CHAPTER

Nanomaterials at the forefront of advances in active and intelligent food packaging

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24.1 Introduction: Definitions and technologic gap

Food packaging has an essential function in the conservation and maintenance of food quality, mainly involving containment and protection functions. Packaging technology has been modifying paradigms to provide product-consumer information (Cheng et al., 2022). Initially, food quality is perceived by some sensory indicators (color, flavor, and texture), but in general, it is difficult to evaluate food quality after the packaging process (Ghaani et al., 2016). In this context, advanced food packaging technology is necessary as it facilitates information on packaged foods to consumers, as well as contributing to the decrease of food waste, spoilage, recalls, and outbreaks of foodborne diseases (Sobhan et al., 2021; Vilela et al., 2018). In addition, innovative packaging with enhanced functionality is also needed to accommodate consumer trends, driven by population growth and the simultaneous increase in demand for healthier foods, with the presence of bioactive compounds, greater digestibility, and nutrient supply, among others (Vilela et al., 2018; Schaefer and Cheung, 2018). The market demand for fresh, high-quality, minimally processed foods with an extended shelf life has created the urgency of modernized packaging technology (Soltani Firouz et al., 2021). In this sense, two main categories of packaging were developed: (1) Active packaging and (2) Smart packaging.

Active packaging (AP) consists of packaging systems with the incorporation of certain additives to keep or prolong the quality and shelf life of the product, guaranteeing its safety and integrity (Kerry et al., 2006; Yildirim et al., 2018). According to the European Commission, AP is created to deliberately incorporate components that would release or absorb substances into or out of the packaged food or the environment around the food. The constituents of this type of packaging were deliberately included in the packaging material or in the headspace of the packaging to improve the performance of the packaging system (Janjarasskul and Suppakul, 2018). These packaging arrangements can be

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divided into active scavenging systems (absorbers), which remove undesired compounds present in food or its environment, for example, moisture and oxygen, and active-releasing systems, which add compounds to the packaged food or into the headspace, such as antimicrobial compounds (Yildirim et al., 2018). Several AP systems have been extensively reported, such as oxygen and ethylene scavengers, moisture absorbers/controllers, flavor absorbing/releasing systems, carbon dioxide emitters/absorbers, antioxidants release, and antimicrobial packaging (Vilela et al., 2018; Soltani Firouz et al., 2021; Prasad and Kochhar, 2014).

Intelligent packaging (IP) consists of food packaging systems with tools to detect changes in the internal and external environmental conditions of the packaged food, to communicate the status of the system to the stakeholders of the supply chains including manufacturer, retailers, and consumers (Yam, 2012). According to the European Food Safety Authority (EFSA), IP materials are articles and materials that monitor the situation of packaged food or the environment surrounding the food. IP facilitates consumers' decisions, enhances safety, and helps to enhance quality by providing information and warning of potential problems (Soltani Firouz et al., 2021). IPs encompass the concepts of AP and IP, being subdivided according to the type of monitoring: (1) Environmental conditions, to identify changes in food quality characteristics, through temperature or time indicators, gas leakage and sensors, or relative humidity; (2) Quality or quality indicator compounds, using biosensors and freshness sensors/indicators; (3) Data carriers to store/transfer data, using indicators/sensors to monitor the external environment and exhibit the information (Müller and Schmid, 2019; Heising et al., 2014).

In this context, for the design of these types of packaging, nanotechnology shows promise (Ashfaq et al., 2022). For example, nanoparticles have several functionalities, namely (1) Antimicrobial activity, (2) Oxygen scavenging capacity, and (3) Impermeability to UV rays, among others, which make them valuable (Ashfaq et al., 2022). Smart packaging and AP designed with nanostructures are being progressively applied in the food industry; even so in many cases, there is still no commercialization (Soltani Firouz et al., 2021). Therefore, it is important to analyze potential challenges and opportunities of nanotechnology in the process of progress of AP and IP to understand and solve gaps in scientific research for these systems. Thus, this chapter is focused on the main mechanisms of smart, active, and intelligent food packaging systems that use nanostructures, as well as their applications for food industries. The content was organized to discuss AP (antimicrobial, antioxidant, and scavenging/absorbent) and IP, with their indicators (temperature, freshness, gas, pH indicators) and sensors (chemicals and biosensors). In addition, products, patents, challenges, and future trends in the use of AP and smart packaging were also considered.

24.2 Active packaging

APs, specifically, are developed from various materials or nanomaterials with compounds of interest intended to maintain and improve food in its most diverse quality indicators (Roy et al., 2021) (Fig. 24.1). The literature has shown several examples of studies of AP using nanotechnology since its formulation, characterization, and application (Sharma et al., 2017). For this, some biopolymers of origin have been widely used (Cheng et al., 2022): vegetable (polysaccharides and some proteins) (Cheng et al., 2022), animal (mainly proteins), microbiological (bacterial cellulose, polyesters/PHAs), or synthesized by biotechnologic processes (polylactides/PLA) (Theóphilo Galvão et al., 2018; da COSTA et al., 2021; Miranda et al., 2021; Germano et al., 2019; Oliveira et al., 2017; Oliveira et al., 2021).

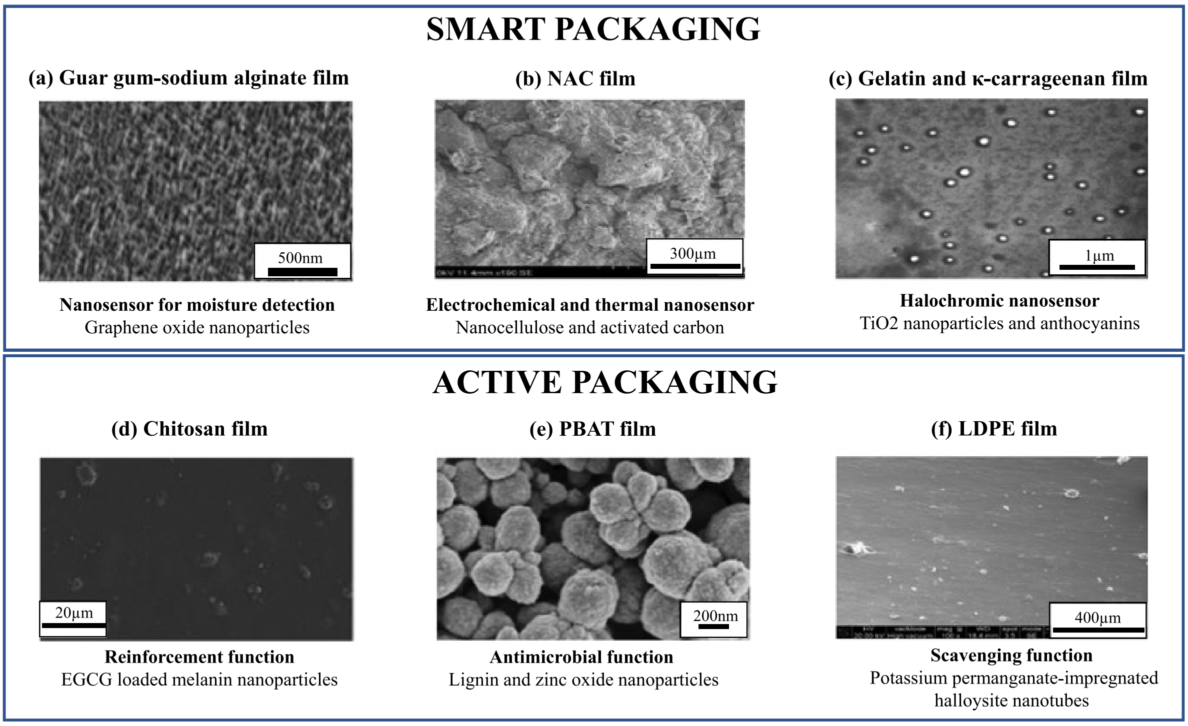

FIGURE 24.1

Different types of nanomaterials applied to smart and active packaging. Examples include the following: (A) Graphene oxide nanoparticles were added to a guar gum—sodium alginate sensor. (B) Activated carbon (NAC) embedded nanocellulose film for nanosensor production. (C) TiO_2 and anthocyanin nanoparticles producing a halochromic gelatin and κ-carrageenan nanosensor. (D) Melanin and epigallocatechin-3-gallate (EGCG) nanoparticles added to chitosan film. (E) Lignin and zinc oxide nanoparticles were incorporated into poly(butylene adipate-co-terephthalate) (PBAT) films. (F) Potassium permanganate-impregnated halloysite nanotubes were incorporated into the low-density polyethylene (LDPE) film.

(A) Adapted from Rahman, S., Chowdhury, D. (2022). Guar gum-sodium alginate nanocomposite film as a smart fluorescence-based humidity sensor: a smart packaging material. Int. J. Biol. Macromol. 216 (June), 571–582. doi:10.1016/j.ijbiomac.2022.07.008. (B) Adapted from Sobhan, A., Muthukumarappan, K., Cen, Z., Wei, L. (2019). Characterization of nanocellulose and activated carbon nanocomposite films' biosensing properties for smart packaging. Carbohydr. Polym. doi: 10.1016/j.carbpol.2019.115189. (C) Adapted from Alizadeh Sani, M., Tavassoli, M., Salim, S. A., Azizi-lalabadi, M., McClements, D. J. (2022). Development of green halochromic smart and active packaging materials: TiO_2 nanoparticle- and anthocyanin-loaded gelatin/κ-carrageenan films. Food Hydrocoll.. doi:10.1016/j.foodhyd.2021.107324. (D) Adapted from Zhao, W., Liang, X., Wang, X., Wang, S., Wang, L., Jiang, Y. (2022). Chitosan based film reinforced with EGCG loaded melanin-like nanocomposite (EGCG@MNPs) for active food packaging. Carbohydr. Polym. 290 (March), 119471. doi:10.1016/j.carbpol.2022.119471. (E) Adapted from Xiao, L., Yao, Z., He, Y., Han, Z., Zhang, X., Li, C., Xu, P., Yang, W., Ma, P. (2022). Antioxidant and antibacterial PBAT/lignin-ZnO nanocomposite films for active food packaging. Indus. Crops Prod. 187 (PB), 115515. doi:10.1016/j.indcrop.2022.115515. (F) Adapted from Joung, J., Boonsiriwit, A., Kim, M., Lee, Y. S. (2021). Application of ethylene scavenging nanocomposite film prepared by loading potassium permanganate-impregnated halloysite nanotubes into low-density polyethylene as active packaging material for fresh produce. LWT, 145(October 2020), 111309. doi: 10.1016/j.lwt.2021.111309.

As mentioned before, AP systems can reduce food waste by providing, in addition to an inert barrier to external conditions, several functions associated with food preservation, such as absorption/elimination, release/emission and removal of properties, temperature, controlling microbes, and quality (Theóphilo Galvão et al., 2018; Kuai et al., 2021; da Costa Mon çã o É Brandão Grisi et al., 2021). In this sense, nanotechnology has been used mainly for the following (Cheng et al., 2022): controlled release of preservatives, antimicrobials, or antioxidants, prolonging the shelf life of the product (Sharma et al., 2017; Ghaani et al., 2016), oxygen uptake, carbon dioxide emitters/absorbers, moisture absorbers, ethylene (Siripatrawan and Kaewklin, 2018; Sobhan et al., 2021), and flavor release/absorption systems, among others (Vilela et al., 2018).

The techniques for manufacturing nanoparticles for packaging development can be subdivided into two basic principles (Cheng et al., 2022), namely bottom-up, which consists of synthesizing a nanomaterial from atomic particles, encompassing different preparation techniques (e.g., sol–gel, vapor deposition, and reverse micelle method) (Nithya et al., 2018; Glatter and Salentinig, 2020; Siddiqui and Alrumman, 2021; Ghaani et al., 2016), and top-down, whose principle is the large-scale division into small-scale compounds (e.g., ablation, nanolithography, milling, sputtering, and electrospinning) (Ashfaq et al., 2022; Siddiqui and Alrumman, 2021; Naser et al., 2019; Aman Mohammadi et al., 2020). See a simplified diagram in Fig. 24.2. Most works follow the top-down principle, with emphasis on the electrospinning technique, which produces fibers with micro- and nano-scale diameters (Zhang et al., 2022a). Electrospinning is a simple, inexpensive, and versatile

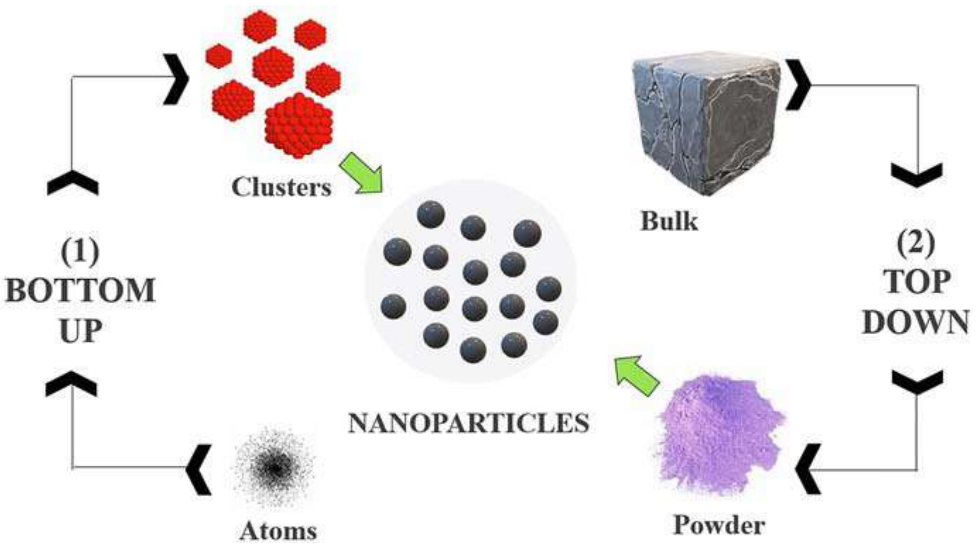

FIGURE 24.2

General methods of producing nanoparticles (Cheng et al., 2022). Bottom-up: nanomaterial synthesis from atomic or molecular species (e.g., chemical vapor deposition, sol–gel, solvothermal, hydrothermal method, reverse micelle) (Ghaani et al., 2016). Top-down: larger compounds are broken down into nanoscale materials by mechanical and chemical forces (e.g., mechanical milling, lithography, electrospinning, etching, sputtering, laser ablation). For didactic reasons, the figure shows only the production of nanoparticles. But there are other structures for the formation of nanomaterials, namely, nanotubes, wires, rods, plates, layered, and polycrystals, among others.

method as it is a promising tool (Hajikhani and Lin, 2022). Nanofiber production occurs when a polymeric solution passes through a fine orifice, e.g., metallic needle, and after application of a voltage, it forms nanofibers (porous, hollow, and in the form of shells, etc.) that are deposited on the collector (Castro Coelho et al., 2021). In terms of packaging, a study on the formation of starch-based nanofiber films by electrospinning showed flexibility compared with films made by the casting method (Zhu et al., 2022). Polylactic acid fibrous membranes with ZnO nanoparticles were developed by electrospinning, showing good results in tensile strength, elongation at break, UV barrier, and antimicrobial activity against *Escherichia coli aureus* (Zhang et al., 2021). The development of nanofibrous packaging of zein–tetradecane–cinnamaldehyde through electrospinning technique without needle, for packaging sausages, managed to keep the temperature cold for about 8 h without refrigeration, in addition to being able to inhibit the growth of *Staphylococcus aureus* (Karim et al., 2022). Polyvinyl alcohol–sodium alginate–polyvinylidene fluoride based on double-layer nanofiber film was developed for the visual monitoring and preservation of pork, resulting in good hydrophobicity, high indicator sensitivity, and ability to extend the meat shelf life, in addition to promoting inhibition against *E. coli* and *S. aureus* (Zhang et al., 2022b). Antibacterial nanofibers of gelatin, chitosan, and 3-phenyllactic acid (PLA) were prepared by electrospinning resulting in a film with good thermal stability and water vapor permeability, which reduced approximately 44 log CFU/mL of *Salmonella enterica Enteritidis* and *S. aureus* in 30 min (Liu et al., 2021a).

Antimicrobial and antioxidant packages have attracted increasing attention and prominence due to their wide use, namely as films/coatings with nanoencapsulated actives and immobilization of active nanoagents on the surface of these food packages (Luo et al., 2022; Oliveira et al., 2020). Especially, the antimicrobial effects in nanostructured APs are basically produced from inorganic materials (nanometric metals, e.g., nano-silver) or organic materials (e.g., nanochitosan) (Oliveira et al., 2021; Liu et al., 2021b; Hosseini et al., 2017). Furthermore, the application of nanocarriers including nanoemulsions and nanoparticles for the protection and delivery of antimicrobials in AP increases the effectiveness, stability, and precision in targeting (Hosseini et al., 2017). In this sense, oxide nanoparticles incorporated into regenerated cellulose-based films, for use in meat products, effectively inhibited the growth of Gram-positive (*B. cereus*, *S. aureus,* and *L. monocytogenes*) and Gram-negative (*E. coli* and *S. typhimurium*) and pathogenic (*V. parahaemolyticus*) items, when the addition was 3% by weight of nanoparticles (Saedi et al., 2021). Recently, a biodegradable film based on whey protein isolate incorporated with chitosan nanofiber and cinnamon essential oil (both in the form of emulsified and nanostructured lipid vehicles) was evaluated (Mohammadi et al., 2020). The nanofiber networks were well distributed along with the films, while the morphologic heterogeneity contributed to the reduction of tensile strength after adding the oil; however, the antibacterial activity was improved (Mohammadi et al., 2020). Another study demonstrated that active films for minimally processed fruits, formed of nanocomposites based on polyethylene oxide and copper nanoparticles, better preserved the color and texture of fruits, which remained acceptable for 3–4 days longer than the control (Sportelli et al., 2019).

When it comes to antioxidant property, films added with silica nanoparticles modified by covalent grafting of gallic acid were evaluated (Dong et al., 2022). The incorporation of nanoparticles significantly increased the antioxidant activity of the films, indicating that these composites have good antioxidant capacity for food packaging (Dong et al., 2022). Bumbudsanpharoke and Ko developed a lignocellulose fiber film, embedded with gold nanoparticles, for packaging gourmet foods (e.g., caviar), which showed good antioxidant properties compared with film without the nanoparticles

(Bumbudsanpharoke and Ko, 2018). Chitosan films containing ZnO nanoparticles and bamboo leaf extract showed strong antioxidant activity, being promising for AP in the food sector (Liu et al., 2021b). In this case, the antioxidant effect was potentiated by the combination of the reaction between the residual free amino groups of chitosan and the flavonoids present in the bamboo extract, resulting in an antioxidant activity of 87.93% (Liu et al., 2021b). AP made of PLA, incorporated with silver nanoparticles and thymol, showed high antioxidant capacity when 1% silver nanoparticles and 8% thymol were incorporated (Ramos et al., 2020). Bionanocomposites (chitosan−guar gum−zinc oxide) and roselle chalice extract were combined to form an active cheese packaging during the ripening period, which was able to protect nutrients against free radicals and photo-oxidation with antioxidant capacity of 72.3% (El-Sayed et al., 2020).

In terms of scavenging/absorbent capacity, AP can be designed to be classified as active scavenging (absorbent) systems that remove unwanted elements from the product such as moisture, carbon dioxide, oxygen, ethylene, and odor (Nikolic et al., 2021). Recently, it was reported that chitosan-based films added to TiO_2 nanoparticles provided ethylene scavenging properties (Siripatrawan and Kaewklin, 2018). Ethylene sequestering film based on corn starch and gum acacia was incorporated from 20% sepiolite to package broccoli (Upadhyay et al., 2022). The active film showed good mechanical, barrier, and ethylene sequestration properties, keeping broccoli packed for 6 days (Upadhyay et al., 2022). A recent study reported the development of an ethylene scavenger based on potassium permanganate (P-HNTs) using halloysite nanotubes as an encapsulating material for tomato packaging. The results showed that 1 g/100 g of P-HNTs increased the thermal stability, crystallinity, and ethylene scavenging capacity (by 1700%) of the nanocomposite film. In addition, the film was able to reduce the rate of ethylene production and respiration for 21 days (Joung et al., 2021).

In general, the incorporation of nanoactive compounds is added to improve the antimicrobial, mechanical, and barrier properties of films and coatings, preventing or hindering the growth of pathogenic and deteriorating microorganisms, improving food quality and safety, in addition to prolonging their shelf life (Kumar et al., 2020). Therefore, AP, associated to nanotechnology, is versatile and very useful for the industry and can be an effective alternative to guarantee food quality.

24.3 Intelligent packaging

As mentioned earlier, IP is endowed with the ability to detect or measure an attribute of the food product or the atmosphere inside and outside the package, with the ability to communicate quality-related information to the production line or to the consumer (Alfei et al., 2020). Communication (food $\rightleftharpoons$ packaging $\rightleftharpoons$ consumer) takes place due to different types of indicators (Table 24.1), which constitute an important part of the smart packaging system. In this context, nanotechnology allows the insertion of nanomaterials (Fig. 24.1) to produce indicators capable of detecting changes due to variations in temperature, freshness, gases, and pH, as seen below (Shao et al., 2021) (Ebrahimi Tirtashi et al., 2019) (Forghani et al., 2021) (Yılmaz and Altan, 2021).

24.4 Indicators

24.4.1 Temperature indicators

Although temperature variation is unavoidable during the production and supply of perishable foods, a modern quality system should be capable of preventing contamination through estimation of freshness,

Table 24.1 Different types of nanoindicators and applications in the food matrix.

Indicator type	Nanomaterial	Food sample	References
Time–temperature indicator	Gold–silver core-shell nanorods	Pasteurized milk	Gao et al. (2021)
	Polydiacetylene–silver nanoparticles	Fruits and vegetables	Saenjaiban et al. (2020)
	Polydiacetylene–silica nanocomposite	Refrigerated foods	Lee et al. (2021)
	Polydiacetylene–silica nanocomposite	–	Suppakul et al. (2018)
	Electrospun nanofibers	Seafood, lamb, poultry, milk, and fruit	Forghani et al. (2021)
Freshness indicators	Chitin nanocrystals	Shrimp and tail	Feng et al. (2020), Ge et al. (2020)
	Cellulose nanocrystals	Shrimp	
	Polydiacetylene–zinc oxide nanocomposite	Milk	Weston et al. (2020)
	Sustainable nanomaterials from wood, sugar cane, and crab shells.	Strawberries	
	Carbon nanotubes	Avocados, bananas, apples, oranges, and pears	Jiang and Liu (2020)
Gas indicator	Methylcellulose nanofibers	Fish fillet	Alizadeh Sani et al. (2022)
	Polyvinyl alcohol nanofibers	Meatball	Yılmaz and Altan (2021)
	Silver nanoparticles	Salmon, chicken, turkey, and beef	Rvspayeva et al. (2019)
	Bacterial cellulose nanofiber	Banana	Pirsa and Chavoshizadeh (2018)
pH indicator	Photoactive nanocarbon	Pork	Koshy et al. (2021)
	Latex glycerol-based dendrimer nanocomposite	Meat and fish	Pounds et al. (2021)
	Nanocellulose	Chicken breast	Lu et al. (2020)
	Methylcellulose–chitosan nanofibers	Lamb meat	Alizadeh-Sani et al. (2021)
	Cellulose acetate nanofibers	Trout	Aghaei et al. (2018)

monitoring and controlling critical parameters. From this perspective, to ensure the quality of food in storage and distribution, it is important to emphasize the role of temperature. The monitoring of this parameter is a crucial factor to guarantee the maintenance of the sensorial, nutritional, and hygienic quality of the product and, consequently, the safety of consumers. In addition, it helps to reduce food waste and environmental pollution (Albrecht et al., 2020; Mohebi and Marquez, 2015; Forghani et al., 2021; Saenjaiban et al., 2020; Manjunath, 2018; Lorite et al., 2017).

In particular, temperature indicators can deliver an important contribution, providing monitoring and recording information about the full or partial history of the product through the supply chain in real time (Mohebi and Marquez, 2015; Pereira et al., 2015). The temperature indicators are classified in three different categories (Barska and Wyrwa, n.d): (1) Critical temperature indicators indicate that the product has, at some point, been exposed to an inappropriate temperature long enough to adversely affect the quality of the product. They are indicated for application in product packaging and show irreversible damage to their properties, even with a single exposure to critical temperatures, as is the case with frozen products, for example, freezing or thawing processes (Barska and Wyrwa, n.d; Ghaani et al., 2016). (2) Critical temperature/time indicators indicate the following: 1exposure to a threshold temperature; (3) The history of changes above threshold temperatures; and (4) The storage time it was exceeded, usually due to an increase in the rate of chemicals, enzymes, and microbiological reactions that cause color changes (Alfei et al., 2020). They are suitable for monitoring any change in temperature, either above or below the critical reference temperature, which causes damage to food properties (Barska and Wyrwa, n.d) (Ghaani et al., 2016) (Taoukis and Labuza, 2003). And (3) time−temperature indicators (TTIs) are the most common indicators on food labels to detect temperature fluctuations at time intervals, providing a quick response or alerts about food quality through color changes, which can help the consumer at the time of purchase (Mohebi and Marquez, 2015) (Barska and Wyrwa, n.d) (Forghani et al., 2021; Saenjaiban et al., 2020). TTIs are considered cheap, economical, small, reliable, and generally eco-friendly. Therefore, they are easy to integrate into the packaging system and can be verified with the naked eye or using electronic devices, with programmable mobile app registration and wireless data transfer (Mohebi and Marquez, 2015; (Manjunath, 2018; (Kalpana et al., 2019).

Over the last few years, several companies have developed commercial TTIs; in the meantime a new perspective is the use of nanomaterials in the production of these indicators, such as the application of nanoparticles of different metals that has been extensively described in the literature, as these materials have unique optical characteristics (Gao et al., 2021). Gold−silver core-shell nanorods were employed to develop a TTI with physicochemical mechanism of measuring with acidity-based responses inducing color changes. This TTI was designed to monitor pasteurized milk in the cold chain and presented some advantages such as high accuracy and sensitivity, mature preparation schemes, stable chemical properties, particular local surface plasmon resonance, and good optical properties (Gao et al., 2021). Another study developed a color change film as a TTI using silver nanoparticles and glycerol in color-changing polydiacetylene−silver nanoparticles embedded in carboxymethylcellulose. The TTI proved to be important mainly for the inspection of fruits and vegetables, presenting advantages such as rapid perception of temperature changes; in addition, silver nanoparticles can increase the surface area of the polydiacetylene, increasing the mobility of the polydiacetylene (Saenjaiban et al., 2020). A TTI with gelatin-templated gold nanoparticles (2%, pH 5) was designed with practical application to monitor foods. The nanoreactor system showed a color change when the sample was exposed to 30°C; however there was no color change while it remained in the freezer for a period longer than 6 months (Lim et al., 2012). Zhang et al. evaluated a gold nanoparticle (TTI) to differentiate TTI reaction time and shelf life, being able to identify and compare the difference between TTI response time and shelf life and being able to determine the rate of food spoilage, without the development of kinetic models (Zhang et al., 2020).

Silica nanomaterials have been used to design temperature indicators. For example, polydiacetylene−silica nanocomposite materials were used in TTIs; e.g., ambient temperature (35°C) triggered color changes (blue → purple → reddish → red) in smart packaging, proving to be potentially applicable for chilled food. Furthermore, the incorporation of silica nanoparticles provided greater overall package stability (Lee et al., 2021). Suppakul et al. also used a polydiacetylene−silica (PDA−SiO_2) nanocomposite to design a low temperature indicator (Suppakul et al., 2018). In this case, PDA−SiO_2 nanocomposites were used as a color developer in a microporous strip of Tween 20, changing from blue to red upon reaching TTI. Other materials have also been used, such as electrospun nanofibers, which are employed for enzyme immobilization. Laccase was immobilized on electrospun chitosan with polyvinyl alcohol fiber to design a TTI, which indicated a change in temperature based on the color alteration of the substrate (Jhuang et al., 2020). Forghani, Almasi, and Moradi summarized some new approaches in smart food packaging, emphasizing electrospun nanofibers as a TTI, being suitable for packaging seafood, mutton, poultry, milk, and fruit (Forghani et al., 2021). Nanofibers for temperature indicators can be produced quickly and at low cost and are versatile and simple.

24.4.2 Freshness indicators

The freshness indicator (FI) is a type of indicator that may deliver semi-quantitative or qualitative data on the quality of packaged foods through physiologic changes or microbiological growth and metabolism during storage, without the need to destroy the food packaging (Ghaani et al., 2016). As with the other indicators, the changes are visually observed after the reaction between the various metabolites formed and the indicator, within the packaging system. Generally, FI provides a color change result that is possible to be monitored and associated with the loss or maintenance of food freshness (Ahmed et al., 2018). The use of FIs in food chain management is efficient and has shown high potential for monitoring perishable foods whether of animal or plant origin. This smart packaging technology is simple, does not require complex operations, and can show if the food is fresh in a direct and specific way (Shao et al., 2021; Yılmaz and Altan, 2021). The main element of an FI is a substance that forms colorful compounds in the presence of metabolites from deterioration reactions, with a consequent change in color or optical properties (Barska and Wyrwa, n.d). The FI can be classified as a chromogenic indicator and data carrier indicator according to mensuration data. Chromogenic ones, habitually, indicate food freshness by changing color, using natural chromogenic pigments (e.g., carotenoids, anthocyanin, curcumin, chlorophyll, and betaine) or chemical chromogenic ones (polyaniline, bromophenol red, phenol red, bromophenol blue, bromocresol green, bromocresol violet, and methyl red) (Bhargava et al., 2020). On the other hand, a data carrier method can supply data associated with food freshness through an optical scanner. For the effective development of a FI, it is necessary to know parameters such as the microbial flora present in food and its possible metabolites, as well as the rate of production of metabolites as a function of variations in storage temperature. The microbial metabolites often used to indicate the freshness of perishables are biogenic amines, volatile nitrogen compounds, sulfuric compounds, glucose, organic acids, carbon dioxide, ethanol, and ATP degradation products (Smolander, 2003; Mohebi and Marquez, 2015).

Nitrogen compounds, e.g., trimethylamine and ammonia, have been employed in FIs to indicate the quality of animal products (beef, chicken, shrimp, and fish). For example, oxidized chitin nanocrystals (O-ChNCs) were incorporated (through noncovalent bonds) into a gelatin and anthocyanin-based film that permitted the visual observation of shrimp and hairtail body freshness. The film with O-ChNCs had its crystalline structure improved; in addition, with an increase of the total volatile basic nutrients, there was a color change (purple → gray → brown) indicating the deterioration of the fish

(Ge et al., 2020). Similarly, cellulose nanocrystals and curcumin nanocapsules were incorporated into soy protein isolate films for detection of freshness in shrimp (Xiao et al., 2021). Due to hydrogen bonding interactions, the addition of nanoparticles improved some properties of the film, such as thermal stability, tensile strength, crystalline structure, decreasing its permeability to water vapor and oxygen and its solubility in water (Xiao et al., 2021). In addition, the application of this film in stored shrimp decreased the total volatile basic nitrogen, and it was able to monitor, visually and in real time, their freshness (Xiao et al., 2021). A polydiacetylene−zinc oxide nanocomposite FI was designed to distinguish fresh milk from spoiled (Weston et al. 2020). For this, the indicator showed a color change in three stages, according to the lactic acid concentration: (1) Blue to indicate freshness with pH range of 6.8 to 6.0, (2) Purple to indicate deterioration at pH 6.0 to 4.5, and (3) Red as an indicator of spoiled milk with pH around 4.5 to 4.0, proving to be efficient in determining the freshness of milk (Weston et al. 2020).

In fruits and vegetables, ethylene has been widely used to identify quality and freshness. Whereas, acetic acid and lactic acid are indicators of fermentative metabolism of acid-producing bacteria. In fruits and vegetables, ethylene has been widely used to identify quality and freshness. Recently, sustainable nanomaterials from wood, sugar cane, and crab shells have been prepared and used as indicators of freshness in strawberries. In addition, the packaging was able to present antifungal property and reduce weight loss and color changes of strawberries compared with control samples. Another study reported the use of indicators based on carbon nanotubes to detect ethylene emission from avocado, banana, apple, orange, and pear. The indicator was able to detect very low concentrations of ethylene ($< 10^{-6}$), showing potential in determining fruit freshness (Liu et al., 2020).

Some microbial metabolites are used as freshness markers, such as sulfide, which can indicate freshness in fruits, vegetables, and meat (Shao et al., 2021). Recently, an FI based on gold nanoparticles supported by Cu^{2+}-modified boron nitride nanosheets was developed (Lin et al., 2022). The indicator was able to measure the amount of hydrogen sulfide released in packaged meats, quickly, easily, and at low cost (Lin et al., 2022). Likewise, when it contains CO_2 in the headspace of packaged foods, it can indicate microbial fermentation or product deterioration. CO_2 is widely used as indicator of freshness and can be detected in any food spoiled by microorganisms. The pH of the medium changes in the presence of CO_2. Therefore several pH-responsive dyes came to be applied to monitor the CO_2 production, such as bromothymol blue/methyl red that was applied in a sugarcane bagasse nanocellulose hydrogel, with color change based on the freshness when used in a chicken sample, relating to CO_2 levels, indicating the growth of microorganisms above what is tolerated to ensure a safe product (Lu et al., 2020). The FI labels produced with only bromothymol blue, by bromothymol blue solutions and methyl red, used alone or mixed, were effective to indicate microbiological changes in green peppers (Shao et al., 2021).

Despite the advantages of applying FI in the food industry, its use is still limited, especially for those using nanomaterials, with few indicators available commercially, namely (1) Insignia CO_2 Detection Pallet Intelligent Labels; (2) Food Fresh; (3) RipeSense, FreshTag, and (4) Toxin Guard (Bumbudsanpharoke and Ko, 2019). Specially, FreshTag from COX Technologies identifies volatile amines in seafood and products requiring storage at refrigeration temperatures. It consists of a plastic chip with a ring inside that contains chemical substances and is in contact with gases diffusing from packaging, including volatile amines. It causes a colorful reaction; then the ring changes from yellow to dark blue (Barska and Wyrwa, n.d) (Ahmed et al., 2018). Toxin Guard can be printed on paper or plastic foil used for food packaging. It is a toxin indicator that uses immobilized lactic antibodies for detection of pathogenic bacteria (Barska and Wyrwa, n.d). It consists of a label containing small blue circles while the product is fresh. After losing the freshness, an “*X*” becomes visible inside the circle.

24.4.3 Gas indicators

Nanoscience offers unique solutions to accurately detect and quantify food safety-relevant analytes, such as gases and vapors. Indicators for gases in packaging are used to determine the quality of food or the level of spoilage, depending on the internal atmosphere of the packaging (Müller and Schmid, 2019; Madhusudan et al., 2018). Gas detection is influenced by time, storage condition, type of food, microbial respiration, materials of constitution, and free space in the package (Shao et al., 2021). In this scenario, gas indicators can detect volatile substances from enzymatic, chemical, and microorganism metabolism (Müller and Schmid, 2019; Matrose et al., 2019).

Some gases such as carbon dioxide, oxygen, ethanol, water vapor, volatile nitrogen, hydrogen sulfide, and other volatile substances can indicate the freshness of foods (Müller and Schmid, 2019; Shao et al., 2021). Of these, carbon dioxide stands out, as it is a product of microbial metabolism and respiration in vegetables, indicating senescence during postharvest (Shao et al., 2021; Tiwari et al., 2021). Specifically, CO_2 sensors can be divided into two classes based on transducer type (Cheng et al., 2022): optical, subdivided into induced fluorescence and pH-based wet optical CO_2 indicator, and (Ghaani et al., 2016) electrochemical, subdivided into potentiometric, amperometric, and conductometric (Puligundla et al., 2012).

For food classes, particularly for fruits and vegetables, in addition to CO_2 release by respiration, maturity can be determined by measuring natural flavor components (such as aldehyde) or by ethylene release (Shao et al., 2021; Kim et al., 2018). In the case of animal products, volatile nitrogen (ammonia, amines, and methyl indole, etc.) produced by the action of bacteria and enzymes during decomposition is commonly detected in gas indicators (Velusamy et al., 2010). On the other hand, compounds such as ammonia, dimethylamine, and trimethylamine are considered the main index to detect the deterioration of meat and seafood and can be recognized by gas indicators (Ahmed et al., 2018).

Recent advances in smart food packaging using nanomaterials to project gas indicators have been reported in the literature. For example, a smart film of chitin and methylcellulose nanofibers was developed to detect increased ammonia vapor concentration, effectively determining freshness in fish fillet (Sani et al., 2021). Polyvinyl alcohol nanofibers optimized with electrospinning polystyrene was used as an oxygen indicator for meatball packaging (Yılmaz and Altan, 2021). The results showed that the gas nanoindicators were effective in controlling oxygen levels inside the package, ensuring the quality and color of the meatballs (Yılmaz and Altan, 2021). Recently, a smart adhesive coated with silica nanoparticles for carbon dioxide detection in packaging, aimed at refrigerated products, performed well compared with the gaseous indicator “After Opening Freshness,” from Insignia Technologies (Wang et al., 2019). The use of carbon dioxide sensors in ready-to-eat foods, such as kimchi, a fermented dish from the Korean tradition, has been reported (Lyu et al., 2019). In this research, branched polyethyleneimine (incorporated with bromothymol blue/tetrabutylammonium ions) was used to measure the changes in CO_2 in the kimchi package during the fermentation process (Lyu et al., 2019). Another research showed that polyetherimide and silver (Ag) nanoparticles were used to form a film to measure the freshness of salmon, chicken, turkey, and beef (Rvspayeva et al., 2019). The film changed color when exposed to gases (0.01 and 0.005 M of 2-mercaptoethanol), released by the meat, as they reduced the Ag ions (Rvspayeva et al., 2019). In terms of plant products, recently, an ethylene gas sensor was developed from a portable bacterial cellulose nanofiber film with potassium permanganate for banana packaging. The gas indicator was able to identify the ethylene released in two ways: (1) during storage (21 days) and (2) due to temperature variations (−4, 5, and 25°C). It is suitable for portable fruit packaging (Pirsa and Chavoshizadeh, 2018). As can be seen, there are several innovative attempts in food packaging using nanotechnology to produce gas indicators that effectively monitor food quality in real time.

It is noteworthy, however, that gas indicators manufactured with nanomaterials can create a regulatory obstacle, as these devices are coupled inside the package, which can lead to the migration of nanoscale components to food (Yang and Duncan, 2021). This becomes a negative point compared with analog sensors that do not have nanomaterials, and it can make the security of nanosensors more difficult (Yang and Duncan, 2021). In addition, gas indicators respond only to volatile compounds. Deteriorating substances on the surface of foods cannot be detected by the indicator, such as nongaseous enzymatic hydrolysis and microbial decomposition products (Shao et al., 2021). However, gas nanosensors, when coupled to packaging, serve as an electronic nose, detecting chemical substances released during food spoilage (Kasaai, 2021).

24.4.4 pH indicators

pH indicators are composed of two parts, i.e., a solid support and a colorant that is sensitive to the change in pH, which can be incorporated into the materials or nanomaterials that make up the packaging (by extrusion and casting) or fixed to the inside or outside of a package (Balbinot-Alfaro et al., 2019; Almasi et al., 2022; Ezati et al., 2019). When the food starts to deteriorate, a pH change occurs, usually caused by extracellular lipolytic and proteolytic enzymes of pathogenic microorganisms of food origin or contaminants (e.g., *Pseudomonas aeruginosa, Pseudomonas fuorescens, Aeromonas caviae, Aeromonas hydrophila, Proteus mirabilis, Proteus vulgaris, B. cereus, Bacillus subtilis, S. aureus, Staphylococcus epidermidis, Micrococcus luteus, C. perfringens,* and *Serratia marcescens*), which cause color changes in the indicator or packaging and provide qualitative information through visual colorimetric changes (Alfei et al., 2020; Shao et al., 2021; Balbinot-Alfaro et al., 2019; Almasi et al., 2022). Different polymers (synthetic or natural) have been proposed as support/carriers for pH-sensitive dyes, including starch−chitosan−polyvinyl alcohol, agar−starch, chitosan−pectin, filter paper, and nanomaterials such as nanocellulose, nanochitosan, and nanostarch, among others (Ezati et al., 2019). The placement of the dye in the smart package determines the three main groups of pH indicators (Cheng et al., 2022): packages with dyes immobilized by physical adsorption on a solid support such as an ion exchanger (Ghaani et al., 2016); packages with dyes covalently bonded to the hydrophilic support (e.g., as cellulose/nanocellulose or glass), and (Sobhan et al., 2021) packages with dyes physically trapped in polymeric matrices (Balbinot-Alfaro et al., 2019). It should be noted that the dyes can be extracted from synthetic sources (e.g., bromocresol green and purple, chlorophenol, methyl red and cresol, bromothymol blue and xylenol) or vegetable sources (e.g., flavonoids: anthocyanins, curcumins, betalains, chlorophyll, and alizarin) (Balbinot-Alfaro et al., 2019; Almasi et al., 2022; Ezati and Rhim, 2020; Ezati et al., 2020; Kanatt, 2020).

The literature has shown the feasibility of producing several pH indicators using nanotechnology. For example, biodegradable colorimetric films of starch and anthocyanins were reinforced with photoactive nanocarbon (1–20 nm) and used as pH indicators to visually monitor pork spoilage (25°C) (Koshy et al., 2021). The color of the indicators changed from pink/purple (fresh stage, t = 0, and pH = 5.8) to green color (damaged stage, t = 48 h, and pH = 8), due to (Cheng et al., 2022) microbial action and (Ghaani et al., 2016) high production of total volatile basic nitrogen, making it a promising smart package for monitoring the freshness of stored pork (Koshy et al., 2021). Another smart film of latex glycerol-based dendrimer nanocomposites has been synthesized, exhibiting high sensitivity to food protonation, to create a real-time pH-responsive meat/fish packaging material. In addition, the film provided a water and heat barrier and resisted deformation (Pounds et al., 2021). Peng Lu et al. (Lu et al., 2020) developed a pH indicator film from a nanocellulose-based hydrogel to monitor chicken breast freshness. The pH indicator changed from green (t = 0) to yellow-orange (t = 24 h) and finally changed to bright red after 2 days (4°C), demonstrating potential application of the film for smart packaging (Lu et al., 2020). Alizadeh-Sani et al. (Alizadeh-Sani et al., 2021) prepared pH-sensitive films based on methylcellulose–chitosan nanofiber and anthocyanins to monitor lamb meat freshness. Protein degradation leads to the formation of volatile nitrogen compounds, such as ammonia and amines, which increase the pH inside the package, so the pH of the lamb meat significantly increased from 5.8 to 7.5 after 72 h, changing the color of the indicator from reddish to pale peach during storage (Alizadeh-Sani et al., 2021). Another research reported the production of cellulose acetate nanofibers containing alizarin and used as an indicator for qualitative evaluation of the deterioration of trout fish (Aghaei et al., 2018). In this study, the color of the indicator visually changed with an increase in the amount of total volatile basic nitrogen (TVB-N), generating a change in the pH value. After the fourth day (pH > 6.3), the indicator showed a very light brick color. On the sixth day (pH = 6.67), the color became darker, representing real changes in pH, and only on the 12th day (pH = 6.94), the sensor color tended to violet, indicating the product deterioration (Aghaei et al., 2018). Therefore, this halochromic indicator can be used in the headspace of food packaging in products that produce gas metabolites (TVB-N or CO_2) (Aghaei et al., 2018). Recently, multifunctional bionanocomposite films (based on konjac glucomannan–chitosan with nano-ZnO and love anthocyanin extract) have been developed, with wide applicability in food. The results showed good sensitivity of the pH indicator, which exhibited relatively wide color differences from red to blue in different buffer solutions (pH 2–12) (Sun et al., 2020).

Sensitive pH indicators in smart packaging systems are crucial for the future of food industries that want to inform consumers about food safety and quality throughout storage; however commercial and safety bottlenecks of nanomaterials are challenges that need to be overcome (Ezati et al., 2019). However, some advances are taking place, such as the high demand in the use of natural dyes, due to their nontoxic and ecologic characteristics, for the manufacture of pH-sensitive indicators.

24.5 Sensors

Nanosensors are devices that contain elements at the nanoscale and are designed to respond to the presence of certain analytes or environmental factors (Table 24.2) (Yang and Duncan, 2021). Nanosensors must have mandatory electronics, which differ from indicators and can be designed to accurately detect and quantify analytes relevant to food safety, quality, or nutritional properties, as will be seen below (Singh et al., 2018).

Table 24.2 Different types of nanosensor to produce indicators and their applications.

Sensor type	Nanomaterial	Applications	References
Chemical sensors	Silver nanoparticles	Meat	Zhai et al. (2019)
	Anthraquinone and azo chromophores	Crabs	Zhang et al. (2019)
	Silver and zinc nanoparticles	Food packaging	Lee et al. (2021)
	Gold nanoparticles	Detection of methylparathion pesticides and other organophosphate pesticides	Deng et al. (2016)
Biosensors	Silicon nanostructure	Detection of *Escherichia coli*	Massad-Ivanir et al. (2016)
	Detection and identification of pathogens: *Salmonella typhimurium* and de *Escherichia coli* JM109 e DH5-α	Silver nanowires	Ali et al. (2018)
	L-lysine-insensitive LysG nanodrop-based biosensor	L-histidine metabolite sensors	Della Corte et al. (2020)
	DNA nanostructure	Detection of *Staphylococcus aureus*	Yu et al. (2020)
	Hexaferrocenium tri [hexa(isothiocyanato)iron(III)] trihydroxonium (HexaFc) complex	DNA biosensor	Ariffin et al. (2021)

24.5.1 Chemical sensors

The word sensor is often used as a synonym for indicator, but they are two words with different meanings. The sensor is distinguished by having an electronic part that transforms chemical, physical-chemical, or biochemical interactions into an electrical signal that can be measurable (Pirsa et al., 2022). The sensors have a transducer with two basic functions: a part that receives a signal or stimulus and the transduction element that converts the input signal of the sensing element into an electrical signal. Afterward, this signal is then processed and transformed into analytical data. Sensors can be classified in different ways, namely the following (Cheng et al., 2022): considering the type of transducer (e.g., optical, electrochemical, thermal) (Ghaani et al., 2016), the application (Sobhan et al., 2021), materials used (e.g., nanoparticles/nanosensors), and (Vilela et al., 2018) the type of interaction with the received signal or stimulus (e.g., chemical sensors) (Azeredo and Correa, 2021). In this section, we will discuss chemical sensors specifically. Considering this type, the input signal can come from a chemical reaction of the analyte or from its direct presence. According to Hulanicki et al. (Hulanicki et al., 1991), the receiving part of the chemical sensors, responsible for selectivity and specificity, can be based on several principles, operating together or separately (Cheng et al., 2022): physical, where no chemical reaction occurs (measurement by absorbance, refractive index, conductivity, temperature, or mass variation) (Ghaani et al., 2016); chemical, where a chemical reaction with the participation of the analyte results in the analytical signal (Sobhan et al., 2021); or biochemical, in which a biochemical process is the source of the analytical signal.

Chemical sensors are commonly used as portable devices and have more recently been integrated into food packaging. The demand for innovative food packaging to ensure food with greater safety, quality, and traceability has encouraged the development of sensors for packaging (Rodrigues et al., 2021). As mentioned earlier, food has come a long way where packaging plays an important role in maintaining quality and safety. During this time, damage to the integrity of the package can occur, causing changes in the atmosphere of the package and exposing the product to physical and pathogen contamination, which can result in a shorter shelf life of the packaged food, health risks, and economic loss to companies through recalls. Sensors appear in this scenario as practical and useful devices that monitor any change in packaging or food quality (Andre et al., 2022).

Sensors can be especially useful for food such as fruit to inform the status of quality related to freshness, ripeness, pathogens, and gases, which can be correlated with food safety (Alam et al., 2021). It is important to keep in mind that for fruits harvested under good sanitary condition, the shelf life may also depend on the gases present in the storage atmosphere. Maintaining the ideal composition of respiratory gases (O_2 and CO_2) and ripening of climacteric fruits (ethylene) inside the package is a determining factor for the freshness of fruits. For fruits, its physiology must be especially considered to integrate sensors to packaging (Dwi Anggono et al., 2022). Traditional methods for evaluating continuous respiration rate can be time-consuming, nonportable, and cost-effective. Therefore, alternative methods are attractive especially if a measure is noninvasive and nondestructive. Because electrochemical and optical sensors are easy to use, measure in real time, relatively low cost, and accurate, they are useful in detecting and controlling the regularity system of gas concentration and other substances indicators of food spoilage (Sobhan et al., 2021; Vilela et al., 2018; Schaefer and Cheung, 2018; Soltani Firouz et al., 2021; Kerry et al., 2006).

The quality of fish has also been widely evaluated by real-time sensors based on electrochemical and optical techniques, Raman spectroscopy, and electronic nose and tongue technologies (Vilela et al., 2018; Yildirim et al., 2018; European Commission, 2004; Janjarasskul and Suppakul, 2018; Prasad and Kochhar, 2014; Yam, 2012). The monitoring of fish freshness can be realized by pH variation, release of biogenic amines, NH_3, CO_2, HCl, and H_2S, substances that are also by-products of the food deterioration from enzymatic reaction or microbial activities. Thus, it is of great significance to determine the quality and safety of fish and other kind of animal meat to reduce health risks. Zhai et al. (Zhai et al., 2019) developed a H_2S sensor based on silver nanoparticles (AgNPs) as a meat spoilage indicator system for IP. AgNPs have been widely used to develop colorimetric sensors, due to their unique local surface plasmon resonance. In this study, the sensor was capable of visually monitoring meat spoilage by selectively detecting the H_2S generated from meats. According to the authors, this novel colorimetric H_2S has great potential for practical application in intelligent food packaging. Zhang et al. (Zhang et al., 2019) developed smart color-changing packaging sensors with pH-sensitive chromophores by printing patterns for monitoring the freshness of the food. The freshness experiment of cooked crabs indicated that they began to deteriorate after 1.5 h and were completely deteriorated after 4 h.

Radio frequency identification (RFID) technology, sometimes called data carriers, is gaining ground as a technology primarily at product traceability and can be integrated into packaging. It can be said that RFID is an advance in barcode technology. Nowadays, it is one of the leading technologies in wireless sensors. The technique consists of a tag composed of a microchip with an antenna and a reader with an antenna (Müller and Schmid, 2019). Tags are designed to take power from a battery (active tag) or electromagnetic induction (passive tag) and send encoded signals to readers. Readers are used to decode tag signals and data management systems (Yildirim et al., 2018). Sensors can be embedded in passive, semipassive, and active tags, directly on the antenna or connected to the chip. The coupling of sensors such as temperature–time, gas, and other kind of sensors to RFID brought new concepts to food by measurements of quantities, as a function of time, or by recording predefined events (Heising et al., 2014; Ashfaq et al., 2022). It is important to note that passive tags are usually smaller, cheaper, and more convenient packages. Nowadays, there are still some barriers to be overcome for the wide use of technology such as cost and possible interference in the RFID signal such as moisture, metals, packaging material, and tag folding.

Different types of materials (conducting polymers, biopolymers, metal oxides, organic dyes, and carbon-based nanomaterials) associated or not with nanoparticles can be used for the development of integrated sensors in food packaging (Roy et al., 2021). Matindoust et al. (Matindoust et al., 2021) showed the use of conductive polymer nanosensors in the food industry and their importance to increase the sensitivity to gases formed during deterioration. The extruded O_2 sensors showed superiority over the sensors previously used in packaged food products. Won and Won (Won and Won, 2021) developed an oxygen sensor based on an electrochemical battery that does not require external energy sources for O_2 detection because it has nanogenerators to collect energy through mechanical energy. The sensor composed of three layers (oriented polypropylene film with deposited silver [cathode], zinc foil [anode], and adhesive gel electrolyte) generated output voltage linearly proportional to the oxygen gas concentration in the range of 0%–21% ($R^2 = 0.99$), being promising for food packaging.

Most sensors and nanosensors used are nonrenewable and made from nonbiodegradable synthetic materials that are incompatible with sustainability objectives. Thus, there is a prospect that new chemical nanosensors, in addition to being sensitive, specific, and relatively low cost, are also eco-friendly (Piro et al., 2020).

24.5.2 Biosensors

The development of biosensors has evolved a lot in recent years thanks to advances in biotechnology. Faster, more specific, and accurate response have been presented in easy-to-handle and affordable devices. Biosensors are interesting analytical tools due to their simplicity of structure and speed of result, in addition to portability that even includes real-time sample data or multiplexed detection (Petrucci et al., 2021).

A biosensor is an analytical device or instrument that comprises a biologic component with a physicochemical transducer that converts a chemical or biologic signal into a quantifiable and processable signal. Biosensors are widely used in different areas as food industry, medicine, agriculture, and environmental analyses (Hassanpour et al., 2018). The biologic component is the bioreceptor being able to be a tissue, microorganism, organelle, cell, enzyme, antibody, or nucleic acid. The transducer can be optical, electrochemical, thermometric, piezoelectric, magnetic, and micromechanical, or combinations of these (Velusamy et al., 2010).

The development of new smart packaging can be boosted with the application of biosensors in its structure and functioning. For example, food contaminants capable of interacting with bioreceptors can be easily detected by biosensors coupled to their packaging, allowing the monitoring of deteriorating microorganisms, pathogens, allergenic molecules, toxins, and other analytes of interest in the food production chain. Biosensors can detect the target analytes in a more effective and specific manner compared to indictors. They are more efficient at sensing the freshness of food better and are also comparable with pathogen indicators (Roya and Elham, 2016). Characteristics such as specificity, simplicity, reliability, sensitivity, and portability are essential in a biosensor.

Food matrices are very diverse and have specific characteristics that can pose challenges to the use of biosensors as strategic packaging tools. Nevertheless, many studies have demonstrated the feasibility of using biosensors in this category of packaging. Nanotechnology is also a strong ally in this field. Deng et al. (Deng et al., 2016) developed a novel acetylcholinesterase biosensor based on nano-porous pseudo carbon paste electrode modified with gold nanoparticles for detection of methyl parathion. This biosensor was based on layer-by-layer assembly of chitosan (CS), gold nanoparticles (GNPs), acetylcholinesterase biosensor (AChE), and Nafion modified nano-porous pseudo carbon paste electrode. This biosensor exhibited a high specificity to the AChE and produced a detectable signal and fast response owing to its excellent electron transfer rate and satisfactory biocompatibility of GNPs/CS. The proposed biosensor exhibited excellent performance parameters, good reproducibility and repeatability, acceptable stability, fast response, low detection limit, and strong anti-interference ability and, therefore, has potential application in detection of methyl parathion pesticides and other organophosphates pesticides.

Fruits and vegetables are foods with a very diverse composition and are very perishable, representing an interesting category for the application of biosensors in their packaging. An important criterion for evaluating fruit freshness and the quickest way to detect microbial contamination is by measuring changes in the gas composition inside packages caused by microbial growth. Alam et al. (Alam et al., 2021) compiled in their study several types of biosensors using different nanostructures (e.g., nanotubes, nanoparticles, nanofibers) to identify fruit quality and detect food safety-relevant pathogens. A relevant initiative in this area is presented by SIRA Technologies, which focuses commercial efforts on the manufacture of biosensors for food packaging, including the Food Sentinel System (SIRA Technologies, CA, USA) and ToxinGuard.

Alternative materials have been explored with a view to their application in smart packaging that use biosensor nanotechnology to show food quality in real time. An interesting example of this line of research has been summarized by Yang et al. (Yang et al., 2021) in the paper showing current status reports on different chitosan (CH)-based nanogels, providing new perspectives and ideas for researchers to develop nanocomposite materials and facilitating the development of potential application of CH-based nanogels in biosensors.

A wide variety of recently developed biosensors have a great potential for application in smart packaging. Hereafter, we list some very interesting examples. Massad-Ivanir et al. (Massad-Ivanir et al., 2016) developed an optical biosensor for the detection of *E. coli*. The biosensor was nanostructured based on oxidized porous silicon that was functionalized with antibodies against *E. coli*. This work proves the application of optical immunosensors without the use of markers for complex matrices. The samples, in addition to a large load of contaminants, also had bacteria at risk of cross-reaction, as well as interferences such as soil particles and plant cell remains. This is the reality presented by many foods, especially chicken, meat, fish, and dairy products. Such foods have a large quantity and variety of microorganisms that make up their microbiota and can interfere with the biosensor's response in an IP.

Ali et al. (Ali et al., 2018) developed a disposable fully printed electronic biosensor for real-time detection and identification of pathogens. Three types of widespread pathogens can be detected:

S. typhimurium and *E. coli* strains JM109 and DH5-α. The biosensor is formed by silver electrodes manufactured by an inkjet material printer and silver nanowires printed on the electrodes through the electrohydrodynamic technique on a polyamide-based polyethylene terephthalate base. The detection time was less than 10 min, and the cost per unit was less than 1 USD/unit. This type of technology can be integrated into food packages that have a high risk of pathogenic bacteria growth. Della Corte et al. (Della Corte et al., 2020) designed a semirational engineering assay applying a fluorescence activated cell sorting (FACS)-based technique/counter sorting strategy to generate an L-lysine-insensitive LysG nanodrop-based biosensor. This device could isolate L-histidine-producing strains by FACS, demonstrating that engineering transcriptional regulators to a more focused spectrum of ligands can expand the application range of such metabolite sensors. These results prove the application of such metabolite sensors, with the potential to detect amino acids of interest in the food chain. A colorimetric biosensor for detection of *Staphylococcus aureus* was developed by Yu et al. (Yu et al., 2020) through DNA nanostructure using an aptamer specific to *S. aureus* by hybridization with the capture probe anchored to the surface of the plate via streptavidin—biotin binding. The biosensor showed wide linear range, low detection limit, high specificity in *S. aureus* detection in real and complex samples. Ariffin et al. (Ariffin et al., 2021) performed a study using HexaFc for the first time as an electroactive indicator for porcine DNA biosensor. The new HexaFc complex produced DNA binding and high selectivity from spectrophotometric studies. The DNA biosensor with this ferrocenium indicator showed an efficient response against porcine DNA. It is an easy and fast method for the determination of swine DNA in food products, being a very useful alternative in the prevention of fraud and in the proof of purity applied to the packaging of meat products. According to Eyvazi et al. (Eyvazi et al., 2021) portable biosensors are small, user-friendly, cost-effective, and movable diagnostic biomedical tools that enable real-time screening of food contamination.

Nanotechnology, microfluidics, smartphones, and biologic design strategies, as well as wireless network resources, emerge as alternatives that help researchers to design and develop portable devices, with easy operation and quick responses (Gharib et al., 2022). During the last years, a wide variety of sensors for the detection of biologic contaminants in food with different principles, such as electrochemical, magnetic, and optical properties, have been developed (Eyvazi et al., 2021). Biosensors are considered efficient due to their sensitivity and selectivity to biologic contaminants and can be extended to other microorganisms by altering the specificity of antibodies or aptamers, thus becoming more versatile options than traditional methods that are widespread in laboratory routine (Arshavsky-Graham et al., 2022). These advances in the development of new biosensors provide great opportunities for application improvement of the nanotechnology applied to IP.

24.6 Products and patents: Smart packaging in food products

In this section, the development of industrial patents in databases is described, namely the following (Cheng et al., 2022): Trademark Office (USPTO) (Ghaani et al., 2016), National Institute of Intellectual Property (INPI) (Sobhan et al., 2021), World Intellectual Property Organization (WIPO), and (Vilela et al., 2018) Spacenet. First, the search terms were more generic “food packaging” AND “nanoparticles” Later, specific terms were used, such as “nanocomposite film for food” AND “active film” AND “nanoparticles” and “food” “biosensors” OR “sensors” OR “nanosensors” for food packaging. Documents that presented the keywords in the title or abstract, published between 2017 and 2021, were considered valid. In addition, the field "IPC" (International Patent Classification) was searched according to the focus of this Chapter. 11 patents were found in the USPTO database, 2 patents from the INPI database and 145 patents in the Espacenet database, including patents of the WIPO and The European Patent Organization (OPE/EPO). Therefore, 158 patents have been published in the last 5 years, 11 of them focused on biosensors. China (CN) leads the ranking of patent applications (Taoukis and Labuza, 2003), followed by the United States (US) (Nithya et al., 2018), WIPO (WO) (Miranda et al., 2021), Republic of Korea (KR) (Yam, 2012), European Patent Organization (OPE/EPO) (EP) (Yildirim et al., 2018), Japan (Soltani Firouz et al., 2021), Australia (AU) (Ghaani et al., 2016), Canada (CA) (Ghaani et al., 2016), Romania (RO) (Ghaani et al., 2016), United Kingdom (UK) (Ghaani et al., 2016), and Brazil (BR) (Ghaani et al., 2016).

These publications include the development of packaging with nanoparticles that act in food safety, and some patent examples are listed (see more in Table 24.3): (Cheng et al., 2022) soy antioxidant protein composite film for food packaging polymeric nanomaterials (CN112280/315A); antibacterial composite film with nano Ag—TiO_2 mixing polylactic acid (CN112812521A); nano-silver immobilized nano cellulose—MXene antibacterial composite film, to inhibit the growth of pathogenic bacteria, prolong food shelf life, and meet the requirements of modern food AP (CN112662014A); and green, biodegradable, and multifunctional collagen-based nanocomposite film that solves the problems of degradation, barrier property, and unique function of food packaging materials (US2021309817A1). In the area of biosensors, the preparation and application of aptamer-quantum dot biosensors for the detection and elimination of *Salmonella* stands out, which belongs to the technical area of detection and inactivation of pathogenic bacteria in food (AU2021100518A4) or the production of a membrane that prevents direct contact of food with the nanomaterial of the optical sensor (US2021010942A1).

New cutting-edge technologies such as nanotechnology promise to offer significant benefits to the food packaging industry. Thus, patents demonstrate the interests of industry and researchers in the sector. It is essential that these trends are catalyzed through proactive measures to improve the R&D environment, initiated through policy interventions, so that investments, including the development of a trained workforce, are planned for growth of (active/smart) packaging using nanotechnology.

24.7 Challenges in the use of AP and IP

There are many works on IP and AP with the perspective of commercial application in food. Although much research focuses more on proof of concept (to work or not to work correctly), they, in general, do not address the following: (1) The safety aspects of food intake protected by these packages, economic viability (including mass and energy balance study), (2) Environmental impact, and (3) Application that considers that the functionality of AP and IP can vary from the product chosen to be protected. Especially are technologies related to nanosensors that are still little explored in market applications due to economic issues and a series of large-scale production challenges. In fact, a complete study requires a high investment and is usually financed by companies seeking to commercialize the product.

The research, focused on proof of concept, especially when the food is in direct contact with the packaging, should also involve aspects of toxicity and allergenicity, in addition to studying the migration of compounds from the packaging to the food over time. When active/functional compounds at the nanometer scale are used in packaging, there may be doubts regarding the reliability of the

Table 24.3 The most recent patents published in 2021.

Title	Inventors	Applicants	Publication number	IPC	Publication
Nisin–chitosan nanoparticle antibacterial film, preparation method, and application	Lin Lin Bai Mei Cui Haiying	Univ Jiangsu	CN106962498A CN106962498B	A23C19/097 D01D5/00 D01F1/10 D01F6/94 D04H1/728	February 12, 2021
Food packaging articles including substrates with metal nanoparticles	Dankovich Theresa [UUS]	Folia water Inc [US]	US2021321496A1	B32B B32B1/02 B32B15/12 B32B3/10	October 14, 2021
Processing method of self-reinforced starch-based multifunctional material	Miao Ming Jia Xue Campanella Osvaldo Jin Zhengyu Zhang Tao Ye Lei	Univ Jiangnan	CN112552554A	A61K47/36 B65D65/46 C08K5/053 C08K5/103 C08K5/11 C08K5/21 C08L3/06 C08L3/08 C08L5/08 C08L77/04	
Composite antibacterial food packaging film and preparation method thereof	Jiang Shanxue Yao Zhiliang Cao Xinyue Wang Fang	Univ Beijing Technology and Business	CN113004568A	C08J7/14 C08K3/08 C08L5/08 C08L67/04	June 22, 2021
Antibacterial food packaging film and preparation method thereof	Yao Zhiliang Jiang Shanxue Cao Xinyue Shen Xianbao	Univ Beijing Technology and Business	CN112940333A	C08F2/00 C08F212/08 C08F212/14 C08F212/36 C08J5/18 C08J7/12 C08J7/14 C08L25/08 C08L25/18	June 11, 2021
Super-hydrophobic thermoplastic films for packaging	Forloni Roberto [IT] Dong Xin [US]	Cryovac Llc [US]	US2021284410A1	B65D65/42 B65D75/30 B65D81/20	September 16, 2021

Nano antibacterial gglucosyl composite particles and processing method and application thereof	Miao Ming [CNCN] Liu Yao [CNCN] Zhi Chaohui [CNCN] Zhang Tao [CNCN] Jin Zhengyu [CNCN]	Univ Jiangnan [CNCN]	WO2021098693A1	A01N43/16 A01P1/00 A23L3/3562 B82Y30/00	May 27, 2021
Vie dill oil—*Pleurotus eEryngii* polysaccharide antibacterial nanofiber as well as preparation method and application thereof	Cui Haiying Li Hong Lin Lin	Univ Jiangsu	CN112391696A	B82Y40/00 D01D5/00 D01F1/10 D01F8/16 D01F8/18 D04H1/728	February 23, 2021
Composite antibacterial food packaging film and preparation method thereof	Jiang Shanxue Yao Zhiliang Cao Xinyue Wang Fang	Univ Beijing Technology and Business	CN113004568A	C08J7/14 C08K3/08 C08L5/08 C08L67/04	June 22, 2021
Preparation method of antibacterial preservative film loaded with nanoscale essential oil	Li Xuehong Lu Yong Gao Suli	Univ Zhengzhou Light Ind	CN113045785A	C08J5/18 C08L5/00 C08L5/08 C08L5/16 C08L91/00	June 29, 2021
High-barrier antibacterial composite film based on nano ccellulose/mxene immobilized nano-silver as well as preparation method and application thereof	Wang Xiaoying Tang Shuwei Wu Zhengguo	Univ south China Tech	CN112662014A	C08J5/18 C08K3/08 C08K5/053 C08K9/02 C08L1/02	April 16, 2021
Low-silver-content fish scale gelatin—agar—AG NPs composite film as well as preparation method and application thereof	Zhang Jinli Chen Zihe Chen Yanting Ren Zhongyang Weng Wuyin Li Qingbiao	Univ Jimei	CN112430341A	A23B7/154 A23B7/16 A23L3/3562 C08J5/18 C08K3/08 C08L5/12 C08L89/00 C09D105/12 C09D189/00 C09D5/14	March 2, 2021

Continued

Table 24.3 The most recent patents published in 2021.—cont'd

Title	Inventors	Applicants	Publication number	IPC	Publication
PETG antibacterial composite film and preparation method thereof	Zhang Qigang Li Yuanhong Wang Weiwei Cai Wenbin	Henan Yinjinda New Mat	CN112812521A	C08J5/18 C08K3/08 C08K3/22 C08K9/12 C08L67/02 C08L67/04 D01F1/10 D01F6/62	May 18, 2021
Lignin nanoparticles as well as preparation method and application thereof	Xiao Lingping Sun Runcang Li Wenxin Li Xiaoying Xiao Wenzhe Guo Yanzhu Chen Xiaohong	Univ Dalian Polytechnic	CN112851977A	A61K8/72 A61Q19/08 B82Y30/00 B82Y40/00 C08J3/14 C08L97/00	May 28, 2021
***Ilex chinensis* Sims essential oil/*Lycium barbarum* polysaccharides antibacterial nanofiber as well as preparation method and application thereof**	Cui Haiying Li Hong Lin Lin	Univ Jiangsu	CN112391697A	B82Y40/00 D01D5/00 D01F1/10 D01F8/16 D01F8/18 D04H1/728	February 23, 2021
Calcium oxide nanoparticles and polymeric nanocomposite comprising same, method for producing same from natural solid food by-products/waste rich in calcium carbonate and method for preparing the polymeric nanocomposite, and use thereof as an antimicrobial agent in films for packaging food or coatings for various surfaces	Zapata Ramirez Paula Andrea [CL] Silva Mendez Cristian Moises [CL] Sepulveda Espinoza Francesca Antonella [CL] Oyarzun Luman	Univ Santiago Chile [CL]	WO2021046661A1	A01N25/34 A01N59/06 C08L23/00 C08L23/06	March 18, 2021

	Claudio Josue [CL]				
Preparation method for environmentally friendly nano-plating solution for tin plate and using method thereof	Liu Changsheng Wang Shuanghong Liu Fengchao Qi Jianjun Zhang Dabao Sun Chao An Zhenqiang Gong Zhiqiang Zhang Peng Li Jianping	Univ Northeastern Hbis group, Hengshui Plate Ind Hbis	CN111172524A CN111172524B	B82Y40/00 C23C22/36 C23C22/76	July 6, 2021
Biosensor using magnetic nanoparticles and detection device and detection method using same	Hyundoo Hwang [KR] Jaekyu Choi [KR]	BBC	US 20210033603 A1	G01N 33/54,346 (20,130,101); G01N 33/553 (20,130,101); G01N 27/3277 (20,130,101); G01N 33/54,333 (20,130,101); G01N 33/54,393 (20,130,101); G01N 27/3278 (20,130,101); G01N 33/5438 (20,130,101)	February 4, 2021
Colorimetric sensor for detection of food spoilage	Sablani Shyam [US] Rasco Barbara [US]	Univ Washington State [US]	US2021010942A1	G01N21/78 G01N33/00 G01N33/04 G01N33/14	January 14, 2021
Fabrication of an aptamer biosensor based on quantum dots for detection and elimination of salmonella	Wu Wei Sun Feifei Yang Qingli Tang Juan	Univ Qingdao Agricultural [cCN]	AU2021100518A4	C12Q1/6825 C12Q1/689 G01N21/64 G01N33/533 G01N33/569	April 22, 2021
Protein—polysaccharide—essential oil nanometer edible film and preparation method therefor	Li Songlin [CN] Yan Yongyong [CN] Cheng Jiayi [CN] Cheng Xiaoming [CN] Ye Hua [CN] Bai Qingyun [CN]	Huaiyin Inst Technology [cCN]	US2021309816A1	C08J5/18 C08L5/00 C08L89/00	7 October2021

Continued

Table 24.3 The most recent patents published in 2021.—cont'd

Title	Inventors	Applicants	Publication number	IPC	Publication
Soybean protein nano antibacterial film and preparation method thereof	Zhao Xiaotong Lyu Yichao Cao Jiahui Zhang Hong Song Guangshuang	Univ Northeast Agricultural	CN112280/315A	B65D65/46 C08J5/18 C08K5/053 C08K5/07 C08L1/02 C08L89/00	January 29, 2021
Preparation method of green, biodegradable, and multifunctional collagen-based nanocomposite film	Shi Jiabo [CN] Zhang Ruizhen [CN] Lv Siqi [CN] Cui Yu [CN] Cao Wenying [CN] Ma Jianzhong [CN]	Shaanxi Univ of science and technology [CN]	US2021309817A1	C08J5/18 C08K5/134 C08K5/3462	October 7, 2021
Method for preparation of thermoplastic polymer—cellulose nanofiber nanocomposite comprising suspension process and melt process using co-solvent system	Shim Jin Kie Kang Dongho Jeong Bich Nam Kim Gi Hong	Korea Inst Ind Tech [KR]	KR20210068641A	B65D81/24 C08J3/18 C08J3/20 C08J5/00 C08J5/18 C08K5/00 C08K5/10 C08L1/02 C08L63/04 C08L7/02	June 10, 2021
Application of lysozyme two-dimensional nanofilm as antibacterial material	Yang Peng	Univ Shaanxi Normal	CN109845761A CN109845761B	A01N63/00 A01N25/34 A01P1/00 A01P3/00 A01N63/50	April 6, 2021
Preparation and application of carboxymethyl cellulose and reduced graphene oxide—molybdenum disulfide—silver antibacterial film	Yu Yadong Wang Shanshan Li Jingchen	Univ Nanjing Tech	CN112175246A	B65D65/38 C08J5/18 C08K13/02 C08K3/04 C08K3/08 C08K3/30 C08K5/053 C08L1/28	January 5, 2021

analysis results, considering the need for adaptations of conventional methodologies and even the development of more sensitive analytical techniques. In this sense, many companies prioritize the use of compounds extracted from renewable natural biomass to minimize the chances of risk to human and environmental health, added to the low cost and easy access. Another strategy that AP and IP companies have most used is to not allow direct contact between the food and the packaging, as is the case with oxygen, ethylene, and aroma scavengers that are often isolated in the corner of the packaging. This fact also reduces costs, as the production line established in the company to produce packaging is maintained, and only an independent functionality is added to the packaging that a partner can manufacture. The cost of new packaging is always a key and decisive factor for investing in emerging technologies. In turn, it is related to commercial value and its potential for adding value. When the objective is to integrate an electronic device into packaging, in addition to the cost and complexity of research, there is an inherent concern in all packaging and new technologies to comply with the premises of carbon footprint reduction, circular economy, and sustainability.

The possibility of scaling biomolecules, interaction with other molecules in the packaging composition, and its stability over time must also be considered in the development of innovative packaging. This problem is common to synthetic molecules and nanoparticles that are also affected by environmental factors and may lose functionality before fulfilling their role of sustaining the quality and prolonging the shelf life of the product. This compatibility of the shelf life of the food and the functionality of the packaging must also be synchronized to avoid waste and, consequently, huge losses. When production and performance conditions are right, the result is a robust, integral, and functional packaging during transport and storage and an excellent consumer quality measurement and tracking tool.

24.8 Conclusion and future trends

Nanotechnology offers a variety of options in improving food packaging based on nanomaterials and is focused on improving food quality and safety to create new packaging functions, also making it possible to avoid loss in the food properties such as healthiness, taste, and nutritive aspects (Primožič et al., 2021).

The main opportunities of nanotechnology in food packaging refer to the release of active agents together with nanosensors (Cheng et al., 2022), such as in antimicrobial packaging that allow monitoring, management, and control of the condition in real time (Ghaani et al., 2016) and in cybersecurity for prevention techniques with effective defense-in-depth strategies and robust cybersecurity systems (Schaefer and Cheung, 2018).

When it comes to nanotechnology applied to food packaging, nanosensors coupled with artificial intelligence are a trend in the development of smart packaging (Soltani Firouz et al., 2021). However, future research is essential to cheapen the raw material and the production processes of these devices, in addition to investigating the following (Cheng et al., 2022): integration difficulties (Ghaani et al., 2016); size and portability (Sobhan et al., 2021); cost; and (Vilela et al., 2018) low accuracy (specificity, sensitivity, detection limit, and stability) (Alam et al., 2021). On the other hand, Petrucci et al. (Ismail and Sulaiman, 2021) noted that newer nanosensor designs are also becoming increasingly sensitive without significant cost increases with an exceptionally user-friendly interface (Petrucci et al., 2021).

The incorporation of nanocomposites must consider the synergism of the barrier and mechanical and thermal properties of the packaging and of the different food matrices to be packaged. This imposes interfering technologies that can lead to false negatives or cross-responses, particularly when it comes to bacterial contaminants. For example, most studies for detecting pathogens in food or environmental samples are tested in liquid butter solution; however, there may be gaps about their effectiveness in complex samples (Kaya et al., 2021).

AP and IP will be increasingly essential for consumers to have accurate information about the quality/safety status of food, especially for very perishable foods such as fresh fruits and vegetables, meats, and seafood, in this sense, future studies, using nanomaterials, should focus on lowering final costs and on commercially viable systems.

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