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Use of Zinc Oxide Nanoparticles Incorporated in Polybutylene Adipate Terephthalate for Food Packaging. A Focus on the Impact in Functional and Physic-Mechanical Properties and on Migration Thereof

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Keywords: active packaging | antimicrobial activity | biodegradability | migration | PBAT | physic-mechanical properties | ZnO NP size and morphology

ABSTRACT

Polybutylene adipate terephthalate (PBAT) and zinc oxide nanoparticles (ZnO NP) are among the most studied when it comes to address biodegradability and antimicrobial properties of materials for food packaging. This work presents a critical review of recent scientific literature with a focus on the impact in functional, physic-mechanical properties, compostability and on safety. The properties of the nanoparticles, such as morphology and size reported are reviewed together with the range of concentrations and methods of incorporation in the PBAT matrix. The effect on antimicrobial and antioxidant activities, thermostability, tensile and optical properties, as well as on mass transfer properties (barrier to moisture and gases and migration into foods) is discussed. The properties of PBAT/ZnO depend on particle size, shape and concentration of the ZnO NP. Particles with higher surface area and smallest size are reported to have the best performance (usually the spherical shaped). However, information on the morphology/size of the nanoparticle is often not provided. The typical concentration of NP incorporation ranges from 0.5 to 5%, but the interpretation of results regarding the effect of concentration is highly limited due to poor information on the statistical significance of the results, particularly for low concentrations of incorporation. Another limitation found regards the film production method, because most of the studies are based in films prepared by solvent-casting and not by extrusion. Antimicrobial activity is directly linked with the concentration of ZnO NP. Most studies applied in vitro experiments and not tests in real foods (in situ). Zinc migration can occur as the whole nanoparticle or in the ionic form. Studies indicate that migration of ZnO in the nano form is not expected if the nanoparticle is completely embedded in a polymeric matrix, which can be confirmed by some well-established techniques. However, studies focusing on migration from other matrices are lacking. Further investigations are needed to achieve a balanced incorporation of ZnO NP considering the particle activity and zinc migration and to evaluate the impact on safety when used in food packaging.

1 | Introduction

Bio-based materials, and biodegradables in particular, are generally perceived by consumers as a solution for more sustainable packaging [1]. The main applications of biodegradable plastics are currently in (food) packaging, food service ware and shopping bags, that is, low demanding applications, with short contact time and often no need for integrity (sealing). Polybutylene adipate terephthalate (PBAT), a synthetic biodegradable polyester, is the main biodegradable material commercially used today for flexible packaging. PBAT and its blends have raised much attention of researchers and industry. They are targeted in many studies highlighting its biodegradability and dedicating efforts to improving its functionality for food preservation, including antimicrobial active packaging.

Zinc oxide (ZnO) is a metal oxide currently found in daily life applications, including cosmetics (for UV light scattering), medical facilities (e.g., antibacterial paints), dentistry (for blocking microbial leakage) and orthopaedics (as a reinforcing material) [2]. ZnO is listed as a Generally Recognised as Safe (GRAS) material by the Food and Drug Administration (FDA) [3]. ZnO nanoparticles (ZnO NP) have received a positive safety evaluation from the European Food Safety Authority (EFSA) for packaging applications as transparent ultraviolet light (UV) absorbers based on the absence of a significant migration in particulate forms [4, 5]. The potential for diverse applications of ZnO NP is due to the variety of shapes, sizes, crystal forms and properties that can be obtained by different synthesis methods [2, 6].

ZnO NP have demonstrated antimicrobial properties with potential applications in food preservation after combination in different materials. The incorporation of antimicrobial agents into the food packaging matrix eliminates the direct addition of these to the food in high concentrations. There is a high number of published studies dealing with the synthesis and the antibacterial potential of ZnO nanostructures, indicating the relevance of this topic [7]. It is known that the antibacterial activity of ZnO is inversely proportional to the particle size (larger surface area) and directly proportional to the concentration [8]. ZnO NP of smaller sizes can easily penetrate bacterial membranes due to their large interfacial area, thus enhancing their antibacterial efficacy. Therefore, controlling the size and concentration of ZnO NP is crucial to achieving the best bactericidal response [9]. ZnO morphology is determined by the synthesis conditions and it is essential to control parameters such as solvents, precursor types and physicochemical settings such as temperature and pH [9].

With the increase in nanoparticles (NP) uses and applications, the concern with safety issues also increases. When incorporated in some food packaging matrixes, NP or their constituents can migrate. The impact this will have on the safety and quality of the product is of most importance. The nanomaterials' potential hazard associated with food (oral route) is addressed in the EFSA Guidance [10]. However, there is a lack of data to estimate exposure needed for risk assessment [11]. NP have a high surface area to volume ratio, and the active surface chemistry of some nanomaterials could give rise to unwanted chemical reactions compared with conventionally sized particles [12]. Nanomaterials should be evaluated case-by-case basis and the properties of the NP, such as particle size, physical form and morphology, chemical and photocatalytic activity, should be known and also characterised after being incorporated in the matrix [13].

Polybutylene adipate terephthalate and ZnO are among the most studied when it comes to addressing biodegradability and antimicrobial properties for food packaging. This work presents a critical review of recent developments of PBAT based materials with ZnO NP for food packaging with a focus on the impact on functional, physical and mechanical properties and on migration thereof. The range of concentrations, methods of incorporation in the matrix and the effect of particles size and shape, are addressed, as well as a brief consideration of the impact on the compostability of the materials at the end-of-life.

2 | Morphology and Crystallinity

ZnO NP can occur in zero-, one-, two- and three-dimensional structures. The zero-dimensional structure includes nanospherical morphology [14]. One-dimensional structures make up the largest group, including nanorods [15], needles [16], rings [17], tubes [18], columns [19] and wires [20]. The two-dimensional structures are the nanopellets [21] and plate/sheet [22]. ZnO can also be obtained in three-dimensional structures such as nanoflower [23] and snowflake [24].

Most of the works used scanning electron (SEM) and transmission electron microscopy (TEM) to study the morphology and size of the NP. Figure 1 depicts ZnO NP with spherical, rod and flower shapes. The variety of sizes and morphologies is due to

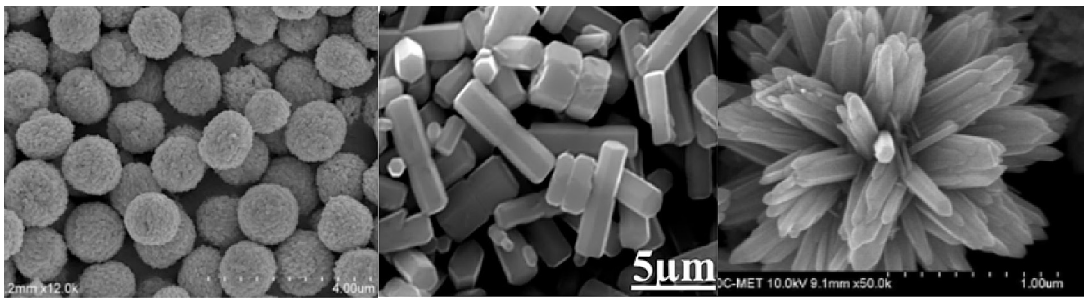

FIGURE 1 | SEM images of ZnO NP. Adapted from: Saikia et al. [25]; Wasim et al. [26]; Yin et al. [27].

the different synthesis techniques. These can be based on chemical, biological or physical methods.

Table 1 presents examples of ZnO NP shapes obtained by different synthesis methods, according to the literature. Chemical synthesis can occur in liquid phase (e.g., precipitation, sol–gel, hydrothermal and solvothermal, sono-chemical) or in vapour phase (e.g., pyrolysis and inert gas condensation). Physical synthesis includes high energy ball milling and laser ablation. The synthesis of ZnO NP mediated by biological resources and wastes is much less explored [37]. Green approaches with plants extracts have received an exponentially growing interest for the ZnO NP synthesis [38]. The use of natural components as stabilising agents can be interesting alternative synthesis methods.

A good dispersion of NP in the polymer matrix is critical for the performance of the nanocomposite. Dispersion can be evaluated by analysing the cross-section of the nanocomposite films by TEM and SEM, as well as the compatibility between the polymer and the nanofiller by observing the adhesion at the particle/polymer interface. Excessively dense distribution (too high concentration) would interfere with the continuity of the matrix and negatively affect the properties of the film [39]. The concentration of NP influences the dispersibility. Ferreira [40] showed that films with 1 wt% of ZnO NP have better NP dispersion compared with those with 3 and 5 wt% of ZnO NP, which show some aggregations. However, in other studies concentrations up to 10% were reported to be well distributed in PBAT matrix [41].

The neat PBAT film usually exhibits a smooth surface, homogeneous and flexible. When NP were incorporated, the film exhibited a rougher surface and agglomerates on the PBAT matrix [42, 43]. Increasing the concentration of ZnO increased the size of the dispersed agglomerates of NP. Nanocomposite film surface roughness increased from 98.3 to 135.5 nm by increasing the ZnO NP content from 2 to 5 wt% [43].

ZnO NP can form a crystal in three different forms: hexagonal wurtzite, cubic zinc blende and rocksalt (Figure 2). ZnO hexagonal wurtzite, in which every zinc atom is tetrahedrally coordinated with four oxygen, is thermodynamically the most stable at ambient conditions [44]. Cubic zinc blende can be stabilised by growing ZnO on cubic substrates. ZnO will only exist in the rocksalt structure at relatively high pressures [45]. ZnO NP usually presents a hexagonal wurtzite structure with very sharp X-ray diffraction (XRD) peaks [42, 43, 46]. However, the ZnO can also present broadband without any typical diffraction peak due to water within the nanoparticle structure, which hinders the crystalline structure. Calcination is usually applied to remove residual water and enhance the NP crystallinity [47].

The crystallinity of polymers influences their performance, namely the mechanical and barrier properties, as well as the biodegradation rate. Therefore, it is of interest to understand the effect of ZnO NP incorporation in the PBAT morphology. PBAT films present a semicrystalline structure with a typical XRD broadband pattern. After incorporation of NP, the diffraction pattern showed no significant changes. The crystallinity index (CI) of PBAT increased from 18% to 18.8% and 22.9%,

TABLE 1 | ZnO methods and resultant size and shape.

Method	Shape	Size	Reference
Precipitation/Coprecipitation	Spherical	—	Purwaningsih et al. [28]
	Rod	Diameter: 30–60 nm Length: 80 nm	Bhadra et al. [29]
	Flower	800 nm	Mohan et al. [14]
	Flakes	—	Kumar et al. [30]
Sol–gel processing	Rod	81–85 nm	Singh and Singh [31]
	Tube	70 nm	Yue et al. [32]
Microemulsions	Rod	Diameter: 22–28 nm Length: 66–72 nm	Yıldırım and Durucan [33]
	Spherical	40–70 nm	He et al. [19]
	Needle	150–200 nm	
	Column	Diameter: 50–80 nm Length: 80–100 nm	
Hydrothermal	Rod	50–200 nm	Lepot et al. [34]
	Rod	100 nm	Panahandeh et al. [35]
	Flower	Diameter: 2 μm Thickness: 2 nm	
Solvothermal	Rod	Width: 70–200 nm Length:300–800 nm	Wang et al. [15]
	Flower	—	Talebian et al. [36]
	Spherical	—	
	Rod	—	

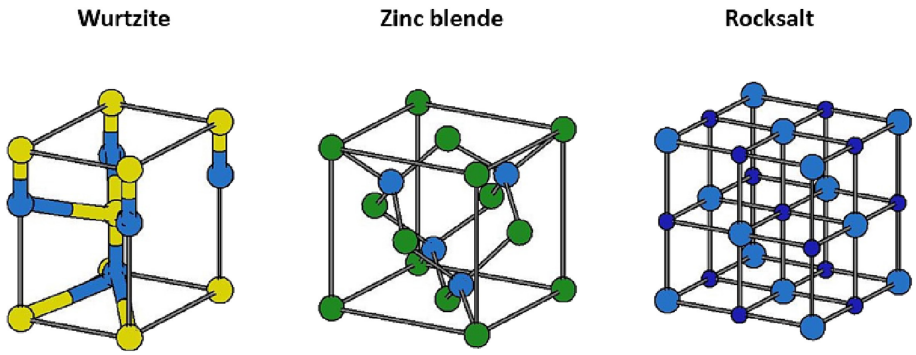

FIGURE 2 | Crystal structures of ZnO.

respectively, when incorporating 0.5% and 1% of ZnO NP. The ZnO NP presented a CI of 27.5% [47]. Therefore, incorporating of ZnO NP seems to increase the crystallinity of the film. It would be interesting to verify this effect on industrially obtained materials as extrusion conditions are known to impact the film crystallinity, due to the inherent chain orientation caused by the die after the extrusion step.

3 | Range of ZnO NP Concentrations and Method of Incorporation

The concentration of ZnO NP incorporated in the PBAT matrix varies between 0.5 and 5 wt% in most of the published works [39–43, 46–57]. The concentration is directly linked with the functional activity of ZnO. Studies have shown that increasing the concentration increases antimicrobial activity [41, 47, 56, 58]. However, ZnO also affects other properties, for example, sealing ability and mechanical properties [57, 58], and therefore, the incorporated concentration should be optimised.

Most of the works reported in the literature have been limited to material development at the laboratory scale only and the solvent-casting method was used to incorporate the ZnO in the film matrix [41–43, 47, 53–57]. The average thickness of these films varies between 80 and 140 μm, except for one study that produced thinner films (between 50 and 60 μm) [53]. Production methods typically used at industrial level have also been reported, although to a much less extend, namely blown extrusion to obtain films of PBAT with 1 to 5% of ZnO NP [30, 50, 59] and injection moulding to obtain articles with up to 1.5% of ZnO combined with lignin [46]. These industrial moulding techniques follow a step of co-rotating twin-screw extrusion to incorporate the particles in the continuous phase, which is then diluted and mixed during blown extrusion. Melt-blending followed by hot-pressing was also used to produce films and testing strips [58] with a film thickness between 100 and 200 μm. Although solvent casting is a very practical method for preliminary tests, it would be interesting to explore with more detail the industrially made films and its properties upon incorporation of nanoparticles.

PBAT with ZnO incorporated as a coating over a paper substrate was also studied. The coating solution was applied to kraft paper using a wire bar coater. The thickness of the coating ranged from 52 to 110 μm [52].

4 | Functional Properties

4.1 | Antimicrobial Activity

ZnO NP have been reported to show antimicrobial activity, reducing the growth of some strains of pathogenic bacteria and spoilage microorganisms, with potential applications, in food preservation. The effects mainly depend on the particle size, shape, concentration and exposure time of the microbial cells [60, 61]. Three processes have been reported for the action of ZnO NP:release of Zn ions [62], direct contact of ZnO NP with the cells resulting in damaging the microbial cells integrity [63], and the formation of reactive oxygen species (ROS) assisted by the effect of light radiation [64]. Some publications indicate that the inhibitory effect of ZnO on Gram-positive bacteria tend to be lower than for Gram-negative. Gram-positive bacteria have a thick cell layer of peptidoglycan in the structure, while Gram-negative bacteria have a thinner layer. Even with an extra layer of lipopolysaccharides in addition to the thin peptidoglycan in Gram-negative bacteria, the cell wall provides less resistance making these bacteria more susceptible to the ZnO effect [65, 66]. However, it must be recognised that the net effect depends on the species/strain and on several factors that are not always controlled or reported in the literature, making it difficult to generalise.

Escherichia coli is the most common microorganism studied in the PBAT nanocomposites incorporated with ZnO NP alone or in combination with other components such as silver nanoparticles (Ag NP) and lignin [46, 47, 51, 55, 58]. Generally, the higher the concentration of ZnO incorporated, the greater the antimicrobial effect and the combination of ZnO with other antimicrobial agents was found to be synergic towards inhibiting of the growth of *E. coli*. The antimicrobial activity against *Staphylococcus aureus* was also studied and contradictory results were reported. These bacteria were found to be less sensitive than *E. Coli* to ZnO particles with lignin [56], but other studies reported a higher effect of particles for *S. aureus* [39, 41, 42]. Another study showed the same inhibitory effect between *E. coli* and *S. aureus* when assessed by the zone of inhibition method, with the halos presenting 13 mm of diameter [54].

ZnO in PBAT matrixes showed less antimicrobial activity against the *Listeria monocytogenes* (Gram-positive) than against the Gram-negative bacteria *E. coli* [52, 53]. Seray [43] demonstrated that the inhibitory effect was higher for

Bacillus subtilis compared with *Pseudomonas aeruginosa* and *S. aureus*.

PBAT/TPS films with an intermediate concentration of 4% ZnO NP showed better antimicrobial effects against total viable count, lactic acid bacteria and yeast and mould, compared with the films containing lower (1–3%) or higher (5%) ZnO NP [50].

The minimum inhibitory concentration (MIC) and minimum bactericidal concentration (MBC) were determined by microdilution method. ZnO NP with reduced graphene oxide (rGO) showed the lowest MIC values against *E. coli* and *S. aureus* ($3.75\,\mu g\,mL^{-1}$) when incorporating the maximum content of rGO (5%). The MBC value was consistent with results decreasing from 25 to $6.25\,\mu g\,mL^{-1}$ for both pathogens with the increase of rGO concentration. Xiao [56] showed that the MIC of lignin-ZnO (LZn) hybrid particles, for the same pathogens, is lower than lignin nanoparticles (LNP) due to the presence of ZnO. It also demonstrated that the antibacterial activity of the hybrid particles gets higher, with the increasing content of ZnO NP.

In practically none of these studies, the different morphologies and sizes of ZnO NP were compared. Usually, only one type of ZnO NP were incorporated into PBAT (sometimes commercially available particles) and the particles were not fully characterised.

It is important to note that many of these studies applied different methods to assess antimicrobial activity, such as the disc diffusion assay and the viable colony count method, among others, making the comparison between them difficult. Soares Silva et al. [65, 66] provided a more comprehensive study using different species and strains, and different methods to assess antimicrobial activity, making comparison possible.

Most of the studies have determined the effect of ZnO NP and not of the effect when the NP is incorporated in the film and/or in films produced by industrial processes, where the actual performance may be different. Additionally, the behaviour of the films in situ (i.e., when actually in contact with the food) may differ from the behaviour in vitro. Only a few works have studied the impact of these materials in contact with real foods [48, 49, 51, 67]. A recent study combined ultraviolet-C light-emitting diodes (UV–C LED) irradiation and antimicrobial PLA/PBAT packaging film with ZnO NP and tested its performance as packaging for fresh-cut vegetables. The films showed higher in vitro antibacterial activity against *E. coli* than *S. aureus*, although the activity was considered to be relatively low due to the slow release of ZnO entrapped by the polymer. Nevertheless, the combined system showed a significant synergistic effect in reducing the bacteria during storage, with a reduction of about 2 log CFU g^{-1} of *E. coli* in onion, cabbage and carrot after 7 days of storage at 10°C [48, 49]. PBAT/PBS films incorporating ZnO at levels higher than 2.7% were shown to delay discoloration of red pigments in packaged minced pork due to efficient inhibition of microbial growth and UV blocking [51]. The incorporation of ZnO NP (together with titanium dioxide) into low-density polyethylene (LDPE) films has also been reported. The bactericidal efficiency tests resulted in the prevention of *E. coli* growth in fresh calf minced meat [67].

4.2 | Other Functionalities

The antioxidant activity of packaging films is important for extending the shelf-life of many packaged foods. Films of PBAT/PLA with 3% ZnO NP exhibited limited antioxidant activity, with results of 4.4% for the 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS) test and 6.9% for the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging activity [48, 49]. Films of PBAT with LZn hybrid particles incorporated at levels of 1%, 2% and 3% showed a radical scavenging activity value (RSA) via DPPH assay, respectively, of 10.6%, 17.6% and 32.5%, relative to the PBAT [56]. However, it is not possible to discriminate the effect of the ZnO itself from the lignin, which is known to be antioxidant. PBAT/TPS films with incorporation of ZnO NP showed a reduction of approximate 50% of the lipid oxidation value determined by the TBARS test (thiobarbituric acid reactive substances) in relation to the control [50]. No significant differences were verified for the films with different concentrations of ZnO NP incorporated (1–5%). However, the study does not include information regarding the conditions of exposure to light of the experiment.

5 | Effect of Size and Shape on Functional Properties of Films

The functional properties of ZnO NP depend on their physical properties, namely on morphology, particle size and specific surface area. These are determined by the synthesis method and conditions used to obtain the particles. Several authors reviewed the physical characteristics of ZnO NP of different morphologies and sizes obtained by different methods [48, 49, 68, 69].

However, only a few studies have assessed the impact of the morphology and size of ZnO NP on their antimicrobial activities. These two parameters are often not independent. Usually, the spherical particles are reported to be more active. The spherical particles are almost always the smallest particles in the studies and consequently have the highest surface area-to-volume ratio. When studies compared NP with different morphologies, generally those that were more toxic to microorganisms were also smaller. Although morphology is a very important parameter, the antimicrobial mechanisms of ZnO NP are size-dependent and in general, when the particle size decreases, its antibacterial activity increases [48, 49, 53, 69].

Stanković [70] studied the influence of ZnO NP shape and size against *E. coli* and *S. aureus*, concluding that spherical particles (with around 30 nm) demonstrated the highest microbial cell reduction rate when compared with hexagonal prism particles (1 nm in length and 100 nm in diameter) and ellipses particles (length of 500–600 nm and diameter of 100 nm). Particles with cuboidal/rod shape of ZnO NP (size of 40–45 nm) were found to have higher antibacterial activity as compared with spherical (size of 60–180 nm) and hexagonal NP (size of 63 nm) [38]. However, in this specific study [38], the spherical shape presents a wide range of sizes (60–180 nm), and the cuboidal/rod shape is the smallest (40–45 nm).

The effect on the antimicrobial activity was the most studied, but the photocatalytic activity was also addressed and largely

influenced by the shape [38]. The photocatalytic activity (PCA) was studied through the degradation of methyl orange dye at 465 nm over time (Figure 3). Spheric-shaped particles showed the lowest PCA for all doses. The spherical- and hexagonal-shaped particles showed higher PCA at intermediate doses, a pattern different than the one presented by the cuboidal NP that increased with the increase of the mass of NP in the range tested. The different morphologies and active surface area may cause different degradation patterns due to interstitial space that mediate the degradation rate.

Most of these studies focused on the activity of the particles *per se*. However, the incorporation of the particles in the packaging matrix (e.g., a film) may also play a role because the physical properties may interfere with the mechanism of action of the ZnO. Tamimi [71] studied the effect of particle morphology (spherical and rod shape) on some properties, including the antibacterial properties of the particles incorporated in starch-based film. Spherical particles not only revealed the best antibacterial effect but also had the best behaviour on mechanical, physicochemical and barrier properties. Rod shape particles showed the highest impact in UV transmission.

Not addressing the impact of different morphologies and sizes in the final properties of the film is a major limitation regarding the studies of functional properties of ZnO/PBAT packaging matrix.

6 | Effect on Physical and Mechanical Properties of Film

6.1 | Thermostability

Most of the biopolymers exhibit low thermal stability because of their weak chemical bonding. The incorporation of fillers, namely metal and metal oxides, is a common approach to improving the thermal properties of nanocomposite films [72]. Most of the studies reported that thermal stability is enhanced by the incorporation of the ZnO NP, compared with that of pure PBAT, resulting in an increase of the temperature of the thermal degradation, although no significant differences are observed when comparing different percentages of ZnO NP incorporation [59]. The endothermic peaks of the differential scanning calorimetry (DSC) thermograms (melting) were recorded at higher temperatures for PBAT/starch incorporated with ZnO NP [39] and PBAT film with ZnO/rGO [42].

However, opposite results were also reported indicating that the addition of ZnO NP reduces the thermal stability of PBAT and that the effect increased with the percentage of ZnO [47, 55]. The effect was attributed to the increase of the oxidative decomposition of the polymer at the ZnO NP interface at high temperatures, accelerating the thermal degradation of the composites.

6.2 | Mechanical

The mechanical behaviour of the nanocomposites is one of the most important properties for packaging applications. The incorporation of nanoparticles in the matrix affects mechanical properties of the film because of interface interactions between the NP and the polymer molecules [39, 72].

In general, the incorporation of NP improves the mechanical properties [41, 51, 53, 54]. The tensile modulus and the yield strength of the film were improved with the incorporation of lignin-ZnO hybrid particles [56] and with the incorporation of ZnO with rGO [42]. Possible synergistic effects of two nanoparticles incorporation, adding Ag NP, were also reported [39, 47].

The effect of NP concentration on the mechanical properties is not linear, and some studies show an optimal intermediate concentration for which the mechanical properties are the best [57, 58]. The tensile strength and elongation at break values decreased when ZnO was added up to 3%, showing a minimum strength, and increased with the addition of ZnO in concentration from 3% to 5% [50]. These results suggested that adding NP into the polymer matrices at a certain concentration decreased the adhesion between the polymer network, causing reduced mechanical strength, while the agglomeration of NP at higher concentrations of NP reduced the dispersion of finer nanoparticles [55]. The elongation at break of the PBAT film decreased with the increase in the ZnO content (1, 3 and 5 wt%) [55]. The tensile strength before the break point increased with the addition of ZnO NP but the film showed a breakpoint at lower elongations when ZnO is incorporated. The yield strength seems to decrease with the incorporation of 1 wt% but then tends to increase for higher loads. Nevertheless, these conclusions are tentative because the results dispersion was not provided so the statistical significance cannot be evaluated.

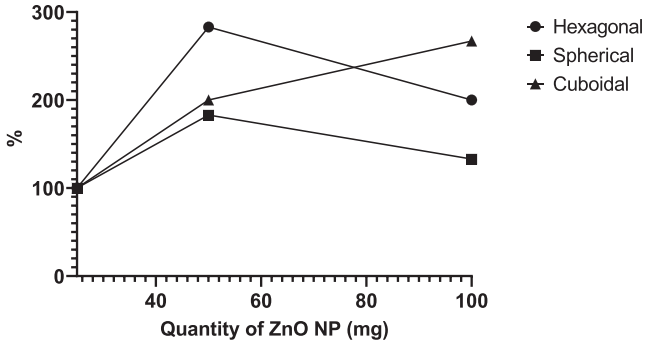

FIGURE 3 | Photocatalytic activity. Effect of ZnO NP amount and shapes. Adapted from: Sharma [38].

6.3 | Optical

Many packaged foods are significantly affected by light, which can cause organoleptic changes, browning and oxidative degradation. Quality changes in foods result from light exposure, particularly UV light, which has high energy. UV irradiation, especially in UV-A (320–400 nm) and UV-B (290–320 nm) regions, is one of the key factors affecting food spoilage and nutrient loss, which greatly accelerated oxidation [50, 73].

ZnO NP is a white powder that can reduce the transmission and transmittance and, therefore, enhance the optical properties of the biodegradable films. The UV-light shielding ability and

transparency of the composite films can be determined by measuring the transmittance at 280 nm and 660 nm [53].

Studies on the light transmission of PBAT/TPS films indicated that the addition of ZnO NP reduced the light transmission, mainly in the UV region (below 380 nm): films containing 4 and 5% of ZnO present less light transmission than films with 1% ZnO NP incorporation, and when compared with the neat PBAT/TPS film [50, 59]. Similar results were obtained with films of PBAT/PBS in UV region below 280 nm [51]. So, the incorporation of ZnO NP clearly provided UV barrier properties as the NP dispersion prevented light transmission through films. The increase in the ZnO NP incorporation improved the haze and decreased the transmittance of the PBAT film: the haze and transmittance of neat PBAT film is 17.3 and 86.4, respectively, and after incorporation of 5% ZnO NP, the haze value increased to 81.5% and the transmittance decreased to 74.6% [55]. However, other studies reported that the incorporation of ZnO NP did not affect the transmission in the UV light region, meaning that the high UV barrier properties of PLA/PBAT film were derived from PBAT polymer [53].

The UV–VIS spectra of neat PBAT/PLA blend film demonstrated high absorption of light at wavelengths below 300 nm, decreasing with the increase of the wavelength, confirming the transparency to visible light [53]. PBAT and PLA/PBAT films showed absorption at wavelength lower than 300 nm and upon ZnO NP was incorporated, the composite films exhibited strong absorption peaks in the region of 360–376 nm due to the absorption of light by ZnO NP [39, 43, 53]. The light absorption of both neat and PLA/PBAT-ZnO NP films gradually decreased above 400 nm, but the light absorption intensity of PLA/PBAT-ZnO NP composite film was higher than that of neat film. This can be due to the crystalline properties of ZnO NP that cause light scattering, which may result in lower light absorption in the visible light range. On the other hand, some reflection of the light can occur at the interface between ZnO NP and the matrix due to the large specific surface area of ZnO NP [53].

In the visible light wavelength, the incorporation of ZnO NP decreased the transmission values in line with the reduction of transparency in this region of the spectra [53]. Furthermore, the transparency to visible light of the nanocomposite films is highly affected by the distribution of the fillers while aggregations and molecular arrangements of the NP structures modify the amorphous-crystalline fraction of the polymers [74].

The colour of the PBAT films is also affected by the incorporation of NP like TiO_2, SiO_2 and ZnO. The Hunter colour values L, a, b (according to ASTM D2244) showed an increment in (a*) redness and (b*) yellowness [53, 54].

6.4 | Permeability and Wettability

Permeability to water vapour (WVP) and oxygen (OP) are barrier properties that greatly affect the quality and shelf life of the packaged products. One of the main goals when adding nanomaterials to biopolymers used for food packaging is to improve their barrier to moisture and oxygen. There are many factors that affect permeability: shape, size and the compatibility between the polymer matrix and the nanoparticle, as well as microstructures and morphology of the matrices [39, 41].

PBAT itself has low barrier properties with reported values for oxygen permeability of 84.0 $cm^3\,mm\,m^{-2}\,day^{-1}\,bar^{-1}$ determined at 23°C and for water vapour permeability of 3.1 $g\,mm\,day^{-1}\,m^{-2}$ determined at 23°C and 50% of relative humidity [75]. These values correspond to $9.7\times10^{-12}\,cm^3\,m\,m^{-2}\,s^{-1}\,Pa^{-1}$ and $25\,g\,m\,m^{-2}\,s^{-1}\,Pa^{-1}$, respectively, when normalised. At the barrier measuring temperature, the material is well above its glass transition temperature (ca. −34°C) where high polymer chain mobility and free volume allow for high rate of gas transfer.

The barrier properties of PBAT/ZnO NP nanocomposites have been addressed in a few studies, with matrices containing PBAT with other components, such as PLA [53, 54] or TPS [50]. Results from different studies were compiled in Table 2 after uniformisation of the units. The films were obtained by different processes (blown extrusion or solvent casting) and the conditions for the permeability measurements (temperature and relative humidity) were also different. For these reasons, the comparison between the results is difficult.

Publications show that the WVP and OP of the nanocomposite films produced in the laboratory by solvent casting are slightly reduced with the incorporation of ZnO NP as compared with neat PBAT film [41, 42, 53, 54]. However, in other works a slight increase in WVP was recorded when adding ZnO NP [48, 49]. The incorporation of 1% ZnO NP in the PLA/PBAT matrix decreased the WVP value by *ca* 20%, from 192 to 152 $g\,m^{-2}\,day^{-1}$ [54]. This has been attributed to the increase in tortuosity of the water molecules path increasing the crossing time through the composite film layer [50, 58]. However, measurements in blown extruded films showed that the addition of ZnO NP increases the value of WVP from 1.4×10^{-12} g m $m^{-2}\,s^{-1}\,Pa^{-1}$ to between values ranging from 1.7 and 2.4×10^{-12} g m $m^{-2}\,s^{-1}\,Pa^{-1}$ when ZnO NP is incorporated at 1–5% [50].

Some studies show that the effect of concentration is not linear and that there is an optimal concentration of incorporated ZnO NP for which barrier properties present the best performance. WVP results increased from 45 to 60×10^{-12} g m $m^{-2}\,s^{-1}\,Pa^{-1}$ when incorporating ZnO NP between 0–1%, although lower values are obtained in the intermediate concentrations [58].

The results for the impact on OP of adding ZnO NP to the PBAT film also depend on the film production technique. For films produced by extrusion, an increase in OP was observed of ca 15% in values measured at 23°C and 50% RH [50] and an increase of 50% in values measured at 38°C and 90% RH [39], upon incorporation of 1% of NP. Results from other authors show a positive effect of adding ZnO NP in films produced by solvent casting with a decrease in OP of 5–10% [41, 48, 49, 54] when incorporating ZnO NP 1 wt %.

It is recognised the difficulty to compare results from these studies because of the different experimental conditions (film production, temperature, relative humidity, type of matrix and components incorporated). Additionally, the precision in the different concentrations of incorporation (e.g., differences of

TABLE 2 | WVP and OP values reported in the literature for PBAT-based films with ZnO NP incorporation.

	Conditions			ZnO NP content (wt%)										
	Film production	T (°C)	Relative humidity	0	0.2	0.4	0.6	0.8	1	2	3	4	5	10	Reference
WVP ($\times 10^{-12}$ g m m^{-2}s^{-1}Pa^{-1})	Blown extrusion	23	50%	1.4	—	—	—	—	1.7	1.8	2.2	2.4	1.8	—	Phothisarattana et al. [50]
	Cast extrusion	25	50%	14					12	13	11	11	12		Promhuad et al. [51]
	Blown extrusion	38	90%	35	30	33	35	35	36	—	—	—	—	—	Zhai et al. [39]
	Solvent casting	25	50%	21	—	—	—	—	—	—	25	—	—	—	The literature [48, 49]
	Solvent casting	25	90%	70	—	—	—	—	56	—	—	—	—	—	Thiyagu et al. [45]
	Melt-blending method	25	55%	45	36	37	45	50	60	—	—	—	—	—	Ge et al. [58]
	Solvent casting	25	50%	24.7	—	—	—	—	—	—	(*)	—	—	—	Shankar et al. [53]
OP ($\times 10^{-12}$ cm^3 m m^{-2}s^{-1}Pa^{-1})	Blown extrusion	23	50%	3.2	—	—	—	—	3.7	6.4	6.6	5.7	3.8		Phothisarattana et al. [50]
	Blown extrusion	23	50%	7.1					6.3	7.4	7.0	7.1	7.2		Promhuad et al. [51]
	Blown Extrusion	38	90%	6.3	5.6	7.0	8.7	9.6	9.9	—	—	—	—	—	Zhai et al. [39]
	Solvent casting	25	90%	5.1	—	—	—	—	4.8	—	—	—	—	—	Thiyagu et al. [54]
	Melt-blending method	25	55%	60	48	45	38	42	46	—	—	—	—	—	Ge et al. [58]
	Solvent casting	23	0%	11.4	—	—	—	—	10.3	—	9.8	—	8.7	7.6	Venkatesan et al. [41]

Note: The asterisk (*) indicates WVP results with different zinc oxide precursors: Zinc acetate = 20.9; zinc chloride = 20.7; zinc nitrate = 32.4×10^{-12} g m m^{-2}s^{-1}Pa^{-1}.

0.2 wt% in series of increasing concentration) is also arguable if one takes into consideration the variability of the industrial processes and the fact that many studies do not provide standard deviations between replicates.

Contact angle (CA) is a measure of the surface wettability and hydrophobicity. Films with CA at 90° and above are considered hydrophobic surfaces [76]. Usually, PBAT alone is nearly hydrophobic. Adding ZnO NP decreases the value of CA and gives a heterogeneous surface that increases wettability explaining what was mentioned before regarding the WVP and OP results [50, 53], but adding a low concentration of nano-ZnO has little effect on the hydrophobicity of the material [57].

6.5 | End of Life

PBAT-based films are among the most used in the increasing market for biodegradable plastics. Biodegradable films can be gradually decomposed in soil by microbial enzymatic actions and bioassimilation [77]. Biodegradable materials are associated with poor physicochemical and mechanical properties such as low barrier properties, low stability during processing and storage and high-water sensitivity. These properties may not suffice the required protection and shelf-life specifications as under today's standards of distribution and supply chains, resulting in shorter shelf-life and increased food losses. Therefore, the benefits of using biodegradable materials as food packaging need to be critically evaluated using recognised tools, such as life cycle assessment (LCA). Furthermore, it is known that the incorporation of active compounds such as nanoparticles, can improve these properties [78] despite there are limited studies focused on interactions between the polymers/nanocomposites and biodegradability.

The biodegradability of PBAT with ZnO NP incorporated at concentrations varying from 0.2–1% was evaluated following the weight loss in soil burial tests for 120 days. At the lowest level of incorporation, a slight reduction in the degradation rate was observed compared with the neat PBAT. However, the statistical significance of this decrease is not discussed. The increase in the degradation rate with the incorporation of NP is more noticed only at 0.6% and higher levels [58]. It is possible that low levels of incorporation yield better dispersibility, which can result in more difficulty to penetrate the matrix by microorganisms. However, it should be considered that studies with sound experimental design are required to obtain results statistically significant.

7 | Effect on Migration

Consumer tends to consider bio-based materials safer than conventional plastics and these natural-based alternatives appear to be a sustainable alternative to conventional plastics. However, their toxicity was compared, presenting similar results [79], which may be a surprise and a disappointment for many. Most often the bio-based materials are evaluated regarding the environmental aspects (such as carbon footprint, end of life) and the impact of the components on safety issues upon human exposure is often neglected.

Migration of nanomaterials from packaging into food is a mass transfer phenomenon and can occur by diffusion, dissolution and abrasion of the packaging surface, causing chemical contamination, affecting the organoleptic properties, and may have negative effects on human health [80]. Uncertainties on the possible health effects and long-term safety of NP when used in food packaging is an important concern, as there are still few studies regarding their toxicity associated with the ingestion, absorption, distribution, metabolism and excretion after oral exposure, and potential interactions of nano-based material with food components [81].

NP have a large surface area-to-volume ratio and may exhibit different physicochemical properties than the bulk-sized material. Consequently, the mechanisms of mass transfer, interaction with the supporting materials and with the food, as well as the potential toxicity may be different [1]. The migration depends not only on the NP type, concentration, size, shape and polymer viscosity, molecular weight and solubility but also on the migration conditions (contact time, temperature) and food composition [82].

According to Regulation (EU) No. 10/2011 [83] and its amendments safety of NP should be assessed on a case-by-case basis considering the physicochemical characteristics and the toxicological profile of the migrate [1]. For metallic NP it is important to determine if the migration occurs in nano or ionic form to understand the toxicological risk. The number of reports on the toxicity of ZnO NP has been increasing in literature with studies addressing cytotoxicity, genotoxicity, hepatotoxicity, pulmonary toxicity, neurotoxicity and immunotoxicity [84]. ZnO NP are studied for their antimicrobial effect, and therefore, it can be expected to also have an effect on microbiome human cells. Creating a framework to predict the toxicity of ZnO NP is challenging due to the diversity and complex nature of the available literature and more studies regarding the effects of NP ingestion on the gastrointestinal tract and other organs of human body are needed.

Migration of ZnO NP can occur through the ionic form (Zn^{2+}) or the whole particle (ZnO NP) as schematically represented in Figure 4. Considering the diffusional properties of NP in polymers and the solubility characteristics of the ZnO NP, ESFA concluded that the substance does not migrate in nanoform. Therefore, the safety evaluation should focus on the migration of soluble ionic zinc (Zn^{2+}) from packaging materials.

Even if migration would occur in the particulate form, this would be dissolved immediately into ionic zinc upon contact with acidic foods or stomach acid [4]. The specific migration limit (SML) for soluble ionic zinc is 5 mg kg^{-1} food) set out by the European Regulation. Regarding safety assessment, EFSA recommends that the Commission impose an exposure upper limit of zinc of 25 mg per person per day [4].

Migration experiments indicate that when NP are completely embedded in the host polymer and the contact surface is not altered by mechanical stress, NP do not have potential to migrate into food as they may be considered immobilised [85]. In cases where NP are not fully embedded but are protruding from and sticking out of the polymer surface, there is a probability of release into food that needs to be considered for risk assessment. Such cases may occur when NP are incorporated at very high use levels in the food contact layer or when the nanocomposite was manufactured under poor technical production conditions. Therefore, it is important to verify whether the NP is fully embedded or freely present at the food contact surface of the nanocomposite [86]. It was concluded that spherical shapes of carbon NP would be the worst-case geometry for diffusion in polymers (quicker diffusion than nonspherical objects due to the geometry) and that only sizes up to 2–4 nm in diameter could migrate [87].

Experimental migration results reported until now are limited and contradictory, reflecting experimental challenges [81]. There are already well-established protocols, especially for the case of metals, to quantify concentration of the elements, such as the wet digestion followed by ICP-MS. However, to distinguish between migration of the dissolved form of the particle and migration of the actual particle elemental techniques only are not enough. Microscopic techniques SEM and TEM, light scattering based such as multiangle light scattering detection (MALLS) are used to characterise the particles after separation by techniques such as Asymmetric-Flow Field Flow Fractionation (AF4) and Centrifugal Field Flow Fractionation (CF3). These altogether are needed to detect and characterise NP and give information on the aggregation state of the particle [81, 88]. The migration of carbon black NP out of plastic materials was investigated and AF4 and MALLS were used to separate, characterise and quantify the potential release of NP. It was concluded that the carbon black does not migrate into food once it is fully embedded in the plastic matrix [89].

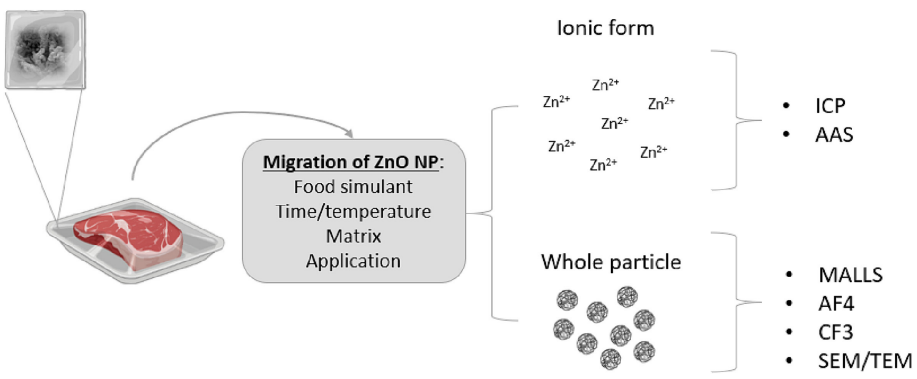

FIGURE 4 | ZnO NP migration: In the ionic form (Zn^{2+}) or as the whole particle (ZnO NP); corresponding techniques for detection and quantification.

Concerning the specific migration (SM) of the Zn^{2+}, the contact conditions and food simulants for the migration tests have been established by Regulation (EU) No. 10/2011 [83] and its amendments. It is known that migration of Zn into food simulants may overestimate the migration into real food.

Migration of ZnO NP has been studied using a variety of matrices, including polypropylene (PP) [90], LDPE [91–94] and polylactic acid (PLA) [95], among others. Migration of Zn^{2+} from PBAT matrix has been much less studied. Studies related to the migration of the whole particle of ZnO NP have not yet been reported. Therefore, research applying adequate detection and quantification techniques is needed. Table 3 summarises the characteristics of the studies analysed, regarding PBAT/ZnO nanocomposites.

Migration of Zn^{2+} increases with the increase in concentration of the particles in PBAT [43, 56]. Increasing the % the rGO, which acts as a shield, decreased the Zn migration in all simulants [42]. The migration is, as expected, the highest for the acetic acid simulant representing acidic food due to the solubility characteristics and induction of changes in the polymer microstructure [59].

The migration profile of Zn^{2+} release from PBAT/ZnO films into food simulants was performed and different mathematical models were fitted to the data to describe the release kinetics [43]. The release of Zn^{2+} ions occurred progressively without reaching equilibrium (Figure 5). An initial linear migration pattern was observed up to around day 5, with a rate that showed not highly affected by the concentration of ZnO incorporated. However, the concentration did affect particularly the level of migration at the longer contact times. At the end of the experiment, more than 70% of the entire load of Zn^{2+} was released from the PBAT matrix due to erosion of the surface.

The Higuchi and Korsmeyer–Peppas models were used to describe the kinetics of the Zn^{2+} release from PBAT/ZnO nanocomposite films [43]. These models have been used to describe drug release from pharmaceutical dosage forms, particularly for controlled-release or sustained-release formulations. Both models described well the release of Zn ions for all concentrations of incorporation presenting high correlation coefficients (R^2).

Most migration studies have been performed with the simulants and research on the migration of NP into actual model foods is limited. The migration of Zn^{2+} into a food model (chicken skin) was determined but the matrix was bacterial nanocellulose nanocomposite [65, 66]. It would be interesting to verify the migration into a real food in contact with PBAT/ZnO packaging films.

Overall migration (OM) is another parameter required for materials food contact compliance, with a European Union's plastic regulation limit of 10 mg dm^{-2} (corresponding to 60 mg kg^{-1} food). OM from PBAT/TPS films was the highest into 3% acetic acid and increased linearly with the increase of ZnO NP concentration (0–5%) with values between 0 and 13 mg dm^{-2}, respectively [59]. The migration into ethanol 10% did not increase with the ZnO NP content and the migration into water was not significantly different than that from the film without incorporation of ZnO. The OM values of films containing 5% ZnO exceeded the limit when in contact with 3% acetic acid simulant and therefore, these films have limitations in the application for packaging acid foods [59]. Similar results were obtained in

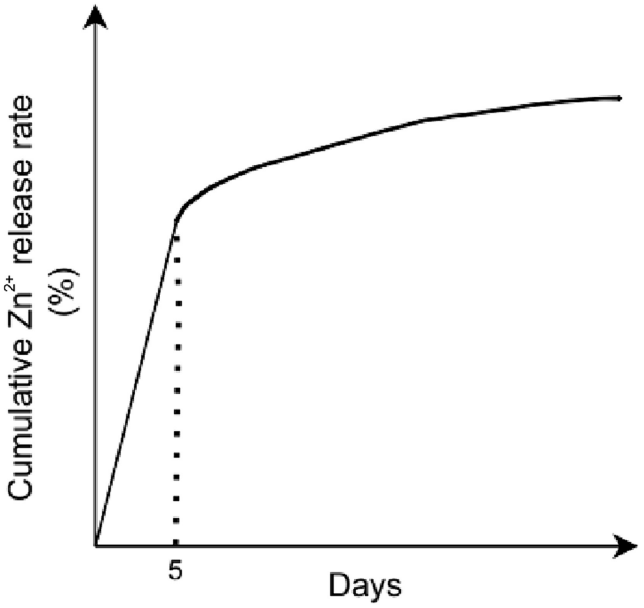

FIGURE 5 | Pattern of Zn^{2+} ions kinetics release from PBAT nanocomposites containing ZnO NPs. Adapted from [43].

TABLE 3 | Studies of migration of NP from PBAT food packaging.

Nanoparticle	Matrix	Food simulant	Time/ temperature	Technique for migration analysis	Reference
Lignin-ZnO hybrid particles	PBAT	Distilled water	18 h; Room T	ICP-MS	Xiao et al. [56]
TiO_2 and ZnO NP	PBAT/TPS	Distilled water 10% ethanol 3% acetic acid	10 days at 5°C	ICP-OES	Phothisarattana and Harnkarnsujarit [59]
ZnO/reduced graphene oxide	PBAT	Distilled water 4% acetic acid 50% ethanol	2 h at 70°C	ICP-MS	Charoensri et al. [42]
ZnO NP	PBAT	Slightly acidified bi-distilled water	45 days	AAS	Seray et al. [43]

PBAT/starch-based films with ZnO NP: the OM limit was exceeded due to the release of starch, although the migration limit of zinc was also exceeded [96].

8 | Conclusions

The purpose of this review was to assess the potential of ZnO NP incorporation in PBAT films for food packaging industry, considering the several properties required to protect and preserve the food. A great amount of research was reported regarding the impact on functional, physic-mechanical properties and migration characteristics. In this review, the range of concentrations, incorporation methods and the effect of particles size and shape are addressed as well as a brief consideration of the impact of ZnO NP on the PBAT-based films biodegradability.

All properties of the materials addressed in the studies covered in this review are affected by the incorporation of ZnO NP. In general, the properties of the PBAT/ZnO nanocomposites are improved, but in some cases, it occurs the opposite, or there is an optimum intermediate concentration. Unfortunately, many studies did not provide information on variability between replicates, which highly limits the interpretation of results due to lack of statistical significance.

In most of the studies, the NP are not fully characterised particularly regarding the morphology and size. The polymeric matrix is also not always fully described or consists of blends of different plastics. Therefore, contradiction between results is often found which cannot be completely justified. Very relevant is that most of the studies are based in PBAT materials prepared at laboratory scale by solvent-casting which is highly convenient but not representative of the industrially made films which suffer the impact of the extrusion process and impact in chain orientation. This is known to have an important effect on properties. On the other hand, other relevant properties such as thermosealing of the films was not addressed in the studies. Therefore, proper validation of the developments is needed.

The active properties (antimicrobial) have been investigated in the studies but applying in vitro strategies, and only a few works have addressed the impact of these materials in contact with real foods. These studies are needed to clarify the efficiency and efficacy of the functional activity of the active materials.

The migration studies of the whole particle or ionic form of ZnO NP incorporate in PBAT are scarce. Further investigations are needed to achieve at an equilibrium of ZnO NP incorporation and its properties be effective but taking in note the concerns regarding the toxicity and health issues when applied in food packaging.

Author Contributions

Ana Rita Mendes: conceptualization, writing, data analysis, writing–review and editing. **Paula Teixeira:** writing–review and editing. **Fátima Poças:** conceptualization, writing, data analysis, writing–review and editing. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

1. F. Poças, "Migration From Packaging and Food Contact Materials Into Foods," in *Reference Module in Food Science* (New York, USA: Elsevier, 2018), https://doi.org/10.1016/b978-0-08-100596-5.21460-1.

2. P. J. P. Espitia, C. G. Otoni, and N. F. F. Soares, "Zinc Oxide Nanoparticles for Food Packaging Applications," in *Antimicrobial Food Packaging* (New York, USA: Elsevier Inc, 2016), 425–431, https://doi.org/10.1016/B978-0-12-800723-5.00034-6.

3. FDA, "Part 182 – Substances Generally Recognized as Safe. In National Archives and Records Administration (NARA)," (2011).

4. EFSA, "Safety Assessment of the Substance Zinc Oxide, Nanoparticles, for Use in Food Contact Materials," *EFSA Journal* 14, no. 3 (2016): 4408, https://doi.org/10.2903/j.efsa.2016.4408.

5. EFSA Panel on Food Contact Materials, E. F. and P. A. (CEF), "Scientific Opinion on the Safety Evaluation of the Substance Zinc Oxide, Nanoparticles, Uncoated and Coated With [3-(Methacryloxy)propyl] trimethoxysilane, for Use in Food Contact Materials," *EFSA Journal* 13, no. 4 (2015): 4063–4071.

6. A. Kolodziejczak-Radzimska and T. Jesionowski, "Zinc Oxide-From Synthesis to Application: A Review," *Materials* 7, no. 4 (2014): 2833–2881, https://doi.org/10.3390/ma7042833.

7. S. Smaoui, I. Chérif, H. Ben Hlima, et al., "Zinc Oxide Nanoparticles in Meat Packaging: A Systematic Review of Recent Literature," *Food Packaging and Shelf Life* 36 (2023): 101045, https://doi.org/10.1016/j.fpsl.2023.101045.

8. K. R. Raghupathi, R. T. Koodali, and A. C. Manna, "Size-Dependent Bacterial Growth Inhibition and Mechanism of Antibacterial Activity of Zinc Oxide Nanoparticles," *Langmuir* 27, no. 7 (2011): 4020–4028, https://doi.org/10.1021/la104825u.

9. A. Sirelkhatim, S. Mahmud, A. Seeni, et al., "Review on Zinc Oxide Nanoparticles: Antibacterial Activity and Toxicity Mechanism," *Nano-Micro Letters* 7, no. 3 (2015): 219–242, https://doi.org/10.1007/s40820-015-0040-x.

10. EFSA Scientific Committee, S. More, V. Bampidis, et al., "Guidance on Risk Assessment of Nanomaterials to be Applied in the Food and Feed Chain: Human and Animal Health," *EFSA Journal* 19, no. 8 (2021): 6768, https://doi.org/10.2903/j.efsa.2021.6768.

11. M. Johansson and Å. Boholm, "Scientists' Understandings of Risk of Nanomaterials: Disciplinary Culture Through the Ethnographic Lens," *NanoEthics* 11, no. 3 (2017): 229–242, https://doi.org/10.1007/s11569-017-0297-2.

12. P. J. P. Espitia, N. F. F. Soares, J. S. R. Coimbra, N. J. de Andrade, R. S. Cruz, and E. A. A. Medeiros, "Zinc Oxide Nanoparticles: Synthesis, Antimicrobial Activity and Food Packaging Applications," *Food and Bioprocess Technology* 5, no. 5 (2012): 1447–1464, https://doi.org/10.1007/s11947-012-0797-6.

13. EFSA Panel on Food Contact Materials, E. F. and P. A. (CEF), "Recent Developments in the Risk Assessment of Chemicals in Food and Their Potential Impact on the Safety Assessment of Substances Used in Food Contact Materials," *EFSA Journal* 14, no. 1 (2016): 4357–4388, https://doi.org/10.2903/j.efsa.2016.4357.

14. S. Mohan, M. Vellakkat, A. Aravind, and U. Reka, "Hydrothermal Synthesis and Characterization of Zinc Oxide Nanoparticles of Various

Shapes Under Different Reaction Conditions," *Nano Express* 1, no. 3 (2020): 1–14, https://doi.org/10.1088/2632-959X/abc813.

15. T. X. Wang and T. J. Lou, "Solvothermal Synthesis and Photoluminescence Properties of ZnO Nanorods and Nanorod Assemblies From ZnO2 Nanoparticles," *Materials Letters* 62, no. 15 (2008a): 2329–2331, https://doi.org/10.1016/j.matlet.2007.11.083.

16. R. Wahab, S. G. Ansari, Y.-S. Kim, H.-K. Seo, and H.-S. Shin, "Room Temperature Synthesis of Needle-Shaped ZnO Nanorods Via Sonochemical Method," *Applied Surface Science* 253, no. 18 (2007): 7622–7626, https://doi.org/10.1016/j.apsusc.2007.03.060.

17. X. Y. Kong, Y. Ding, R. Yang, and Z. L. Wang, "Single-Crystal Nanorings Formed by Epitaxial Self-Coiling of Polar Nanobelts," *Science* 303, no. 5662 (2004): 1348–1351, https://doi.org/10.1126/science.1092356.

18. J.-J. Wu, S.-C. Liu, C.-T. Wu, K.-H. Chen, and L.-C. Chen, "Heterostructures of ZnO–Zn Coaxial Nanocables and ZnO Nanotubes," *Applied Physics Letters* 81, no. 7 (2002): 1312–1314, https://doi.org/10.1063/1.1499512.

19. X. Li, G. He, G. Xiao, H. Liu, and M. Wang, "Synthesis and Morphology Control of ZnO Nanostructures in Microemulsions," *Journal of Colloid and Interface Science* 333, no. 2 (2009): 465–473, https://doi.org/10.1016/j.jcis.2009.02.029.

20. Y. Ding and Z. L. Wang, "Structures of Planar Defects in ZnO Nanobelts and Nanowires," *Micron* 40, no. 3 (2009): 335–342, https://doi.org/10.1016/j.micron.2008.10.008.

21. W. S. Chiu, P. S. Khiew, M. Cloke, et al., "Photocatalytic Study of Two-Dimensional ZnO Nanopellets in the Decomposition of Methylene Blue," *Chemical Engineering Journal* 158, no. 2 (2010): 345–352, https://doi.org/10.1016/j.cej.2010.01.052.

22. J. Jang, C. Yu, S. Choi, S. Ji, E. Kim, and J. Lee, "Topotactic Synthesis of Mesoporous ZnS and ZnO Nanoplates and Their Photocatalytic Activity," *Journal of Catalysis* 254, no. 1 (2008): 144–155, https://doi.org/10.1016/j.jcat.2007.12.010.

23. M. D. Rao and P. Gautam, "Synthesis and Characterization of ZnO Nanoflowers Using Chlamydomonas Reinhardtii: A Green Approach," *Environmental Progress & Sustainable Energy* 35, no. 4 (2016): 1020–1026, https://doi.org/10.1002/ep.12315.

24. M. Przybyszewska and M. Zaborski, "The Effect of Zinc Oxide Nanoparticle Morphology on Activity in Crosslinking of Carboxylated Nitrile Elastomer," *Express Polymer Letters* 3, no. 9 (2009): 542–552, https://doi.org/10.3144/expresspolymlett.2009.68.

25. L. Saikia, D. Bhuyan, M. Saikia, B. Malakar, D. K. Dutta, and P. Sengupta, "Photocatalytic Performance of ZnO Nanomaterials for Self Sensitized Degradation of Malachite Green Dye Under Solar Light," *Applied Catalysis A: General* 490 (2015): 42–49, https://doi.org/10.1016/j.apcata.2014.10.053.

26. M. Wasim, F. Shi, J. Liu, et al., "An Overview of Zn/ZnO Modified Cellulosic Nanocomposites and Their Potential Applications," *Journal of Polymer Research* 28, no. 9 (2021): 338, https://doi.org/10.1007/s10965-021-02689-6.

27. J. Yin, F. Gao, C. Wei, and Q. Lu, "Water Amount Dependence on Morphologies and Properties of ZnO Nanostructures in Double-Solvent System," *Scientific Reports* 4, no. 1 (2014): 3736, https://doi.org/10.1038/srep03736.

28. S. Y. Purwaningsih, S. Pratapa, Triwikantoro, and Darminto, "Nano-Sized ZnO Powders Prepared by Co-Precipitation Method With Various pH," *AIP Conference Proceedings* 1725, no. 1 (2016), https://doi.org/10.1063/1.4945517.

29. P. Bhadra, M. K. Mitra, G. C. Das, R. Dey, and S. Mukherjee, "Interaction of Chitosan Capped ZnO Nanorods With Escherichia Coli," *Materials Science and Engineering: C* 31, no. 5 (2011): 929–937, https://doi.org/10.1016/j.msec.2011.02.015.

30. K. M. Kumar, B. K. Mandal, E. A. Naidu, M. Sinha, K. S. Kumar, and P. S. Reddy, "Synthesis and Characterisation of Flower Shaped Zinc Oxide Nanostructures and its Antimicrobial Activity," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* 104 (2013): 171–174, https://doi.org/10.1016/j.saa.2012.11.025.

31. G. Singh and S. P. Singh, "Synthesis of Zinc Oxide by Sol-Gel Method and to Study it's Structural Properties," *AIP Conference Proceedings* 2220, no. 1 (2020), https://doi.org/10.1063/5.0001593.

32. S. Yue, Z. Yan, Y. Shi, and G. Ran, "Synthesis of Zinc Oxide Nanotubes Within Ultrathin Anodic Aluminum Oxide Membrane by Sol–Gel Method," *Materials Letters* 98 (2013): 246–249, https://doi.org/10.1016/j.matlet.2013.02.037.

33. Ö. A. Yıldırım and C. Durucan, "Synthesis of Zinc Oxide Nanoparticles Elaborated by Microemulsion Method," *Journal of Alloys and Compounds* 506, no. 2 (2010): 944–949, https://doi.org/10.1016/j.jallcom.2010.07.125.

34. N. Lepot, M. K. Van Bael, H. Van den Rul, et al., "Synthesis of ZnO Nanorods From Aqueous Solution," *Materials Letters* 61, no. 13 (2007): 2624–2627, https://doi.org/10.1016/j.matlet.2006.10.025.

35. N. Panahandeh, H. Torabzadeh, M. Aghaee, E. Hasani, and S. Safa, "Effect of Incorporation of Zinc Oxide Nanoparticles on Mechanical Properties of Conventional Glass Ionomer Cements," *Journal of Conservative Dentistry* 21, no. 2 (2018): 130–135, https://doi.org/10.4103/JCD.JCD_170_17.

36. N. Talebian, S. M. Amininezhad, and M. Doudi, "Controllable Synthesis of ZnO Nanoparticles and Their Morphology-Dependent Antibacterial and Optical Properties," *Journal of Photochemistry and Photobiology B: Biology* 120 (2013): 66–73, https://doi.org/10.1016/j.jphotobiol.2013.01.004.

37. A. Naveed Ul Haq, A. Nadhman, I. Ullah, G. Mustafa, M. Yasinzai, and I. Khan, "Synthesis Approaches of Zinc Oxide Nanoparticles: The Dilemma of Ecotoxicity," *Journal of Nanomaterials* 2017 (2017): 1–14, https://doi.org/10.1155/2017/8510342.

38. S. Sharma, K. Kumar, N. Thakur, S. Chauhan, and M. S. Chauhan, "The Effect of Shape and Size of ZnO Nanoparticles on Their Antimicrobial and Photocatalytic Activities: A Green Approach," *Bulletin of Materials Science* 43, no. 1 (2020): 20, https://doi.org/10.1007/s12034-019-1986-y.

39. X. Zhai, S. Zhou, R. Zhang, W. Wang, and H. Hou, "Antimicrobial Starch/Poly(butylene adipate-co-terephthalate) Nanocomposite Films Loaded With a Combination of Silver and Zinc Oxide Nanoparticles for Food Packaging," *International Journal of Biological Macromolecules* 206 (2022): 298–305, https://doi.org/10.1016/j.ijbiomac.2022.02.158.

40. C. Ferreira, P. Souza, H. Toma, E. Silva, M. I. Tavares, and G. Iulianelli, "Influence of ZnO Nanoparticles on the Properties of Biodegradable PBAT Matrix for Food Packaging Application," *Concilium* 23, no. 18 (2023): 226–240, https://doi.org/10.53660/CLM-1923-23N12.

41. R. Venkatesan and N. Rajeswari, "ZnO/PBAT Nanocomposite Films: Investigation on the Mechanical and Biological Activity for Food Packaging," *Polymers for Advanced Technologies* 28, no. 1 (2017a): 20–27, https://doi.org/10.1002/pat.3847.

42. K. Charoensri, C. Rodwihok, S. H. Ko, D. Wongratanaphisan, and H. J. Park, "Enhanced Antimicrobial and Physical Properties of Poly (Butylene adipate-co-terephthalate)/Zinc Oxide/Reduced Graphene Oxide Ternary Nanocomposite Films," *Materials Today Communications* 28 (2021): 102586, https://doi.org/10.1016/j.mtcomm.2021.102586.

43. M. Seray, A. Skender, and A. S. Hadj-Hamou, "Kinetics and Mechanisms of Zn2+ Release From Antimicrobial Food Packaging Based on Poly(butylene adipate-co-terephthalate) and Zinc Oxide Nanoparticles," *Polymer Bulletin* 78, no. 2 (2021): 1021–1040, https://doi.org/10.1007/s00289-020-03145-z.

44. Ü. Özgür, Y. Alivov, C. Liu, et al., "A Comprehensive Review of ZnO Materials and Devices," *Journal of Applied Physics* 98, no. 4 (2005): 2–103, https://doi.org/10.1063/1.1992666.

45. C. B. Ong, L. Y. Ng, and A. W. Mohammad, "A Review of ZnO Nanoparticles as Solar Photocatalysts: Synthesis, Mechanisms and Applications," *Renewable and Sustainable Energy Reviews* 81 (2018): 536–551, https://doi.org/10.1016/j.rser.2017.08.020.

46. A. Mtibe, L. Hlekelele, P. E. Kleyi, et al., "Fabrication of a Polybutylene Succinate (PBS)/Polybutylene adipate-co-terephthalate (PBAT)-Based Hybrid System Reinforced With Lignin and Zinc Nanoparticles for Potential Biomedical Applications," *Polymers* 14, no. 23 (2022): 5065, https://doi.org/10.3390/polym14235065.

47. A. G. de Souza, L. G. H. Komatsu, R. F. S. Barbosa, D. F. Parra, and D. S. Rosa, "The Effect of ZnO Nanoparticles as Ag-Carrier in PBAT for Antimicrobial Films," *Polymer Bulletin* 79, no. 6 (2022): 4031–4048, https://doi.org/10.1007/s00289-021-03681-2.

48. H.-J. Kim, C.-L. Lee, K.-S. Yoon, and J.-W. Rhim, "Synergistic Effect of UV-C LED Irradiation and PLA/PBAT-Based Antimicrobial Packaging Film on Fresh-Cut Vegetables," *Food Control* 138 (2022a): 109027, https://doi.org/10.1016/j.foodcont.2022.109027.

49. I. Kim, K. Viswanathan, G. Kasi, S. Thanakkasaranee, K. Sadeghi, and J. Seo, "ZnO Nanostructures in Active Antibacterial Food Packaging: Preparation Methods, Antimicrobial Mechanisms, Safety Issues, Future Prospects, and Challenges," *Food Reviews International* 38, no. 4 (2022b): 537–565, https://doi.org/10.1080/87559129.2020.1737709.

50. D. Phothisarattana, P. Wongphan, K. Promhuad, J. Promsorn, and N. Harnkarnsujarit, "Blown Film Extrusion of PBAT/TPS/ZnO Nanocomposites for Shelf-Life Extension of Meat Packaging," *Colloids and Surfaces B: Biointerfaces* 214 (2022): 112472, https://doi.org/10.1016/j.colsurfb.2022.112472.

51. K. Promhuad, D. Phothisarattana, Y. Laorenza, N. Bumbudsanpharoke, and N. Harnkarnsujarit, "Zinc Oxide Enhanced the Antibacterial Efficacy of Biodegradable PBAT/PBS Nanocomposite Films: Morphology and Food Packaging Properties," *Food Bioscience* 55 (2023): 103077, https://doi.org/10.1016/j.fbio.2023.103077.

52. S. Shankar and J.-W. Rhim, "Effects of Poly(butylene adipate-co-terephthalate) Coating on the Water Resistant, Mechanical, and Antibacterial Properties of Kraft Paper," *Progress in Organic Coatings* 123 (2018): 153–159, https://doi.org/10.1016/j.porgcoat.2018.07.002.

53. S. Shankar and J.-W. Rhim, "Effect of Types of Zinc Oxide Nanoparticles on Structural, Mechanical and Antibacterial Properties of Poly(lactide)/Poly(butylene adipate-co-terephthalate) Composite Films," *Food Packaging and Shelf Life* 21 (2019): 100327, https://doi.org/10.1016/j.fpsl.2019.100327.

54. T. T. Thiyagu, G. Gokilakrishnan, V. C. Uvaraja, T. Maridurai, and V. R. A. Prakash, "Effect of SiO_2/TiO_2 and ZnO Nanoparticle on Cardanol Oil Compatibilized PLA/PBAT Biocomposite Packaging Film," *Silicon* 14, no. 7 (2022): 3795–3808, https://doi.org/10.1007/s12633-021-01577-4.

55. T. Wang, Y. Shi, Y. Li, and L.-Z. Liu, "The Effects of ZnO Nanoparticle Reinforcement on Thermostability, Mechanical, and Optical Properties of the Biodegradable PBAT Film," *Journal of Polymer Engineering* 41, no. 10 (2021): 835–841, https://doi.org/10.1515/polyeng-2021-0150.

56. L. Xiao, Z. Yao, Y. He, et al., "Antioxidant and Antibacterial PBAT/Lignin-ZnO Nanocomposite Films for Active Food Packaging," *Industrial Crops and Products* 187 (2022): 115515, https://doi.org/10.1016/j.indcrop.2022.115515.

57. T. Yi, M. Qi, Q. Mo, et al., "Ecofriendly Preparation and Characterization of a Cassava Starch/Polybutylene Adipate Terephthalate Film," *Pro* 8, no. 3 (2020): 329, https://doi.org/10.3390/pr8030329.

58. F.-F. Ge, C.-H. Tsou, S. Yuan, et al., "Barrier Performance and Biodegradability of Antibacterial Poly(butylene adipate-co-terephthalate) Nanocomposites Reinforced With a New MWCNT-ZnO Nanomaterial," *Nanotechnology* 32, no. 48 (2021): 485706, https://doi.org/10.1088/1361-6528/ac1b52.

59. D. Phothisarattana and N. Harnkarnsujarit, "Migration, Aggregations and Thermal Degradation Behaviors of TiO_2 and ZnO Incorporated PBAT/TPS Nanocomposite Blown Films," *Food Packaging and Shelf Life* 33 (2022): 100901, https://doi.org/10.1016/j.fpsl.2022.100901.

60. X. Hu, S. Cook, P. Wang, and H. Hwang, "In Vitro Evaluation of Cytotoxicity of Engineered Metal Oxide Nanoparticles," *Science of the Total Environment* 407, no. 8 (2009): 3070–3072, https://doi.org/10.1016/j.scitotenv.2009.01.033.

61. K. S. Siddiqi, A. ur Rahman, N. Tajuddin, and A. Husen, "Properties of Zinc Oxide Nanoparticles and Their Activity Against Microbes," *Nanoscale Research Letters* 13, no. 1 (2018): 141, https://doi.org/10.1186/s11671-018-2532-3.

62. K. Kasemets, A. Ivask, H.-C. Dubourguier, and A. Kahru, "Toxicity of Nanoparticles of ZnO, CuO and TiO_2 to Yeast Saccharomyces Cerevisiae," *Toxicology in Vitro* 23, no. 6 (2009): 1116–1122, https://doi.org/10.1016/j.tiv.2009.05.015.

63. L. Zhang, Y. Ding, M. Povey, and D. York, "ZnO Nanofluids – A Potential Antibacterial Agent," *Progress in Natural Science* 18, no. 8 (2008): 939–944, https://doi.org/10.1016/j.pnsc.2008.01.026.

64. R. Jalal, E. K. Goharshadi, M. Abareshi, M. Moosavi, A. Yousefi, and P. Nancarrow, "ZnO Nanofluids: Green Synthesis, Characterization, and Antibacterial Activity," *Materials Chemistry and Physics* 121, no. 1–2 (2010): 198–201, https://doi.org/10.1016/j.matchemphys.2010.01.020.

65. F. A. G. Soares Silva, T. Bento de Carvalho, F. Dourado, M. Gama, P. Teixeira, and F. Poças, "Performance of Bacterial Nanocellulose Packaging Film Functionalised In Situ With Zinc Oxide: Migration Onto Chicken Skin and Antimicrobial Activity," *Food Packaging and Shelf Life* 39 (2023a): 101140, https://doi.org/10.1016/j.fpsl.2023.101140.

66. F. A. G. Soares Silva, M. Carvalho, T. Bento de Carvalho, M. Gama, F. Poças, and P. Teixeira, "Antimicrobial Activity of In-Situ Bacterial Nanocellulose-Zinc Oxide Composites for Food Packaging," *Food Packaging and Shelf Life* 40 (2023b): 101201, https://doi.org/10.1016/j.fpsl.2023.101201.

67. A. Marcous, S. Rasouli, and F. Ardestani, "Low-Density Polyethylene Films Loaded by Titanium Dioxide and Zinc Oxide Nanoparticles as a New Active Packaging System Against *Escherichia coli* O157:H7 in Fresh Calf Minced Meat," *Packaging Technology and Science* 30, no. 11 (2017): 693–701, https://doi.org/10.1002/pts.2312.

68. S.-E. Jin and H.-E. Jin, "Antimicrobial Activity of Zinc Oxide Nano/Microparticles and Their Combinations Against Pathogenic Microorganisms for Biomedical Applications: From Physicochemical Characteristics to Pharmacological Aspects," *Nanomaterials* 11 (2021): 263, https://doi.org/10.3390/nano11020263.

69. B. Lallo da Silva, M. P. Abuçafy, E. Berbel Manaia, et al., "Relationship Between Structure and Antimicrobial Activity of Zinc Oxide Nanoparticles: An Overview," *International Journal of Nanomedicine* 14 (2019): 9395–9410, https://doi.org/10.2147/IJN.S216204.

70. A. Stanković, S. Dimitrijević, and D. Uskoković, "Influence of Size Scale and Morphology on Antibacterial Properties of ZnO Powders Hydrothemally Synthesized Using Different Surface Stabilizing Agents," *Colloids and Surfaces B: Biointerfaces* 102 (2013): 21–28, https://doi.org/10.1016/j.colsurfb.2012.07.033.

71. N. Tamimi, A. Mohammadi Nafchi, H. Hashemi-Moghaddam, and H. Baghaie, "The Effects of Nano-Zinc Oxide Morphology on Functional and Antibacterial Properties of Tapioca Starch Bionanocomposite," *Food Science & Nutrition* 9, no. 8 (2021): 4497–4508, https://doi.org/10.1002/fsn3.2426.

72. R. Venkatesan, T. T. Thiyagu, and N. Rajeswari, "Zinc Composite Materials and Food Packaging," in *Composites Materials for Food*

Packaging (Hoboken, New Jersey: Wiley, 2018), 153–173, https://doi.org/10.1002/9781119160243.ch4.

73. M. J. Mehta and A. Kumar, "Ionic Liquid Assisted Gelatin Films: Green, UV Shielding, Antioxidant, and Antibacterial Food Packaging Materials," *ACS Sustainable Chemistry & Engineering* 7, no. 9 (2019): 8631–8636, https://doi.org/10.1021/acssuschemeng.9b00423.

74. M. Zolfi, F. Khodaiyan, M. Mousavi, and M. Hashemi, "The Improvement of Characteristics of Biodegradable Films Made From Kefiran–Whey Protein by Nanoparticle Incorporation," *Carbohydrate Polymers* 109 (2014): 118–125, https://doi.org/10.1016/j.carbpol.2014.03.018.

75. A. Pietrosanto, P. Scarfato, L. Di Maio, M. R. Nobile, and L. Incarnato, "Evaluation of the Suitability of Poly(lactide)/Poly(butylene-adipate-co-terephthalate) Blown Films for Chilled and Frozen Food Packaging Applications," *Polymers* 12, no. 4 (2020): 804, https://doi.org/10.3390/polym12040804.

76. E. E. Ubuo, I. A. Udoetok, A. T. Tyowua, I. O. Ekwere, and H. S. Al-Shehri, "The Direct Cause of Amplified Wettability: Roughness or Surface Chemistry?" *Journal of Composites Science* 5, no. 8 (2021): 213, https://doi.org/10.3390/jcs5080213.

77. J. Jian, Z. Xiangbin, and H. Xianbo, "An Overview on Synthesis, Properties and Applications of Poly(butylene-adipate-co-terephthalate)–PBAT," *Advanced Industrial and Engineering Polymer Research* 3, no. 1 (2020): 19–26, https://doi.org/10.1016/j.aiepr.2020.01.001.

78. S. M. Chisenga, G. N. Tolesa, and T. S. Workneh, "Biodegradable Food Packaging Materials and Prospects of the Fourth Industrial Revolution for Tomato Fruit and Product Handling," *International Journal of Food Science* 2020 (2020): 1–17, https://doi.org/10.1155/2020/8879101.

79. L. Zimmermann, A. Dombrowski, C. Völker, and M. Wagner, "Are Bioplastics and Plant-Based Materials Safer Than Conventional Plastics? In Vitro Toxicity and Chemical Composition," *Environment International* 145 (2020): 106066, https://doi.org/10.1016/j.envint.2020.106066.

80. C. V. Garcia, G. H. Shin, and J. T. Kim, "Metal Oxide-Based Nanocomposites in Food Packaging: Applications, Migration, and Regulations," *Trends in Food Science & Technology* 82 (2018): 21–31, https://doi.org/10.1016/j.tifs.2018.09.021.

81. Y. Picó, "Safety Assessment and Migration Tests," in *Nanomaterials for Food Packaging* (New York, USA: Elsevier, 2018), 249–275, https://doi.org/10.1016/B978-0-323-51271-8.00009-7.

82. C. Sharma, R. Dhiman, N. Rokana, and H. Panwar, "Nanotechnology: An Untapped Resource for Food Packaging," *Frontiers in Microbiology* 8 (2017): 1735, https://doi.org/10.3389/fmicb.2017.01735.

83. S. Keerthana and A. Kumar, "Potential Risks and Benefits of Zinc Oxide Nanoparticles: A Systematic Review," *Critical Reviews in Toxicology* 50, no. 1 (2020): 47–71, https://doi.org/10.1080/10408444.2020.1726282.

84. A. Störmer, J. Bott, D. Kemmer, and R. Franz, "Critical Review of the Migration Potential of Nanoparticles in Food Contact Plastics," *Trends in Food Science & Technology* 63 (2017): 39–50, https://doi.org/10.1016/j.tifs.2017.01.011.

85. R. Franz, J. Bott, and A. Störmer, "Considerations for and Guidance to Testing and Evaluating Migration/Release of Nanoparticles From Polymer Based Nanocomposites," *Nanomaterials* 10, no. 6 (2020): 1113, https://doi.org/10.3390/nano10061113.

86. J. Bott, A. Störmer, and R. Franz, "A Comprehensive Study Into the Migration Potential of Nano Silver Particles From Food Contact Polyolefins (pp. 51–70)," (2014a), https://doi.org/10.1021/bk-2014-1159.ch005.

87. I. K. Ventouri, S. Loeber, G. W. Somsen, P. J. Schoenmakers, and A. Astefanei, "Field-Flow Fractionation for Molecular-Interaction Studies of Labile and Complex Systems: A Critical Review," *Analytica Chimica Acta* 1193 (2022): 339396, https://doi.org/10.1016/j.aca.2021.339396.

88. J. Bott, A. Störmer, and R. Franz, "Migration of Nanoparticles From Plastic Packaging Materials Containing Carbon Black Into Foodstuffs," *Food Additives & Contaminants: Part a* 31, no. 10 (2014b): 1769–1782, https://doi.org/10.1080/19440049.2014.952786.

89. J. Liu, J. Hu, M. Liu, G. Cao, J. Gao, and Y. Luo, "Migration and Characterization of Nano-Zinc Oxide From Polypropylene Food Containers," *American Journal of Food Technology* 11, no. 4 (2016): 159–164, https://doi.org/10.3923/ajft.2016.159.164.

90. N. Bumbudsanpharoke, J. Choi, H. J. Park, and S. Ko, "Zinc Migration and Its Effect on the Functionality of a Low Density Polyethylene-ZnO Nanocomposite Film," *Food Packaging and Shelf Life* 20 (2019): 100301, https://doi.org/10.1016/j.fpsl.2019.100301.

91. H. Huang, K. Tang, Z. Luo, H. Zhang, and Y. Qin, "Migration of Ti and Zn From Nanoparticle Modified LDPE Films Into Food Simulants," *Food Science and Technology Research* 23, no. 6 (2017): 827–834, https://doi.org/10.3136/fstr.23.827.

92. W. Lee, N. Bumbudsanpharoke, G.-H. Gawk, J.-M. Oh, and S. Ko, "Effect of Repeated Contact to Food Simulants on the Chemical and Functional Properties of Nano ZnO Composited LDPE Films for Reusable Food Packaging," *Polymers* 15, no. 1 (2022): 9, https://doi.org/10.3390/polym15010009.

93. B. Panea, G. Ripoll, J. González, Á. Fernández-Cuello, and P. Albertí, "Effect of Nanocomposite Packaging Containing Different Proportions of ZnO and Ag on Chicken Breast Meat Quality," *Journal of Food Engineering* 123 (2014): 104–112, https://doi.org/10.1016/j.jfoodeng.2013.09.029.

94. M. Heydari-Majd, B. Ghanbarzadeh, M. Shahidi-Noghabi, M. A. Najafi, P. Adun, and A. Ostadrahimid, "Kinetic Release Study of Zinc from Polylactic Acid Based Nanocomposite into Food Simulants," *Polymer Testing* 76 (2019): 254–260, https://doi.org/10.1016/j.polymertesting.2019.03.040.

95. S. Srishti, P. Joel, G. Patricia, et al., "Safety Profile of ZnO Active Packaging PBAT Based Biomaterial for Food Packaging. First Tier Evaluation," *Food Control* 161 (2024): 110389.

96. "Commission Regulation (EU) No 10/2011 of 14 January 2011 on Plastic Materials and Articles Intended to Come Into Contact With Food," (forthcoming). In Official Journal of the European Union.