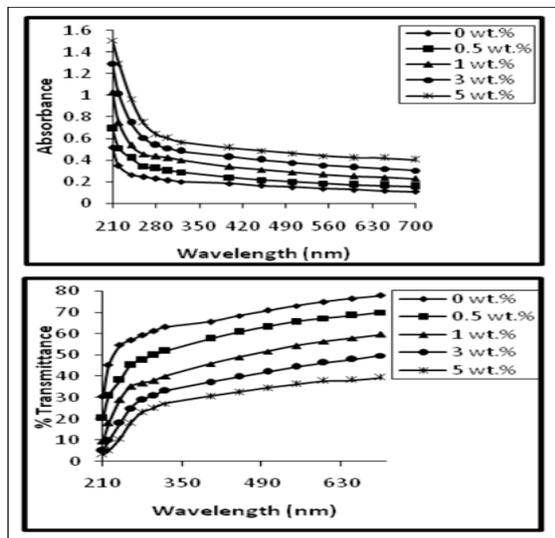


**Figure 7.** Scanning Electron Micrograph of LDPE/ZnO nanocomposite at 5% by weight [27]



**Figure 6.** DSC test curve of composite materials [26]

In that study, thin films of low-density polyethylene/zinc oxide nanocomposites at different weight contents (0, 0.5, 1, 3, and 5%) were prepared by Vijaya S. et al.[28]. In addition, thin films of ZnO/LDPE nano composites were elaborated by solvation. The LDPE was dissolved in xylene using a magnetic stirrer on a hot plate at 100 °C for 2 hours. ZnO was added to the LDPE solution with varying weight proportions (0, 0.5, 1, 3, and 5%). The solutions were dried in an oven at 80 °C for 1 hour. According to the obtained spectra, the absorption was more significant for the progressively loaded LDPE-ZnO samples (meaning that the absorption increases as the loading percentage of ZnO increases) than for the virgin LDPE samples. The absorption spectra reveal that the chemical structure of the polymer underwent no changes following the incorporation of ZnO nanoparticles. It has been illustrated that with the increase in the concentration of ZnO nanoparticles, the transmittance decreases significantly, indicating the effect of ZnO incorporation in the LDPE/ZnO nano composites.



**Figure 8.** The optical transmittance spectra of LDPE and ZnO/LDPE thin films with different concentrations of ZnO in LDPE [28]

The strawberry is a fruit highly susceptible to mold and fungi. Its shelf life once harvested is approximately 5 days at low temperatures (0–4 °C). Up to 40% of strawberries can experience quality loss and spoilage during storage. For this reason, numerous strategies have been developed to address this issue. Furthermore, a study conducted by A. Emamifar and M. Mohammadzadeh [29] studied the enhancement of the preservation and prolongation of the shelf life of fresh strawberries at 4 °C by employing LDPE packaging materials infused with ZnO nanoparticles. In a twin-screw extruder from GmbH & Co. KG, Duisburg, Germany, nanocomposites with different percentages of antimicrobial compounds (1, 3, and 5% by weight) along with 10% by weight of PE-g-MA were prepared. The recovered samples were dried overnight in a vacuum oven at 50 °C. Samples were then compressed using a heated press at 180 °C to obtain various films. Structural characterization was conducted using a transmission electron microscope (TEM), and mechanical characterization was performed using a tensile testing machine (ASTM D882-12). Fresh strawberry packaging was created using a manual heat sealer, incorporating antimicrobial nanocomposites and pure LDPE films, each measuring 15 cm × 10 cm.

The TEM images of LDPE/ZnO nanocomposites with 1, 3, and 5% by weight of ZnO showed that the nanoparticles of the filler exhibited aggregates, likely attributed to strong attractions between ZnO nanoparticles. The TEM image of the LDPE/ZnO nanocomposite (3% by weight of ZnO + 10% PE-g-MA) revealed well-dispersed nanoparticles within the polymeric matrix. The effective dispersion is facilitated through the inclusion of PE-g-MA, which serves not only as a compatibilizer but also prevents the formation of aggregates. Tensile strength, elongation at break, and Young's modulus were determined based on the stress-strain curves of the LDPE/ZnO nanocomposites. A notable enhancement is evident in the Young's modulus of all LDPE/ZnO nanocomposites, accompanied by a substantial reduction in elongation at break across all tested films (indicating improved rupture stress). In addition, the introduction of PE-g-MA in the nanocomposites, containing 3% by weight of ZnO as a compatibilizer, resulted in improved rupture stress, attributed to significant interfacial adhesion and interaction between the LDPE matrix and ZnO nanoparticles.

It was observed that after packaging fresh strawberries, colonies of yeast, mold, and bacteria developed (microbial population) during 16 days of storage at 4 °C. The microbial population of strawberries increased rapidly day by day in all packaging films based on pure LDPE. The incorporation of ZnO nanoparticles into the packaging films hindered the growth of the microbial population during storage [30–32], with an increase in the mass fraction of ZnO nanoparticles to 5%, the antimicrobial activity of the films was further heightened. Moreover, the introduction of 10% PE-g-MA to LDPE films with 3% by weight of ZnO led to a marked reduction in the microbial population compared to films with an equivalent ZnO nanoparticle fraction. This reduction is ascribed to the effective dispersion of nanoparticles in the matrix and the robust interaction between the LDPE matrix and ZnO nanoparticles.

Conversely, A.A. Alluhaibi and R. Kh.Sendi [33] carried out a study elucidating the impact of plasticizers and the concentration of zinc oxide nanoparticles on the properties of polyethylene thin films. Researchers have expressed interest in the combination of nanomaterials with polymers. The use of nanostructured block copolymers in advanced systems has exhibited the ability to achieve structuring at nanoscales, leading to the emergence of novel properties and potential synergistic effects. This renders such systems intriguing and effective [34]. Due to ZnO's ability to filter UV rays, it has found extensive application in cosmetic products, particularly sunscreen lotions.

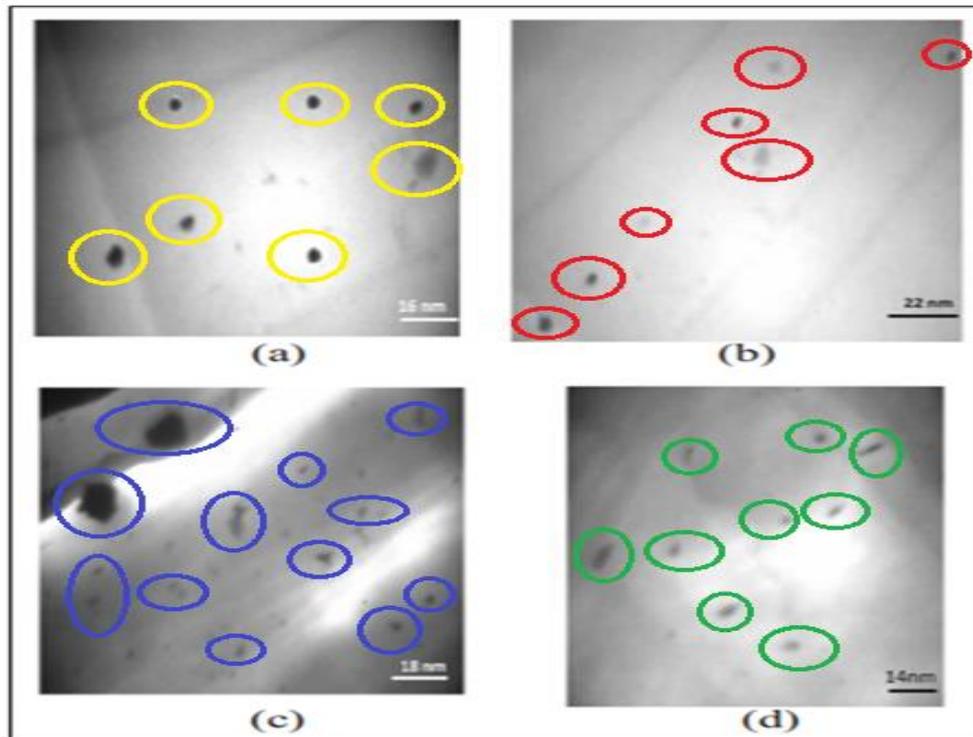
LDPE, acknowledged for its semi-crystalline nature, is employed in manufacturing insulating materials for electric power cables, capitalizing on its distinctive blend of mechanical and electrical characteristics [35]. However, other polymeric composite materials are utilized in various industrial applications due to their beneficial mechanical, electrical, and thermal properties [36–40]. Investigating the optical absorption spectra of both non-crystalline and crystalline materials provides valuable insights into their band structure and energy gap [41, 42]. ZnO nanoparticles were added to LDPE samples in concentrations ranging from 1% to 5%. The LDPE was weighed, dripped onto ZnO nanocomposites, and the mixture was homogenized after heating to 160 °C. Thin films were produced by coating

the mixture onto a slide using a rotary device and annealing at 120°C. The thin films obtained were subsequently cut into smaller pieces for analysis.

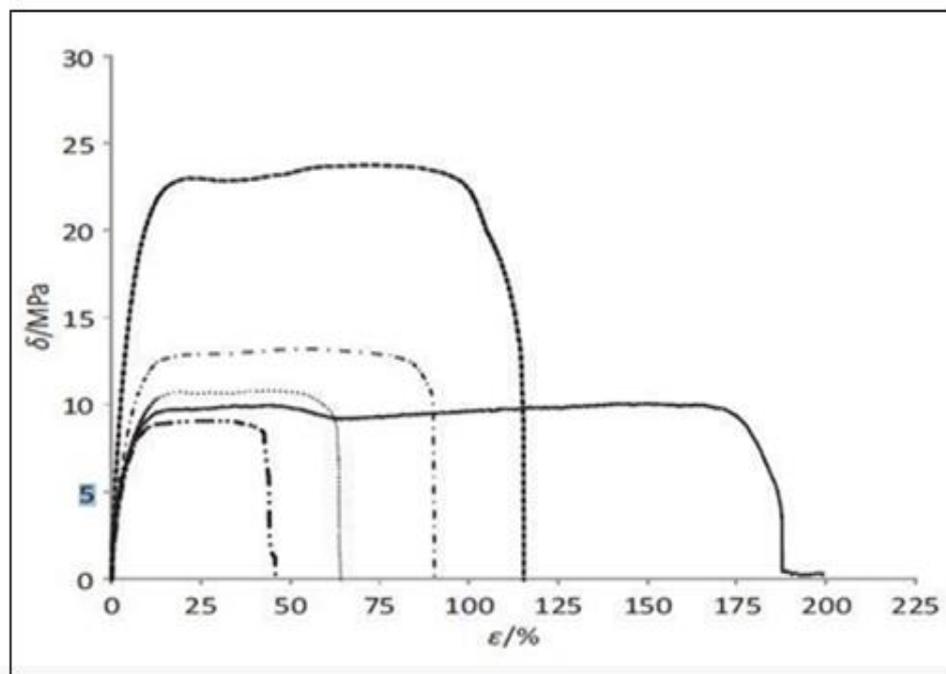
### 2.7. Scanning Electron Microscopy (SEM)

The films prepared in the study displayed excellent physical stability and adherence to LDPE substrates, with

no observed cracking or peeling after deposition. SEM micrographs depict LDPE/ZnO thin films, presenting as white dots against a grey background. The grain size exhibited an increase with thickness, reaching 100 nm. Remarkably, the thin film showcased a surface featuring discernible ZnO, mitigating the roughness observed on the pre-plasticized polymer's surface.



**Figure 9.** TEM Images of Films: a) LDPE/ZnO Film (1% by weight), b) LDPE Film (3% by weight), c) LDPE Film (3% by weight) + 10% PE-g-MA, and d) LDPE Film (5% by weight) [29]



**Figure 10.** Stress-strain diagrams for pure LDPE film (—), LDPE film with 1% by weight of ZnO (- • • -), LDPE film with 3% by weight of ZnO (- • -), LDPE film with 5% by weight of ZnO (•••), and LDPE films with 3% by weight of ZnO + 10% PE-g-MA [29]

2.8. Transmission Electron Microscopy (TEM)

This microscopy revealed that ZnO nanoparticles are extensively distributed in NPs. However, at a 1% mass fraction of ZnO nanoparticles, slight agglomeration occurred (Fig. 1a), likely due to strong nanoparticle attraction. TEM images of an NP LDPE film containing 3%

ZnO nanoparticles exhibited a uniform distribution, with aggregates spanning from 10 nm to 20 nm (see Figs. 1c and 1d). Upon increasing the mass fraction to 5%, the quantity of agglomerates rose, and their volume exhibited greater irregularity (see Fig. 1d). This occurrence is attributed to the diverse morphological and chemical properties of the nanoparticles.

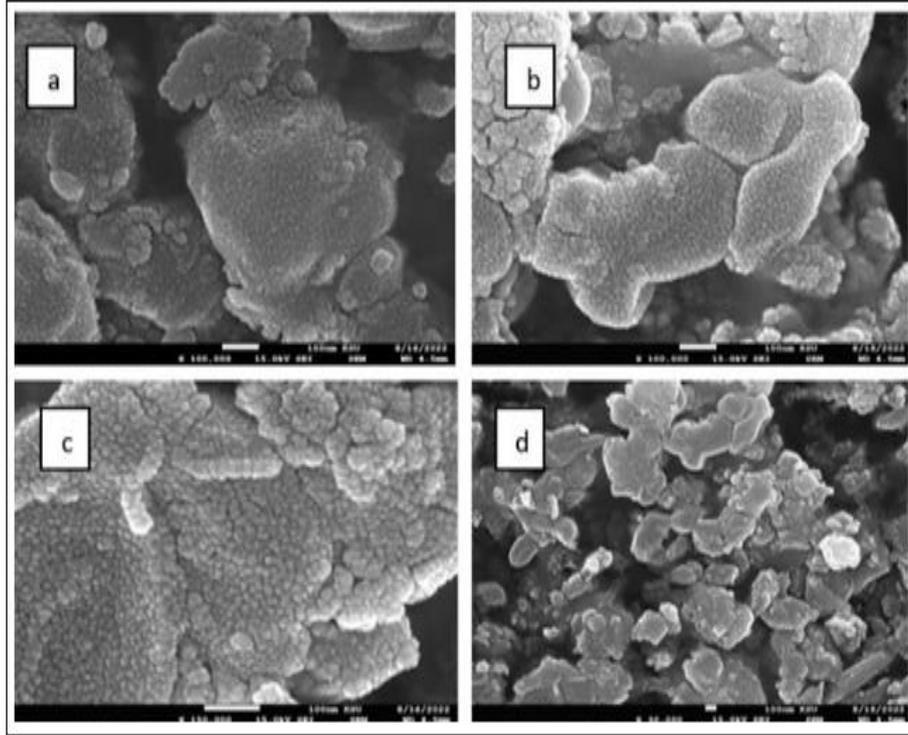


Figure 11. (a, b) white clouded clots with irregular distribution for ZnO/LDPE thin Film.(c, d) SEM micrographs for the surface of LDPE/ZnO for 100nm of LDPE/ZnO thin film [33]

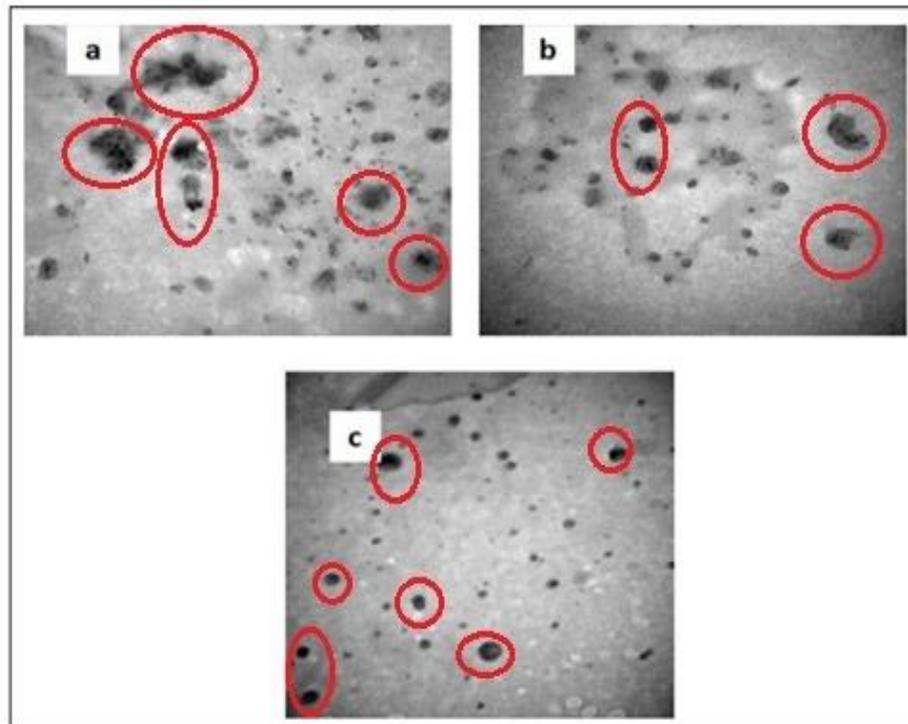


Figure 12. TEM images of thin film: a) LDPE films with 1 % ZnO nanoparticles, b) LDPE films with 3 % ZnO nanoparticles, c) LDPE films with 5 % ZnO nanoparticles[33]

### 2.9. Electrical Properties of LDPE/ZnO composites

The electrical properties of polymeric composites with organic and inorganic fillers are of paramount importance for various industrial applications [43-45]. To explore the impact of inorganic nanoparticle fillers on the electrical properties of low-density polyethylene (LDPE), Fuqiang Tian et al. [46] prepared LDPE/ZnO nanocomposites with varying filler loadings through a melt blending method and examined their electrical characteristics. The study revealed that the trap density in the nanocomposites increased 3 to 5 times compared to pure LDPE. Additionally, the conduction current of the nanocomposites was reduced to 0.5 to 0.25 of that in LDPE across both low and high electric fields. Electrical breakdown strength was observed to improve at lower filler contents but decrease at higher concentrations (>1 wt%). Furthermore, the electrical treeing lifetime of the nanocomposites was extended up to 50 times compared to pure LDPE. Both homo- and hetero-space charges were significantly suppressed in nanocomposites with 0.5 and 7 wt% ZnO content. These findings suggest that enhancements in electrical properties, particularly in charge transport and space charge suppression, are closely associated with the substantial increase in deep trap density. Additionally, S.C. Tjong et al. [47] investigated the dielectric and conductivity properties of LDPE/ZnO composites as a function of ZnO volume fraction and frequency. This study utilized ZnO powders of 200 nm and 2 µm as reinforcing particles, and the composites were prepared using a melt mixing process.

The results indicated that the dielectric constant of the composites, which were reinforced with micro- and nanoparticles, increased almost linearly with the ZnO volume content. In contrast, the resistivity of the microcomposites decreased sharply when the ZnO content reached or exceeded 18 volume %. For the nanocomposites, resistivity began to decrease at a lower ZnO content, specifically 2.8 volume %. The critical interparticle distance between adjacent ZnO particles required to achieve low resistivity was found to be 400 nm. Based on the concept of interparticle distance, the percolation threshold for both micro- and nanocomposites was determined to be 52 volume%.

### 3. The use of zinc oxide (ZnO)-LDPE composites

The ZnO-LDPE composites are generally used as films for the extended storage and shelf lives [18]. Because of the high and very effective antimicrobial activity, the nanocomposites ZnO-LDPE films are used as packaging materials and antimicrobial packages. Moreover, the ZnO/LDPE nanocomposites films have a very effective antimicrobial activity because of good dispersion of nanomaterials in the polymer matrix [18, 33]. Since ZnO nanoparticles are thermally stable, the thermal processing is used to produce the LDPE films. In addition, the nanoZnO-LDPE composites are used in the electrical insulation [26] as the ZnO/LDPE composites exhibited a higher dielectric constant and dielectric loss factor than pure LDPE.

## 4. Conclusions

The systematic review of the ZnO LDPE nanocomposites shows clearly that adding treated nanoparticles to polymer matrices makes a big difference in the properties of the materials. The utilization of stearic acid-treated titanium dioxide in recycled polypropylene demonstrated enhanced rheological, mechanical, and thermal characteristics, showcasing its potential for diverse industrial applications. Moreover, the addition of zinc oxide nanoparticles in polyethylene matrices, especially when aided by PE-g-MA for dispersion, exhibited promising changes in optical and mechanical properties. In the context of strawberry packaging, LDPE films incorporating ZnO showcased antimicrobial features, effectively inhibiting microbial growth during storage, with additional benefits observed through the inclusion of PE-g-MA. These findings collectively emphasize the versatility and performance enhancements achievable by tailoring formulations with treated nanoparticles in polymer matrices, opening avenues for advanced applications across various industrial sectors. Consistent with the hypothesis, incorporating ZnO into polyethylene films was undertaken to produce a thin film endowed with water-resistant features and a resilient moisture barrier. This engendered enhanced combined effects due to the combined optical and mechanical properties of the materials. The research clarified how nanoparticles were organized within the polymer to form the structure of the thin film. SEM analysis unveiled a relatively uniform dispersion of nanoparticles across the fracture surfaces of the polymer.

### Declarations

**Ethics approval and consent to participate** Not applicable

**Consent for publication** Not applicable

**Availability of data and materials** Not applicable

**Competing interests** The authors declare that they have no competing interests

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