A review on nanomaterials and nanohybrids based bio-nanocomposites for food packaging

Kalpani Y. Perera[1,2], Swarna Jaiswal*[1,2] and Amit K. Jaiswal[1,2]

[1]School of Food Science and Environmental Health, Technological University Dublin - City Campus, Central Quad, Grangegorman, Dublin D07 ADY7, Ireland.

[2]Environmental Sustainability and Health Institute (ESHI), Technological University Dublin - City Campus, Grangegorman, Dublin D07 H6K8, Ireland

***Corresponding author:**

Email: swarna.jaiswal@TUDublin.ie (Swarna Jaiswal)

Abstract

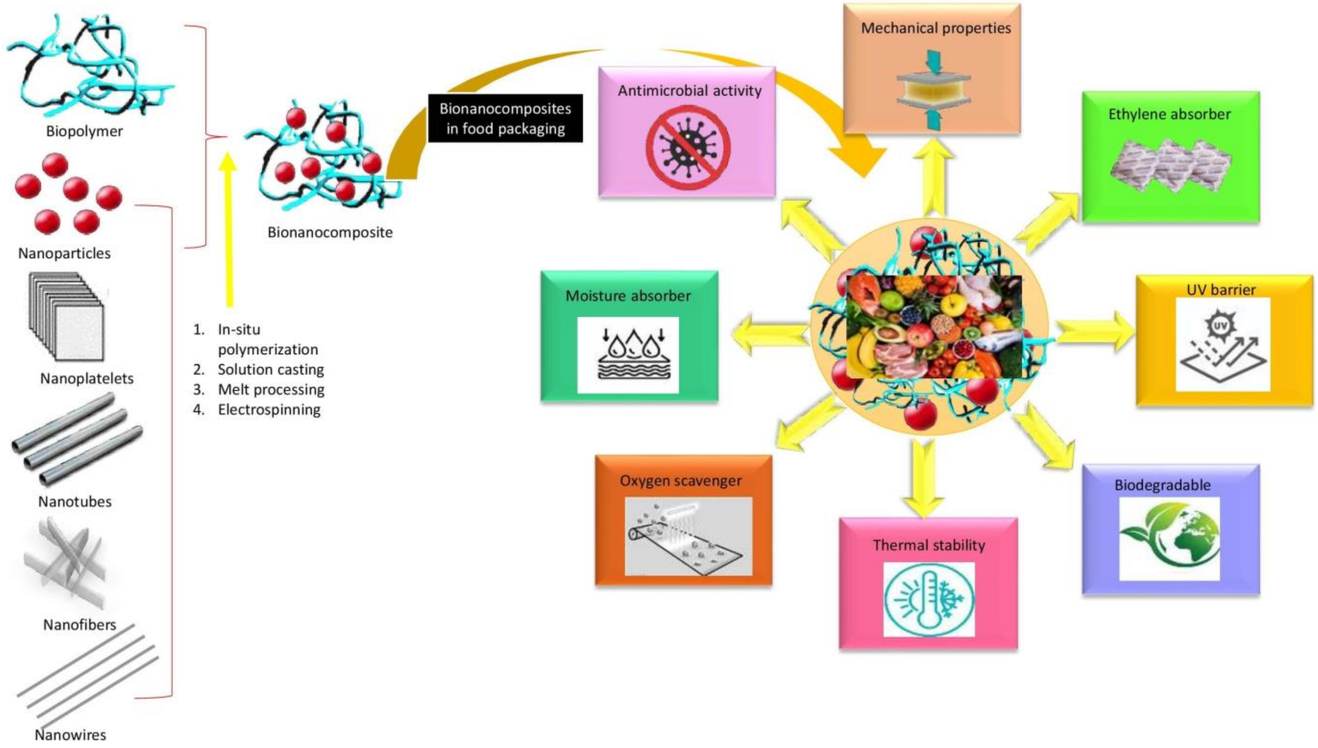
With an increasing demand for a novel, eco-friendly, high-performance packaging material "bio-nanocomposites" has attracted great attention in recent years. The review article aims at to evaluating recent innovation in bio-nanocomposites for food packaging applications. The current trends and research over the last three years of the various bio-nanocomposites including inorganic, organic nanomaterials, and nanohybrids, which are suitable as food packaging materials due to their advanced properties such as high mechanical, thermal, barrier, antimicrobial, and antioxidant are described in detail. In addition, the legislation, migration studies, and SWOT analysis on bio-nanocomposite film have been discussed. It has been observed that the multifunctional properties of the bio-nanocomposite~~s~~ materials, has the potential to improve the quality and safety of the food together with no /or fewer negative impact on the environment. However, more studies need to be performed on bio-nanocomposite materials to determine the migration levels and formulate relevant legislation.

Keywords: Bio-nanocomposite; Organic nanomaterials; Inorganic nanomaterials; Nanohybrid; Biodegradable Polymers; Food Packaging; SWOT analysis; Legislation

Highlights

- Bio-nanocomposites has attracted a great attention in recent years as packaging materials.
- Article presents recent progress in bio-nanocomposites for food packaging application.
- Bio-nanocomposites showed significant barrier, mechanical, thermal properties.
- Multifunctional properties of bio-nanocomposites can improve the food quality and safety.
- Migration, regulations, toxicity, and SWOT of bio-nanocomposite has been discussed.

Graphical abstract



Overview of bio-nanocomposite material in food packaging

1. Introduction

Food packaging plays an important role in food safety, quality, and shelf-life. The food packaging systems protect food from environmental contamination, odors, shocks, dust, temperature, mechanical forces, breakage, moisture, gases, physical damage, light, microorganisms, and humidity, during transport, processing, storage, and marketing. It plays an important role in maintaining the basic attributes of food such as color, temperature, taste, texture, quality of the food product, increase self-life and subsequently reducing food waste. The main causes of food deterioration such as oxidation, microbial spoilage, and metabolism, can be avoided from a good food packaging system which will result in increased food quality and shelf-life. Oxidation of food products can result in decreased nutritional value, energy content, flavor, and color thus decreasing the quality of food. The presence of pathogenic microorganisms increases the risk of food-borne diseases in humans. Packaging also allows us to glamorize the food product for marketing and provides an opportunity to deliver important information to the consumer (Braga et al., 2018). Thus, food packaging is essential for protection, containment, convenience, and communication (Al-Tayyar et al., 2020). In recent years, there has been a significant advancement in the packaging field, and it is now commonly referred to as active and intelligent packaging.

Food packaging material selection is an important factor in the food packaging industry. The packaging material can isolate the product from the external environment and has to be a non-toxic, impermeable physical barrier. The important properties of a food packaging material are physical, chemical, mechanical, thermal stability, antimicrobial activity, and water and light barrier properties (Al-Tayyar et al., 2020). Packaging materials such as natural packaging materials (gourds, shells, grass, wood, etc.), paper, glass, metal, plastic, biopolymer, and bio-nanocomposites are utilized as the food packaging materials (Risch, 2009). Natural materials, paper, glass, and metal have successfully been used as packaging materials for centuries as they are cheap, lightweight, and environmentally friendly. The trend of using plastic has massively increased in the past decades since it has become an effective packaging material due to its light weight, cost-effectiveness, high transparency, versatility, and ease of the process. Moreover, these synthetic polymers have good mechanical, thermal, and barrier properties. However, the merits of plastic may be the high production volume, short usage time, and the non-biodegradable nature that has ~~have~~ caused major concerns worldwide (Horst et al., 2020; Matthews et al., 2021; Rocha et al., 2020; Zubair & Ullah, 2020). As concerns grew, the use of biopolymers as packaging materials has begun recently due to their lower environmental impact and at the same time, they can mimic the properties of conventional polymers. In addition to the most valuable property of biodegradability, biopolymers exhibit properties such as excellent physical, mechanical and barrier properties. These properties allow the biopolymers to increase self-life, food quality, convince, and consumer attraction. Additionally, they are of low cost and easily accessible (Indumathi et al., 2019; Mangaraj, 2018; Sharma et al., 2020; Valerini et al., 2018). As biopolymer research progressed, nanotechnology was incorporated into these packaging materials to form a “bio-nanocomposites” material as an eco-friendly alternative packaging material. Bio-nanocomposites are typically constructed on biopolymer matrixes reinforced by nanofillers.

Bio-nanocomposites have increased barrier, mechanical, thermal, and antimicrobial properties which attributes to the presence of the nanomaterials in the packaging matrix. The nanoparticles introduced to the polymer matrix, acts as reinforcement, resulting in a complex diffusion path that results in reduced permeability of gas and water. Nanoparticles also create linkage with the biopolymers resulting in reducing the interaction of the water molecules with the polymer. These factors result in increased barrier properties of the packaging materials. The bond between the biopolymer and the nanoparticle results in the increased mechanical properties of the packaging materials. Finally, the high aspect ratio and homogeneous dispersion of the nanoparticles can strengthen the mechanical and thermal resistance of the packaging material due to the molecular mobility and relaxation of the polymer (Garcia et al., 2018). The various nanomaterials are used for food packaging such as silver NPs (Ramos et al., 2020), copper NPs (Wu et al., 2020), zinc oxide NPs (Sruthi et al., 2018), titanium dioxide NPs (Kaewklin et al., 2018), silicon dioxide NPs (Guo et al., 2018), nanocellulose (Niu et al., 2018), nanoclays (Asdagh & Pirsa, 2020), chitosan NPS (Rizeq et al., 2019), etc. Oxygen scavengers such as ZnO can be used for the packaging of cooked meat products, cheese, bakery products, fruit, vegetable, seeds, nuts, etc. to prevent discoloration, mold growth, rancidity, and for the retention of vitamin C. Ethylene absorbers such as Zeolite, Ag, TiO_2, ZnO can be used for climacteric fruits and vegetables food packaging by the reduction in ripening and senescence, thereby enhancing the quality and prolonging shelf-life. Antioxidant releasers such as Ag, ZnO, CuO, Graphene are mostly used for fresh fatty fish, meat, seeds, nuts, and fried products packaging to improve the oxidative stability of the product. Finally, nanoparticles (NPs) (such as Ag, TiO_2, ZnO, Cu, Graphene) with antimicrobial activity are used for fresh meat, fish, vegetable, fruits, dairy products, grain, cereals, and bakery products, ready-to-eat meals packaging to prevent microbial growth (Yildirim et al., 2018). There are about 400 companies in the world that focus on nanoparticles in food and food packaging. Nanocor is a USA-based AMCOL International Corporation which is specialized in the production of nanoclay-based plastic bio-nanocomposites with the trademark Nanomer®. These products have improved thermal, barrier, and physical properties. However, Plantic Technologies Limited, Australia designed starch-nanoclay-based ‘thermoformed plantic trays’ for Cadbury, Dairy Milk, and Mark

& Spencer Swiss chocolate. These packaging materials are biodegradable, non-toxic, have improved rheological, mechanical, and moisture properties (Bumbudsanpharoke & Ko, 2015).

There are several reviews on nanomaterial applications in food packaging. The published review in this area are focused on metal oxide-based nanocomposites in food packaging (Garcia et al., 2018), trends and challenges of biopolymer-based nanocomposites (Taherimehr et al., 2021), the concepts, and the future outlook of bio-nanocomposites materials for food packaging (Youssef & El-Sayed, 2018), applications and challenges of nanotechnology in food packaging (Enescu et al., 2019), production cost and safety of nanomaterials in food packaging (Tsagkaris et al., 2018), antimicrobial bio-nanocomposites (Sharma et al., 2020), and metal and inorganic nanoparticles in food packaging (Hoseinnejad et al., 2018; Jafarzadeh & Jafari, 2021). However, their scopes vary from this review article, and as per the authors' knowledge, none of the reviews has highlighted the most recent advances in organic, inorganic nanomaterials and nanohybrid based bio-nanocomposites for food packaging application. This review aims to evaluate the current trends and research articles published over the last three years on the various bio-nanocomposites, including inorganic, organic nanomaterials, and nanohybrids for food packaging applications. In addition, this review article discussed the migration of nanoparticles, legislation, and SWOT analysis of bio-nanocomposite-based food packaging materials.

2. Bio-nanocomposites in food packaging

Bio-nanocomposites are composites that ~~contains~~ a bio-based polymer matrix and an organic/ inorganic filler with at least one nano scale material (Saini et al., 2018; Arfat et al., 2017). Bio-nanocomposites are also known as bio-composites, nanocomposites, bio-nanocomposites, green composites, biohybrids, or bioplastics. Bio-nanocomposites exhibit beneficial properties such as biocompatibility, antimicrobial activity, biodegradability, mechanical, optical, and barrier.

Importantly, they are cost-effective, eco-friendly, and renewable. Bio-nanocomposites are categorized based on the matrix used, origin, shape, and the size of reinforcements (1) particulate (iso-dimensional nanoparticles used as filler) (2) elongated particles (nanotubes and cellulose nanofibrils used as filler) (3) layered structure (flocculated/phase-separated nanocomposites, intercalated nanocomposites, exfoliated nanocomposites). Bio-nanocomposites are prepared using various techniques such as in situ polymerization, solution casting, melt processing, electrospinning, and processing under supercritical conditions (Saini et al., 2018; Sharma et al., 2020).

The beneficial properties of bio-nanocomposites are studied in detail when determining the most suitable bio-nanocomposites for food packaging. The surface morphology of the bio-nanocomposite is studied through particle distribution, particle size, clarity of film, smoothness of surface, and texture of the material (Jayakumar et al., 2019). It is vital for a bio-nanocomposite to have high mechanical strength and dimensional stability, as the packaging system is essential to secure the food during stress conditions such as storage, handling, processing, and distribution. Therefore, it is essential to study the mechanical properties and the thickness of the films (Indumathi et al., 2019; Swaroop & Shukla, 2018). In addition, the bio-nanocomposite should have increased optical and barrier properties that are essential to avoid photo degradation and oxidation of food during storage and transport. Transparency, color, and clarity of the packaging material are important in terms of consumer attraction to the food product (Mangaraj, 2018; Sharma et al., 2020). Moreover, the increased thermal properties of the bio-nanocomposite are essential if the packaging material is exposed to high/ low temperatures during food packaging, transportation, and storage. The bio-nanocomposite film usually has high antimicrobial activity, which is vital to regulate unwanted microorganisms in a food packaging system (Mathew et al., 2019). A detailed description of the different bio-nanocomposites in food packaging is addressed in the following sections by categorizing it based on the type of nanomaterial incorporated.

3. Nanomaterials used in bio-nanocomposite and their applications

The use of nanotechnology in the food packaging industry has immensely increased in the past few years. The interest in using nanotechnology is due to the high reactivity, adherence, improved bioavailability, bioactivity, and surface effect of the nanoparticles (NPs). Before applying nanomaterials into food packaging, it is extremely important to understand the structure, environmental effects, safety, and toxicity of NPs. These factors depend on the properties of NPs, such as physical (size, shape, crystallinity), state of dispersion, and surface properties (Sharma et al., 2019). The incorporation of nanotechnology in food packaging enhances properties such as barrier (against O_2, CO_2, moisture, UV radiation, volatile substances, and light), mechanical, hydrophobicity, physical, thermal, antimicrobial, antioxidant, and rheological (Enescu et al., 2019). In addition, it decreases the weight of the packaging material while it is eco-friendly (Nile et al., 2020).

Nanotechnology is important in food packaging because it increases self-life, food quality, enhances food security, and enhances the texture, flavor, color, and nutrient availability of the food product. Therefore, nanotechnology is highly appreciated in food packaging, storage, and distribution in active and smart packaging. The NPs are used in the food industry in the forms of nanotubes, nanoliposome, nanoemulsions, nanolaminate, nanocomposites, nanospheres, nanocapsules, and nanofibers (Sharma et al., 2019). The most used NPs as food packaging materials can be either inorganic or organic. The inorganic particles can be transition metals (e.g. silver, copper, gold), alkaline earth metals (e.g., calcium, magnesium), non-metals (e.g. selenium, silicates), and metal oxides (e.g. zinc oxide, silicon dioxide, and titanium dioxide (Enescu et al., 2019). Other inorganic nanomaterials such as nanoclay and graphene oxide are commonly used in food packaging materials.

The most used organic NPs in food packaging are nano-cellulose, chitosan NPs, and starch NPs. Further, nanofibers, nanoplatelets, nanotubes, and nanowires are also used in bio-nanocomposite films. To incorporate these NPs to the polymer matrix, different methods such as electrospinning, steam coating, ion implantation, sputtering and electrochemical deposition are utilized (Braga et al., 2018). However, the use of nanotechnology in the food and beverage industry has become a controversial issue. This is due to the lack of knowledge of safety issues and the gap in ~~of~~ toxicology information. The migration and toxicity studies performed on the various nanomaterials are discussed in detail~~ed~~ in different section.

3.1. Inorganic nanomaterials and packaging applications

3.1.1. Metal based nanomaterials for food packaging.

Silver nanoparticles (AgNPs) have a strong antimicrobial activity towards a broad variety of pathogenic microorganisms such as viruses, fungi, yeasts, and bacteria. This property allows AgNPs to be used in water/air purifications, wound dressing, textile consumer products, therapeutic agents, biosensors, drug delivery systems, and medical devices. Moreover, AgNPs are easily processed due to their low melting points, making them useful for food packaging applications (Nakamura et al., 2019).

The studies carried out on AgNPs bio-nanocomposite films implied that AgNPs have ~~has~~ high antimicrobial activity towards both gram-positive and gram-negative bacteria (Cao et al., 2020; Lin et al., 2020; Motlagh et al., 2021; Ramos et al., 2020). Sonseca et al. (2020) carried out a study on green synthesized eco-friendly AgNPs with a Chitosan mediated method. These synthesized

AgNPs were incorporated into an oligomeric lactic acid and poly lactic acid matrix, which enhanced the antimicrobial activity, mechanical (toughness increased from 1.8 to 5.2 MJ/m^3 with the addition of 0.5% AgNPs), and thermal properties of the packaging material. The newly developed packaging material showed enhanced degradation of the packaging film with visible disintegration after 28 days. Migration studies were carried out by Ramos et al. (2020) to determine the release kinetics of AgNPs and Thymol from a poly-lactic acid matrix in an ethanol 10% (v/v) aqueous food simulant at 40°C. Inductively coupled plasma mass spectrometry (ICP-MS) was used to determine the total amount of silver released from films, with limits of detection and limits of quantification values of 1.19 µg kg^{-1} and 3.98 µg kg^{-1}, respectively. These results are well within the regulatory limits for silver, which is 0.01 mg Ag kg^{-1} food (EuropeanCommission, 2009).

The cytotoxicity effects of AgNPs synthesized by the green method from the extract of leaves of goldenrod (Solidago) were evaluated on H4IIE-*luc* (rat hepatoma) cells and HuTu-80 (human intestinal) cells using an xCELLigence real-time cell analyzer (Botha et al., 2019). This study confirmed the cytotoxicity of AgNPs at a concentration of 50µg/mL. Although the migration of AgNPs to food products/stimulants is very low, the toxicity effect of AgNPs develops a concern when used in food packaging in a higher concentration. Further, the increased use of AgNPs in different industries has led to the increased levels of AgNPs in the environment. These AgNPs are easily migrated as Ag^+ into liquids and soil, thus entering microorganisms, animals and causing toxicity effects on the whole eco-system (Yu et al., 2013).

Copper NPs (CuNPs) are extensively used as sensors, catalysts, surfactants, antimicrobials, and antifouling paints in the industry. However, increased usage of CuNPs showed a toxicity effect, especially on aquatic animals (Wu et al., 2020). Limited studies have been carried out on CuNPs bio-nanocomposite films mainly due to this toxicity effect. Li et al. (2020) developed an antimicrobial food packaging film with copper sulfide nanoparticles (CuS NPs) designed through a photothermal effect incorporated into a carrageenan matrix by casting method. This film resulted in increased transparency, a slight increase in mechanical properties, and thermal stability with a maximum decomposition at 250°C. The combined CuS NPs and near-infrared light irradiation of the packaging material reduced viable bacterial counts packaged beef. Further, studies on have been performed on copper oxide nanoparticles (CuONPs). Here different ratios of CuO NPs were incorporated into sodium alginate and cellulose nanowhisker matrix. This film with CuO NPs at 5 mM, has exhibited antimicrobial activity with a high zone of inhibitions against *S. aureus* (27.49 ± 0.91 mm), *E. coli* (12.12 ± 0.58 mm), *Salmonella sp*. (25.21 ± 1.05 mm), *C. albicans* (23.35 ± 0.45 mm) and *Trichoderma spp*. (5.31 ± 1.16 mm). Further, this film was able to prevent microbial growth in freshly cut pepper for up to 7 days (Saravanakumar et al., 2020). Many of the limited migration studies on CuNPs has been performed on synthetic polymers (Ahari & Lahijani, 2021).

Gold nanoparticles (AuNPs) are employed in the food packaging industry because of their medicinal, oxidative catalytic, and antibacterial properties, as well as their inert and nontoxic nature. AuNPs might be employed to fix other NP defects due to their nontoxic properties. Considering Au NPs are costly; hence a little research has been done on them. As a result, Au NPs are widely recommended as antibacterial agents for high-end, premium items like Caviar (Paidari & Ibrahim, 2021). Bumbudsanpharoke and Ko (2018) developed a packaging film using a green synthetic route to produce AuNPs that were directly produced and incorporated into a lignocellulose fibre matrix. The film showed enhanced radical scavenging activity thus showing a boost of antioxidant properties.

Sulphur NPs (SNPs) shows antibacterial effect against a variety of microorganisms while posing no toxicity to human cells (Saedi et al., 2020). Furthermore, many companies emit sulfur as by-product trash as sulfite in the form of air emissions or sulfates in the form of runoff or gypsum. Because of their restricted usage and disposal techniques, these chemicals have substantial environmental consequences. As a result, incorporating SNPs in food packaging may help to minimize the world's waste impact. The type and size of SNP have been shown to have a significant impact on its antibacterial action (Shankar & Rhim, 2018). Priyadarshi et al. (2021) SNPs were produced by sodium thiosulfate pentahydrate acidification and incorporated in alginate films. The tensile and water vapor barrier characteristics improved by 12 % and 41%, respectively, while the UV barrier properties increased by 99%. The antimicrobial activity of *E. coli* was 60%, while a complete bactericidal effect was observed for *L. monocytogenes* in 12 hours.

There are several studies carried out on metal nanomaterial incorporated bio-nanocomposites are depicted in Table 1.

3.1.2. Metal oxide-based nanomaterials for food packaging

Zinc oxide NPs (ZnONPs) have received remarkable global interest in industrial and biomedical applications due to their unique electrical, optical, catalytic, and photochemical properties. The UV-blocking, antimicrobial and antifungal properties make ZnONPs important in the food packaging and cosmetic industry (Sruthi et al., 2018). These nanocomposites containing ZnONPs exhibited advantageous properties for food packaging such as increased thermal properties (Indumathi et al., 2019; Kumar et al., 2019; Kumar et al., 2020; Rukmanikrishnan et al., 2020), mechanical properties (Jayakumar et al., 2019; Wang et al., 2020a), barrier properties (Indumathi et al., 2019; Shankar et al., 2018), antimicrobial activity (Abdollahzadeh et al., 2018) and decreased moisture content (Rukmanikrishnan et al., 2020). Moreover, ZnONPs incorporated bio-nanocomposites were able to increase the self-life of different food products including black grapes

(Indumathi et al., 2019), green grapes (Kumar et al., 2019), Ras cheese (El-Sayed et al., 2020), raw meat (Wang et al., 2020a), and minced fish (Abdollahzadeh et al., 2018; Shankar et al., 2018).

Kumar et al. (2020) developed a bio-nanocomposite packaging material utilizing green synthesized ZnONPs incorporation into a chitosan- gelatin polymer matrix. This film showed antimicrobial activity against both gram-negative (*E. coli*) and gram-positive (*S. aureus*) bacteria and improved thermal stability. The tensile strength of the ZnONPs incorporated film decreased slightly from 32.02 MPa (chitosan-gelatin film) to 26.39MPa while elongation at break increased from 20.24% (chitosan-gelatin film) to 35.65%.

While Jayakumar et al. (2019) developed an intelligent pH sensing wrap by incorporating ZnONPs and phytochemicals into a starch-poly vinyl alcohol matrix by solvent casting technique. This packaging material showed higher water barrier, mechanical and antimicrobial properties along pH sensing ability. The ZnONPs have been modified slightly for food packaging applications, such as Valerini et al. (2018) studies on aluminum-doped ZnONPs incorporated polylactic acid matrix and the film showed-high antimicrobial activity.

ZnONPs migration study was performed by Bumbudsanpharoke et al. (2019) on a low-density polyethylene -ZnO nanocomposite film in distilled water, 4% acetic acid (w/v), 50% ethanol (v/v), and n-heptane food stimulants. While the migration levels increased with time, the concentration of ZnONPs, and the acidic nature, the migration levels for all stimuli were between 0.006 and 3.416 mg L^{-1} which is much lower than the European Union specific migration limit of 5 mg kg^{-1} food (EuropeanCommission, 2016).

Titanium dioxide (TiO_2) is traditionally used as a food coloring additive. Therefore, TiO_2NPs are used in nanocomposite films as a safe, economical, and abundant material. The TiO_2NPs possess stable, biocompatible, dispersible, hydrophilic, photocatalytic, UV blocking properties, excellent ethylene scavenging, and antimicrobial activities (Kaewklin et al., 2018; Riahi et al., 2021; X. Zhang et al., 2019). TiO_2NPs incorporated bio-nanocomposite films have been tested against fruit products such as tomato (Kaewklin et al., 2018), green bell pepper (Salama & Aziz, 2020) and it prolonged the self-life of food products with the aid of the ethylene photodegradation ability of TiO_2NPs. Hosseinzadeh et al. (2020) developed TiO_2NPs incorporated chitosan-cymbopogon citrus essential oil bio-nanocomposite film by solvent casting method for the storage of minced meat at refrigerated conditions. The TiO_2NPs incorporated film increased water vapor permeability by 28%, while reducing elongation at break from 4.77 to 2.94 %. It also prevents the growth of total bacteria in minced meat during storage. Liu et al. (2020) developed a pH indicator intelligent packaging bilayer film in combination with agar- κ-carrageenan–anthocyanin. Here, the protective layer consists of agar and TiO_2NPs, while the sensor layer contains κ-carrageenan–anthocyanin. This film showed an increased UV–vis light barrier with a transmittance close to zero, and mechanical properties~~ were~~ in terms of tensile strength increased from 10.6193(c MPaontrol film) to 16.8668 MPa, and elongation at break increased from 30.9023% (control film) to 57.0802%. Results exhibited a colour change in buffer solutions, ammonium vapour and pork spoilage trails. Although TiO_2NPs has excellent ethylene absorbance, UV barrier properties and is ~~an ~~approved food additive, it may have potential migration and cytotoxicity issues which are studied in a limited-number in food packaging studies. Chen et al. (2019) studied the TiO_2NPs migration to food stimulants in polymer-laminated steel- TiO_2NPs composite film. The migration test was carried out at increasing temperature using acetic acid, ethanol, and ester food simulants, and the TiO_2NPs migration levels were determined by inductively coupled plasma optical emission spectrometry (ICP-OES). Results showed that the effect of increased temperature increases TiO_2NPs migration levels when compared to the time in the food stimulants.

Magnesium Oxide NPs (MgONPs) are nanoparticles with high reactivity, excellent stability, high surface area, less toxicity, and low cost. Thus, it is used mainly as a catalyst and as an antibacterial agent (Jagadeesan et al., 2019). The mechanism of action of the antimicrobial activity of MgONPs depends on the production of reactive oxygen species (ROS) (Castillo et al., 2019). Swaroop and Shukla (2018) developed a bio-nanocomposite for food packaging application with reinforcing MgONPs into a polylactic acid matrix to develop a food packaging material by solvent casting method. These films have enhanced mechanical (increase tensile strength to 29%), oxygen barrier up to 25%, UV barrier, and antimicrobial properties. However, in the presence of MgONPs, the water vapour barrier properties decreased by approximately 25%. A study was carried out by incorporating MgONPs into a matrix of carboxymethyl chitosan to form a waterproof and antibacterial food packaging material (Wang et al., 2020b). The developed packaging material ~~It~~ showed improved thermal stability, mechanical properties (87.45 % increase of elastic modulus and 171.13 % increase of elongation at break for 1% MgONPs incorporated films), UV shielding ability, water insolubility, and good antimicrobial activity against *L. monocytogenes* and *S. baltica*. Both the films showed superior UV shielding performance and antimicrobial performance when MgONPs were incorporated into packaging films.

Silicon dioxide NPs (SiO_2NPs) are used in several industries such as the food industry, material packing, textile, biomedical applications, paints, inks, and pharmaceutical industry because of their adhesive, catalytic, reinforcing agent, anti-binding, anti-foaming, viscosity controller, and desiccant properties (Guo et al., 2018, Emamverdian et al., 2020). Bi et al. (2020) developed an antioxidant and antimicrobial food packaging material with the incorporation of SiO_2NPs into a chitosan and D-α-tocopheryl polyethylene glycol 1000 succinate matrix. When compared with the control films, the SiO_2NPs film showed the increased tensile strength (from 27.28MPa (control film) to 32.99MPa) and elongation at break (from 20.59% (control film) to 40.53%) while it showed the lowest moisture content, water vapour and oxygen permeability. It also showed enhanced free radical scavenging activity and increased antimicrobial activity. Further, this packaging material was able to increase the oxidative stability of soybean oil during storage. Qiu et al. (2021) also developed a bio-nanocomposite film with the incorporation of SiO_2NPs into poly (3-hydroxybutyrate-co-3-hydroxyhexanoate) packaging matrix through a solvent casting method. The SiO_2NPs were able to accelerate crystallization, increase thermal stability, mechanical properties, and barrier properties. The migration levels of SiO_2NPs were determined by the food stimulant test. The overall migration levels increased with the increased concentration of SiO_2NPs and the acidity of stimulants, however, was much below the specific migration limit of SiO_2NPs which is 60 mg/kg of food.

Zeolite is composed of crystalline metal oxides (eg, Si, P, Al, Ti, B, Ga, Ge, Fe, etc.) consisting of a tetrahedral atom. Although zeolite is available in abundant, its industrial applications are limited because of the impurities and chemical composition diversity. They are currently used in industry as catalysis, adsorption, and ion-exchange agents. Zeolites nanoparticles as promising antidiarrheal agents, antitumor adjuvants, antibacterial agents, and drug carriers (Derakhshankhah et al., 2020). Marzano-Barreda et al. (2021) developed an active food packaging material for fresh broccoli florets by incorporating Zeolite NPs into a polybutylene adipate terephthalate, citric acid, and cassava starch matrix by the blown extrusion method. Zeolite reduced Young's modulus from 19.74MPa to 18.41MPa and enhanced elongation at break from 74.84% to 97.74%, while it did not influence the water vapour permeability. This packaging material reduced the metabolism of fresh broccoli florets for 7 days, keeping the colour and vitamin C content intact. Alp-Erbay et al. (2019) developed Zeolite NPs, silica microparticles, poly (ε- caprolactone) based electrospun films by solution electrospinning with high histamine-binding capacity. The designed films have increased transparency and mechanical properties. The tensile strength of the film increased from 19.6 MPa (control) to 193.3 MPa (zeolite 5 wt %), 195.2 MPa (zeolite 10 wt %) and 203.1 MPa (zeolite 15 wt %). Further, the Zeolite NPs containing films had increased histamine entrapment performance due to the improved porous structure and better adsorption selectivity. This film can thus be utilized as an active-scavenging packaging material to capture heat-stable histamine and other biogenic amines emitted by fish and fishery products. Zeolite NPs were modified and doped with silver (Ag^+) or gold (Au^{+3}) cations and incorporated into a carboxymethyl cellulose matrix to enhance the mechanical, antimicrobial activity, water vapour transmission, and gas transmission rate (Youssef et al., 2019).

Some recent applications of metal oxide bio-nanocomposite films are explained in Table 2.

3.1.3. Other inorganic nanomaterials

Nanoclays is a type of purified clay that is classified into sepiolite (sepiolite and palygorskite), illite (illite and glauconite), smectite (montmorillonite, beidellite), chlorite (chlorite), and kaolinite (kaolinite, halloysite). Nanoclays are utilized in a wide variety of applications in industries including medicine, pharmaceuticals, cosmetics, catalysts, food packaging, and textile (Asdagh & Pirsa, 2020). Additionally, it is useful in environmental protection as it is used as an adsorbent for volatile organic compounds and organic/mineral pollutants. The diverse application attributes for nanoclays to be modified into various changes (Asdagh & Pirsa, 2020). Nanoclays have some important properties such as easy adaptation with different polymers, diffusion in polymer layers, production, and low cost. Hence, it has been highly studied for food packaging application in the past few years.

Lee et al. (2019) developed a montmorillonite nanoclay, agar, and gellan gum-based ternary nanocomposite films by the solution casting method. The film showed improved tensile strength (from 29.9MPa to 44 MPa), thermal stability, and rheological properties, while the water barrier (from 1.90×10^{-9} g/m^2 Pas to 1.7010^{-9} g/m^2 Pas) and contact angle were reduced. A packaging material was developed with organo nanoclay, polycaprolactone, and chitosan through a solvent casting method using glycerol monooleate or oleic acid as plasticizers. Here, organo nanoclay showed antimicrobial activity against *E. coli*, *P. aeruginosa*, and *C. albicans* (yeast).

Asdagh and Pirsa (2020) also developed an intelligent packaging system with the incorporation of montmorillonite nanoclay into the pectin-carum capsicum essential oils-β-carotene matrix. This packaging nanocomposite material had high antibacterial activity, antioxidant activity, flexibility, and firmness. Further, this material increases the oxidative stability of butter during storage, while the packaging system is a color indicator (due to the presence of β-carotene) to detect the oxidation of butter and expiration time. Pires et al. (2018) developed a bio-nanocomposites packaging material based on chitosan and two distinct montmorillonites nanoclay (cloisite®Na$^+$ and cloisite®Ca^{+2}) that were infused with rosemary essential oil or ginger essential oil and evaluated for fresh poultry meat packaging. These packaging materials were able to increase antimicrobial, antioxidant properties and reduce lipid oxidation while increasing the self-life of packed poultry meat.

While Giannakas (2020) developed two nanocomposite films: sodium montmorillonite-essential oils-low density polyethylene (NaMtEO) film and organically modified montmorillonite as thyme, oregano, and basil with low density polyethylene (OrgMt) film. The properties of these two nano clays were evaluated and compared. Both the bio nanocomposite films had increased antioxidant activity, tensile strength, and barrier properties. However, OrgMt based film had higher tensile strength and barrier properties than NaMtEO based film, making OrgMt the most suitable packaging—material. Additionally, recent studies were also performed on modified montmorillonites; silver modified montmorillonite (Makwana et al., 2020), and organically modified montmorillonite (Giannakas, 2020). These nanocomposites were found to have improved mechanical, thermal barrier, and antimicrobial properties.

According to a study carried out by Salarbashi et al. (2018) Cloisite 30B nanoclay when incorporated into soluble soybean polysaccharide increases in tensile strength (12.71MPa to 14.28 MPa), surface roughness, melting temperature (80.6°C to 104.1 °C), and selective high antimicrobial activity. In this study, the migration of nanoclay was evaluated in deionized water and a bread sample. The findings showed that nanoparticles could migrate into deionized water as a food simulant, but they could not migrate into bread as a food model. Further, it was found through the cytotoxicity studies of cloisite 30B nanoclay on intestinal epithelium and heart cells that it has a cytotoxic effect at a 7% cloisite 30B concentration. Since there is a migration of 0.76 ppm Al and 1.23 ppm Si into the epithelial cells layer and 0.08 ppm Al and 0.11 ppm Si migration into heart cells. Therefore, it was recommended by Salarbashi et al. (2018) to only use this packaging material for solid food packaging.

Graphene oxide (GO) is a single-atom thick layer of sp2 bonded carbon atoms with a very large surface area consisting of functional groups such as hydroxyl, epoxide, and carbonyl groups. Due to the unique thermal, mechanical, and electronic properties of GONPs, it is used as a reinforcing nanofiller in bio-nanocomposites (Jamroz et al., 2020). Studies of de Paiva et al. (2020) designed GO-chitosan-based biodegradable bags to extend the self-life of melon. The bags were prepared by making bio-nanocomposite films through a solvent casting method and then sealing two films together. The incorporated GONPs increase tensile strength of films from 0.063MPa (control) to 0.083MPa, decrease water vapor permeability from 0.33 g/m.s.Pa (control) to 0.27 g/m.s.Pa and decreased the microbiological growth. Further, it was an effective packaging material for melon fruits by keeping a better external appearance and prolonging the self-life of fruits. Additional studies on GONPs were performed by Shekhar et al. (2020) on GO incorporated poly (D-glucosamine) matrix. For this study, GO was synthesized by modified Hummer's method and then the film was formed by reinforcing into chitosan by matrix a solution casting method. The film demonstrated an increase in tensile strength, thermal stability, and good electrical conductivity with low resistance.

Some recent applications of other inorganic nanoparticle incorporated bio-nanocomposite films are listed in the Table 3.

3.2. Organic nanomaterials and packaging applications

3.2.1. Nanocellulose

Nanocellulose can be mainly classified into cellulose nanocrystals (CNC), cellulose nanofibers (CNF), and bacterial cellulose. Nanocellulose materials have become a growing field of industry and study due to their abundance, high aspect ratio, mechanical properties, renewability, biocompatibility, and lack of toxicity (Trache et al., 2020). Nano-cellulose is employed in several industries as biomedical products, wood adhesives, electroactive polymers, textiles, food coatings, barrier/separation membranes, antimicrobial films, paper products, cosmetic and nanocomposite materials (Trache et al., 2020). The incorporation of nanocellulose into a polymeric matrix enhances the tensile strength, thus increasing mechanical properties, thermal stability, and decreasing the elasticity (Niu et al., 2018; Noorbakhsh-Soltani et al., 2018; Ramesh & Radhakrishnan, 2019).

Niu et al. (2018) developed polylactic acid- chitosan-based food packaging with the incorporation of cellulose nanofibers. Here a two-layer composite film was developed, out of which the first layer was the cellulose nanofiber modified by rosin and the second layer was a poly-lactic acid matrix coated with chitosan. This film showed enhanced mechanical properties which progressively increased up to 32.3 MPa with 8% cellulose nanofiber concentration while it also showed significant antimicrobial activity against *E. coli* and *B. subtilis*. Studies on bio-nanocomposite films with cellulose nanofibers were performed with the incorporation of chitosan-curcumin (Zhang et al., 2021) and chitosan/tannic acid (Huang et al., 2019) that showed high crystallinity, improved the oxidation resistance, UV blocking properties, and excellent antibacterial activity. In addition, Chen et al. (2020) designed an intelligent pH-sensitive food packaging with the combination of cellulose nanofiber-purple sweet potato anthocyanin-oregano essential oil. This film also had improved antimicrobial activity, barrier performance to ultraviolet and visible light, tensile strength, and elasticity.

3.2.2. Chitosan nanomaterials

Chitosan NPS has become one of the most promising nanomaterials made from polymer matrix due to its distinctive characteristics, biodegradability, nontoxicity, and antimicrobial properties. Thus, chitosan NPs have a wide variety of applications in pharmaceutical and biomedical engineering such as tissue engineering, drug delivery, gene therapy, cancer therapy, biomolecule monitoring, and an antimicrobial agent (Rizeq et al., 2019). The chitosan NPs are valuable nanofillers that are synthesized by the chitosan biopolymer using methods such as tripolyphosphate crosslinking, microemulsion, precipitation, coacervation, reverse micellar, selfassembly, nano spray-drying, supercritical fluids, electrospraying, and emulsification (Garavand et al., 2020). Chitosan NPs are currently used in food packaging applications as demonstrated by Zheng et al. (2018) on starch- Litsea cubeba oil. Here, antioxidant and antimicrobial chitosan - Hardleaf oatchestnut starch-Litsea cubeba oil edible films were designed by the solution casting method. These nanocomposite films have increased tensile strength (from 27.33 MPa to 33.54 MPa), scavenging ability (from 20.67% to 52.34%), antimicrobial activity, and reduced moisture absorption. In addition, the water vapor permeability decreased from 1.531×10^{-11}g $m^{-1}pa^{-1}s^{-1}$ to 1.491×10^{-11}g $m^{-1}pa^{-1}s^{-1}$.

Further, food packaging studies by Shapi'i et al. (2020) on starch films with the incorporation of chitosan NPs synthesized *via* ionic gelation. The developed packaging film had significant antimicrobial activity and also efficiently inhibited microbial growth in cherry tomatoes for up to 10 days. Further, chitosan NPs are more effective in inhibiting gram-positive bacteria in comparison to gram-negative bacteria. The antimicrobial potency of chitosan NPs was also visible in the studies of Cui et al. (2020) on chitosan NPs-zein- pomegranate peel extract. Pomegranate peel extract was encapsulated into chitosan NPs by an ionic gelation method and then incorporated into the zein matrix. Further, cold nitrogen plasma was used to modify the surface of the film to provide a uniform release of pomegranate polyphenols. The film showed better thermal properties and reduced growth of *L. monocytogenes* bacteria during pork storage. The plasma treatment effectively maintained the prolonged release of pomegranate polyphenols, resulting in high antibacterial action against *L. monocytogenes* throughout storage.

Vahedikia et al. (2019) also formed chitosan NPs incorporated zein and cinnamon essential oil film for food packaging applications. Here the chitosan NPs were formed by the ionotropic gelation method and the films were prepared by the casting method. The incorporation of the chitosan NPs in these films showed a significant increase in the tensile strength from 0.95 MPa (control sample) to 2.15 MPa. While an inhibitory area was not detected in the chitosan NPs incorporated film in both *E. coli* and *S.aureus*. Although chitosan NPs have antimicrobial properties, the concentration of NPs used in film creation is insufficient to form sufficient physical connections with the bacterial cell wall to provide antibacterial effects.

Recent advancements in bio-nanocomposite materials were made using chitosan nanofiber as the nanofiller. Lin et al. (2018) studied the bio-nanocomposite film chitosan nanofiber -ε-polylysine which was prepared by using an electrospinning apparatus. Increased moisture content and water solubility were observed in this film. Additionally, it successfully inhibits *S. typhimurium* and *S. enteritidis* microbial growth in chicken and acted as an antimicrobial packaging material. Alizadeh-Sani and colleagues developed two intelligent pH-sensitive packaging systems; ~~C~~chitosan nanofibers- methyl cellulose- Saffron petal anthocyanin (Alizadeh-Sani, et al., 2021a) and ~~C~~chitosan nanofibers- methylcellulose- barberry anthocyanins (Alizadeh-Sani, et al., 2021b). For these studies, anthocyanin was extracted from saffron petal and barberry respectively then the anthocyanin contents (cyanidin-3-glucoside based) were calculated via a pH differential method. Anthocyanin-chitosan nanofiber-methylcellulose bio-nanocomposite films were then prepared by the solution casting method. Increased mechanical, water barrier properties, antimicrobial activity, antioxidant activity, and UV-vis barrier properties were observed in these films while prolonging the shelf-life of meat and seafood products.

3.2.3. Starch NPs

Starch NPs are novel material that has different physicochemical and biological properties when compared to starch biopolymers. The starch NPs are biocompatible, biodegradable, cost-effective, renewable, and non-toxicity. Further, they have a high absorptive capacity, solubility, reaction surface, and biological penetration rate. The starch NPs can be formed through enzymatic, chemical, or physical methods by using the methods such as enzyme debranching, milling, sonication, thermosonication, nanoprecipitation, ultrasound, and non-thermal plasma (Campelo et al., 2020).

A few studies have been conducted with the incorporation of starch NPs into a biopolymer matrix. Oliveira et al. (2018) designed a packaging film with the incorporation of starch nanocrystals into a mango kernel starch matrix. Here starch nanocrystal increased the tensile strength by 90% and youngs modulus by 120% when compared to the control film. Furthermore, the water vapor permeability was reduced by 15%. In a different study on starch NPs were conducted by Condes et al. (2018), where starch nanocrystals and starch granules were incorporated into a matrix amaranth protein isolate. With the incorporation of nanocrystals, Young's modulus and tensile strength enhanced, while the elongation at break reduced. With the addition of starch nanocrystal, the solubility of films decreased from 80% to 40% and reduced the water vapour permeability of the film.

Recent applications of organic bio-nanocomposite films are highlighted in Table 4.

4. Nanohybrids

Nanohybrid nanocomposite films are materials that are formed with the combinations of inorganic nanomaterials (metal and metal oxides) and organic nanomaterials. Nanohybrids can overcome individual demerits of the nanoparticles. Hybrid nanoparticles are synthesized using various approaches such as microwave irradiation, laser ablation, electrochemical method, electrospinning, mechanical method, chemical method, and biological methods (Oun et al., 2020).

Research has been performed with the combination of inorganic-inorganic, organic-organic, and organic-inorganic nanomaterial for improved performance and quality of the bio-nanocomposite films (Abolghasemi-Fakhri et al., 2019; Salmas et al., 2020; Vizzini et al., 2020; Yeasmin et al., 2020).

When discussing inorganic-inorganic nanohybrids in recent years, ZnNPs have been combined with MgONPs (Vizzini et al., 2020) and ZnONPs (Mellinas et al., 2020) to form bio-nanocomposite films. Vizzini et al. (2020) designed an active packaging film with the incorporation of Zn-MgO NPs and Alginate to control the *L. monocytogenes* contamination in cold-smoked salmon. The developed film showed no bacterial growth in salmon at 4°C for 4 days. The cytotoxicity of Zn-MgO NPs was assessed using human macrophage-like cells U937 and differentiated human promyelocytic leukemia cell line HL-60. According to the cytotoxicity analysis, Zn-MgO at concentrations of less than 1 mg/mL could be safely used to form active packaging. While Mellinas et al. (2020) developed a packaging film with ZnO-ZnNPs and cocoa bean shell extract incorporation into a Pectin. At this point, microwave-assisted extraction was used to extract cocoa bean shell extract (polyphenols) from cocoa bean shell wastes and the ZnO-ZnNPs were synthesized by a microwave heating method. Finally, the film was prepared by combining the solution using a casting technique. The film showed increased thermal properties, oxidative stability, UV barrier properties reached 98%, a decrease in oxygen transmission rate up to 50%, and photodegradation efficacy of 90% after 60 min. The combination of AgNPs with MgONPs-Poly butylene adipate-co-–terephthalate-based ternary bio-nanocomposite film was investigated (Zhang et al., 2020). MgO-Ag NPs were synthesized by stirring at room temperature for 24 hours and then combined with Poly butylene adipate-co-terephthalate to form the film through a solvent casting method. The mechanical, water barrier and oxygen barrier properties of the film were increased. Whereas Yeasmin et al. (2020) developed a nanohybrid film from cellulose nanofibrils - montmorillonite clay and pullulan through a solution casting method. The composite was degraded with increased burial time in soil due to the presence of pullan, however, montmorillonite clay slowed the degradation process. While retaining transparency, the film increased tensile strength, thermal stability, water barrier properties, and reduced moisture susceptibility.

Limited research on organic-organic nanohybrids research has been carried out by the scientists on nanocrystalline cellulose- chitin nano whiskers- poly lactic acid (Xu et al., 2020),. Xu et al. (2020) developed a packaging film using the poly lactic acid matrix and nanocrystalline cellulose and chitin whiskers nanofillers for packaging application through the melt extrusion method. This film possessed enhanced properties such as enhanced thermal, mechanical, and barrier properties.

Most of the recent studies on nanohybrids-based bio-nanocomposite films for food packaging were performed with the combination of inorganic-organic nanomaterials. The studies of He et al. (2021) focus on a novel paper coating for strawberry packaging application was produced using carboxymethyl cellulose and varying concentrations of cellulose nanocrystals immobilized AgNPs. With increasing cellulose nanocrystals-AgNPs content, the tensile strength increased 1.26 times, while the water vapor decreased 45.4 %. There was a 93.3 % reduction of air barrier properties, and the antimicrobial properties of coated paper improved. Further, the quality of packed strawberries increased, and the shelf-life extended up to 7 days. While,

An increase in antimicrobial activity was observed when CuONPs were combined with cellulose nano whisker (Saravanakumar et al., 2020) and chitosan nanofibers (Almasi et al., 2018). Saravanakumar et al. (2020) designed a packaging material for fresh-cut pepper packaging. This biopolymeric film was designed by the incorporation of the two NPs CuONPs and cellulose nanowhiskers. These two NPs had different applications in this packaging material were, CuONPs prevented the microbial contamination of the food product and cellulose nano whiskers enhances the barrier properties of the material. The resulted film showed antimicrobial activity against *S. aureus, E. coli*, *Salmonella sp.*, *C. albicans,* and *Trichoderma spp*. Further, this film showed increased antioxidant activity and prevention of microbial growth in fresh-cut pepper for 7 days.

As observed by all the above-mentioned studies (as depicted in Table 5) most of the nanohybrid bio-nanocomposite films had superior properties which are beneficial for food packaging applications.

5. Migration of nanoparticles into food products from bio-nanocomposite films

There are many merits of active packaging when compared to the traditional packaging material. However, one of the main challenges faced by NP incorporated active packaging is the migration of NPs into the packaged food product (Enescu et al., 2019). The NPs on the surface of the packaging material is mostly not harmful to humans and does not cause environmental pollution. Nonetheless, if it migrates into the food, it may cause human health problems. The process of migration begins with the transfer of materials from one location to another during usage, storage, or disposal. Migration of the NPs to food can be caused because of diffusion, dissolution, and abrasion of the packaging surface (Garcia et al., 2018; Nile et al., 2020). In bio-nanocomposites, NPs diffuse, desorb, dissipate, or are transferred from the composite materials into the food; this is accomplished by matrix decomposition (Azizi-Lalabadi et al., 2021). The release of NPs from the surface and the oxidative dissolution of NPs are the two steps of this migration process (Ahari & Lahijani, 2021)

The migration of NPs to the food product is dependent upon several factors such as properties of NPs (e.g., particle size, molecular weight, solubility, and diffusivity in the polymer), environmental conditions (temperature, mechanical stress), food conditions (pH value, composition), packaging characteristics (polymer structure and viscosity, position of the NPs); interaction between the NPs and the material and contact time. When humans consume the food product, the migrant NPs enter the human body, causing toxicity. The main reason for toxicity is its persistent, non-dissolvable, and non-degradable nature. NPs can easily cross membrane barriers and capillaries, leading to different toxicokinetic and toxicodynamic properties. NPs may prompt toxicity in the human gastrointestinal tract, liver, kidneys, and immune system (Enescu et al., 2019; Nile et al., 2020).

The toxicity of the migrant material may be different based on the surface morphology, composition, charge, and ~~the~~ chemistry of the NPs. The toxicity increases with the increased size of the metal NPs. The risks associated with nanotechnology in food packaging warrant better understanding, consumer awareness, government guidelines, policies, and detection methods (Garcia et al., 2018; Nile et al., 2020). Based on EU regulations for food-related applications of nanotechnology including bio-nanocomposites, special considerations have been declared for a series of tests and requirements. The migration tests are performed on food stimulants such as Ethanol (10, 20, 50 % v/v), Acetic acid (3 % w/v), vegetable oil, or food products such as fresh/frozen fruits and vegetables (unpeeled and uncut or peeled and/or cut). The migration tests are performed at different temperatures and different periods up to 30days. The methods used to determine the migration levels of NPs should be sensitive to very low concentrations of NPs, selective and they should be accurate. atomic emission (ICP-AES), inductively coupled plasma mass (ICP-MS) and optical emission spectrometry (ICP-OES) are techniques used to determine the migration of NPs.

There are specific migration levels for each substance or group of substances which are expressed as mg substance per kg food. In some of the substances, the migration levels are not detectable, while for other substances no migration is permitted (EuropeanCommission, 2011a). Some nanoparticles are not permitted for usage in the European Union due to a lack of knowledge on their toxicity. The standard regulations included some suggestions about the release and migration of NPs from nanoproducts, but they were not particularly successful due to the lack of a specific mechanism for recognizing migrated NPs. As a result, the standard regulations have concentrated on nanomaterial labeling. Thus, International Standards Organization (ISO) has updated guidelines (ISO TC 229) on nano product labeling to ensure that consumers are informed of the items they purchase. However, numerous researches showed that the majority of NPs incorporated into polymers, tend to agglomerate and remain firmly fixed in the polymeric matrix, and hence do not migrate (Garcia et al., 2018).

As explained in detail in the above sections, migration studies were performed in food stimulants or food products for AgNPs (Motlagh et al., 2021; Ramos et al., 2020), ZnONPs (Bumbudsanpharoke et al., 2019), TiO_2NPs (Chen et al., 2019), SiO_2NPs (Qiu et al., 2021) and Cloisite 30B nanoclay (Salarbashi et al., 2018). Most of the studies conducted on migration is focused on AgNPs and synthetic polymers, and the studies are limited for other NPs (Garcia et al., 2018).

6. Legislation associated with bio-nanocomposites in food packaging.

Regulations and legislation play a key role in the food, human and environmental safety of nanomaterial-based products when industrializing them and making the bio-nanocomposite based packaging materials available to the public. They play as official sources and references for public knowledge and awareness regarding the bio-nanocomposite food packaging films. The Food and

Agriculture Organization of the United Nations (FAO), World Health Organization (WHO), Europe Union (EU), Food and Drug Administration (FDA) of the USA, and many other global, government, and non- governmental organizations have put forward nanotechnology-related regulations and legislation with the food industry. Although, regulations and legislation directly related to bio--nanocomposite-based food packaging are very limited. The safety and inertness for all Food Contact Materials (FCMs) were put forward in 2004 in Commission Regulation (EC) No 1935/2004 (EuropeanCommission, 2004). The food contact material is required not to release constitutes to food products that are harmful to health and not change the composition, taste, odor in an unacceptable manner. In addition, the EU put forward a regulation on active and intelligent materials and articles intended to come into contact with food on Commission Regulation (EC) No 450/2009 (EuropeanCommission, 2009). It stated that it's essential a case-by-case analysis on packaging materials containing nanomaterials. The EU regulations on plastic materials and articles intended to come into contact with foodstuffs; Commission Regulation (EC) No 975/2009 (EuropeanCommission, 2009), Commission Regulation (EU) No 1282/2011 (EuropeanCommission, 2011b) and Commission Regulation (EU) No 202/2014 (EuropeanCommission, 2014) is not directly related to bio-nanocomposite based food packaging. However, it contains details on migration levels and that nanomaterial must be only used if stated.

Commission Regulation (EU) No 10/2011 has explained the migration limits of certain chemical substances, or for a group of substances and also expressed the migration test in detail (EuropeanCommission, 2011a). The detection limit for the chemical is 0.01 mg per kilogram of food or food simulant. This regulation has been amended at different stages in Commission Regulation (EU) 2016/1416 (EuropeanCommission, 2016) and Commission Regulation (EU) 2020/1245 (EuropeanCommission, 2020). Specific migration limits according to these regulations for some of the chemical substances are as below; Cu =5 mg/kg food or food simulant, Zn = 5 mg/kg food or food simulant, and Mg = 0.6 mg/kg food or food simulant. Further, the authority has confirmed that SiO_2NPs don't have any safety concerns when the aggregates are larger than 100 nm and when no migration is detected. ZnONPs do not migrate in nanoform from polyolefins or unplasticized polymers, according to authority and the specific migration level is 0.05 mg/kg (EuropeanCommission, 2016). In the opinion of the authority, montmorillonite clay modified with hexadecyltrimethylammonium bromide does not raise any safety concerns when used as an additive in plastic food contact materials, and no migration is observed when the nanoparticle range < 100nm (EuropeanCommission, 2020). The authority concluded that TiO_2NPs, when used as an additive at up to 25.0 % w/w in all polymer types in contact with all food types during any time and temperature circumstances at a NP range of less than 100nm, pose no risk to consumers (EuropeanCommission, 2020).

7. SWOT analysis of bio-nanocomposites in food packaging

7.1. Strengths

As discussed throughout the review, bio-nanocomposites have many strengths that are essential for food packaging. Most importantly, bio-nanocomposites are biodegradable and renewable, thus environmentally friendly. Bio-nanocomposites are novel, low-weight, high-performance materials which can be used as alternatives for plastic-based packaging materials. They consist of many beneficial properties such as increased mechanical, thermal, and barrier properties. Further, bio-nanocomposites tend to have high antimicrobial properties, oxygen scavenging properties, and ethylene absorption activity. These properties are essential in increasing self-life, reducing microbial growth, increasing the quality, and safety of food products during storage and transport.

In addition, sensor bio-nanocomposites have been produced using intelligent packaging to detect food spoilage/quality during storage.

7.2. Weaknesses

Bio-nanocomposite-based packaging materials are novel. Research must be conducted to make them industrial-level global food packaging materials. They are not as cheap and readily available as the currently used traditional packaging materials. The up-scale production and processing methods are yet to be confirmed for bio-nanocomposites. A bio-nanocomposite based food packaging system is yet to be developed which can be used as a universal packaging material for all types of food products. There is limited research carried out on bio-nanocomposutes thus it has limited industrial applications. The developed bio-nanocomposite based food packaging materials are specific to the food type whereas plastic can be used in all food packaging applications. Finally, there are insufficient regulations and legislation when considering bio-nanocomposites in food packaging.

7.3. Opportunities

The use of bio-nanocomposite creates an opportunity to develop biodegradable, sustainable materials and eco-friendly to replace non-biodegradable food packaging materials such as polyethylene terephthalate, polyvinyl chloride, polystyrene, and polymethyl methacrylate. The use of bio-nanocomposite films provides an opportunity to improve active and smart packaging to create the most suitable packaging materials for food products.

7.4. Threats

The risk of bio-nanocomposite-based food packaging provides a unique challenge for food safety. It is a concern that the nanomaterial incorporated in bio- nanocomposite film may migrate into the food or drink which is in contact with the material. This may pose a risk to human health, animals, and the environment. Nanomaterials enter the body in a variety of ways and can cause damage to human cells by altering mitochondrial function, producing active oxygen, increasing membrane permeability, causing toxic effects leading to chronic diseases such as allergies, asthma, various inflammations, cardiovascular disease, and cancer. Furthermore, working with nanomaterials results in the release of nanoparticles into the environment, resulting in environmental pollution.

The migration levels and the cytotoxicity effects of nanomaterials are still not fully understood, since the use of bio-nanocomposite as a food packaging material is a novel area. The future it holds is unforeseen and unpredictable, just like plastic when it was invented decades ago.

8. Future perspectives and conclusion

The use of different types of packaging materials has evolved through history and bio-nano composite materials are the most suitable materials that are developed with essential characteristics such as biocompatibility, antimicrobial activity, biodegradability, mechanical, optical, and barrier properties. The use of nanoparticles in food packaging is an immense challenge due to the migration of nanoparticles into food products. This may ultimately lead to the cytotoxicity of human cells during consuming food products or being in contact with this. Thus, it is extremely important to study the migration and the cytotoxicity effect of the NPs before the industrialization of the packaging materials. However, there is a lack of studies on the impact of NP on human health, and the migration of NPs into food warrants further studies. The biodegradation of bio-nanocomposite-based food packaging materials and the environmental impacts caused by migration into soil and water are also fields that are under investigations. Furthermore, the development and categorization of the most suitable packaging material for different food types are not established. Also, the combination of active and intelligent packaging materials using NPs are some upcoming perspectives in this field of study. With the combination of active and intelligent packaging, the packaging material used especially for long transportation and large packaging can be taken to the next level by inserting sensory robotics into the bio-nanocomposite material. These sensors can be directly linked to a computer system to observe the changes of material such as antimicrobial, chemical, or thermal properties.

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Table 1: Some recent applications of metal-based nanomaterials in bio-nanocomposite films

Nanomateria	Packaging metrics	Food product	Properties of bio-nanocomposite films	Reference
Silver (Ag) NPs				
Ag NPs	chitosan/gelatin	None	• Improved the tensile strength and water vapor resistance. • High antibacterial activity against *S. aureus* and *E. coli*	Cao et al. (2020)
	Poly (lactic acid) - Thymol	None	• Increased antioxidant properties and antibacterial activity against *Escherichia coli* and *Staphylococcus aureus*	Ramos et al. (2020)
	Konjac glucomannan-poly(ε-caprolactone)	None	• Increased thermal stability, elongation at break and relatively hydrophobic • Excellent antibacterial activities against *S. aureus* and *E. coli*	Lin et al. (2020b)
Chitosan mediated Ag NPs	Poly lactic acid	None	• Increased mechanical and thermal properties • Excellent antibacterial activities against *S. aureus* and *E. coli*	Sonseca et al. (2020)
Copper NPs (Cu-NPs)				
Cu-NPs	Low-density polyethylene	Peda (Indian sweet dairy product)	• Improved mechanical properties and decreased water vapour permeability • Excellent antimicrobial activity against *S. Aureus* and *E. coli*	Lomate et al. (2018)
CuS NPs	Carrageenan	Beef	• High transparency, enhanced mechanical properties and thermal stability • Antibacterial activity against *E. coli* (99.2%) and *S. aureus* (99.9%) • Photothermal effect can inhibit the growth of bacteria on the packaged beef product	Li et al. (2020)
CuO NPs	sodium alginate-cellulose nano whisker	Cut pepper	• Antibacterial activity against with high zone of inhibition against *S. aureus* (27.49 ± 0.91 mm), *E. coli* • (12.12 ± 0.58 mm), *Salmonella sp.* (25.21 ± 1.05 mm), *C. albicans* (23.35 ± 0.45 mm) and *Trichoderma spp.*(5.31 ± 1.16 mm). • Enhanced antioxidant activity • Prevent microbial contamination in fresh cut pepper	Saravanakumar et al. (2020)

Gold nanoparticles (AuNPs)				
AuNPs	Lignocellulose fiber	None	• Enhanced radical scavenging activity thus showing a boost of antioxidant activity	Bumbudsanpharoke and Ko (2018)
Sulphur nanoparticles (SNPs)				
SNPs	Alginate	None	• The tensile and water vapor barrier characteristics both improved by 12 % and 41%, respectively. • UV barrier properties increased by 99%. • The antimicrobial activity of *E. coli* was 60% while a complete bactericidal effect was observed for *L. monocytogenes* during a 12 hour time period.	Priyadarshi et al. (2021)
SNPs	Chitosan	None	• Enhanced hydrophobicity, mechanical strength, and water vapor barrier property • Antimicrobial activity against *E. coli* and *L. monocytogenes*	Shankar and Rhim (2018)

Table 2: Some recent applications of metal oxide based nanomaterials in bio-nanocomposite films

Nanomaterial	**Packaging metrics**	**Food Product**	**Properties of bio-nanocomposite film**	**Reference**
Zinc oxide (ZnO) NPs				
ZnO NPs	Chitosan - cellulose acetate phthalate	Black grapes	• Increased thermal stability and barrier properties, low surface wettability and high contact angle • Extended the shelf-life of black grape fruits • Strong antimicrobial activity against *S. aureus* and *E. coli*	Indumathi et al. (2019)
	Chitosan-guar gum-Roselle calyx extract	Ras cheese	• Improved mechanical, permeability, antimicrobial and antioxidant properties • Ras cheese coating with bio-nanocomposite film protects surface yeasts, molds and other bacteria growth for three months	El-Sayed et al. (2020)
	Chitosan-gelatin	None	• Improved thermal stability, elongation-at-break, and compactness properties • Significant antimicrobial activity against *E. coli*	Kumar et al. (2020)
	Chitosan-potato protein-linseed oil	Raw meat	• Improved transparency, tensile strength, elasticity and moisture barrier capability • Raw meat samples showed excellent acceptable sensory properties during 7 days storage while reducing the speed of increasing pH and total bacterial counts	Wang et al. (2020a)
	Agar	Green grape	• Improved thermal stability, elongation and film thickness, whereas tensile strength and transparency decreased • Grapes packaged in composite films showed fresh appearance up to 14 and 21 days	Kumar et al. (2019)
	Agar- nisin-cinnamon essential oil	Minced fish	• Increased antimicrobial activity against; L. monocytogenes, *S. aureus* than *E. coli* and *P. aeruginosa* • Film effectiveness against *L. monocytogenes* is dependent on *L. monocytogenes* (seven strains) strains	Abdollahzadeh et al. (2018)
	Gellan- xanthan gum	None	• Increased tensile strength, thermal stability, glass transition temperature, ultra-violet light shielding and decreased the water vapor permeability	Rukmanikrishnan et al. (2020)

	Polylactic acid	none	• Increased mechanical properties, UV and visible light barrier performances • Strong antibacterial activity shown by inhibition zones against *E. coli* and *S. aureus*	Zhang et al. (2021b)
	Polylactic acid	minced fish paste	• Increased thickness, tensile strength, and water vapor barrier, UV-light barrier property and decrease in the transparency • Strong antibacterial activity against *E. coli* and *L. monocytogenes* • Showed strong antibacterial function in minced fish paste	Shankar et al. (2018)
Aluminum-doped ZnO NPs	Polylactic acid	none	• Uniform coverage, high visible transparency and strong antibacterial activity against *E. coli*	Valerini et al. (2018)
Titanium oxide (TiO_2) NPs				
TiO_2 NPs	Chitosan	tomato	• Better tensile Strength, barrier properties and ethylene photodegradation ability	Kaewklin et al. (2018)
	Chitosan-*Cymbopogon citratus* essential oil	minced meat	• Increased WVP, improved mechanical properties and decreased solubility in water • Prolonged minced meat shelf-life by suppressing the growth of total bacteria, *Enterobacteriaceae*, *psychrotrophic* bacteria, *S. aureus*, and lactic acid bacteria and also decreasing TVB-N value	Hosseinzadeh et al. (2020)
	Gelatin-grapefruit seed extract	none	• Increased surface roughness, mechanical strength, water contact angle and complete prevention of UV light transmission • Showed some antioxidant activity and strong antibacterial activity against *E. coli* and *L. monocytogenes*	Riahi et al. (2021)
	Chitosan-anthocyanin-rich black plum peel extract	none	• Increased mechanical properties higher barrier properties (water vapor and UV–vis light) • Strong free radical scavenging, high ethylene scavenging ability and antimicrobial activity • Films were pH-sensitive due to anthocyanins	Zhang et al. (2019b)
	Carboxymethyl cellulose - chitosan	green bell pepper	• Increased thermal stability, tensile strength, Young's modulus, UV-barrier properties, antimicrobial activity and reduced water vapor permeability • Shelf-life studies on green bell pepper showed excellent resistance to mass loss and spoilage during storage	Salama and Aziz (2020)
	Starch - pectin	none	• Increased thermal stability, mechanical, UV barrier and moisture barrier properties	Dash et al. (2019)

	Agar-κ-carrageenan–anthocyanin	pork	- Enhanced the mechanical properties, colour stability - UV–vis light barrier property, pH sensitivity, and physical properties - Exhibited visual colour changes in the buffer solution (pH 2.0–12.0), ammonia vapour (80 M), and pork spoilage trials.	Liu et al. (2020)
Magnesium Oxide (MgO) NPs				
MgO NPs	Polylactic acid	none	- Improved tensile strength and oxygen barrier properties - Decreased vapor barrier properties - Exhibit superior antibacterial efficacy against *E. coli*	Swaroop and Shukla (2018)
MgO NPs	Carboxymethyl chitosan	none	- Improved thermal stability, UV shielding ability, and water-insolubility - Excellent antimicrobial activity against *L. monocytogenes* and *S. baltica*	Wang et al. (2020b)
Silicon dioxide (SiO_2) NPs				
SiO_2 NPs	Chitosan, D-α-tocopheryl polyethylene glycol 1000 succinate	soybean oil	- High tensile strength, elongation at break, low moisture content, water vapor and oxygen permeability - Strong free radical scavenging activity and high antimicrobial activity - Increase in oxidative stability of soybean oil	Bi et al. (2020)
SiO_2NPs	Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate)	none	- Increased thermal stability, mechanical properties and barrier properties - Accelerate crystallization	Qiu et al. (2021)
Zeolite				
Zeolite	Poly(ε-caprolactone)	fish and fishery products	- Transparent films with reduced porosity, improved mechanical strength and high histamine-binding capacity - High antimicrobial activity against *S. aureus* - Acts as an active-scavenging packaging materials for fish and fishery products	Alp-Erbay et al. (2019)
zeolite	Poly (butylene adipate-co-terephthalate)-glycerol- citric acid-starch	broccoli florets	- Increased elongation at break, decreased Young’s modulus, and unaltered tensile strength - Reduce fresh broccoli florets metabolism preserving the color, and vitamin C content for 7 days	Marzano-Barreda et al. (2021)

zeolite doped with silver (Ag+) or gold (Au+3) cations	Carboxymethyl cellulose	none	• Increased gas transmission rate, water vapor transmission, mechanical and antimicrobial properties	Youssef et al. (2019)

Table 3: Some recent applications of other inorganic nanoparticle incorporated bio-nanocomposite films

Nanomaterials	**Packaging metrics**	**Food product**	**Properties of bio-nanocomposite films**	**Reference**
Nonoclay (Montmorillonite(MTT), Cloisite Na+, Cloisite 30B, Cloisite 20A)				
Nanoclay (montmorillonite)	Pectin/ Carum copticum Essential oils/β-Carotene	Butter	• Flexible, firm and high antioxidant activity • High antimicrobial activity against *B. cereus* than *E. coli* • Low microbial load, high oxidative stability and low color change during butter packaging • Smart color indicator to detect the butter oxidation and expiration time	Asdagh and Pirsa (2020)
montmorillonites (MMT´s) (Cloisite®Na+ and Cloisite®Ca+2)	Chitosan - rosemary essential oil or ginger essential oil	fresh poultry meat	• Extend shelf-life of the fresh poultry meat by reducing lipid oxidation and microbiological contamination	Pires et al. (2018)
Cloisite 30B	Soluble soy bean polysaccharide	none	• Increase tensile strength, melting temperature surface roughness and decreased elongation at break • Inhibit growth of *S. typhi*, *Staphylococcu*s, and *Listeriamonocytogenes*	Salarbashi et al. (2018)
Montmorillonite	Agar/gellan gum	none	• Enhanced thermal stability, tensile strength, and rheological properties	Lee et al. (2019)
Silver modified montmorillonite	Agar-carboxymethyl cellulose	none	• Exhibited a great antibacterial activity against *B. subtilis* and *E. coli*	Makwana et al. (2020)
Graphene oxide nanoparticles (GONPs)				
GONPs	Chitosan	Melon	• Increased tensile strength, Young's modulus decrease in water vapor permeability and microbiological growth • Prolong shelf-life of melon fruits while keeping good external appearance	Paiva et al. (2020)
	Poly (D-glucosamine)	None	• Increased tensile strength, thermal stability and good electrical conductivity with low resistance	Shekhar et al. (2020)

Table 4: Some recent applications of organic nanoparticle incorporated bio-nanocomposite films

Nanomaterials	**Packaging metrics**	**Food product**	**Properties of bio-nanocomposite films**	**Reference**
Nano-cellulose				
Nano-cellulose	Chitosan-Gelatine-Starch	none	• Increased tensile strength and decrease elongation at break • Good thermal stability up to 100 °C and low air permeability	Noorbakhsh-Soltani et al. (2018)
	Chitosan-Polylactic acid-Rosin	none	• Increased hydrophilicity and mechanical properties • Exhibited excellent antimicrobial performance against *E. coli* and *B. subtilis*	Niu et al. (2018)
Cellulose Nanofiber	Chitosan-Curcumin	none	• Increased crystallinity, oxidation resistance, UV blocking properties, and antibacterial activity • Slight decrease in the water vapor barrier properties and mechanical strength	Zhang et al. (2021)
	Purple sweet potato anthocyanin-oregano essential oil	none	• Increased barrier performance, tensile strength, elongation at break, and elastic modulus • Excellent antimicrobial activity and the inhibition rate against *E. coli* and *L. monocytogenes* • pH sensitive packaging material	Chen et al. (2020)
	Chitosan-tannic acid	none	• Increased hydrophilicity and mechanical properties • High antibacterial activity against *E. coli* and *S. aureus*	Huang et al. (2019)
Chitosan nanoparticles (CSNPs)				
CSNPs	Hardleaf oatchestnut starch-Litsea cubeba oil	None	• The tensile strength and scavenging ability of films increased • Water vapor permeability and moisture absorption decreased • Antimicrobial activity increased significantly against *S.aureus* and *E. coli*	Zheng et al. (2018)
	Starch	Cherry tomatoes	• Antimicrobial activity against *B. cereus, S. aureus, E. coli* and *S. typhimurium* • Inhibit the microbial growth in cherry tomatoes during packaging	Shapi'i et al. (2020)

	Zein-pomegranate peel extract	Pork	• Increased thermal stability and mechanical properties. Antimicrobial against *L. monocytogenes* in pork sample	Cui et al. (2020)
Chitosan nanofiber	ε-polylysine	Chicken	• Enhanced moisture content, water solubility and reduced transparency • Successful inhibit *S. typhimurium* and *S. enteritidis* in chicken	Lin et al. (2018)
	Methyl cellulose-Saffron petal anthocyanin	Lamb	• Increased tensile strength, light screening properties • Increased antimicrobial activity against *E. coli* & *S. aureus* , and antioxidant activity • Smart packaging materials for monitoring changes in the freshness of lamb during storage	Alizadeh-Sani et al. (2021a)
	Methylcellulose-barberry anthocyanins	meat and seafood products	• Increased mechanical, water barrier properties and reduced UV–vis light transmittance • pH-sensing color indicator film for real-time freshness monitoring of meat and seafood products	Alizadeh-Sani et al. (2021b)
Starch NPs				
Starch nanocrystals	Mango kernel starch	None	• Increased the tensile strength by 90% and youngs modulus by 120% • Reduced water vapor permeability by 15%	Oliveira et al. (2018)
Starch nanocrystals	Amaranth protein isolate	None	• Increased Young's modulus and tensile strength enhanced • Reduced elongation at break reduced • Decreased water solubility from 80% to 40%	Condes et al. (2018)
β-Carotene loaded starch nanocrystals	Chitosan-gelatin	None	• Showed radical scavenging activity of 1.5 ± 0.3% • sustained release (≈51.5 ± 0.7%) of β-Carotene for 12 days	Hari et al. (2018)

Table 5: Some recent applications of nanohybrids in bio-nanocomposite films

Nanomaterials	Packaging metrics	Food product	Properties of bio-nanocomposite films	Reference
Inorganic-Inorganic nanohybrids				
Zn NPs-MgO NPs	Alginate	Smoked salmon	• No *L. monocytogenes* growth in packed Cold-smoked salmon for 4 days	Vizzini et al. (2020)
Zn NPs-ZnO NPs	Pectin- Cocoa Bean Shell Waste Extract	None	• increased thermal properties, UV barrier properties oxidative stability and decrease in oxygen transmission rate	Mellinas et al. (2020)
Ag NPs- MgO NPs	Poly (butylene adipate-co-terephthalate)	None	• Increased mechanical, water barrier property, and oxygen barrier property	Zhang et al. (2020)
Ag NPs-TiO_2 NPs	Fish gelatin-chitosan	None	• Significantly increase in water solubility • High antibacterial activity	Lin et al. (2020a)
Au NPs-Ag NPs	Cellulose		• Strong antimicrobial activity against *E. coli*	Tsai et al. (2017)
Organic-Organic nanohybrids				
Nanocrystalline cellulose- chitin and whiskers	Poly lactic acid	None	• Increase tensile strength, mechanical and barrier properties • Reduced oxygen transmission.	Xu et al. (2020)
Inorganic-Organic nanohybrids				
Ag NPs-nanocellulose	Grape seed extract	None	• Increased mechanical properties, low water vapor permeability, low oxygen permeability and strong antioxidant activity • Good antimicrobial activity against *E. coli* and *S. aureus*	Wu et al. (2019)
Ag NPs- cellulose nanocrystals	Carboxymethyl cellulose	Strawberries	• Increased mechanical strength, water vapor and air barrier properties • Antibacterial activities against *E.coli* and *S.aureus* compared with uncoated paper • Maintain better strawberries quality and extend the shelf-life up to 7 days	He et al. (2021)

CuO NPs- cellulose nano whisker	Sodium alginate	Fresh cut pepper	• Promising antibacterial activity against *S. aureus*, *E. coli*, *Salmonella sp*., *C. albicans* and *Trichoderma spp*. • Increased antioxidant activity • Prevent the microbial contamination in fresh cut pepper	Saravanakumar et al. (2020)
CuO NPs - chitosan nanofibers	Bacterial cellulose	None	• Considerable release controlling ability and increased antimicrobial activity	Almasi et al. (2018)
Cellulose nanofibrils-montmorillonite	Pullulan	None	• Improved tensile strength, thermal stability and, water barrier properties • Decrease moisture susceptibility.	Yeasmin et al. (2020)