



# Tailoring of Polymer and Metal Nanobiocomposites Corroborated with Smart Food Packaging Systems—A Review

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Received: 1 May 2023 / Accepted: 14 June 2023 / Published online: 12 July 2023

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## Abstract

There is a robust industrial drive for resolving the issues related to packaging materials for contributing a pivotal essence towards circular economy via their recycling. Nanotechnology is one of them, which is able to contribute and develop a prime impact on both quantitative and qualitative manufacturing of safe and effective foods/food components (perishable or semi-perishable) with a pragmatic assessment of enhanced shelf life. In this review, a brief overview was perceived on nanocomposite materials towards food packaging. Numerous polymeric/metallic nanocomposite systems (natural and synthetic) were explored for their migration issues; antioxidant, antimicrobial, and barrier properties; recyclability; consumer acceptability; toxicity; and regulatory aspects. The discussion was also extended with some recent trends and future perspectives of cutting-edge nanomaterials (nanosensors) in smart food processing, packaging, security, storage, quality evaluation of preserved foods, and the methods arrayed for assessing the nanomaterial impact over the biological systems.

**Keywords** Nanotechnology · Nanocomposite · Nanosensors · Food packaging

## Introduction

In today's era, the nanomaterial demand has been expanding significantly and can be recognized as a fastest growing market. Nanostructured materials have vast applications compared to their bulk counterparts. Nanotechnology covers numerous fields like medical, agriculture, environment, and food segments with having an emerging research scope in agri-food industries revealing a remarkable global growth rate in food production with a superior quality, safety, and nutritional value (Sekhon, 2014; Thiruvengadam et al., 2018). Engineered nanomaterials can accelerate testing and

monitoring of adulteration in food products (He et al., 2019). Nanosized particles were improved by their biological efficiency and surface to volume ratio (Naghdi et al., 2019). The revenue generated by the nanotechnological market has already been achieved up to \$38.5 billion in 2020 and is being expected to reach an annual growth rate of 12.2% between 2021 and 2026 (Chausali et al., 2022). The current demand for biocentered food packaging elements is rising at an annual rate of 18.3% (Cerqueira et al., 2016).

In packaging, nanocomposites overwhelmed the foot-races of orthodox packaging pattern and provoking antimicrobial, thermal, barrier, mechanical, and degradation tendency followed by nanosensing for enhancing consumer alertness towards varied circumstances (temperature, gas, moisture, contaminants, etc.) essential for maintaining the safety of food products (Pereda et al., 2017). Incorporating nanocomposites and nanoparticles (nanometal, nanofillers, nanosensors, bioactive compounds, metal oxides, antioxidants, mixed polymers, and oxygen scavengers) has advanced smart/intelligent, active, and biobased packaging systems (Cerqueira et al., 2018; Primožič et al., 2021). Biobased packaging via biocompatible/biodegradable (polylactic acid (PLA), ethylene terephthalate (bio-poly), polybutylene succinate (PBS), polyester amide (PA), starch and cellulose thermoplastics, and polyvinyl alcohol (PVA))

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bionanomaterials turned to be a viable substitute against conventional packaging (plastic). Incorporating lipid-based coatings films (edible) (acylglycerols, fatty acids, essential oils, waxes, and extracts), proteins (zein, gelatin, myofibrillar proteins, milk, and soy proteins), and polysaccharides/biopolymers (cellulose, starch, chitosan, pectin, alginate, kefiran, carrageenan, and pullulan) in food packaging ensured food safety and safeguard the environment from pollution (Primožič et al., 2021). Conversely, nanocomposite enabled smart packaging (based on temperature–time integrating systems), and sensors (e.g., gas detectors) reveal contamination and spoilage via pathogens, microorganisms, air/gases, and organic molecules (Ranjan et al., 2014). Thus, the current food packaging trends are associated with nanoreinforcement, nanocomposite, nanosensing, and biodegradability for providing nutritious and safe food products to the consumers (Dasgupta & Ranjan, 2018). The migrational peril of nanoparticles in food items, along with their presumed toxicity, is a matter of apprehension. Inadequate risk assessment and deficient clinical trial outcomes followed by a few studies seem to barricade the social and commercial acceptability of nanopackaged food products.

Despite the predominant public opinion against the utility of nanotechnology in food segment, there has been a desperate interest on nanocomposites being a material of choice towards food packaging since the last two decades (Duncan, 2011; Parisi et al., 2015; Taherimehr et al., 2021; Huang et al., 2023; Nile et al., 2020). Several polymers with multi-layered structures are utilized in food packaging to obtain adequate barrier, mechanical, and sealing properties (Alias et al., 2022). Despite the non-recyclable tendency of multi-layered structures, a very strong industrial urge is there for developing packaging components which could target green consumerism and sustainable development at a sound economy, for pivoting the recyclability of the packaging components with additional safety (Asim et al., 2022; Hopewell et al., 2009; Wandosell et al., 2021). However, polymer nanocomposites (PNCs) as packaging materials could fulfill the above domain with an optimum recyclability. PNCs are usually developed by dispersing the inert, nanoscale fillers like silica nanoparticles (Perera et al., 2023; Zou et al., 2008), carbon nanotubes, silicate and clay nanoplatelets (Jamali et al., 2023; Singh et al., 2023a, b), starch nanocrystals (Borriello et al., 2009), chitin/chitosan nanoparticles (Chen et al., 2008), graphene (Wang et al., 2022), nanowhiskers, and inorganics or cellulose-based nanofibers (Bilbao-Sainz et al., 2010; Cao et al., 2008; Chadha et al., 2022), throughout a polymeric matrix (Duncan, 2011). The PNC showed an improved strength (Li et al., 2009), thermal properties (Yang et al., 2008), and flame resistance (Popescu et al., 2023).

In compliance with the green revolution/go green concept, industries are moving towards developing biobased

packaging materials, restraining the utility of fossil-based plastics. However, biodegradable/biobased materials either from synthetic origins (e.g., polyhydroxyalkanoates (PHA), poly-(butylene succinate) (PBS)) and PLA or from natural resources (e.g., alginate, starch, gelatin, or chitosan) have usually shown an inferior mechanical and barrier properties (brittleness, low heat distortion temperature, and pitiable resistance on the way to deformation) compared with the conventional plastics (fossil based) (Garavand et al., 2022; Kuswandi, 2017). Hence, a significant research effort was being amended for developing biomaterials that could meet the standards for several food products regarding fat and water resistance, gas barrier ( $O_2$ ,  $CO_2$ , and  $H_2O$ ), and mechanical properties, along with possessing utmost industrial efficiency. The nanotechnology could be a better option for developing biomaterials which could fulfill the industrial scale-up along with cost-efficiency (Othman, 2014; Rhim et al., 2013a, b). The biopolymer nanocomposites recently explored for their packaging applications include thermoplastic starch and derivatives, PBS, PLA, and polyhydroxybutyrate (PHB) (Othman, 2014). The research focused on biopolymers, such as chitosan, cellulose, and gelatin, for the advancement of food packaging with cellulosic paper-based materials (Youssef & El-Sayed, 2018).

The current review is dealing with many nanomaterial-mediated packaging with a special importance for biobased packaging components/systems. The article has emphasized on the current trends in the development of biobased packaging with several improved features, smart and intelligent packaging, and next-generation packaging (polymers derived from agro-food waste for development of eco-friendly packaging) for restricting the usage of plastic waste and their degradation issues. In this review, we have summarized the significance of nanocomposites towards food packaging with a special emphasis towards antimicrobial effect, barrier properties, migrational issues, antioxidant properties, consumer acceptability, toxicity, evaluation, and regulatory standards for packaging components, including their recyclability aspects.

## Nanotechnology in Food Packaging

In the current scenario, the contribution of packaging industry to the world economy is very high (around 55–65% of \$130 billion) (Berrabah et al., 2023). In muscle-based food products, the involvement of intelligent and active packaging systems has immensely increased in order to suppress spoilage, enhance the enzyme-based tenderness, and bypass contamination, retention of the originality/freshness (cherry red color) in red meats, and reduction in weight loss (Attaran et al., 2017). The nanosensors are applicable for detection of pesticides, microbial contamination, and toxins in the food

products (based on the production of color and flavor) which can be helpful in providing contamination or food spoilage alarm to the consumers (Sahoo et al., 2021). The nanoparticle (NP)-based packaging systems in the food industry showed a potential antimicrobial efficiency, considered carriers (antimicrobial) for polypeptides, which protects against microbial spoilage and improves shelf life of products (Jafarzadeh et al., 2023; Zhang et al., 2023). The packaging material is coated with starch containing antimicrobial agents acting as a barrier towards the microbes (Cai et al., 2022).

The metals and metal oxide NPs like silver, iron, carbon, zinc oxides, silicon dioxide, titanium oxides, and magnesium oxide are widely explored with their antimicrobial potentials and also incorporated as food ingredients/additives (Sahoo et al., 2021; Kaur & Sidhu, 2021). Enriched resistance to heat, mechanical strength, and low weight along with an enhanced barrier against CO<sub>2</sub>, O<sub>2</sub>, UV radiation, volatiles, moisture, and the development of reactive oxygen species (ROS) via TiO<sub>2</sub> (pernicious towards microbes of pathogenic origin) can be achieved by the help of nanocomposites.

Abundant NPs like silicate nanoplatelets, clay, SiO<sub>2</sub>, graphene, starch nanocrystals, carbon nanotubes, chitosan or chitin NPs, cellulose-mediated nanofibers, and many inorganics can be applied as fillers in a polymeric matrix, thereby making it as fire-resistant, highly reactive, and lighter with enhanced thermal properties followed by the low permeability towards gases which are commonly used for coating and packaging purposes (Berrabah et al., 2023; Kumar & Gaikwad, 2023; Mihindukulasuriya & Lim, 2014; Pinto et al., 2013). The presence of silver in the silver zeolite showed antimicrobial potential through generation of ROS. Silver zeolite coated with ceramics is useful to preserve food, disinfection of medical products, and decontamination of materials (Inobeme & Adetunji, 2023; Singh et al., 2017). Carbon nanotubes enable the acclimatization of unpleasant flavors by eliminating CO<sub>2</sub>. The food packaging components and bottles developed from nanoclay-based nanocomposites (bentonite) significantly augment the features of gas barrier and, thus, inhibit the diffusion of moisture and oxygen, spoilage of food materials, and drink destabilization (Egger et al., 2009; Mylvaganam & Rathnayake, 2020). The polymeric nanomatrices enhanced the concert of packaging material for food and offer several functional attributes like antimicrobial and antioxidant, as well as scavenging that leads to a prolonged shelf life of packed food materials (Cai et al., 2022). The amalgamation of clay NPs into the polylactic acid (PLA) and ethylene vinyl alcohol has shown progress in mechanical strength and oxygen barrier, increases the shelf life of food materials, and is a barrier towards volatiles, moisture, and gases (Chaudhry et al., 2008; Kaur et al., 2023). PLA bionanocomposites showed a faster biodegradation rate compared to PLA without nanofillers (Berrabah et al., 2023; Garcia, 2022).

Chitosan-based nanocomposite films also have shown potential antimicrobial activity, especially with silver-based nanocomposites (Lindström & Österberg, 2020). The phytylglycogen octenyl NPs along with the ε-polylysine and PEG-coated NPs enhanced the shelf life of essential oil from garlic (Scheffler et al., 2010; Yang et al., 2009). In food packaging, silicate NPs decrease the drying and spoilage of food by acting as a barrier for moisture and gases (Neethirajan & Jayas, 2011). In nanomicelle-based products, the addition of glycerin removes residues of pesticide from vegetables and fruits, as well as oil/dirt from cutlery (Kausar, 2020; Lindström & Österberg, 2020). Nanoemulsions can easily control several food pathogens like gram-negative bacteria, along with intelligent and active packaging systems.

### Nanotechnology Overwhelming Dominance over Conventional Packaging Technologies

The appropriate analysis of polymeric properties (mechanical, thermal, and barrier) leads to an assessment of maneuverability and product-package shelf life via incorporation of NPs/nanofillers/nanocomposites/nanocoatings/surface biocides (with high surface area and aspect ratio) (Rhim et al., 2013a, b; Sharma et al., 2020) transforming the biopolymers into bionanocomposites for exploring the potentials in three-dimensional (3D) pattern (Hoseinejad et al., 2018, Joz Majidi et al., 2019). The permeability of polymeric materials usually depends on (i) polymer/material characteristics (crystallinity, molecular orientation, chain stiffness, and free volume); (ii) properties of the permeants (nature and molecule size of the polymers) for restraining oxygen and water vapor; and (iii) environmental factors (such as moisture content and temperature) (Bahrami et al., 2020). Though polysaccharides denoted an effective barrier property towards gas transference (O<sub>2</sub> and CO<sub>2</sub>), preventing surface-browning and oxidative rancidity, but still, they cannot convey barrier properties towards water vapor and moisture. Polyelectrolyte complexes via electrostatic interactions can also reveal barrier and mechanical properties (elastic modulus, tensile strength, and elasticity). For intriguing the biopolymer-based food packaging system from hypothetical to reality, it needs some sort of improvisation (such as improved barrier properties of food packaging components/materials for declining moisture and gas transfer along with protection against UV light exposure, to enhance thermal, mechanical, and antimicrobial properties via reinforcement of biopolymers like nanocellulose and montmorillonite) via incorporation of organic or inorganic NPs (such as metal oxide NPs, metal NPs, mixed metal oxide NPs, carbon nanomaterials (graphene), and nanoclay) to correct the shortfalls. Despite immense potentials, certain challenges like cost of production associated with bionanocomposites and lack of information regarding the toxicity/ecotoxicity of nanofillers

along with their migration into food components add constraints towards the growth rate of bionanocomposites in the current era (Jafari et al., 2015; Pilevar et al., 2019; Pires et al., 2021; Vahedikia et al., 2019).

### Impact of Metal NPs for Influencing the Barrier Properties of Food Packaging Composites

Nanocomposites carry the combo of polymer/biopolymer and NPs (of either inorganic or organic fillers with particle size < 100 nm) to develop an impermeable tortuous path, restricting the gaseous (O<sub>2</sub> and CO<sub>2</sub>) diffusion (Duncan, 2011, Ture et al., 2013). The NPs in the composite also develop nucleation of heterogeneous crystals in the polymer matrix, diminishing the permeability of the contaminants (Jafarzadeh et al., 2016). The reinforcement of metallic NPs like Ag and nano-SiO<sub>2</sub>, into different biodegradable polymers such as carrageenan and gelatin, leads to limitation of the water vapor permeability (WVP) by developing impermeable barrier matrix (Jafarzadeh et al., 2016, 2018; Kanmani & Rhim, 2014b; Rane et al., 2014; Reddy et al., 2018; Rhim & Wang, 2014; Shankar et al., 2015; Tabatabaei et al., 2018). The reinforcement of the polymers such as gelatin, soybean polysaccharide, and poly (3-hydroxybutyrate-co-3-hydroxyvalerate) with NPs like ZnO, TiO<sub>2</sub>, and Ag leads to restriction of the permeability of oxygen (OP) (by developing the impermeable matrix) into the food materials, enhancing their shelf life, which is equally supported by various research outcomes (Castro-Mayorga et al., 2016; Jafarzadeh et al., 2017; Nafchi et al., 2013). NPs (such as TiO<sub>2</sub>, CuO, ZnO, and Ag) incorporated into various polymeric films develop hydrogen bonding (between NPs and the matrix) resulting in an elevated surface interaction which improves the thermal, morphological, chemical, and mechanical/tensile strength of the packaging composite along with provision of optical protection against UV light to preserve transparency, color, and UV absorbance/transmission (Hasheminya et al., 2018; Jafarzadeh et al., 2017; Kanmani & Rhim, 2014b; Shaili et al., 2015; Zolfi et al., 2014).

In view of the migration, safety, and toxicological aspects of metal NPs towards food products, the reinforcement of polymer materials with NPs usually develops filler-matrix interaction (nanocomposites) resulting in particle size reduction and develops a matrix-filler compatibility (Jafarzadeh & Jafari, 2021). Thus, the fabricated nanocomposite packaging materials can lead to the development of protection against the contaminants and toxins and boost up preservation, stability, dimensional stiffness, strength, communication, and marketing of numerous food items (Jafarzadeh et al., 2021). However, it was reported that the free NPs lead to the development of oxidative stress and inflammatory conditions along with organ damage by crossing the cellular barriers (Maisanaba et al., 2015a, b). Hence, further research

is needed to understand and resolve the migrational issues of NPs into food products from the nanocomposite packaging (Jafarzadeh & Jafari, 2021).

### Barrier Properties of Polymer Nanocomposites

The nanofillers distributed uniformly all through the polymer matrix modify the molecular diffusion rate by developing tortuous pathway resulting in an enhanced barrier-like properties. Moreover, the polymer nanocomposites revealing barrier properties can also be influenced by restricting the mobility of the polymer strands (Wu et al., 2021). Several polymers unveil numerous barrier properties, such as polyethylene terephthalate (PET) that proposed virtuous barrier properties on the way to oxygen, paralleled to high-density polyethylene (HDPE). Likewise, HDPE reveals a superior barrier property against water vapor than that of PET (Yam, 2010). Usually, the polymeric barrier properties are relying on several factors, like hydrogen bonding, polarity, branching, crystallinity, and cross-linking (Duncan, 2011). Moreover, one migrant could be exaggerated with its permeability in the presence of another, e.g., a significant reduction of oxygen barrier properties of ethylene vinyl alcohol (EVOH) in the presence of high-humidity conditions because of the polymeric plasticization and swelling (Yam, 2009).

### Stability and Antimicrobial Approaches of NPs/Nanocomposites

Several factors like processing condition, nature of the product, storage and distribution, and type of package specifically affect the shelf life of foods and food products (Emblem, 2012). Moreover, the intrinsic factors (e.g., microbes, pH, water activity, level of reactive compounds, and enzymes) along with certain extrinsic factors (e.g., temperature, total pressure, relative humidity, light, partial pressure of various gases, and mechanical stress) impel the food material degradation at storage condition (Fedotova et al., 2010). The lethality of the existing microbes over the surface of food and packaging materials was greatly influenced by the generation of NP-mediated ROS. The antimicrobials (natural) or NPs of CuO, Cu, Ag, MgO, Pd, ZnO, Fe, and TiO<sub>2</sub>, contained in nanoemulsions/nanoencapsulations, could be adhered either by hydrogen, electrostatic, or covalent bonding to develop antimicrobial packaging elements. The disability of *Salmonella enterica*, *Listeria innocua*, and *Escherichia coli* via potential surface charge of engineered water nanostructures (EWNS) can be efficiently carried out on the tomato and stainless steel surface without manipulating the quality (sensory) of food through production of ROS,

which thus leads to the development of water vapor which sinks the risk of environmental hazards (Pyrgiotakis et al., 2015). The organic (chitosan and essential oil) nanomaterials are also implemented for preservation of food products. The NPs of silver restrained in collagen and cellulose sausages' casings revealed their bactericidal potential against *Staphylococcus aureus* and *E. coli* without harming the environment and humans (Fedotova et al., 2010). Unlike silver NPs, silver–polyamide nanocomposites also unveiled their antimicrobial potentiality against *E. coli* and *S. aureus* for a period up to 28 days. The silver NP–coated films via a layer-by-layer technique revealed a specific antimicrobial potential against *Pseudomonas fluorescens* (gram-negative) and *S. aureus* (gram-positive) (Azlin-Hasim et al., 2016). A significant antimicrobial effect against *Salmonella typhimurium*, *E. coli* O157:H7, *Listeria monocytogenes*, and *S. aureus* was witnessed by chitosan silver nanocomposite (De Silva et al., 2015; Qamar et al., 2020). Enhanced antimicrobial and mechanical properties were witnessed for packaging films composed of ZnO-encapsulated halloysite–polylactic acid nanocomposites (De Silva et al., 2015). LDPE/ZnO + Ag nanocomposites are responsible for deactivation of several pathogenic bacteria in different meat products, thereby enhancing the shelf life of the meat product (chicken breast fillets) (Lee et al., 2023). The pullulan films amalgamated through metal NPs (silver or ZnO) and oregano/rosemary essential oils were studied for both stability and antimicrobial activity at altered temperatures (4, 25, 37, and 55 °C) for 7 weeks against the food pathogens such as *L. monocytogenes* and *S. aureus* (Khalaf et al., 2013). The results unveiled the accelerated antimicrobial efficacy of nanocomposite films of pullulan at a reduced temperature (<25 °C), and it was diminished with an increased temperature (>25 °C).

Several other inorganic nanocomposites for food packaging include nano-zinc oxide, nanoclay (Chaudhary et al., 2020), titanium nitride NPs (nano-TiN), and nano-titanium dioxide (nano-TiO<sub>2</sub>) (Mohanty et al., 2009; Rubilar et al., 2014). The nanomaterials such as titanium dioxide and zinc oxide are often applied as a photocatalyst for degrading the organic molecules as well as the microorganisms; meanwhile, nanoclays, AgNPs, and layered silicates could be acting as antimicrobials (Majeed et al., 2013). The photocatalytic tendency of nano-TiO<sub>2</sub> and nano-ZnO subsidizes ROS production, ensuring the bacterial cell lysis by their cytoplasmic oxidation (Bodaghi et al., 2013). It has also been reported that ZnO is comparatively more attractive and effective than AgNPs because of cost-effectiveness and less toxicity (Silvestre et al., 2011). The European Food Safety Authority (EFSA, 2018) approved nano-TiN, a food contact material (Deng et al., 2011), and it was found to be widely applicable as processing aid and enables mechanical strength especially for polyethylene terephthalate (PET) (Chaudhry & Castle, 2011; Sharma et al., 2017).

## Polymer Nanocomposites

It is a multiphase solid (hybrid) material containing nanoscale fillers in one of the phases and must possess at least one dimension < 100 nm dispersed in a polymeric matrix (Wypij et al., 2023). Such nanocomposites conspicuously unveil improved thermal, mechanical, physicochemical, and optical properties over the original polymer or their composites with having a very low loading of fillers (5 Wt% or below). Numerous studies have revealed a positive impact on the barrier properties of the polymer nanocomposite later with their reinforcement by nanofillers. Thus, they confer their highest potential as the advanced technology for food packaging by maintaining the overall quality, safety, and shelf life of packed foods (Karimi et al., 2023; Rhim et al., 2013a, b). Polymer nanocomposites are composed of nanofillers, plasticizers, compatibilizers, and polymer matrix.

## Types of Polymer Used for Several Studies

### Biopolymers of Natural Origin

Biopolymers of natural origin are carbohydrates such as cellulose, starch, alginate, chitosan, carrageenan, and agar and proteins from natural sources like corn zein, soy protein, gelatin, wheat gluten, whey protein, casein, and collagen (Shankar & Rhim, 2016).

### Synthetic and Biodegradable Polymers

Synthetic and biodegradable polymers are poly(glycolic acid) (PGA), poly(l-lactide) (PLA), poly(butylene succinate) (PBS), poly( $\epsilon$ -caprolactone) (PCL), and poly(vinyl alcohol) (PVOH) (Shankar & Rhim, 2016).

### Microbial Polyesters

Microbial polyesters are poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), poly(hydroxyalkanoates) (PHAs), and poly( $\beta$ -hydroxybutyrate) (PHB) (Shankar & Rhim, 2016).

### Non-biodegradable Polymers

Non-biodegradable polymers are polyamide, nylon, polyethylene terephthalate, polyolefin, and polyurethane (Rhim et al., 2013a, b).

## Bionanocomposites

Nanotechnology oriented fabrication of biopolymers (like proteins, chitosan, starch, and cellulose-based

nanocomposites) leads to a reduction in their cost with enhanced efficacy while being considered food packaging components. Plasticizers are usually added for an upliftment of mechanical and properties of the biopolymers (Espitia et al., 2014). Numerous materials like clay and metals along with their oxides such as nano-zinc oxide (nano-ZnO), silver NPs (AgNPs), and nano-titanium dioxide (nano-TiO<sub>2</sub>) (Bumbudsanpharoke & Ko, 2015) are implemented for their corresponding antimicrobial, barrier, thermal, and mechanical efficacy. Moreover, the enclosure of antioxidants, aroma, oxygen scavengers, and colors may enhance the biochemical food packaging efficiency of bionanocomposites (Gupta, 2023; Majid et al., 2018; Sarfraz et al., 2020).

### Bionanocomposites from Natural Origin

**Nanocomposites Based on Starch** Starch, a fabulous polysaccharide (cheap, abundant, eco-friendly, and recyclable), is commonly implemented in fabricating several biocentered packaging systems. However, it has certain drawbacks, such as low barrier and mechanical properties and highly sensitive towards moisture and UV radiations (Flores et al., 2007). NPs of poly(methyl methacrylate-*co*-acrylamide), TiO<sub>2</sub>, ZnO, and graphene grafted with starch showed an improved barrier and mechanical properties (Goudarzi et al., 2017; Jayakumar et al., 2019). It appears to be in diversified morphology, with a particle size range from 9 to 400 nm along with crystalline/amorphous form (Xie et al., 2013). The hydroxylation and acetylation of starch lead to improved mechanical/barrier properties (Altaf et al., 2022; Chaudhary et al., 2008; Volkert et al., 2010). The development of starch-based nanocrystals can be applicable as reinforcement agent (Le Corre et al., 2010). Similarly, the modified thermoplastic films of starch are widely applicable in food wrapping.

**Chitosan-Based Nanocomposites** Chitosan is an abundant biocompatible/biodegradable as well as antimicrobial polysaccharide derived from deacetylation of chitin, suitable for fabrication of numerous nanocomposites (Wang et al., 2018). Chitin-based nanowhiskers/NPs were developed by either deproteinization, sonication, or ionotropic gelation (using sodium tripolyphosphate) (Chang et al., 2010a, b; Riseh et al., 2023). Chitin incorporated with nano-/microreinforcements usually resulted in the formation of layered nanosilicates and thus improves the mechanical strength of chitosan-based packaging material (Abdollahi et al., 2012; Casariego et al., 2009; Hsu et al., 2012; Lavorgna et al., 2010).

**Cellulose-Based Nanocomposites** A natural biocompatible/degradable homoglycan is comprised of the monomers of glucose. Cellulose-based nanocomposites (CNC), nanofibrils (with a diameter of 2–20 nm), metal and metal oxide

(Fe<sub>3</sub>O<sub>4</sub>, Ag, and TiO<sub>2</sub>), nanoclay, and cellulose nanowhiskers (obtained from a crystalline region of cellulose fibrils) were widely reinforced in polymeric matrices as fillers for enhancing the barrier, mechanical, and thermal properties of polymer-based packaging (Brinchi et al., 2013; Dufresne, 2010; Duran et al., 2011; Eichhorn et al., 2010; El-Sayed & Youssef, 2023; Velásquez-Cock et al., 2014). Moreover, like plant cellulose, bacterial cellulose (BC) was highly engaged in the development of light weight, economic, and strong nanocomposite-based packaging films with a better mechanical strength (Wan et al., 2009). Current applications of different nanoclay composites in food packaging are depicted in Table 1 (Perera et al., 2023).

**Protein-Based Nanocomposites** Protein-based nanocomposites are believed to be an outstanding food packaging material. Plant proteins from lectins, wheat gluten, soy, corn zein, and sunflower were being implemented for the development of bionanocomposite as well as biobased packaging. However, because of the poor barrier and mechanical properties, protein-based packaging is no longer popular except keratin and corn zein. (Fortunati et al., 2018). Hence, incorporation of plasticizers into the protein-mediated polymer matrices helps in troubleshooting of the physicochemical inadequacy (Zubair & Ullah, 2020). The hydrophobicity of proteins made them a potential corroborator for designing of safe packaging components by the Food and Drug Administration (FDA) (Chuacharoen & Sabliov, 2016).

**Nanocomposites Based on Nanoclay** Nanoclays, specifically montmorillonite (MMT), abundantly incorporated (via solution intercalation, polymerization, or melt intercalation) as nanofiller in the layered phyllosilicate clays in order to improve the properties such as elastic modulus, Young's modulus, and water and gas barrier as well as thermal stability of polymeric matrix towards several folds (Cui et al., 2015). The literature revealed that the presence of nanoclays has boosted the oxygen barrier, resulting a prolonged shelf life of food materials along with PLA and ethylene vinyl alcohol (EVOH) polymer composites/matrix. By reinforcement, nanoclay (of saponite, MMT, laponite) develops a torturous path towards diffusion and resulted in a reduction in permeability (80–90%) (Sachdeva, 2021). It was also observed that the natural polymers such as starch, cellulose, proteins, and chitosan with added plasticizers/nanofillers are being considered potential substitutes to their conventional ancestors as packaging components with crucial applications (Qin et al., 2016; Wakai & Almenar, 2015). Graphene nanometals along with their oxides could result in enhanced performance with potent antimicrobial efficacy (Goudarzi et al., 2017; Tang et al., 2019).

**Table 1** Current applications of nanoclay composites in food packaging

Sl No	Packaging materials	Type of nanoclay	Packaged food product	Packaging material with their characteristics	Superiority compared to conventional packaging
1	Poly(lactic acid)	BT	N/A	Materials for active food packaging	Improved mechanical and barrier properties
2	Xylan-alginate	BT	N/A	Edible films	Eco-friendly, edible, and Biodegradable
3	Carnauba wax	BT 32A	Orange	Coating leads to preserve nutritional and sensory quality	Eco-friendly, edible, biodegradable with enhanced shelf life
4	Starch–low-density polyethylene–date palm seed extract	Cloisite 20A	N/A	Active packaging film with antimicrobial efficacy	Antimicrobial, biodegradable, and eco-friendly
5	Polyvinyl alcohol	Cloisite Na+	N/A	Multilayer nanocomposites coated with base paper	Improved mechanical and barrier properties
6	Poly(butylene succinate-cobutylene adipate)–poly(lactic acid)	Cloisite 30B	N/A		Eco-friendly, biodegradable, improved mechanical and barrier properties
7	Polypropylene -based cellulose nanofiber	Cloisite 20A	N/A	Active packaging material	Improved active, mechanical, and barrier properties
8	Gelatin–whey protein isolate–orange peel extract	Cloisite 30B	N/A	Food packaging material with antimicrobial potential	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
9	Xylan-alginate	Halloysite	N/A	Edible films	Eco-friendly, edible, and biodegradable
10	Chitosan	MMT	Gouda cheese	Active food packaging with antimicrobial approach	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
11	Rice flour–gelatin	MMT	Pork belly	Antimicrobial active food packaging along with prolonged shelf life	Biodegradable, antimicrobial, eco-friendly, with enhanced mechanical and barrier properties
12	Low-density polyethylene	MMT	Sugarcane juice	Extended shelf life of the active packaging film	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
13	Chitosan–CuO	MMT	N/A	Improved antimicrobial efficacy	Eco-friendly, antimicrobial, biodegradable, with enhanced mechanical and barrier properties
14	Pectin	MMT	N/A	Bilayer food packaging material	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
15	Wheat gluten	MMT	N/A	Active food packaging material	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
16	Poly(butylene succinate-cobutylene adipate)–poly(lactic acid)–carvacrol	MMT	N/A	Active packaging film with antimicrobial impact	Mechanical, antimicrobial, and barrier properties
17	Chitosan–rosehip seed oil	MMT Cloisite C30B	N/A	Active packaging film with antimicrobial response	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties

Table 1 (continued)

Sl No	Packaging materials	Type of nanoclay	Packaged food product	Packaging material with their characteristics	Superiority compared to conventional packaging
18	Gelatin–chitosan	MMT	N/A	Active food packaging material	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
19	Poly(lactic acid)–thymol	MMT	N/A	Active packaging film with antioxidant and antimicrobial approach	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
20	Polyvinyl alcohol	MMT-Na+	N/A	Coating of packaging material against moisture sensitivity	Barrier properties
21	Pectin–methylene blue	Nanoclay	Oranges, tangerines and kiwi	Smart active film and antioxidant to measure vitamin C levels	Intelligent, antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
22	Starch–tragacanth gum	Nanoclay	N/A	Active food packaging material	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
23	Poly(lactic acid)	Nanoclay	N/A	Active food packaging material	Biodegradable, antimicrobial, eco-friendly, with enhanced mechanical and barrier properties
24	Starch loaded with methyl orange and bromocresol green	Nanoclay	Milk	Intelligent packaging material for preventing milk spoilage	Eco-friendly, antimicrobial, biodegradable, with enhanced mechanical and barrier properties
25	Polyvinyl alcohol–red cabbage extract	Nanoclay–hydrophilic bentonite	N/A	Intelligent packaging film with pH-responsiveness	Intelligent, antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
26	Pectin– <i>Carum copticum</i> essential oils– $\beta$ -carotene	Nanoclay Cloisite® Na+	Butter	Smart active film for determining oxidation behavior of butter	Antimicrobial, biodegradable, eco-friendly, with enhanced mechanical and barrier properties
27	k-carrageenan–cellulose nanocrystals	Organically modified MMT	N/A	Active packaging film	Biodegradable, antimicrobial, eco-friendly, with enhanced mechanical and barrier properties
28	Cassava starch–clove essential oil	MMT	Strawberries	Active packaging film	Eco-friendly, antimicrobial, biodegradable, with enhanced mechanical and barrier properties

BT bentonite, MMT montmorillonite



## Synthetic Bionanocomposites

Bioplastics (renewable or biodegradable), a potential alternative to conventional plastic, are found to be one of the prime attractions for numerous researchers throughout the globe in the recent era. Bioplastics/biopolymers undergo microbial transformation to develop carbon dioxide, organic compounds, water, and hydrogen (Luzi et al., 2015; Peelman et al., 2013). The reports obtained from numerous literature have ensured the safety and efficacy of such nanocomposites as packaging materials towards food components (Sarfraz et al., 2020).

**Nanocomposites Based on PLA** Because of the transparency, easy availability, and mechanical strength, PLA is prioritized among the materials of choice towards packaging of food components (Jamshidian et al., 2012; Jonoobi et al., 2010) despite few drawbacks. However, the drawbacks can be overwhelmed by using nanofillers. PLA in combination with nanoclays, metal oxides ( $\text{TiO}_2$ ,  $\text{Fe}^{3+}$ , Ag,  $\text{SnO}_2$ ,  $\text{Ce}_2\text{O}_4$ ), lignocellulosic nanofillers, cellulose nanowhiskers, CNC, lignin NPs, or chitosan were applied for packaging of numerous food materials/products with enhanced barrier, mechanical, and antimicrobial properties (Bonilla et al., 2013; Busolo & Lagaron, 2013; Man et al., 2012; Sanchez-Garcia & Lagaron, 2010a, b; Sikong et al., 2010; Svagan et al., 2012; Yang et al., 2016; Zhang et al., 2008; Zhu et al., 2011).

**Nanocomposites Based on PHA** PHAs (in association with PHBV and PHB) are biodegradable components, usually isolated from the microbes and recognized to be one of the best choices for biomediated packaging. However, it is associated with numerous shortfalls like brittleness, thermal instability, and low strength. In order to overcome such discrepancies, the elements like ZnO, nanoclay, carbon nanotubes, and fullerenes are incorporated into them. PHA along with cellulose nanowhiskers denoted significant boosting in barrier properties and mechanical strength (Martínez-Sanz et al., 2016). The incorporation of Ag and ZnO to PLA-based nanocomposites featured antimicrobial response to the packaging (Castro-Mayorga et al., 2014; Díez-Pascual & Díez-Vicente, 2014). The addition of carbon nanotube (CNT) to PHBV matrix resulted in an enhancement of antibacterial and crystallization behavior followed by improved gaseous and thermal resistance (Sanchez-Garcia & Lagaron, 2010a, b).

**Mixed Polymer-Based Composites** The hurdles of individual polymers (like thermal stability, mechanical strength, UV and water barrier, economic crisis as well as oxygen permeability) can be overcome by a combination of synthetic or biopolymeric composites (Fortunati et al., 2018). For

example, PLA is extensively added with polyhydroxyalkanoates (PHA), PHBV, polybutylene adipate terephthalate (PBAT), and polycaprolactone (PCL) to give an enhanced efficacy of bionanocomposites with better mechanical strength and biodegradability (Briassoulis et al., 2021; Dasan et al., 2017; Li et al., 2011; Moustafa et al., 2017; Sabet & Katbab, 2009). PLA/PBAT composite incorporated with cellulose (nanocrystal)-silver nanohybrids revealed a better toughness, mechanical strength, and thermal and crystallization properties as well as antimicrobial properties (Sarazin et al., 2008).

## Protein NPs

Protein-based NPs are usually applied in food packaging for betterment of the tensile strength as well as aqueous barrier properties (Ahmad & Ghosh, 2020). The incorporation of peanut protein NPs in starch-based biocomposites results in improvement of temperature resistance, moisture barrier properties, and mechanical strength (Li et al., 2015). Similarly, incorporation of zein NPs to whey protein isolate-based films boosted mechanical strength and moisture barrier properties (Ashfaq et al., 2022).

## Biobased and Biodegradable Nanocomposite

Nanofillers combined with antioxidants/antimicrobials revealed an additional hurdle towards oxidative spoilage bacteria, making a constraint on the stability of a product packed with plastic materials. Incorporation of montmorillonite along with numerous essential oils into the biopolymers (e.g., essential oils of ginger/chitosan/montmorillonite or rosemary/chitosan/montmorillonite or montmorillonite/soy protein/clove) (Echeverría et al., 2018; Souza et al., 2018, 2019) and abridged lipid oxidation of the meat increased barrier property against UV light and oxygen (Pires et al., 2018).

The antimicrobial impact and biodegradability efficacy of metal oxides (e.g., biocomposites of ZnO/cellulose acetate phthalate/chitosan and mucilage/ZnO/CMC corroborated with nanobiocomposites like ZnO/alginate, cellulose nanofiber/WPI/ $\text{TiO}_2$ /rosemary essential oil, and ZnO/nanorods/gelatin/clove essential oil) and polymer composite have been studied (Ejaz et al., 2018; Indumathi et al., 2019; Mohammadi et al., 2019; Rezaei & Shahbazi, 2018; Sani et al., 2017). The composite coating (chitosan/ $\text{TiO}_2$  nanocomposite) made over the paper packaging material was found to enhance the mechanical strength of the paper and also inhibited the surface growth of microbes (Tang et al., 2016). Similarly, PET substrate coated with chitosan-vermiculite nanoclay resulted in a substantial declination in oxygen permeability (Essabti et al., 2018).

The nanotechnology-reinforced materials require very less amount of polymeric concentration and thereby revealed a reduction in production cost (Kanmani & Rhim, 2014a, b). However, it has also been reported that nanofillers were used to alter the polymer biodegradability along with their microbial degradation because of the altered crystallinity (Mishra et al., 2018; Souza & Fernando, 2016). Figure 1A presents the biobased materials used for antimicrobial food packaging application (Tan et al., 2021).

### Mechanism of Action of Metal and Metal Oxide NPs

Since the previous decade, both metals along with the mixed metal oxides are exhaustively analyzed for their exposure into food packaging. By imparting an oxygen partial pressure, metal oxides (MOs) lead to the development of rigorous alterations in their composition and lattice structure. The d-block MOs (CuO, ZnO, MgO, and TiO<sub>2</sub>) revealed antimicrobial properties because

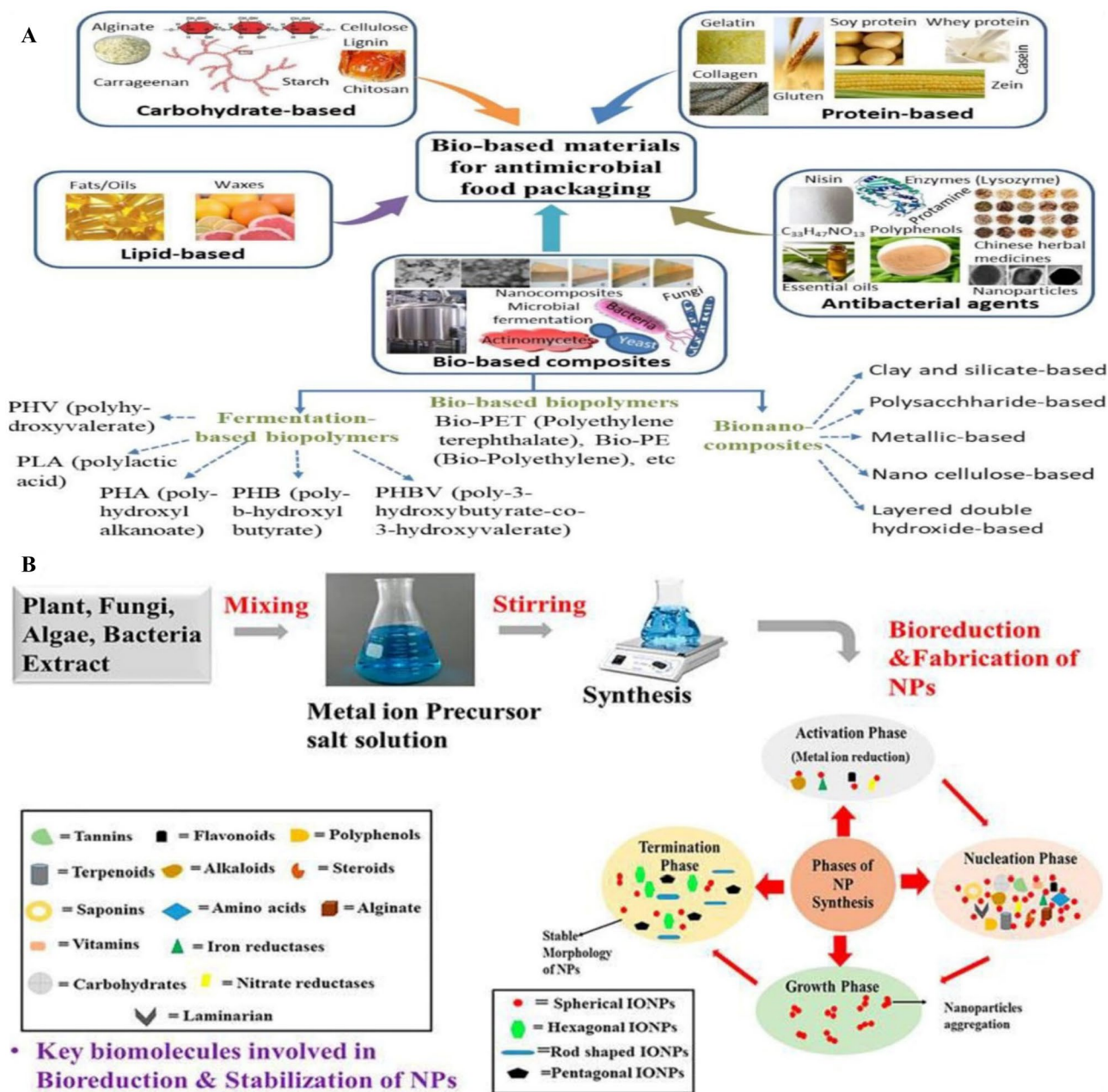


Fig. 1 A Various types of biobased materials used for antimicrobial food packaging application. B Schematic representation of the mechanism of synthesis of metal oxide NPs by using plants, algae, fungi,

and bacteria. Reproduced with permission from references Tan et al. (2021) and Nile et al. (2020)

of a minute variability in stoichiometry of O-atom (Abdel-Karim et al., 2020). Polymer-MO-NPs alleviate preeminence in the activities of nanocomposites (Peponi et al., 2014). However, oxides of Ag and Au received the least priority because of their instability (Suchomel et al., 2018). Ag and Au-NPs denoted antimicrobial activities via free metal ion toxicity and oxidative stress (Singh et al., 2022). The NPs induce alteration in membrane potential by binding to the bacterial membrane and, hence, cause a reduction in ATP level, thereby inhibiting tRNA binding into the ribosome (Ahmad et al., 2017). Owing to the internalization of facetious bacteria (gram-negative), Au-NPs showed a provoked antibacterial response against gram-positive bacteria rather than its ancestor (Cui et al., 2012). Modified polymer matrix incorporated with Ag-NPs showed an antibacterial potential along with the gas permeability (Kumar et al., 2021). Figure 1B presents the mechanism of the synthesis of metal oxide NPs (schematic representation) by using algae, fungi, plants, and bacteria (Nile et al., 2020).

## Inorganic Nanomaterials Applied in Packaging

### Carbon Nanotubes

These are carbon allotropes, in cylindrical form, and are of two types, namely, single-walled and multi-walled nanotubes encompassing numerous concentric cylinders (Huang et al., 2015a, b). They are responsible for the advancement of antimicrobial and mechanical strength of polymers used for packaging as well as in the formation of oxygen sensors to monitor the altered atmospheric packaging and detection of food spoilage (Biswal & Misra, 2020; Rezić et al., 2017; Singh et al., 2023a, b; Zhu et al., 2017).

### Silver Nanoparticles

AgNPs are extensively explored as antimicrobial agents (broad spectrum) towards enhancement of the shelf life of food packaging (Biswal & Misra, 2020). AgNPs are applicable in biodegradable as well as non-biodegradable polymers towards fabricating food packaging by considering their migrational toxicity (Ahmad et al., 2021; Carbone et al., 2016; Istiqola & Syafiuddin, 2020). The NPs potentially restrict the utilization of preservatives in the food elements up to a quiet extent (Kraśniewska et al., 2020; Taghinezhad & Ebadollahi, 2017). The metal-based nanocomposites and NPs as food packaging components are depicted in Fig. 2A and B (Hossain et al., 2021; Kumar et al., 2021).

### Zinc Oxide Nanoparticles (ZnO NPs)

ZnO is also considered an essential micronutrient (antimicrobials) being added into numerous fortified dietary and food supplements (McClements & Xiao, 2017;

Stuparu-Cretu et al., 2023).  $Zn^{2+}$  induces ROS to carry out generation of cell organelles and causes cell lysis (Kim et al., 2022). The NPs of ZnO facilitate antibacterial, barrier, and mechanical skills of the composite films while being added into the polymer matrix (Abbas et al., 2019; Kim et al., 2022).

### Titanium Dioxide Nanoparticles ( $TiO_2$ NPs)

$TiO_2$  NPs are considered being white metal oxides, useful in blocking UV radiations, as coloring agents and vastly applied as food additives and as nanocomposites towards packaging of food. They are supposed to enhance the mechanical, barrier, and chemical impacts of the films, corroborated by cost-effectiveness, chemical stability, non-toxicity, and eco-friendly nature (Baranowska-Wójcik et al., 2020; Mohr et al., 2019; Sungur et al., 2020). The antimicrobial efficacy of such elements is dealt with the generation of ROS and free radicals (Venkatasubbu et al., 2016).

### Nanoclays

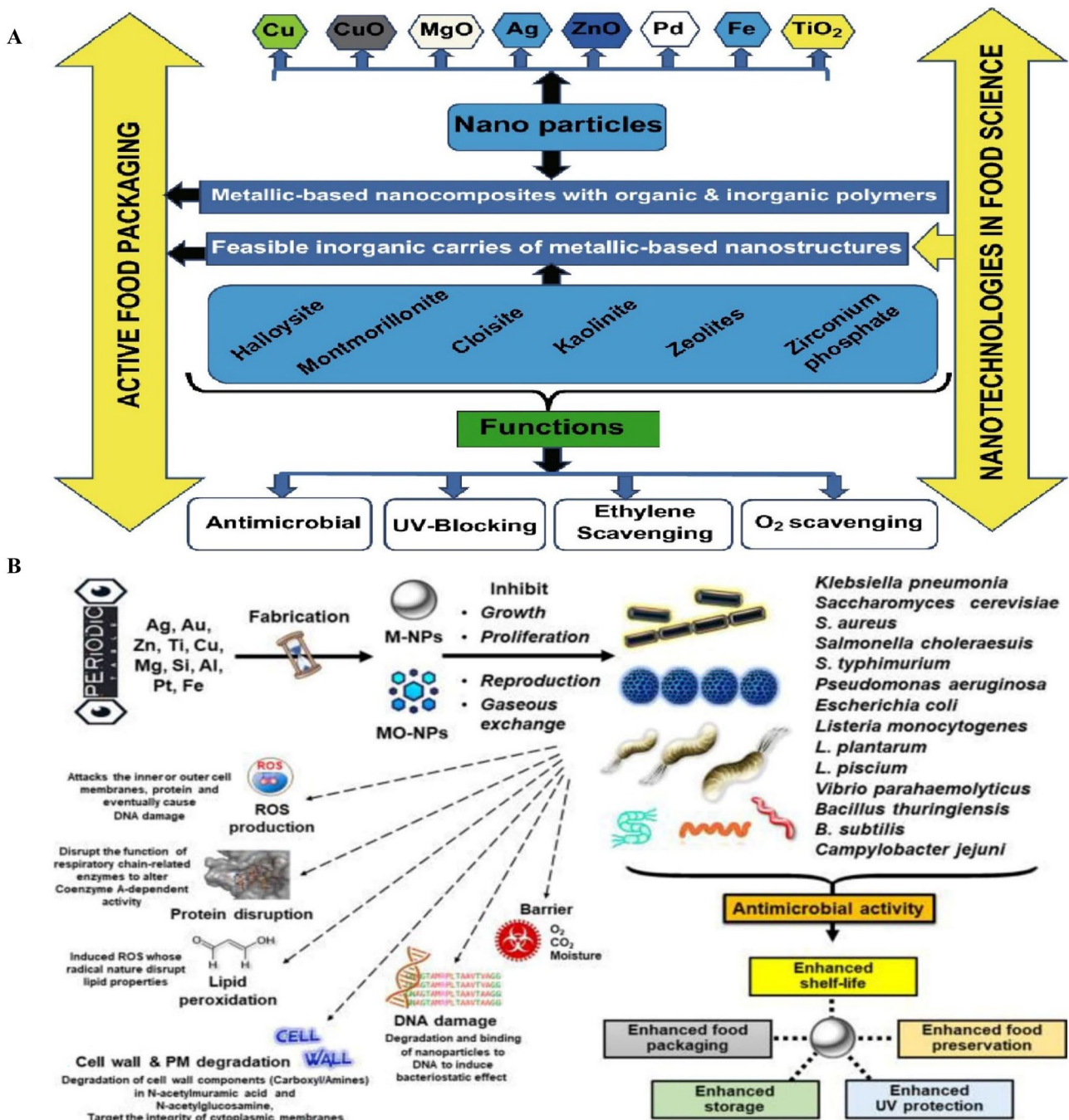
Presently, nanoclay has gained some popularity for its application in food packaging (Guo et al., 2018a, b; Nath et al., 2022). They usually appear in platelet form along with a soft flaky texture, with a high aspect ratio and low specific gravity. The clay in the form of organophilic MMT and montmorillonites denoting a high aspect ratio and thermoplasticity is widely applied in packaging. The migrational aspect of the nanoclay usually depends on the interaction between nanoclay and polymer along with temperature, food, and time of exposure (Bandyopadhyay & Ray, 2019).

### Nanosilica

They are usually incorporated into the hydrophobic coatings, especially for the materials meant for self-cleaning. They impart a non-adhesive coating (e.g. coating with Aerosil<sup>®</sup> silica NPs) on the food components, thereby resulting in a development of free-flowing tendency which enhances their packing speed into the containers (e.g., powdered soup, beer, and wine) (Agriopoulou et al., 2020; Chen et al., 2013; Kumari et al., 2019).

## Active Packaging Systems for Food

The active packaging of any material comprised of  $CO_2$  scavengers, moisture regulating agents, antimicrobials, and oxygen scavengers. Based on the purpose/requirement, the active packaging systems are designed for storing of food materials (Dias et al., 2013) such as short-term chilled storage acclimatizing either by overwrap packaging systems or by vacuum packaging or via modified



**Fig. 2** A The potentials for the development of metal-based nanocomposites in active food packaging. **B** Metal-based NPs as food packaging components. Reproduced with permission from references Hossain et al. (2021) and Kumar et al. (2021)

atmosphere packaging (MAP) systems. Gases like O<sub>2</sub> and CO<sub>2</sub> were adapted for long-term chilled storage by maintaining bulk gas flushing for meat products (Attaran et al., 2017). The polymeric films such as polypropylene (PP) and a low-density polyethylene (LDPE) with surface modification were commercially used for packaging of foods (Attaran et al., 2017). Numerous patents have

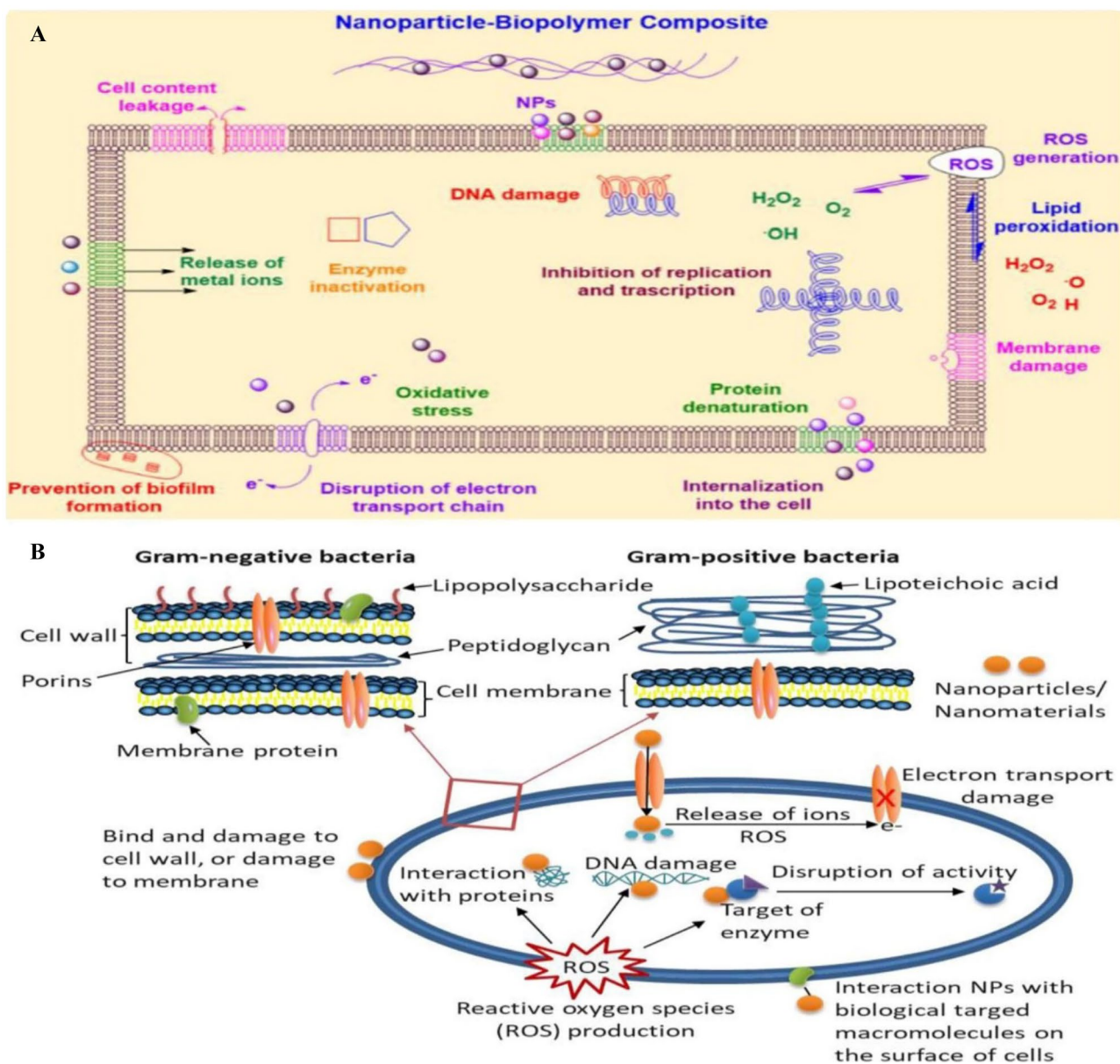
been established for utilization of nanosilver and nanoclays highlighting their impact on food packaging which were being filed under Europe, the USA, and Asia (Hagen & Drew, 2016). Addition of carbon nanotubes and allyl isothiocyanate into the active packaging systems boosts up the qualitative and quantitative aspects of food packaging composites (Dias et al., 2013). The antimicrobial

mechanisms of the action of nanocomposites (schematic representation) and nanoparticle/nanomaterial food packaging are denoted in Fig. 3A and B (Basavegowda & Baek, 2021; Tan et al., 2021).

### Intelligent/Smart Packaging Systems for Food

The intelligent systems of packaging lead to an environmentally based stimulus system dealing with an altered repairing of the packaging system as per the pathogenic existence. NP-based nanosensors are engaged for the detection of food contaminants

whereas the custom-made nanosensors are amended for the analysis of food, detection of colors/flavors, detection of quality of the drinking water, and the clinical diagnosis of toxins, chemicals, and food pathogens (Augustin & Sanguansri, 2009; Berekaa, 2015; Li & Sheng, 2014). NPs based on barcodes, i.e., nanobarcodes, are utilized as ID tags (Branton et al., 2008). Such kind of packaging leads to improvement of the shelf life of the food products (Azeredo & Correa, 2021) and can also able to detect different temperatures, chemical contaminants, gases, aromas, intensity of light, metabolic by-products of microbiota, and pathogens (King et al., 2018). Moreover, it



**Fig. 3** **A** The schematic representation of antimicrobial mechanisms of action of nanocomposites designed for food packaging. **B** The potential antimicrobial mechanisms of nanoparticles/nanomaterials

als in food packaging. Reproduced with permission from references Basavegowda and Baek (2021) and Tan et al. (2021)

was also associated with more effective real-time packaging processes such as food rotting with the impact of biosensors towards detection of different odors followed by an assessment of quality, safety, and efficacy of food products (Attaran et al., 2017; Dasgupta et al., 2015; Morris et al., 2017). Some advanced nanobiosensors deployed in quality evaluation of food packaging system include radiofrequency identification (RFID), temperature time integrators (TTIs), freshness indicators, O<sub>2</sub> sensors, moisture indicators, microbial detectors, and gas sensors (Shankar & Rhim, 2016).

### Migration of Packaging Materials

Usually, it has been noticed that the NPs can be released via diffusion, dissolution, and desorption ([https://food.ec.europa.eu/safety/chemical-safety/food-contact-materials/legislation\\_en](https://food.ec.europa.eu/safety/chemical-safety/food-contact-materials/legislation_en)). The European legislation on plastic materials (EU) No. 10/2011 has developed regulations regarding the configuration of plastic food contact materials (FCMs) and provided an approved list of permissible materials to be added for manufacturing of FCMs ([https://food.ec.europa.eu/safety/chemical-safety/food-contact-materials/legislation\\_en](https://food.ec.europa.eu/safety/chemical-safety/food-contact-materials/legislation_en)). European Commission, 2011, & European Legislation (EU) No 10/2011. The regulation also provided the information regarding several standardized test conditions, their timings, temperature, and testing medium (food simulant) (<http://europa.eu>). A highest proposed limit of migration of food from an un-authorized material via any kind of functional barrier is 0.01 mg/kg. Nanotechnology in the food packaging system has reformed the packaging standard, limiting the migrational studies of polymer nanocomposites along with a limitation towards quantitative and qualitative analysis (Han et al., 2011; Huang et al., 2015a, b). Despite numerous merits, the parameters/aspects of NPs like particle size, concentration, solubility, molecular weight, diffusivity in the polymer, mechanical stress, temperature, composition, and pH, polymer viscosity and structure and contact time should be considered (Song et al., 2011). However, it has turned to be a difficult as well as a tedious task for estimating the migration of particles into the food matrices quantitatively. Hence, the natural food stimulants are believed to be the potential alternatives to measure the specific and overall migrational tendency of materials in food packaging (Honarvar et al., 2016).

### Characterization Methods of Polymer Nanocomposites

The polymer nanocomposites along with their surface morphology and microstructure can be analyzed (e.g., internal structure, spatial distribution, and nanofillers dispersion in a polymer matrix) by a transmission electron microscope

(TEM) and scanning electron microscopy (SEM). X-ray diffraction (XRD) analysis was conducted for studying the crystalline size, crystallinity, and composite structures (e.g., intercalated, tactoid, or exfoliated structures of polymer/clay composite and/or polymer nanocomposite); Fourier transform infrared spectroscopy (FTIR) was conducted for determination of functional groups as well as the chemical changes associated with polymeric interaction in the nanofillers. In addition to the above, a number of analytical techniques such as thermogravimetric analysis (TGA), atomic force microscopy (AFM), and nuclear magnetic resonance (NMR) could be amended for the characterization of the polymer nanocomposites (Okamoto, 2023). Several nanofillers along with their properties in bionanocomposite films are depicted in Table 2 (Sharma et al., 2020).

### Properties and Evaluation of Polymeric Nanocomposites

The polymeric nanocomposites frequently unveil significantly enriched physical and mechanical properties (because of the sturdier interfacial interaction among the layered silicate and the matrix) such as improved strength and heat resistance, higher modulus with reduced gaseous permeability, and augmented biodegradability of the polymeric composites compared to their pristine moieties.

### Mechanical Properties

#### Tensile Strength and Modulus

Toyota researchers were the first to report tensile strength and modulus for the first time towards polyamide 6-clay hybrid nanocomposite structures (Pandey, 2020). In a polymer matrix, the nanocomposite recital is associated with exfoliation of the clay up to a certain extent, resulting in an elevated interaction between the polymers and clay (Baek et al., 2022; Tang et al., 2011).

#### Compression

The compression behavior of nanocomposites (glassy epoxy-clay) was quite associated with the changes in interfacial interactions, layer charge densities, and platelet aspect ratios (Rabothata et al., 2021).

#### Fracture

An increase in modulus to the delaminated nanocomposite structure leads to bring a considerable fracture toughness (Yassin, 2023). It has been noted that a change (reduction) in

**Table 2** Numerous properties of nanofillers in bionanocomposite films

Type of nanofiller	Biopolymer with their types	Several properties of bionanocomposite films along with their antimicrobial activity	Country
AgNPs, clay (Cloisite 30B)	Gelatin	The addition of organoclay and AgNPs to gelatin/AgNPs/clay nanocomposite film revealed a potential antimicrobial efficacy (zone of inhibition at 12 and 13 mm <sup>2</sup> ) against food-borne pathogens (like <i>E. coli</i> and <i>L. monocytogenes</i> ) along with improved hydrophobicity and improved barrier properties against water vapor and UV followed by provoked tensile strength of films	Korea
AgNPs	Gelatin	The nanocomposite films of gelatin/AgNPs exhibited potential antimicrobial activity towards food-borne pathogens like <i>S. typhimurium</i> and <i>Bacillus cereus</i> (at a concentration of 30 and 40 mg). Apart from the above, they also revealed the enrichment with properties like intensity of color, improved moisture content, UV barrier, elongation, and thermal stability as well as hydrophobicity followed by a reduction in permeability of water vapor (WPV), elastic modulus, and tensile strength	Korea
ZnO-rich nanorod (nr)	Sago starch, bovine gelatin	The zone of inhibition with respect to an elevated concentration of ZnO-nr contents gets increased. Similarly, the incorporation of gelatin and sago starch-based biopolymers revealed a better zone of inhibition (80 and 65 mm <sup>2</sup> ) with the incorporation of a minimum concentration (5%) of ZnO-nr against <i>E. coli</i> O157:H7. The above matrix also leads to enhance the heal-seal and mechanical properties of the biofilms; exhibition of UV absorption; reduction in oxygen permeability; and reduction in the water absorption capacity and moisture content of the films	Iran
AgNPs	Agar	The composite leads to improve the properties like enhanced WVB and surface hydrophobicity; and improved mechanical strength; composite films with elevated AgNP concentration (0.5–2%) demonstrated pivotal antibacterial activity (broad spectrum) against pathogenic microbiotas like <i>L. monocytogenes</i> and <i>E. coli</i> O157:H7	Korea
AgNPs	Hydroxy propyl methylcellulose (HPMC)	Improve mechanical and barrier properties and tensile strength. HPMC in combination with AgNPs revealed impactful zone of inhibition in variable concentration against pathogenic microbes like <i>E. coli</i>	Brazil
Halloysite nanotubes	Carboxymethyl cellulose (CMC)	Revealed an impactful mechanical property along with WVB and thermal stability. Both Hal-AAg and CMC/Hal-AAg composite films denoted better antimicrobial efficacy against a broad spectrum of microbiota	China

**Table 2** (continued)

Type of nanofiller	Biopolymer with their types	Several properties of bionanocomposite films along with their antimicrobial activity	Country
Cellulose nanocrystals	CMC	The developed films revealed improved thermal stability and transparency followed by a competent mechanical strength	Korea
Lignin-rich rice straw nanofibers	Cellulose acetate	The cellulose acetate composite film was demonstrated with improved water vapor permeability (WVP) and hydrophilicity along with a reduced thermal stability	Egypt
Montmorillonite (MMT)	Cellulose acetate/polyethylene glycol	Improved thermal stability, mechanical strength, and storage potentiality along with the barrier properties. The zone of inhibition was found to be effective against the wide range of bacteria on increasing the concentration of MMT. The loading capacity of the film was also enhanced	India
Unmodified MMT (NaMMT), organically modified MMT (Cloisite 30B), AgNPs, and a Ag-zeolite (Ag ion)	Chitosan	Modified chitosan films with improvised mechanical and WVBP properties. The nanocomposite films incorporated with Cloisite 30B exhibited antimicrobial activity only against <i>S. aureus</i> and <i>L. monocytogenes</i> (gram-positive bacteria), and the antibacterial efficacy was reported to be much better compared to Na-MMT incorporated film	Korea
Silver oxide	Chitosan	The CS–Ag <sub>2</sub> O in solution form [(with an aqueous dilution (50%) containing acetic acid (1%)] denoted a very clear and greater zone of inhibition (14–24 mm) against <i>E. coli</i> , <i>S. aureus</i> , <i>Pseudomonas aeruginosa</i> , and <i>Bacillus subtilis</i> compared to CS–Ag <sub>2</sub> O nanocomposite film. Thus, the nanocomposite of chitosan could be an excellent choice for food packaging	India
AgNPs	Chitosan	The composite revealed an altered thermal stability, as a barrier against UV light along with development of rugosity in the material; chitosan-PVA films altered by AgNPs demonstrated a better antifungal and antibacterial activity	Colombia
TiO <sub>2</sub>	Chitosan	CS-Ti nanocomposites declined the tensile strength and found to be highly effective against <i>S. aureus</i> with/without the presence UV irradiation	Indonesia
Hydrophobic nanoclays	Tilapia skin gelatin	Improved barrier and mechanical properties	Thailand
Brucite nanoplates	Starch	Enhanced properties of starch-based plastics; useful in food contact and packaging	Brazil
Halloysite nanoclay	Potato starch	Declination of the permeability of the gaseous moieties, followed by improvement in mechanical and barrier properties towards potato starch films	Iran
Lignin NPs	Wheat gluten	The water uptake tendency of gluten gets reduced with improved tensile strength and thermal stability	Italy



**Table 2** (continued)

Type of nanofiller	Biopolymer with their types	Several properties of bionanocomposite films along with their antimicrobial activity	Country
Chitosan NPs	Fish gelatin	Shelf life of the food products gets extended; improved tensile strength, elastic modulus, and WVVB; development of edible films towards packaging of food materials; and FG/CSNP composite films containing varied concentration of OEO revealed improved zone of inhibition (range between 17.66 and 33 mm) against wide varieties of microbiota	Iran
Halloysite nanoclay	Soluble soybean polysaccharides	The developed film was associated with poor/reduced oxygen permeability, reduced moisture uptake, WVP, and water solubility of the films	Iran
Ag-MMT	Agar-CMC	The modified properties are like poor/reduced oxygen permeability, reduced moisture uptake, WVP, and water solubility of the films along with enhancement of mechanical properties and surface hydrophobicity of the films	India
ZnO	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)	Resulting broad spectrum antimicrobial activity against few foodborne pathogens such as <i>Bacillus subtilis</i> and <i>E. coli</i> ; prolongation of the shelf-life of packaged food. Prominent zone of inhibition was observed by increasing the concentration of Ag-MMT nanocomposites (from 1 to 3 wt %) Hence, the composite (Agar-CMC/Ag-MMT nanocomposite) was considered to be a good choice for antibacterial food packaging material	Spain
AgNPs and cellulose nanocrystals	Poly(lactic acid)	Showed potentiality against <i>E. coli</i> and <i>S. aureus</i> . The ZnO incorporated in PHBV resulted a positive impact against both migration and barrier properties; the improved biodegradability approach of the materials denoted great impact for its consideration towards development of containers towards oxygen as well as the food products of maximum moisture sensitivity	Italy

plastic deformation (via the presence of microscale aggregates) into the polymer matrix leads to the development of a brittle nanocomposite (Yassin, 2023).

### Permeability, Barrier Properties, and Solvent Resistance

A significant decrease in permeability was accredited to a high level aspect ratio of the clay layers, resulting in tortuosity of the nanocomposite concerning with the gaseous outcome (Jose et al., 2012; Yassin, 2023). The elevated intensity of gas barrier for the films of polymer nanocomposites is associated with disseminated silicate layers (ordered) resulting in very high aspect ratios of the polymer matrix (Bakar et al., 2012; Gokkurt et al., 2012). The toppled barrier

properties associated with polymer–clay nanocomposites revealed an augmented solvent resistance, especially associated with organic solvents like toluene, alcohol, and chloroform (Wang et al., 2023).

### Thermal Properties

#### Thermal Stability

The thermal stability of the nanocomposites increases according to the content and length of silicate layers and by the addition of copper nanoparticles (not more than 5%) (Adegbola et al., 2020; Molefi et al., 2009).

## Flammability

The data associated with polymeric combustion of nanocomposites (Vinyas et al., 2019) revealed their flammability approach via formation of a residue followed by a better thermal stability which can be acting as a protective barrier through reduction of mass and heat transfer within the polymer and flame (Vinyas et al., 2019).

## Optical Properties

Due to the light scattering tendency of the particles or fibers, the traditional composites seem to be opaque (Białkowska et al., 2023). The Maglite nanocomposites, an epoxy matrix, were found to be a transparent smectite-nanocomposites matrix (Youssef, 2013).

## Polymeric and Nanobased Systems Applied in Food Packaging

### Nanoparticles with Their Antimicrobial, Antioxidant, and Biocatalytic Efficacy

The data obtained from the literature revealed that nanosilver, i.e., NanoCid<sup>®</sup> L2000 developed by Nano Nasb Pars Company, Tehran, Iran, efficiently controls the growth of *S. typhimurium*, *E. coli* O157:H7, *L. monocytogenes*, and *Vibrio parahaemolyticus* (Vance et al., 2015; Zarei et al., 2014). It has also been reported that carboxymethylcellulose-enabled AgNPs film showed better antibactericidal potential compared to the solo AgNPs (Siqueira et al., 2014). Nanoengineered surfaces (via TiO<sub>2</sub>, nanosilver, and ZnO) were used to suppress the microbial growth and enhance the safety and quality of the food materials. The biocontamination associated with food transportation, poultry farming, and food processing was overcome by UV-C, ultraviolet light-activated TiO<sub>2</sub>, and silver-coated nanofiber mats (Khan et al., 2015). Nanophotocatalysts, nanoadsorbents, and nanoenabled membranes are applied to wastewater treatment (Rodrigues et al., 2017). The antimicrobial efficacy of nanomaterials is also explored in marine transport, textile industry, food packaging, and medicine (Sharma et al., 2020; Suvarna et al., 2022). The elements such as organic acids, essential oils, enzymes, peptides, and biopolymers conjugated with MgO, AgNP, Cu, CuO, Cd selenide/telluride, ZnO, carbon nanotubes, and chitosan are acting as antimicrobial composites. Similarly, several plant products such as clove, thyme, rosemary oil, tea tree, inner bark of pine trees (*Pinus sylvestris*), and sea buckthorn (*Hippophaë rhamnoides* L.) leaves

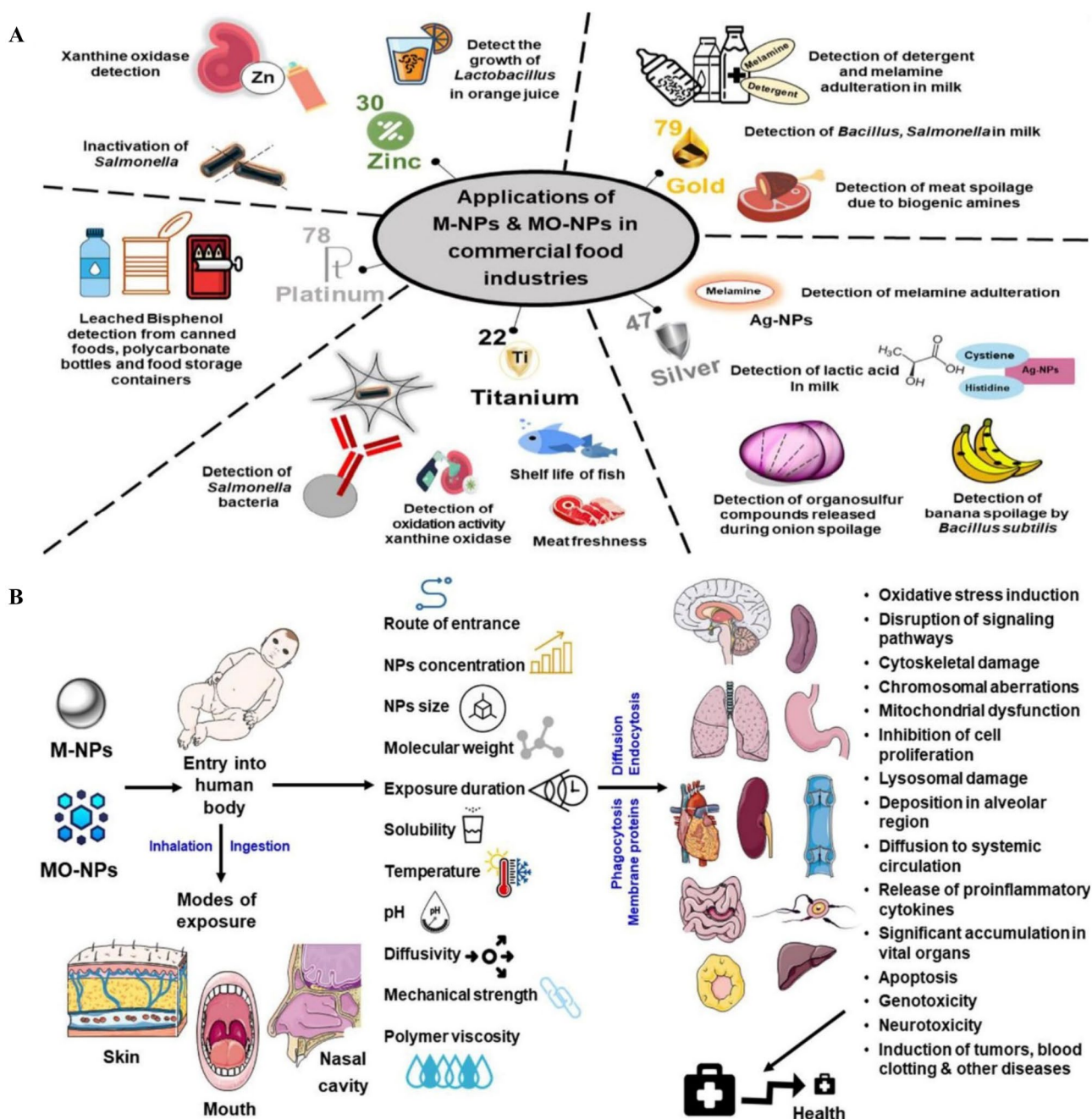
(Brobbe, 2017; Vasile et al., 2017a, b) can be applied for food packaging. The proposed mechanisms associated with the antimicrobial response of NPs include (a) either by disruption/penetration into the cell envelope or interruption with trans-membrane electron transfer or oxidation of cell components or production of reactive oxygen species (ROS). Moreover, the combination of nisin/chitosan with  $\alpha$ -tocopherol showed an antimicrobial and antioxidant potential (Lai, 2023; Vasile et al., 2013, 2017a, b). Figure 4A represents the positivity associated with metal NPs along with some of their salient features associated with food industries on a commercial basis, and Fig. 4B denotes some of the negative impacts of metal NP-modulated packed foods over human health (Kumar et al., 2021).

The oxidation of food products (during storage and transport) revealed development of color loss, rancidity of lipids, and degradation of vitamins. The antioxidants such as metal chelators, free radical scavengers, oxygen scavengers, and singlet oxygen quenchers are usually amalgamated in the food packaging along with polymeric blending (Couto & Almeida, 2022; Gómez-Estaca et al., 2014; Tian et al., 2013). Migratory and non-migratory coatings of antioxidants may be applied via covalent immobilization without disturbing the sensorial impact of packaged food materials (Nerín et al., 2008; Stoleru et al., 2016; Yemmireddy et al., 2015).

Biocatalysts (enzymes) are usually applied for preserving optimal pH, thermal/solvent stability, in order to avoid any alteration during the period of proceeding, transport, and storage of packed food materials. Lysozymes are usually unified into the packaging coatings for a covalent immobilization and controlled release, thereby maintaining the pH and thermostability (Barbiroli et al., 2012; De Souza et al., 2010; Vasile, 2018).

### Nanobiosensor-Based Detection of Toxins, Pathogens, Pesticides, Contaminants, and Heavy Metals

Numerous toxins, microbiotas, and pathogens deteriorating the quality of food materials are detected by fluorescent NPs (Burriss & Stewart, 2012). Nanobiosensor-based bioreceptors are responsible for detection of toxin developed by several pathogens. Similarly, enzymes, gaseous sensors, monoclonal antibodies, aptamer sequences, and ssDNA probes are engaged in the identification of food pathogens (Lotfi et al., 2019). Graphene particles, Au/AgNPs, and peptide bionanosensors revealed a better sensitivity and miniaturization and helps in pathogenic detection. (Mustafa & Andreescu, 2020). A presence of a multiple number of common contaminants in food/food materials such as drugs, heavy metals, veterinary antibiotics, pesticides, and allergens is found to be affecting human health adversely. Several platforms were applicable for detection of such contaminants,



**Fig. 4** **A** The positivity associated with metal NPs and their features used in commercial food industries. **B** The negative impacts for consumption of metal NP-based packed foods on human health. Reproduced with permission from reference Kumar et al. (2021)

namely, (i) magnetic NPs ( $\text{Fe}_2\text{O}_3$  NPs) towards enrichment and separation of analyst, (ii) electrochemical and optical sensors (nanomaterials like Au and AgNPs with good conductivity and surface platform resonance (SPR) characteristics), and (iii) graphene and carbon nanotube (CNT)-based electrochemical sensors (for a better electrical conductivity) (Mustafa & Andreescu, 2020). For detecting heavy metals like  $\text{Cu}^{2+}$ ,  $\text{Hg}^{2+}$ , and  $\text{Fe}^{3+}$ , Wang et al. utilized

nitrogen-doped carbon dots as fluorescent sensor (Wang et al., 2017).  $\text{CuO}$  NPs coupled with indium tin oxide (ITO) were used for detecting organophosphate pesticides (Tunisi et al., 2018). Further, paraoxon (a pesticide) was identified via Pt NP-based bionanosensor and usually offers better enzyme loading tendency, surface area, and conductivity (Hondred et al., 2018). A brief overview of a diffusion and chemical reaction-based time-temperature indicator (TTI)

as well as time–temperature-related deterioration of nanoparticulate elements is depicted in Fig. 5A whereas Fig. 5B reveals several characteristics and applications of nano-derived metallic components/materials in food packaging (Bajpai et al., 2018; Choi et al., 2020).

## Characterization and Toxicological and Safety Features of Nanomaterials Associated with Food

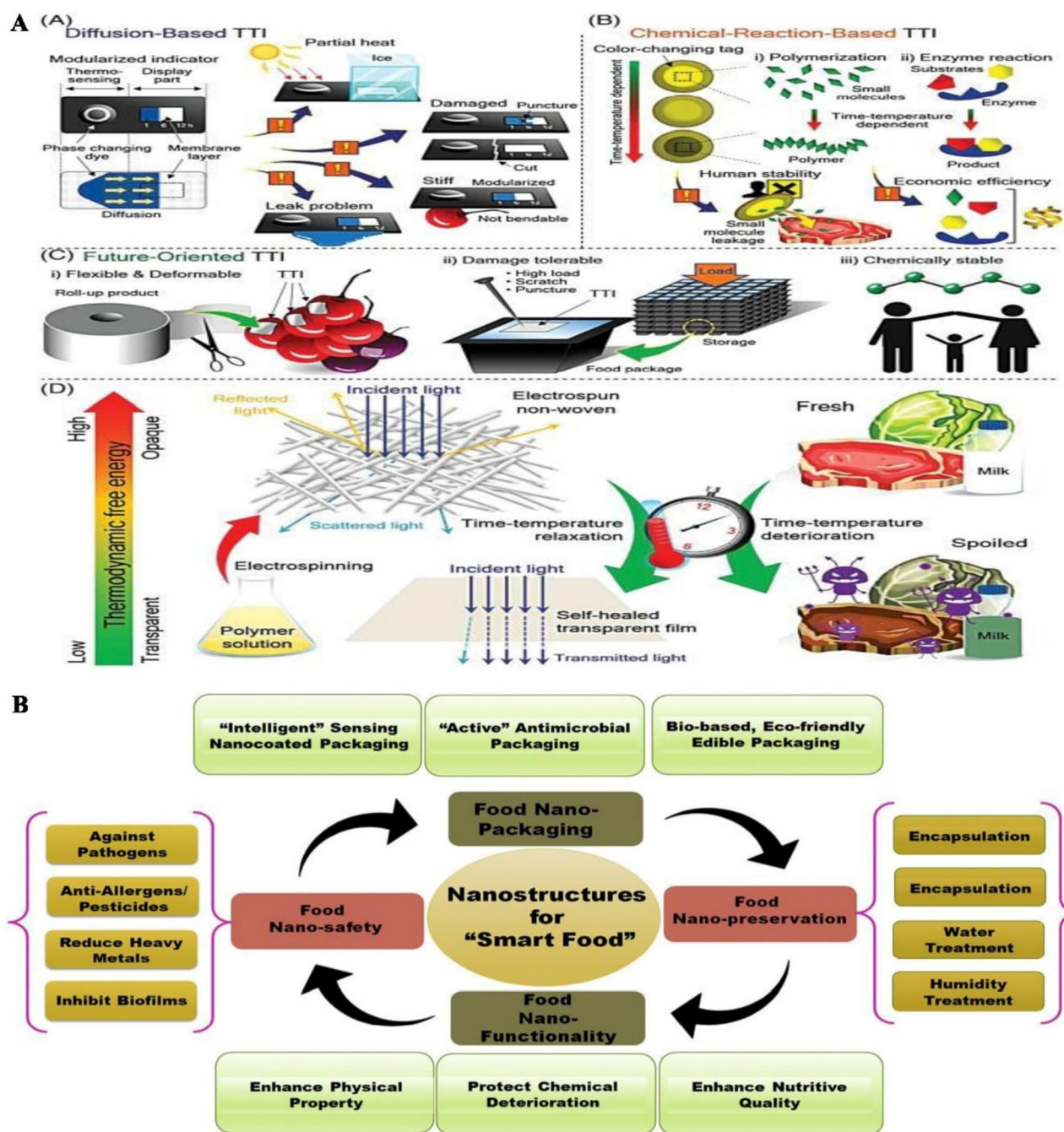
Despite the global exploitation of nanotechnology, the public concern is increasing simultaneously related to their environmental impact and toxicity. The toxicity of NPs is stirred via catalytic, dynamic, and kinetic impact on their functionalization, agglomeration, functional environment, and net particle reactivity (Chen et al., 2015; Zou et al., 2016). NPs in the peripheral surface of the packaging components were not problematic, but their integration and translocation into food materials impeded human health (cytotoxicity and genotoxicity) (Teow et al., 2011). The toxicokinetic issues of NPs are basically associated with their non-degradable, persistent, and non-dissolvable nature (Tiede et al., 2008). The deficit in government guidelines, consumer awareness, policies, and detection of nanotechnological risk assessment provides stringency in familiarization of nanobased toxicity evaluation and regulatory processes. Because of the tiny particle size (Schrand et al., 2010), NPs are highly reactive and can readily move across the capillaries and membrane barriers, resulting in numerous toxicodynamic and toxicokinetic outcomes. The enzyme and protein binding tendency of NPs leads to stimulation of oxidative stress and cellular apoptosis via production of ROS (Hajipour et al., 2012). The evidences of the literature revealed that NPs induce severe toxicity to numerous vital organs such as the kidneys and liver along with the immune system. Researchers from all round the world have reported several toxicological issues (both in vitro/in vivo) towards the metal NPs such as Ag and TiO<sub>2</sub> (Botelho et al., 2014; Mao et al., 2016; Valdiglesias et al., 2013). The scanning electron microscopy (SEM) images of graphene–epoxy composites, bacterial nanocomposite fibers, and carbon-based nanomaterials along with their suitable modifications are depicted in Fig. 6 (1–3) (Ibrahim et al., 2021; Mitura et al., 2021; Raul et al., 2022).

International Conference for Harmonization (ICH) and the Organization for Economic Co-operation and Development (OECD) anticipated for numerous methods for genotoxicity detection towards NPs. The uptake of exogenous nanomaterials such as nanosilver, particulates of asbestos, quartz, and crystalline silica leads to the development of primary genotoxicity (via ROS generation) (Kumar & Dhawan, 2013; Mitura et al., 2021). Genotoxicity was induced by ZnO, C<sub>60</sub>, and TiO<sub>2</sub> via the development of peroxynitrite (Kumar et al., 2011;

Wojewódzka et al., 2011; Xu et al., 2009). The ZnO NPs are also associated with the induction of nutritive and oxidative stresses, provoking secondary genotoxicity and inflammatory reactions in human monocytes (Senapati et al., 2015). The physicochemical parameters affecting induction of genotoxicity along with generation of ROS include particle charges, surface, size, particle dissolution, shape, the metal oxide ions and nanometals, UV-modulated induction, aggregation, route of interaction with cells, inflammatory conditions, and the pH of the medium (Fu et al., 2014). MOs are incorporated into the food industry/sector as smart/intelligent packaging components and/or nutritional additives owing to their antimicrobial potentiality. Moreover, it was also observed that NPs of ZnO are less toxic compared to the rest of nanomaterials pertinent to the food industry (Kumbıçak et al., 2014; Kwon et al., 2014). 3-Mercaptopropanoic acid–CdSe/ZnS quantum dots at a very low concentration (10 nM) induced genotoxicity and cytotoxicity in plants. The induction of oxidative stresses (ROS, RNS) along with lipid peroxidation was governed by uptakes of NPs in biological systems which causes membrane disintegration, DNA damage, and cell death. The metabolic trends, bioavailability, toxicity, and disposition of NPs towards the environment must be studied in order to exterminate nanotechnology modulated problems in the food industry (Badgley et al., 2007). Biobased nanocomposite packaging associated with their antimicrobial impact is presented in Table 3 (Ramos et al., 2018).

As per as the regulation for food contact materials (plastic) (EU) 10/2011, only those NPs authorized under Annex I of the regulation (e.g., carbon black (at 2.5 wt%), titanium nitride, and silica) are to be incorporated in plastic packaging meant for food products (Reig et al., 2014; Simoneau et al., 2012; Wyser et al., 2016). Titanium nitride NPs (at a concentration of 20 mg/kg) are explored as additive in manufacturing of polyethylene terephthalate (PET) bottles; however, the overall unambiguous migration limit and specific migration limit (SML) for the same were not specified in the regulatory guidelines. Moreover, the fillers/particles other than the above list to be included in food contact plastic packaging must be submitted to the European Food Safety Authority (EFSA) for approval based on the assessment of their toxicity, migration, and viable exposure tendency (Störmer et al., 2017; Vrček et al., 2016).

The detection and characterization of nanomaterials within a polymer matrix are usually carried out via X-ray fluorescence (XRF), X-ray diffraction (XRD), transmission electron microscopy (TEM), asymmetric flow field-flow fractionation (AF4) associated with a dynamic light scattering (DLS), or a multi-angle light scattering (MALS) detector (for determination of particle size) (Barage et al., 2022; Sarfraz et al., 2021). Similarly, AF4 in association with inductively coupled plasma-mass spectrometry (ICP-MS) or single-particle (sp)-ICP-MS is used for physicochemical

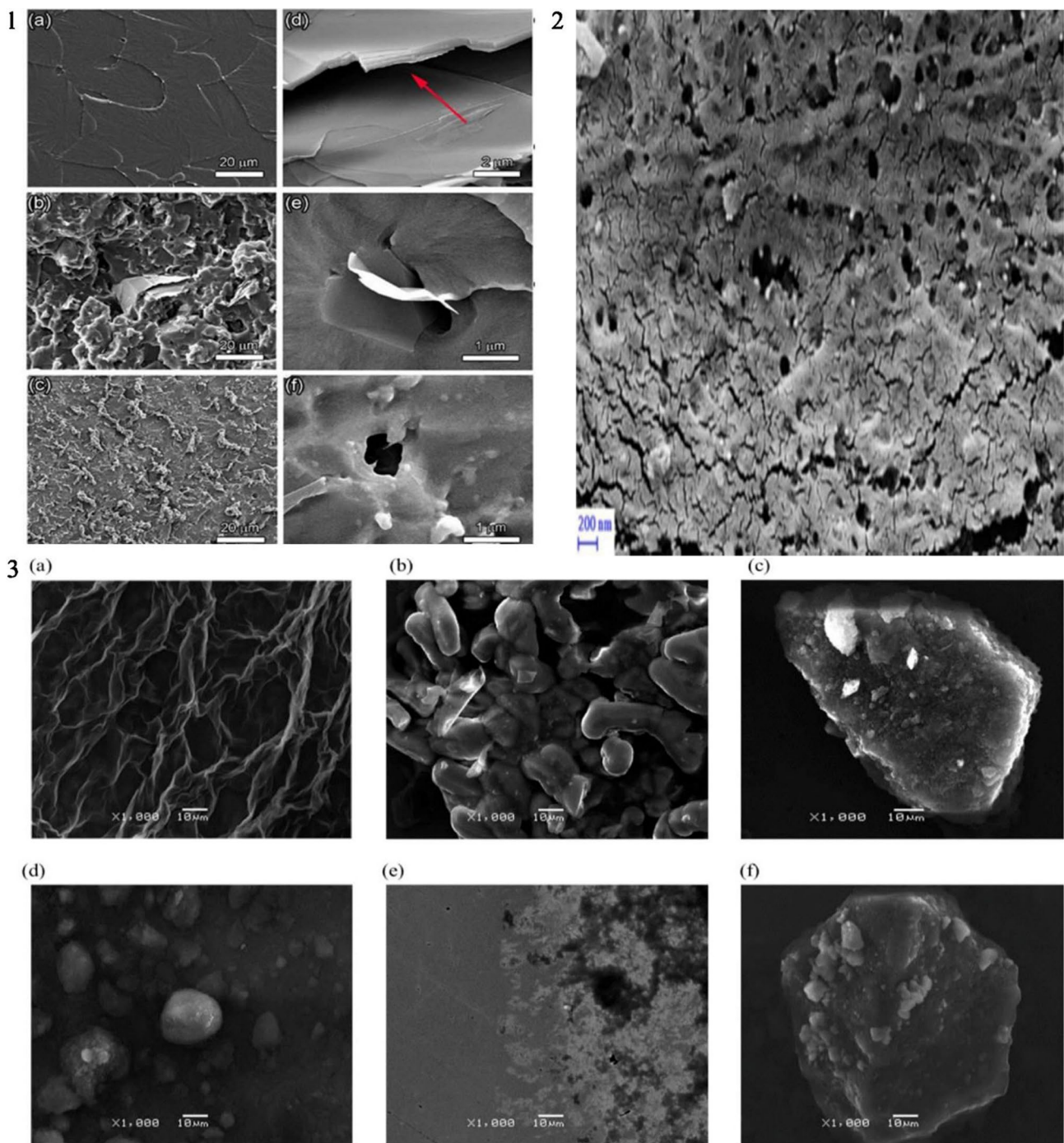


**Fig. 5** A Time–temperature indicators (TTIs), their operating mechanisms, and expected problems. (A) A modularized diffusion-based TTI with thermal sensing and display components. (B) A chemical-reaction-based TTI that uses color-changing chemicals. (C) The outstanding characteristics of the desired future-oriented TTI. (D) An innovative type of TTI fabricated using self-healing nanofibers. The

time–temperature-dependent change in surface area and corresponding light transmittance is driven by the flow of thermodynamic free energy and operates on the same timescale as the deterioration of perishable foods. **B** The systematic approach of nanoparticles in various areas of food industry. Reproduced with permission from references Choi et al. (2020) and Bajpai et al. (2018)

analysis (Bustos et al., 2013; Fabricius et al., 2014; Laborda et al., 2020; Olesik & Gray, 2012). However, sp-ICP-MS measurements possess difficulties while measuring complex samples as incompatible matrices or nanodispersions containing broad particle size (Olesik & Gray, 2012). The

risk of NP exposure at the time of development and processing stages could be reduced by following the protocols as detailed in ISO/TS 12,901 series (Ramos & Almeida, 2022; ISO: Geneva, Switzerland, 2014). However, there is an abridged exposure possibility towards the post-production



**Fig. 6** **1** Mechanical characterization and morphology of 1.0 wt.% graphene–epoxy composites. **(a–c)** SEM images of fracture surfaces of epoxy resin, ultrasonic-treated nanocomposites, and the HP- and ultrasonic-treated nanocomposites, respectively. **(d)** High-magnification image of graphene block in **(b)**. The big gap between the graphene sheets, as indicated by the arrow in **(d)**, implies that the graphene sheets slide over each other during the bending test. **(e, f)** High-magnification images of wrinkled and bridging graphene in **(c)**. **2** SEM micrographs of bacterial nanocomposite fibers. **3** SEM images of the

surface morphology carbon-based nanomaterials (CBNs): **(a)** graphene oxide (GO) film; **(b)** amorphous carbon powder formed using radio frequency plasma activated chemical vapor deposition method; **(c)** fluorescent nanodiamond (MDCHF); **(d)** plasma–chemically modified detonated nanodiamond particles (MDP1); **(e)** chemically modified detonated nanodiamond particles with hydroxyl functional groups (MDCHPOH); **(f)** pure detonated nanodiamond particles (DND). Reproduced with permission from references Ibrahim et al. (2021), Raul et al. (2022), and Mitura et al. (2021)

**Table 3** Bio-based nanocomposite packaging tested for their antimicrobial properties

Packaging materials	Nanomaterials	Active compounds	Active compound concentration	Observations on developed bionanocomposites
Chitosan	MMT	Rosemary EO	0.5, 1, and 1.5% (v/v)	1.5% (v/v) EO showed antibacterial activity against <i>L. monocytogenes</i> , <i>E. coli</i> , <i>P. putida</i> , <i>Streptococcus agalactiae</i> , and <i>Lactococcus lactis</i>
Agar	Cellulose nanoparticles	Savory EO	0.5, 1, and 1.5% (v/v)	1.5% (v/v) EO demonstrated the highest inhibition capacity against <i>L. monocytogenes</i> , <i>Staphylococcus aureus</i> , <i>Bacillus cereus</i> , and <i>Escherichia coli</i>
Chitosan/quinoa protein	Chitosan tripolyphosphate nanoparticles loaded with thymol	Thymol	4.4–0.44 mg/mL	Films exhibited good antimicrobial activity against <i>L. innocua</i> and <i>S. aureus</i> and high activity against <i>Salmonella typhimurium</i> , <i>Enterobacter aerogenes</i> , and <i>E. coli</i>
HPMC	Chitosan nanoparticles	Oleic acid	1% (w/w)	Oleic acid showed higher antibacterial activity against <i>Bacillus</i> and <i>Klebsiella</i> , inhibition against <i>Pseudomonas</i> and no inhibition for <i>Salmonella</i>
Fish gelatin	Chitosan nanoparticles incorporated with <i>Origanum vulgare</i> L. EO	<i>Origanum vulgare</i> L. EO	0.4, 0.8, and 1.2% (w/v)	1.2% (w/v) EO showed antibacterial efficacy against <i>S. aureus</i> , <i>L. monocytogenes</i> , <i>S. enteritidis</i> , and <i>E. coli</i>
Pectin/papaya puree	Cinnamaldehyde nanoemulsions	Cinnamaldehyde	1% (w/w)	Cinnamaldehyde exhibited higher activity against <i>L. monocytogenes</i> and <i>S. aureus</i> than <i>E. coli</i> and <i>S. enterica</i>
PLA	Lipid bilayers of PDANHS Nanoliposomes	Cinnamaldehyde	60, 120, 180, 240 mg/mL	Cinnamaldehyde PLA films did not show antibacterial activity against <i>E. coli</i> W1485 and <i>B. cereus</i>
PLA	Cellulose nanocrystals and lignin nanoparticles	Lignin	1 and 3% (w/w)	Lignin PLA films showed antibacterial activity against <i>Pseudomonas syringae</i> pv. <i>Tomato</i>
Starch	Halloysite	Nisin	2 and 6 g/100 g	Nanocomposite film containing nisin effectively inhibited <i>Clostridium perfringens</i> , <i>S. aureus</i> , and <i>L. monocytogenes</i>
PLA	PLA nanofibers loaded with cinnamon EO/-CD	Cinnamon EO	11.35 mg/mL	Cinnamon EO films displayed excellent antimicrobial activity against <i>E. coli</i> and <i>S. aureus</i>
WPI	Cloisite 30B organoclay	Cloisite 30B	5, 10, and 20 g/100 g WPI	WPI-Cloisite 30B films showed bacteriostatic effect against <i>L. monocytogenes</i>
Starch	Cellulose nanofiber	Cellulose	2–10% (w/w)	Cellulose nanofiber increased inhibition effect towards gram-positive but not against gram-negative bacteria

*β*-CD cyclodextrin, EO essential oil, HPMC hydroxypropyl methylcellulose, PDA-NHS polydiacetylene-N-hydroxysuccinimide

and transportation of nanomaterials/formulation as it is usually carried out in thoroughly sealed containers.

The mutagenicity and cytotoxicity study of organomodified clays revealed an induced alterations at a concentration range of 0–250 g/mL, whereas the unmodified clay remains claim free towards any sort of toxicity within a concentration range of 0–125 g/mL (Maisanaba et al., 2015a, b). Majority of the *in vitro* toxicity reports claimed cell death; but the preclinical and clinical data demonstrated a very low toxicity (Maisanaba et al., 2015a, b). Guo et al. conducted the toxicity study on silica particles on HT9-MTX and Caco-2 co-cultures (Guo et al., 2018a, b) and reported an altered intestinal epithelial cell functionality. Moreover, the report published by the European Food Safety Authority revealed non-toxicity of silica (E 551) (Younes et al., 2018). The nanoparticulate toxicity is prominently prejudiced by their size, dispersion, morphology, and concentration.

### Regulatory Aspects, Consumer's Perception, Acceptability, and Safety Consideration

Although nanotechnology is extensively applicable in packaging of foods and nutraceuticals with an advanced functional properties such as improved thermal, mechanical, sensing, antimicrobial, and barrier properties of the packaging components, still, consumer acceptability evaluation is highly prioritized towards its deployment in food packaging (Gupta et al., 2012; Nasir et al., 2022). The nanomediated products are toxic towards animals and plants. So far, no such standard regulatory guidelines were strictly implemented for regulating their exploration in both food and agriculture sectors. In the USA, the USFDA is enthusiastically involved in the regulation of nanofood and their packaging. Food Standards for Australia and New Zealand (FSANZ) is also actively involved in regulating nano-food ingredients and additives in Australia (Cubadda et al., 2016; Siddiqui et al., 2022). Similarly, the Scientific Commission on Emerging as well as Newly Identified Health Risks (SCENIHR) is widely associated with assessment of risk associated with nanotechnology in the European Union and emphasized the safety assessment of nanotechnology-based food ingredients prior to their authorization for clinical use (Tinkle et al., 2014). The European Union Novel Foods Regulation (EC 258–97) usually covers food or nanofood components. The EFSA suggested that the nanoadditives and food packaging materials authorized prior to 2009 must be treated with respect to the reevaluation program. Moreover, China and Japan, the superpower of major nanomaterial production, have no such specific regulations associated with nanotechnology-based products (O'Brien & Cummins, 2010). The deficiency of regulations for food

in various countries is because of their less availability, exposure, and clinical toxicity. Because of the issue of emerging regulatory guidelines, most of the countries were demanding a genuine regulatory system/protocol towards efficient handling of risks concomitant with the nanofood. Numerous nanotechnology-based guidelines/protocols have been designed by regulatory agencies throughout the world for assessing the safety of nanomaterials and recommend their future concerns applicable in several commercial products, like food ingredients, cosmetics, drug products, and animal food (Zhang et al., 2018). The scientific guidelines were released by the EFSA Scientific Committee towards handling of nanoscience-mediated potential risks towards feed and food (Barlow et al., 2009) and industry recommendations for using contrived nanomaterials (NMs) (EFSA, 2011). The guideline includes (1) requirement of physico-chemical characterization for enzymes, food additives, novel foods, pesticides, and feed additives and (2) testing protocols for identification of hazards in NMs such as genotoxicity (*in vitro*), pharmacokinetics, and oral toxicity studies in rodents. On Contaminants in the Food Chain (CONTAM), the EFSA Panel released a statement regarding the presence of nanoplastics and microplastics in sea food reporting the non-toxicity and toxicokinetic aspect of micro- and nanoplastics on human health (EFSA, 2011, 2016).

However, more research is needed to establish the analytical protocols for toxicity evaluation of micro-/nanoplastics in food. Society acceptance and willingness to pay (WTP) towards nanofood and packaging are the topics of interest. The societal perception of nanofood/packaging was studied in Germany and France. The report revealed a reluctance of consumers to accept the incorporation of nanotechnology in their food products (Peters et al., 2014). The United States Food and Drug Administration (USFDA) promotes and supports the safe use of nanotechnology (Wyrwa & Barska, 2017). As per the European Parliament and the Council Regulation 1169/2011, foods containing nanomaterials should be labeled (Grieger et al., 2016). The survey report of nanotechnology-based products conducted in the USA indicated an acceptance of the technology by the US citizen wide inclusion of additional information regarding nanotechnology (Brown & Kuzma, 2013; Giles et al., 2015). Moreover, the studies need transparency in the development of nanotechnology-based regulation and the consumers should be informed about its benefits and/or risks (Mustafa & Andreescu, 2020).

In the case of silver-based packaging materials, the release of silver ions leads to a major consumer concern towards their safety and sustainable impact over human health (Gupta et al., 2012). As per the evaluation report of Siegrist et al. (Nasir et al., 2022) concerning about the recognition of nanotechnology in food and packaging materials,



it has been hypothesized that nano-outside (e.g., packaging) is more acceptable by the consumers than nanoinside (e.g., foods) (Siegrist et al., 2008). However, the awareness and perception of the consumers on nanotechnology and food packaging are transformed in the recent era and might be turning into a positive way in the coming future. Rigorous toxicological screening along with complete government legislations/guidelines and protocols is highly essential for legalized nanotechnological approaches. A globally recognized and accepted regulatory system is promptly needed for observing and regulating the exploitation of nanoparticles in the food industry (González-Nilo et al., 2011; Osmani et al., 2022). Based on the safety aspects of nanomaterials, the research report (limited data) posed many health hazards because of their toxicity (migration from food packaging to food contents. Though nanoclays are recognized as safe, some studies reported the migration of aluminum and silicon from nanoclay. Moreover, the organomodified clay like octadecylamine and aminopropyltriethoxysilane revealed toxicity (Jiang et al., 2019; Oh et al., 2019; Salgado et al., 2021; Wagner et al., 2017). Similarly, nanocellulose is found to be non-toxic and certified by ISO Standard 109,993–5 (Camarero-Espinosa et al., 2016; Oh et al., 2019; Salgado et al., 2021; Shatkin & Kim, 2015), whereas the dry powder inhalation developed from nanocellulose showed stickiness towards alveolar region (Lindström & Österberg, 2020; Wagner et al., 2017). Because of the chances of formation of ROS (reactive oxygen species) via retention, carbon nanotube particles were found to be unsafe (Francis & Devasena, 2018; Kobayashi et al., 2017; Mohanta et al., 2019; Shatkin, 2020). The agencies like the EFSA and FDA established different regulations towards migration of nanoparticles from different packaging materials (Paidari et al., 2021). Similarly, the migration associated with the clay nanoparticles was reported and found to be directly associated with time of contact and temperature (Huang et al., 2015a, 2015b). The toxicity of NPs was elucidated in a binary ways: (i) via reactive oxygen species-mediated toxicity and (ii) chemical composition of NP-mediated toxicity. Metal or metal oxide NP recrystallization and/or crystallization lead to alteration of functionality of several protein molecules, while metal alloys (Fe/Pt alloy, Co/Cr alloy, ZnO, SiO<sub>2</sub>, and TiO<sub>2</sub>), multi-walled nanotubes (MWCNTs), and single-walled nanotubes (SWCNTs) usually reveal genotoxicity (Brennan et al., 2023; Han et al., 2011; Kitz et al., 2022; Landsiedel et al., 2009). Moreover, physicochemical parameters like pharmacokinetics, osmotic concentration, toxicity, and pH of nanomaterials could be governed (both qualitative and quantitative) for a non-toxic and commercial viability (He et al., 2014, 2015a, b). Till date, the USFDA and European union (EU) are found to be the only segment which reflects a clear regulation towards handling and incorporation of nanomaterials in food legislation (He & Hwang, 2014). Further

studies are much needed for a better toxicological investigation of nanocomposites and bionanocomposites for their social and commercial engagement.

## Recyclization of Nanoreinforced Plastic Packaging

Recyclization of the polymer nanocomposites is considered being another important factor for managing the nanotechnological functionalization. Though mechanical recycling protocols are there for plastic nanocomposites (Khalid et al., 2022; Welle, 2023), still, advancement in research in such area is in high demand. Researchers have performed experiments towards recyclization of nanoreinforced plastic packaging (PE, PP, and PET monolayer films reinforced by 4 wt% of nanoclay, CaCO<sub>3</sub>, Ag, and ZnO NPs) (Sánchez et al., 2014). The outcome does not reveal any impactful changes in the material properties (like degradation fumes, pinholes, haze, smells, and elongation at break) as well as material quality of nanoreinforced recycled materials compared to their conventional ancestors (Welle, 2023). Bionanocomposites in association with organic wastes were used to develop safe and good quality of compost via microbial decay (Armentano et al., 2018; Kijchavengkul & Auras, 2008; Zhao et al., 2008). Collaborating with the ideal prospects of individual polymers (natural and synthetic) and nanoparticles of clay, cellulose nanocrystals, layered silicate, and TiO<sub>2</sub>, they are potentially applied in the food industry (Lu et al., 2009). The compostability and degradability of bioplastics varied on the basis of their type (e.g., biocomposites of PLA/Laponite revealed the highest microbial attachment towards the surface resulting in biofilm formation, whereas PLA/organo-montmorillonite (OMMT) showed the lowest biofilm formation because of its inhibitory impact over biodegradation. Nanoclays influence the microbial degradation of natural/synthetic polymers (Castro-Aguirre et al., 2018; Goodwin Jr et al., 2018).

## Challenges Restraining the Impact of Nanocomposites on Food Packaging and Their Prevention Measures

Despite a promising and innovative tool for food packaging, the lack of knowledge towards consumer acceptability, migration, toxicity, and recyclability restricts the application of nanotechnology in food packaging. As per the authorization of “Plastic Food Contact Materials Regulation (EU) 10/2011,” nanoparticles developed from titanium nitride, silica, and carbon blocks show some possible environmental consequences (Sarker et al., 2023). However, nanoparticles of hazardous moieties reveal negative concerns towards

environmental and individual well beings. Therefore, the disposal, reusability, and individual toxicity analysis of the polymer nanomaterial should be prioritized before involving them into the sustainability and circular economy. In order to proceed with commercial approach, long-term in vivo NP studies along with food intolerance and accidental contaminations need to be resolved (Sarker et al., 2023). The NPs and their risk of toxicity broadly associated with their ingestion, inhalation, and skin absorption are a growing issue and need an urgent development of reliable analytical tools (for qualitative and quantitative estimation) towards their evaluation in food products (Chaudhary et al., 2020). The screening techniques like mass spectroscopy (test material composition), dynamic light scattering (particle size distribution), optical emission spectrometry (trace level elemental analysis), positron emission tomography (radio-tracing), electromagnetic signal enhancement, and chemical signal enhancement via chemisorption can be implemented with the approved regulatory information/guidelines for better analysis and sensing of the NPs in food products. Raman Nano Chip™, a nanorod-mediated safety detection system either/or coated with metallic, oxide, or polymeric NPs, is applicable in early detection of NP migration from food contact materials to avoid any other complications (Chaudhary et al., 2020).

## Future Prospective and Conclusion

Nanocomposites are recognized as a promising asset towards food packaging. With an advent of demand for sustainability, materials from numerous renewable resources are reinforced to contribute to overwhelming the hurdles concerned with biomaterials towards their industrial applicability. In this review, we summarized the development of numerous polymeric/metallic nanocomposite systems (natural and synthetic) and explored for their antioxidant, antimicrobial, and barrier properties; recyclability; migration issues; consumer acceptability; toxicity; and regulatory aspects. The discussion was also extended with some recent trends and future perspectives of cutting-edge nanomaterials (nanosensors) associated with smart food processing, packaging, security, storage, quality evaluation of preserved foods, and the methods arrayed for assessing the nanomaterial impact over the biological systems.

- Nanocomposites such as PLA/MMT, PHBV/graphene, PHBV/MMT, and MMT/chitosan nanosheets on PLA substrate coated with PVA/MMT solution exhibited impressive oxygen/water vapor barrier impact with the grading of compostable food packaging elements.
- Nanomaterials with modification in their persistent hurdles could be the potential material for the food industry and lead to a prolonged global economic hike with an altered food productivity rate at an enhanced stability. By implementing nanofillers, the barrier properties associated with the polymers can be provoked significantly via uniform distribution. Nanoclays are considered being playing a pivotal role towards improvement of the barrier properties of the fossil-based polymers and biodegradable and biobased materials.
- Layer-by-layer assembly and surface grafting techniques are able to overcome agglomeration and achieve uniformity in nanofiller distribution. Thermal, mechanical, and gas barrier properties of silica, Cu, and Zn NP-based polymer matrices are found to be enhanced significantly.
- Moreover, the physicochemical aspects like morphology, size, and surface chemistry of nanofillers need to be optimized on behalf of their specific solicitation. Along with antioxidant and antimicrobial properties, nanocomposites are capable of safeguarding and shelf life perpetuation of food materials. Silica particles, along with nanoclays, are efficient in encapsulating natural components like essential oils.
- The circular economy, sustainability, recyclability, and waste management of the polymer nanocomposite materials are to be prioritized. The available information is still very limited towards the mechanical recycling tendency of the plastic composite materials; hence, an extended research is highly essential. The biodegradable food packaging materials available in the market are still hindered by ampoules of challenges. Techniques implemented for improving the water and oxygen barrier properties (like multi-layer coextrusion, nanocomposite fabrication, and coating which have been promising in developing biodegradable, high oxygen/water vapor barrier systems towards food packaging) are ideal in dealing with our lively hood products.
- The current and future research is deeply associated with the design and development of active and intelligent packaging for enhancing the shelf-life of foods (via incorporation of liquid and moisture absorbers, oxygen scavengers, antimicrobials, and indicators/sensors for time–temperature and freshness detection of biodegradable packaging) during the period of storage and transportation. Such a system will definitely enhance the efficacy of the packaging components in both downstream (like transformation towards post-consumption compostable) and upstream (preconsumption monitoring of shelf-life and their extension).
- Universal compostability (including home and marine compostability of different polymers) draws a special attention towards biodegradable packaging design. In the countries where the facilities for industrial compostability are limited, the home composting opportunity is playing a key role in domestic biodegradation of waste

stream. With a tremendous exploration of plastic waste (specifically the microplastics gathered in the marine environment), it is very crucial and exciting to go with marine biodegradation trends thereby developing a solution for the plastic pollution control. However, the discovery of biodegradable plastics is playing a critical role in mitigating the robustness of plastic pollution.

However, there is a shortfall of knowledge associated with toxicity, migration, recyclability, and consumer acceptability of nanoreinforced plastic packaging components. The nanomediated food packaging is associated with ampoules of important issues which need to be resolved like their industrial scale-up, safety concerns (migration), and recyclability. As per the “Plastic Food Contact Materials” Regulation (EU) 10/2011, NPs only from carbon black, titanium nitride, and silica are considered being authorized. Materials besides the above to be used for food contact plastic packaging needed to be submitted to the government food safety authorities along with all specific data associated with toxicology, migration, and possible exposure towards their authorization. We are confident that the improvisation in biodegradable polymers with numerous aspect leads to offer opportunities for the discovery of next-generation sustainable food packaging by substituting the single-use food packaging films and containers, with an abridged utility of plastic waste and footprint of carbon.

**Author Contribution** The contribution of all authors is as follows: Manoj Kumar Sarangi: conceptualization, manuscript writing, data collection and compilation, editing, revision, correction, and review. Sasmita Padhi: data collection and compilation, correction, and review. L.D. Patel: conceptualization, correction, and review. Goutam Rath: conceptualization, correction, and review. Sitansu Sekhar Nanda: correction and review. Dong Kee Yi: correction and review.

**Data Availability** Data sharing is not applicable to this article, as no datasets were generated or analyzed during the current study.

## Declarations

**Conflict of Interest** The authors declare no conflict of interest.

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