

Review

Applicability of Agro-Industrial By-Products in Intelligent Food Packaging

Silvia Amalia Nemes ¹, Katalin Szabo ¹  and Dan Cristian Vodnar ^{1,2,*} 

¹ Institute of Life Sciences, University of Agricultural Sciences and Veterinary Medicine, Calea Mănăştur 3–5, 400372 Cluj–Napoca, Romania; amalia.nemes@usamvcluj.ro (S.A.N.); katalin.szabo@usamvcluj.ro (K.S.)

² Faculty of Food Science and Technology, University of Agricultural Sciences and Veterinary Medicine, Calea Mănăştur 3–5, 400372 Cluj–Napoca, Romania

* Correspondence: dan.vodnar@usamvcluj.ro; Tel.: +40-747341881

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Abstract: Nowadays, technological advancement is in continuous development in all areas, including food packaging, which tries to find a balance between consumer preferences, environmental safety, and issues related to food quality and control. The present paper concretely details the concepts of smart, active, and intelligent packaging and identifies commercially available examples used in the food packaging market place. Along with this purpose, several bioactive compounds are identified and described, which are compounds that can be recovered from the by-products of the food industry and can be integrated into smart food packaging supporting the “zero waste” activities. The biopolymers obtained from crustacean processing or compounds with good antioxidant or antimicrobial properties such as carotenoids extracted from agro-industrial processing are underexploited and inexpensive resources for this purpose. Along with the main agro-industrial by-products, more concrete examples of resources are presented, such as grape marc, banana peels, or mango seeds. The commercial and technological potential of smart packaging in the food industry is undeniable and most importantly, this paper highlights the possibility of integrating the by-products derived compounds to intelligent packaging elements (sensors, indicators, radio frequency identification).

Keywords: smart packaging; by-products; antioxidant properties; indicators; sensors; zero-waste; food quality; shelf-life

1. Introduction

The food packaging concept arose with the desire of humans to conserve food for a longer time, and it was adapted gradually to the industrialization and commercialization processes [1].

Since the earliest times, people consumed fresh food on the same days when raw materials were hunted or reaped from the garden without any food preservation issues. With the evolution through time and the trend of the population to live in communities, the need for food preservation appeared and food packaging solutions were found. Glass is found as the first material for food packaging in written history, as a precedent of paper, which is now widely used in the food industry [1]. Moving to one of the most debated packagings of the contemporary era, plastic material is found in specific studies from the year 1870 [1]. Back then, brothers John W. Hyatt and Isaiah S. Hyatt had patented the first commercially available plastic material, which was a mixture containing pyroxylin and camphor used in the manufacture of objects such as dental plates or shirt collars [1]. The evolution of this packaging material is spectacular, reaching 380 million metric tons in 2015 globally, covering 40% of the materials used for packaging. From this ratio, 60% is used only in the food packaging industry, and the rest is used in areas such as healthcare, cosmetics, or household [2].

Regardless of specific domains (e.g., medical, pharmaceutical, automobile, construction), constant progress can be observed in all fields, as well in the food packaging industry, and a significant evolution is perceived, according to global digitalization. If in the past few years researchers were focused on developing new packaging materials, in the present moment, the spotlight relies on developing new packaging concepts, such as electronic devices (e.g., indicators, sensors) incorporated in the package, providing more information about the food inside.

Nowadays, the challenge in the food industry is to fulfill consumer demand, which is focused on healthy, fresh, and the least processed food products. With the need to have relevant options that enhance their lifestyle, concerning the shorter time for shopping in the supermarket, and less time spent cooking the food, consumers are also aware of the effects of packaging on the environment. Globalization has allowed many products from all around the world to be more accessible, and consumers are used to obtaining these products at any time and relatively at a low cost [3]. As a result of these reasons and others such as safety, marketing, or cost-effectiveness, producers pay much attention to the packaging they choose for the products.

At a global level, the food packaging area is one of the most advanced industries, which makes people think about the economic impact of this manufacturing sector [4]. Advancing this idea, the price of the packaging is found in the final price of the food, and consumers know this aspect. This is one of the reasons why it is difficult to introduce a new concept of packaging in the market place, only if its cost is lower or if the consumer is informed and knows its benefits [5].

In 2009, the food packaging industry was estimated at the value of US\$380 billion, and it is one of the largest packaging industries, representing more than 50% of all packaging industries at a global level. Plastic is the most imported packaging material, and it is evaluated at \$9.5 billion, followed by paper with \$4 billion, glass with \$1.6 billion, and wood \$0.3 billion. The food packaging market has a huge economic impact also on the developing countries taken in the study by The Food and Agricultural Organization of the United Nations (FAO), and its value was estimated at US\$15.4 billion [4].

Generally, the package of a product has a substantial role in its journey, beginning with transport, the distribution process, retailing, and most important, protection and preservation. With all these efforts, a report published by the World Packaging Organization shows that more than 25% of food products are wasted because of improper packagings such as inappropriate dosage, an absence of reclosing function, insufficient storage protection, or smaller package sizes [5]. To solve these specific problems strongly related to the global concern of food waste, the technologies of food packaging keep advancing. Furthermore, producers from the packaging market are permanently challenged to develop new packaging models in line with consumers' request, which is in a continuous changing process.

Consumer requests are in a continuously changing process; therefore, producers from the packaging market are permanently challenged to develop new packaging models. The latest trends on the supermarket's shelves are the ready-to-eat products with improved shelf life based on the newest methods, such as active, intelligent, and smart packaging concepts.

Besides the safety improvements for food quality or marketing solutions that smart packaging offers for consumers and producers, it has an impact also for the economic domain, especially due to the global market which is expected to be doubled until 2021 [6], and to reach US\$26.7 billion until 2024 [7]. For example, active packaging, which is characterized by an active function to extend the shelf life of the food product through bioactive compounds, was estimated in 2013 to occupy 26.9% from the global market of the packaging industry, which is evaluated at US\$6.4 billion, while intelligent packagings were estimated at US\$2.3 billion [8]. The most relevant countries that develop active and intelligent packaging are the US, Japan, and Australia. Figure 1 presents the growth rate predicted by 2026 [7].

The main purpose of food packaging is to prolong the shelf life of the food by protecting it from the outside environment [9]. Improving this aspect and maintaining the quality of the products supposed to use new strategic methods of fabrication or a new strategy of packaging. Various strategies can be used for improving the shelf life of a product, such as time-temperature devices, nanomaterials, the addition of chemicals, carbon dioxide emitters/absorbers, oxygen indicators, and barcode label

biosensors [3,10–12]. This kind of packing can be part of two categories, active or intelligent packing, and if these two methods are combined, it can be called smart packaging [13].

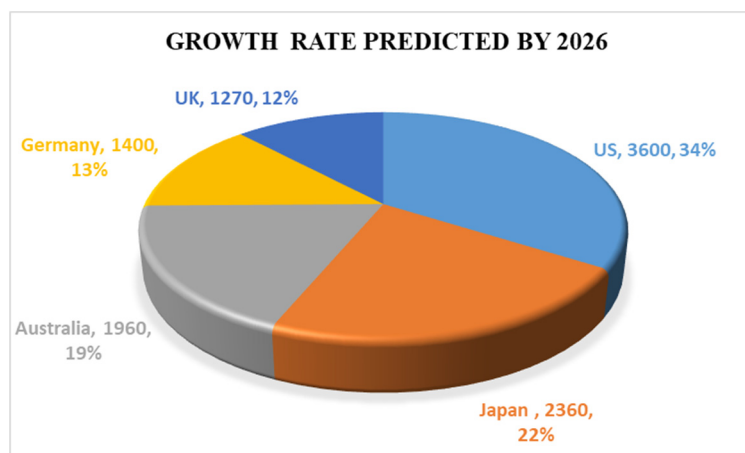


Figure 1. Global growth rate predicted by 2026 for the largest market (Data Sources from Ref. [7]).

The present review paper is focused on bringing answers and opening future research directions for the most common problems of the food packaging industry, such as food safety and control managing, food waste, and foodborne diseases. In addition, further attention is needed to the outstanding, unresolved, research issues such as reusability, biodegradability, and environmental protection against the waste resulted from food packaging industries. In this context, the present review points out possible solutions for developing alternative food packaging materials and recovering bioactive compounds from agro-industrial by-product sources that can support the “zero waste” agenda by integrating it in innovative food packaging.

The paper also aimed to make a classification of the newest smart packaging, including how this technology can be a solution for food waste and how compounds recovered from agro-industrial wastes can be used in active and intelligent packaging.

2. Concepts Definitions

Beginning with the middle of the 20th century, important evolution and major changes have been made in the development of new food packaging concepts, such as aseptic processing and packaging, modified atmosphere packaging, microwaveable packaging, and smart, active, and intelligent packaging systems [1]. Besides, the three advanced packaging technologies are all derived from the artificial intelligence technology associated with recent advances of computer science, which are found to serve in several practical applications, for example, porous materials [14], medical science [15,16], and the food industry [17,18].

Innovative packaging can be a way to transfer and apply intelligent science from the food industry to consumers. The newest concepts, active, intelligent, and smart packing exceed the usual attributes of a food pack. Intelligent packaging is considered a system that communicates with the consumer [19]. Active packaging is considered one of the most innovative concepts due to its precise functions that enable reading, feeling, seeing, or smelling the food inside [20]. Smart packaging is considered an update of the active pack to grow the industry of food products, especially the food packaging industry, with precious functions such as chemical and electrical-driven functions such as electronic displays storage temperature, self-heating or self-cooling containers, updated nutritional data, and the application of high-voltage pulsed electric fields [20–22]. Innovative packaging is a hot topic with an increased research interest; therefore, the concepts are described differently by scientists, and clear definitions have not been established yet. Some of these definitions are presented in Table 1.

Table 1. Definitions found in the literature for active, intelligent, and smart packaging.

Innovative Packaging	Definitions	Reference
Active packaging	- “active food contact materials and articles (hereinafter referred to as active materials and articles) means materials and articles that are intended to extend the shelf life or to maintain or improve the condition of packaged food. They are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food”.	[23]
	- A system that makes the interaction between the food, packaging, and the environment.	[24]
	- Packages that increase the shelf life of the food product by using natural compounds incorporated in the packaging.	
Intelligent packaging	- “ ‘intelligent food contact materials and articles’ (hereinafter referred to as intelligent materials and articles) means materials and articles which monitor the condition of packaged food or the environment surrounding the food”	[23]
	- Systems that provide the user with information on the conditions of food and should not release their constituents into the food.	[25]
	- A system adept to “sense, detect or record external or internal changes in the product”	[19]
	- A system added to packaging with the purpose of monitoring (food quality, critical control points) and giving information about the supply chain.	[3]
	- “The term “intelligent” involves an “ON/OFF” switching function on the package in response to changing external/internal stimuli, to communicate the product’s status to its consumers or end users”	[26]
Smart packaging	- “Innovative packaging system that combines the benefits of measuring, estimating, or predicting different aspects of food quality or safety with the release of an active substance that extends the product shelf life”.	[27]
	- The packaging capable of monitoring the changes that can appear in the product, packaging, or environment.	[24]
	- Upgraded packaging system with functional attributes that bring benefits to the food products and consumers.	[21]

The packaging has a role in preventing contamination of the food product and maintaining its freshness also. The food validity period can be determined depending on food product specifications such as the content of saturated and unsaturated fatty acids, enzyme activity, water activity, pH, or protein content [10,28–30], and the packaging type used as a barrier layer from contamination [19].

The shelf life of a food product is determined as the time between when the product was packaged and the last day that it can be consumed without any health risks and at the same quality. The expiration date can be limited by numerous intrinsic and extrinsic factors that are presented in Table 2, which can influence sensorial, textural, and microbial characteristics [13].

Table 2. Intrinsic and extrinsic factors that can influence the shelf life.

Intrinsic Factors	Extrinsic Factors	Reference
Water activity	Time–temperature profile	[9]
pH value	Temperature control	[31]
Redox potential	Relative humidity	[13]
Available oxygen	Exposure to light	[32]
Nutrients	Microbial environment	[32]
Natural microflora	Environment in packaging	[9,13]
Biochemical products	Heat treatment	[13]
Preservative	Purchaser handling	[13]

Distance between the place of production and place of sale is another issue raised with globalization that accentuated the drawbacks related to shelf life. Most of the foods are perishable and for this reason, producers have chosen different methods to improve the product’s shelf life such as cooling, heat treatment, or modified atmosphere [33]. With all these, the microorganisms are not eliminated, and they stimulate reducing the quality and food safety. To solve and control this issue, the concept of active, intelligent, and smart packaging was developed.

3. Smart Packaging

According to many studies, smart packaging is defined as the packaging that includes both active and intelligent systems acting synergistically, as illustrated in Figure 2. It is capable of monitoring the changes during storage (increases/decreases in temperature or humidity) and acts to slow down the quality degradation. Using the compounds from active packagings, such as antioxidants, emitters of carbon dioxide, antibacterial agents, humidity, ethylene, and oxygen scavengers together with intelligent devices has obtained the concept named “smart packaging” [7,24,34,35].

Smart packaging includes devices that are capable of heating or cooling food inside and show in real time the nutritional information on the electronic display [21]. Some of the smart packaging belonging to the canning and beverage industries has been shown in a study [36]. A device incorporated in the package that can change the temperature of the food inside was developed for bottles, cans, or carton packages. It can lower the product’s temperature with 18 °C in a short time (two or three minutes) before consumption. The principle of this packaging is based on absorbing the heat from the liquid inside, using the vapors obtained by releasing from a vinyl bag a quantity of pressurized water that evaporates immediately [37].

Another example of smart packaging is dedicated to heat coffee, tea, soup, and hot chocolate cups. In this case, the exothermic reaction between water and calcium oxide is the basis. Inside the cup, calcium and water are placed separately. Consumers are asked to invert the cup and mix the components, thus activating an exotherm reaction. The material that the cup is made of allows keeping the temperature for approximately 20 min [36].

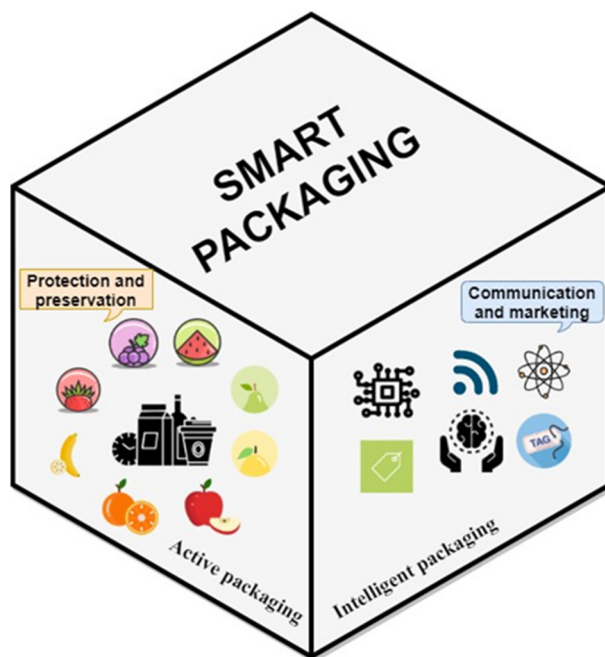


Figure 2. Smart packaging concept.

A new principle improved is an electronic tracking tag, which helps to have better control on the food distribution chain. Applying the Radio Frequency Identification (RFID) technology was advanced as a pack that is useful to prevent fraud, decrease economical losses, and enhance the fundamental operation such as storage, distribution, and marketing. RFID packaging can be considered smart packaging only if it is used alongside a sensor or biosensor that can provide the specific location of the product [35].

Oxygen-absorbing technology involves the combination of oxidation of certain compounds (iron powder, ascorbic acid, photosensitive polymers, enzymes), which are able to reduce the oxygen level inside the package, from the regular value between 0.3% and 3%, which is found in the conventional packaging systems such as modified atmosphere, vacuum, or substitution of internal atmosphere to an oxygen value of 0.01% [30]. Ascorbic acid and sodium bicarbonate were used for the development of a smart oxygen-absorbing label used in the meat industry. It is working to release carbon dioxide while absorbing the oxygen from the packaging, which is a reaction that preferably happens only when it is required. This action offers protection and prolongs the shelf life of the food inside. It can be made to operate at different temperatures (refrigeration or freezing) and a specific humidity [38].

The area of smart packaging is in full development and can provide solutions to reduce foodborne diseases and also help to reduce environmental problems. Research in this field is growing, evolves, and matures continuously, and smart packing is expected to improve food safety and quality soon.

4. Active Packaging

The concept of active food packaging is defined as innovative packaging that interacts with the food and absorbs derived chemicals to prolong the shelf life and ensure safety and quality at the same time. Active packaging creates a barrier between food and environmental space and stays in touch with the food inside, offering protection [29]. An important characteristic of active packaging is the capability of maintaining a low concentration in oxygen to slow the lipid oxidation. A low oxygen atmosphere can be achieved by the incorporation of antioxidant compounds (that act as protectors against oxidation processes) such as vitamins C and E [11], propolis, tocopherols, or plant extracts [29].

Overall, active packaging involves active compounds from different sources that can be in contact with the food or can be incorporated in the coating material [8]. Active compounds should be safe for

human health; if combined active compounds are used, the mixture must be evaluated and authorized by the European Food Safety Authority [25].

4.1. Nanomaterials

The latest trends in the active packaging industry are to develop new active materials for preventing degradation and maintaining the quality for a longer time [28]. Good candidates to evolve in this direction are nanomaterials given their mechanical, optical, thermal, and antimicrobial attributes [11].

Nano packaging materials have an advantage in the food industry due to the protection they can assure against the foodborne pathogens as *Escherichia coli*, *Staphylococcus aureus*, and *Salmonella*, which are known for food poisoning [39]. Researchers have claimed that nanotechnology can solve the most common food poisoning symptoms such as fever, diarrhea, nausea, vomiting, and abdominal pain, which can even cause death in the case of children, pregnant women, and old people [40–42]. According to many research papers, nano packaging can also come with solutions for reducing environmental waste by using bionanocomposites (chitosan, starch, alginate, carboxymethyl cellulose, pectin) [43–45]. To make biocomposite materials an efficient solution for the food packaging industry, they should be improved for having high antimicrobial activity, better mechanical properties, and gas barrier functions. These improvements are possible using nanotechnology and active and intelligent packaging solutions [46]. A good mechanical amendment and antimicrobial activity for the most common pathogens found present in food were identified as zinc oxide (ZnO) nanoparticles and titanium dioxide (TiO₂) nanoparticles [39,43]. Both biocompatible materials have been tested for their efficacy against *Salmonella typhi*, *Klebsiella pneumoniae*, and *Shigella flexneri*. The results showed that TiO₂ and ZnO have inhibitory action against the bacteria mentioned above [39,47].

4.2. Polymers

The research on sustainability and the circular economy bring to the front the zero-waste processes that require the re-integration of by-products as a priority for the zero-waste agenda [48]. In this context, many studies have been made for developing new methods to integrate compounds extracted from by-products in usual technologies, such as food packaging.

The most important fact is that those compounds can be recovered from food by-products, such as seafood by-products (heads, gills, skin, trimmings) [49,50] for chitosan extraction, and fruit and vegetable industries by-products (pumpkin seeds and peels, grapefruit peel, sunflower head, sisal waste, pomegranate peel, eggplant peel, sour orange peel) for alginate and pectin [51–54]. They present high properties of biocompatibility, bioadhesion, and biodegradability, which is why their applicability is persistently growing [44].

As a coating material, a mixture of chitosan with 0.5% apple peel polyphenols or chitosan–proanthocyanidins combination has shown better mechanical properties and tensile strength [55,56]. Better flexibility and water resistance presented the films based on chitosan and grape seed extract or chitosan and apricot kernel essential oil in a ratio of 1:1 [57,58]. The application of starch in the packaging industry is limited by the disadvantage of reduced plasticity [59]. A mixture form by urea and ethanolamine can act as a plasticizer to prepare thermoplastic starch and succeeds to growth its mechanical properties and solve the issue of low plasticity [60]. Packaging mixtures presented behind do not cause environmental problems, are biodegradable and not toxic, and can be obtained from sustainable sources [61,62].

4.3. Antioxidants

Polyphenols are parting from the class of phytochemicals, and they can be found in plant-derived products such as coffee, tea, wine, fruits, vegetables, or chocolate [63,64]. They can be recovered from plant residues such as peels, bran, husks, and skins, which usually are wasted in their processing [65,66]. Many studies highlight the benefits that polyphenols can bring to human health, beginning with the preventive effects against cardiovascular disease [67–69], anti-inflammatory, anticoagulant, anticancer, and antioxidant proprieties [70–76]. For those functions and others (e.g., the prebiotic effect [64]), polyphenols are used in several areas such as drugs and the food industry or packaging area.

However, active packaging is still a studied area with great potential, which can improve the food packaging industry considerably.

5. Intelligent Packaging

The food industry is in continuous progress and one of the most revolutionary concepts developed recently is intelligent packaging [26]. Intelligent packaging can be represented by a small tag that is capable of monitoring the quality of food and can notice the consumer if there is a contamination problem with the food product [77]. In comparison with active packaging, intelligent packaging has the advantage to communicate directly with the consumers through an incorporated device [78].

With the technology evolution, indicators used in intelligent packaging were classified into two categories: indirect indicators and direct indicators, as presented in Figure 3. Researchers are now focused to develop more the second category (direct indicators) because of their ability to maintain the quality of the product and also to give more targeted information about volatile compounds of microbial origin, biogenic amines, toxins, or pathogenic bacterias [3].

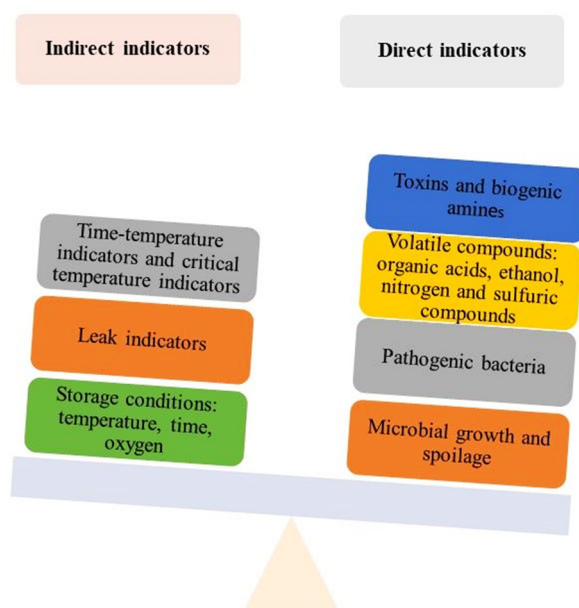


Figure 3. Food quality and safety indicators used in intelligent packaging.

The latest consumer demands of the 21st century are the products that can help them saving time, such as “ready-to-eat” and “heat and eat” meals. Another requirement registered is represented by the products’ microwavable, easy opening, reusable characteristics [79]. An easy-to-use food package label (Figure 4a) for pork was developed using pH indicators (bromocresol purple, bromothymol blue, and a mixture of bromothymol blue and methyl red) for monitoring freshness and shelf life [80]. A self-adhesive label, named Fresh-Check (Figure 4b), was developed to ensure consumers about the freshness of the perishable food products. “The active center circle of the Fresh-Check darkens irreversibly, faster at higher temperatures and slower at a lower temperature”; in this way, consumers

can know for sure if the product is fresh or not [81]. Another study has developed an on-package colorimetric sensor label (Figure 4c) for monitoring the ripeness of the apple after packing. This sensor was able to detect the aldehyde emission, in solution or vapor, of apples based on Methyl Red. As the apples mature, the sensor label changes its color in yellow, orange, and red in the end [82].

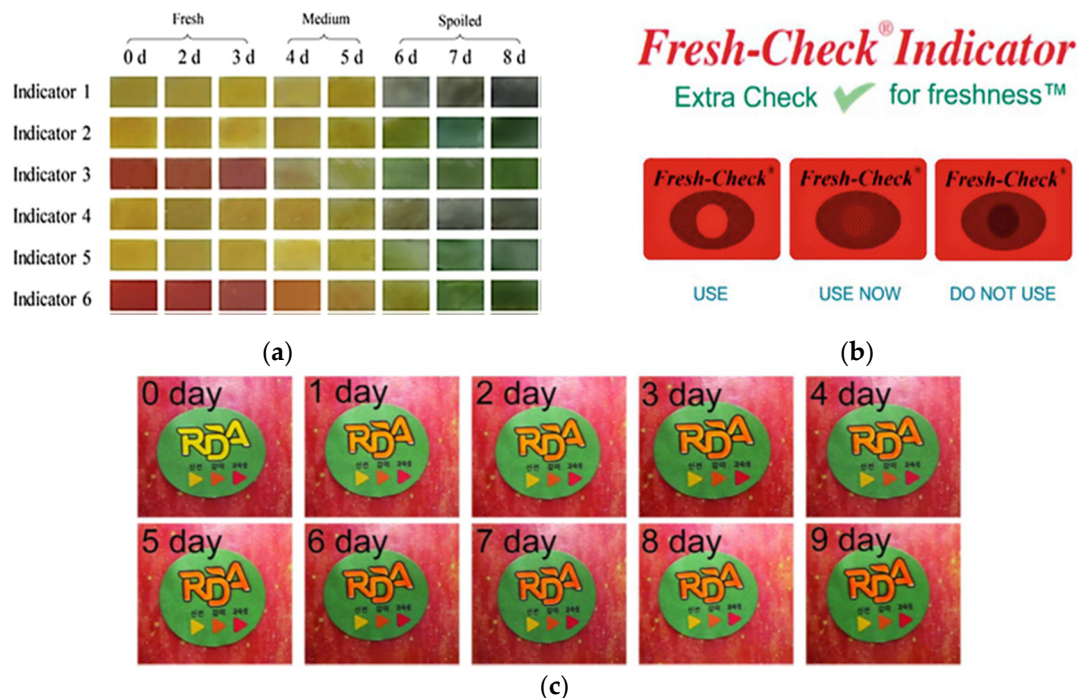


Figure 4. (a) Color changes of indicator labels for lean pork, (b) Fresh-Check label, (c) Color changes of a sensor label after exposure to apple flavor.

However, carefully watching the action of the intelligent labels, an important aspect to note is that these labels used as indicators are in direct contact with the food, which can be dry (coffee, fruits, and vegetables, bakery ware), liquid (beer, beverage), or semisolid (meat, fish). The migration of compounds from food contact labels according to food safety needs to be tested agreeing to the specific European directives (Regulation 450/2009) that regulate that active and intelligent packaging materials require authorization [3].

Intelligent packaging can give information about the state of food inside and can present the entire lifecycle of the product beginning with packing and distribution up to selling [78,83]. It can be used along with active packaging for watching the effect of the active compounds and their efficacy.

Intelligent packaging has a specific role: to improve safety and quality issues and to control the traceability of the food products. To fulfill this task, systems such as sensors, indicators, and Radio Frequency Identification (RFID) are used [26].

5.1. Sensors Used in Food Packaging

Sensors are one of the most studied electronic devices from the intelligent packaging field. Their main function is to detect and convert a signal form to another, using a transducer [19]. Sensors can be classified into active or passive sensors. If the transducer needs external power for measurement, the sensor is active; if it measures without help, the sensor is passive [24]. A traditional sensor is capable of measuring parameter changes such as temperature, pH, humidity, light exposure, or color changes. Research is advancing in the improvement of chemical sensors that can monitor package integrity and food quality and safety [19,24,84]. Among the chemical sensors found in the scientific literature are those detecting volatile organic compounds, compounds with high sensitivity, and gas molecules (H_2 , CO, NO_2 , O_2 , H_2S , NH_3 , CO_2 , CH_4), which have high importance to the food sector [24].

Biosensors are part of chemical sensors that differ by the biological components used as detectors such as cells, antibodies, bacteria, yeast, fungi, plant and animal cells, biological tissue or enzymes, which are obtained by isolation and purification from biotechnological processes [24]. The most successful type of biosensor is the glucose sensor for diabetics, which is from the medical part, but they are found also in the pharmaceutical industry, food and process control, environmental monitoring, defense, and the security area [85].

In the intelligent packaging industry, chemical sensors, biosensors, and others can be used and incorporated in films. Flex Alert Company Ltd. with Vancouver-based partners has developed a commercially available biosensor that is capable of detecting pathogens (*E. coli*, *Salmonella*, and aflatoxins) in coffee beans, dried nuts, seeds, wine barrels, and fresh fruit. An active biosensor, using wireless communication, alerts manufacturers, consumers, and distributors to some toxin presence by sending information in real-time during storage and in packaged products [86].

Following the potential of sensors in the food industry, it can be seen that they may be a solution for combating food waste and also for reducing the risk of diseases caused by altered foods.

5.2. Indicators Used in Food Packaging

Indicators are part of the intelligent packaging group of devices. They have a different method of action than sensors do, by providing immediate qualitative visual information such as color change, color intensity change, or diffusion of colors, which are irreversible changes about the product [24,87]. Indicators are the most commonly used in commercial form, and a few of them are presented in Table 3.

Table 3. Commercially available indicators for intelligent packaging.

Type	Trade Name	Manufacturer	Information	Reference
Gas indicators	O ₂ Sense	Freshpoint Lab	<ul style="list-style-type: none"> - aims to alert consumers, producers, or sellers if the integrity of the packaging is damaged by detecting oxygen inside, which can lead to the degradation of quality and safety of the product inside. - the indication is based on a color change. 	[24,87,88]
	Novas	Insignia Technologies Ltd.	<ul style="list-style-type: none"> - specially made for products packed in plastic material, in a controlled atmosphere. - when the packaging is degraded, the pigment used as an indicator changes its color. 	[24,87]
	Ageless Eye	Mitsubishi Gas Chemical Inc.	<ul style="list-style-type: none"> - an in-packaging indicator that monitors the presence/absence of oxygen. - if the oxygen level is 0.1% or less, the color of the indicator is pink; when the oxygen level attains 0.5% or more, the color indicated will be blue. 	[88–91]
Freshness indicator	Freshtag	COX Technologies	<ul style="list-style-type: none"> - fish and seafood products are emitting a special odor (caused by volatile amines) when they lose their freshness. - the dye-based indicator interacts with the odor-causing chemicals and produces a change color reaction. - the intense pink color created indicates the lack of freshness. 	[24,92]
	Sensorq	DSM NV and Food Quality Sensor International	<ul style="list-style-type: none"> - it is placed inside of the packaging. - is applied for meat and poultry products. - detect the gaseous by-products of bacterias that cause food poisoning. 	[87,93]
Time-temperature	Timestrip	Timestrip UK Ltd.	<ul style="list-style-type: none"> - it monitors time and temperature for products where temperature can be a critical control point. - the adhesive label is applied to the food packaging and once that is activated, it starts to monitor the temperature (days, weeks or months). - the device records the time when the temperature recommended was not respected. - it is applicable for fresh seafood, fresh produce, airline catering, school meals, home delivery diets, food retailing, restaurants, hub, and spoke production. 	[87,94]
	Monitormark	3M	<ul style="list-style-type: none"> - it is used as part of the secondary packaging and monitors the storage and transportation condition. - it is an adhesive label that easily attaches to the packaging and visually shows exposure and relative time over which exposure happened. - it has an irreversible action, even if the temperature during storage or delivery returns to a normal value. - it is applied for food products such as bakery, beverage, confectionery, and meat products that have critical temperature points beginning with −15 to 26 °C. 	[19,87,95,96]

Table 3. Cont.

Type	Trade Name	Manufacturer	Information	Reference
Time–temperature indicators	Onvu	Ciba Specialty Chemicals and FreshPoint	<ul style="list-style-type: none"> - the system monitors the freshness of a food product and is specially made for products sensitive to temperature. - after the label is activated, it becomes dark and then grows progressively lighter with time if the temperature rises. - the product has reached the end of the shelf life when the color reaches the reference color tone. - is commercially available for products as meat, fish, and dairy products with a shelf life of 5–6 days at 5 °C. 	[97,98]
	L5-8 Smart TTI Seafood Label	Vitsab	<ul style="list-style-type: none"> - specially made for seafood products and is improved for the recognition of <i>Clostridium Botulinum</i> toxin formation; - the second most common pathogen (25%) in seafood is <i>Clostridium Botulinum</i>, and its growth and multiplication are directly influenced by temperature, for example, at 60 °F, multiplication is 10–12 times more rapid than at 40 °F or at 72 °F, it is even 25–27 times more rapid than at 40 °F. - this label is based on an enzymatic reaction, which is given by the enzyme mix and substrate from the center of the label; the mixture is activated by applying moderate pressure on the “window” and is recognized by a homogenous green color in the “window”. - the shade of green color can be changed in four ways (25%, 40%, 65%, 85%), and this is correlated with the ending of the product’s validity. - if the green color changes and is replaced by an orange color (100%) or red (120%), it means that the product is no longer safe for consumption. 	[24,99,100]
	Cook-Chex	Pymah Corp	<ul style="list-style-type: none"> - it comes in the form of a cardboard tag. - the label contains a purple color chemical indicator (chromium–chloride complex) which, at various conditions of time and temperature in a pure steam atmosphere, changes its color to green. - this type of indicator is used commercially to verify the sterilization or autoclaving operation in the canning industry. 	[87,101]
	Evigence sensors	EVIGENCE SENSORS	<ul style="list-style-type: none"> - it is a visual indicator that undergoes color changes after exposure at a higher temperature than the one established by the manufacturer. - the speed of changing the color of the indicator (from silver to white) increases directly in proportion to the increase in the temperature of the product’s the environment. - indicator labels can be calibrated for products with a shelf life from some hours to several years. - labels are improved so that their application and activation is automatically done on the food packaging line. - using the SMART DOT™ app for mobile, consumers can scan the label before, during, and post-acquisition, for having more details (the remaining shelf life, time to repurchase). 	[102,103]

Table 3. Cont.

Type	Trade Name	Manufacturer	Information	Reference
Time-temperature indicator/freshness indicator	Fresh-Check	Temptime Corp	<ul style="list-style-type: none"> - monitors the food in the store or at home. - it is an adhesive label, which has a central reactive part, represented by a circle; the circle darkens at an accelerated rate if the product is not stored at the appropriate temperature, and consumers can see the freshness of the food products in real time. 	[19,104]
Disinfection Indicator	Thermostrip DL	LCR HALLCREST LLC	<ul style="list-style-type: none"> - a waterproof and self-adhesive label that irreversibly changes its color depending on the temperature of dishware in dishwashers. its main purpose is to indicate if the proper dishwasher temperature has been reached for sanitation. - it can be used also as proof for HACCP (Hazard Analysis and Critical Control Point). The temperature range covered by the indicator is beginning with 29 to 290 ° C. 	[105]

5.3. Radio Frequency Identification

Radio Frequency Identification (RFID) is an automatic identification technology based on remote data storage and retrieval using devices named labels, which are improved with wireless sensors for product identification. The RFID system is made of specific elements presented in Figure 5. In the food industry, RFID technology is part of the intelligent packaging sector. An RFID tag tracks food traceability in real time and monitors the entire cold chain for food applications (e.g., intercontinental fresh fish logistic chain) [106].

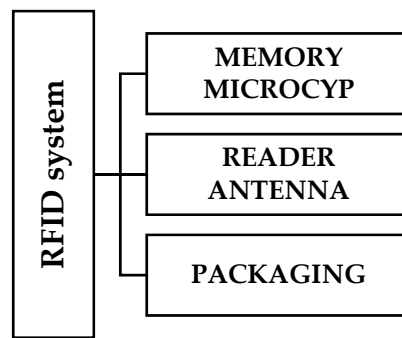


Figure 5. Radio Frequency Identification system elements.

RFID tags can be part of two categories, passive and active tags; the category depends on the use or not of external or integrated batteries for the transfer of information in the memory microchip. Passive tags are using a magnetic field created between the reader antenna and tag; thus, the intelligent tag takes the energy and transfers the encoded data to the memory chip of the label. Active RFID tags are using batteries for energy power to make the circuit between the microchip and the reader antenna [87]. Taking back to first principles, the working process of the RFID system is explained in Figure 6.

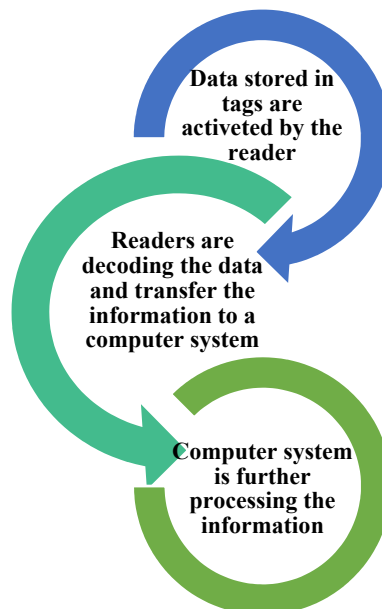


Figure 6. The working principles of an RFID system.

Therefore, RFID tags can increase the efficiency of supply chains and provide suppliers with an advanced method of food product monitoring until they become available to consumers.

6. Food Packaging Materials

6.1. Typical Packaging Materials

Since the Industrial Revolution from the 18th–19th century, industries had lost the value of reusing the packaging or the packaging material [24]. Over the years, this fact caused unwanted waste, which created much damage to the environment. The Food and Agricultural Organization of the United Nations (FAO) made a short report and a SWOT (Strengths, Weaknesses, Opportunities, and Threats) analysis of the food packaging industry in developing countries. They have presented a global view of this industry, with special attention in its size, structure, and the materials used. The most applied materials for packing are represented by paper, rigid plastic, metal, and glass, as seen in Figure 7 [4].

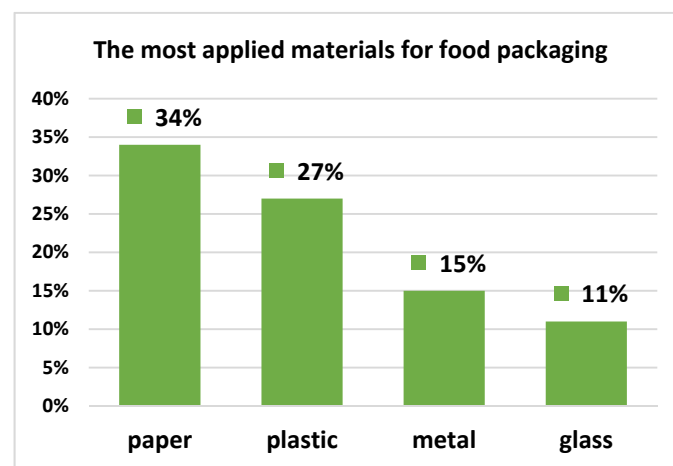


Figure 7. The most applied materials for food packaging (Data Sources from Ref. [4]).

In 2015, an amount of 322 M tons of plastic waste was registered, of which 49 M tons was from the food packaging industry [43]. Packaging materials used in the modern era such as glass, metals, paper, plastic, and alternative biomaterials are presented in Table 4 with their advantageous and disadvantageous characteristics.

Table 4. The advantages and disadvantages of typical and alternative packagings.

Advantage		Disadvantage	References
Typical Packaging Materials			
Glass	Inorganic and chemically inert;	High manufacturing temperatures; Big energy consumption at manufacturing; High net mass; Breaks easily and it is nonbiodegradable.	[1,13]
	Transparent, recyclable and it does not pollute;		
	Easy to model;		
	It can be sterilized at high temperatures;		
	It offers protection against water and gas vapor;		
	It does not deteriorate by oxidation or other forms;		
	It does not react with food or the environment.		
Metals (steel, tin, chromium, and aluminum)	It is a barrier to gasses and humidity;	Has high costs of manufacturing; Energy intensive;	[1,13]
	It can be sterilized and it is recyclable;		
	Covers a larger area of the food industry;		
	Low toxicity;		
	Good mechanical strength and resistance to working.		
Paper and cardboard	Low costs, low weight, recyclable, and biodegradable;	Barriers to moisture and vapor are weak; Shock protection is low.	[1,13]
	The resulting wastes can be reused for energy recovery;		
	It has good printing properties.		
Plastic	Economically efficient;	Replaces glass, metal, and paper packaging; Pollute the soil; It is not degradable and harms marine life; The debris cannot be eliminated and can be ingested from seafood.	[1,13]
	Good mechanical properties and a barrier to gasses and humidity;		
	It is flexible, transparent, or can be colored.		
Alternative Packaging Materials			

Table 4. Cont.

	Advantage	Disadvantage	References
Chitosan-based biofilms	Non-toxic and biodegradable;	Restricted scope.	[13,107–109]
	Environmental friendly;		
	Antimicrobial and bacteriostatic proprieties;		
	Preserve the taste;		
	Improve physical, mechanical, and chemical properties;		
	Maintain tissue firmness; inhibit the increase of respiration rate.		
Starch-based biofilms	Frequency in nature, renewable;	High water solubility; Insufficient water barrier properties; Weak mechanical properties; Reduced plasticity.	[1,59,107,110]
	Environmentally friendly;		
	Low cost, non-toxic;		
	Biodegradable;		
	Suitable for foods with low humidity (confectionery and biscuit trays).		
Films and coatings based on mango by-product	Good properties of permeability;	Low surface tension.	[111,112]
	Color stability;		
	Antioxidant activity;		
	Hydrophobicity;		
	Decreases in manufacturing cost.		

6.2. Alternative Materials

Alternative biomaterials can refer to different compounds (e.g., poly-lactic acid, starch, cellulose) incorporated in coating materials. The most important aspect of the food industry is the material used for packaging. To solve the issues of plastic pollution caused by the food packaging or pharmaceutical industries, the researchers on this domain are focused on finding alternatives packaging solutions [43].

The latest trends are to use bioplastic such as biopolymers and also plant extract incorporated for their antimicrobial properties [107]. Natural biodegradable polymers are obtained in a complex metabolic process. Biopolymers belong to the class of proteins, polysaccharides, lipids, phenolic compounds, and other classes, and they can be extracted from biomass or obtained from the vegetal, animal, or microbial sources presented in Figure 8 [45].

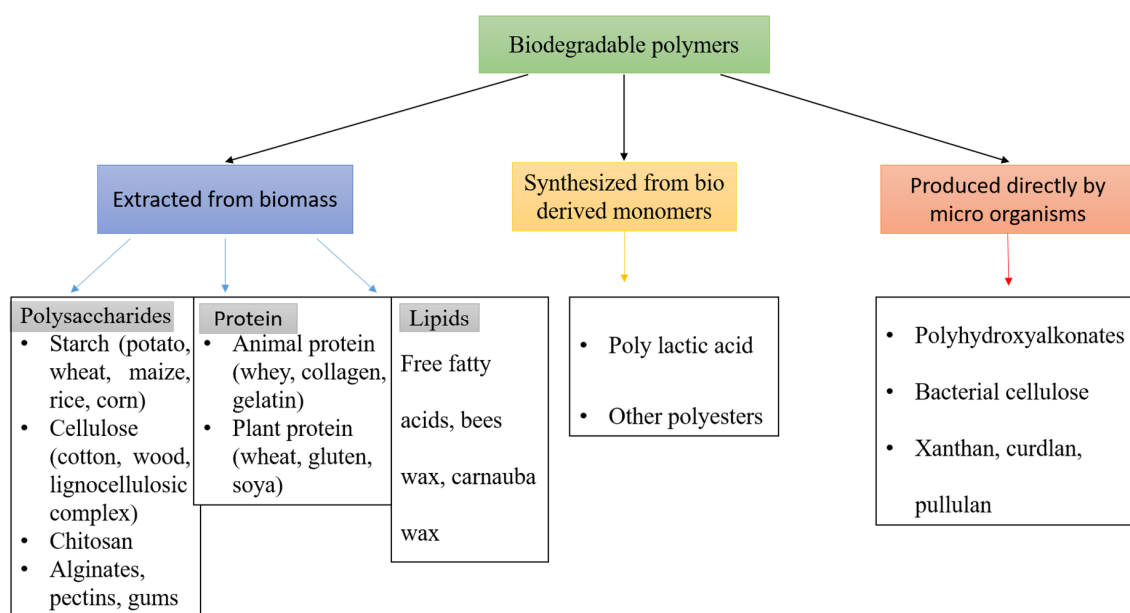


Figure 8. Classification of natural biodegradable polymers.

Starch is one of the most used natural biodegradable polymers at the moment, and it is being part of the carbohydrates class; its chemical structure is presented in Figure 9. The most important sources of starch are cereals (wheat, rice, corn) and tubers (potato) [107,113]. Starch contains two fundamental compounds: amylose, which is responsible for the structure and the rigid proprieties, and amylopectin, which helps digestion through its precise functions such as the enhancing of solubility of the polymer [114]. More than this, using starch-based biofilms is offering food package antioxidant activity, especially if gallic acid is added, 0.3 g/g starch [110]. A Systematic Review and Meta-Analysis has also shown recently the potential of using potato starch in the development of biofilms used for food packaging because it is an important raw material derived from many industries such as fruits and vegetable processing, has a low cost, and is the most wealthy biomaterial [115].

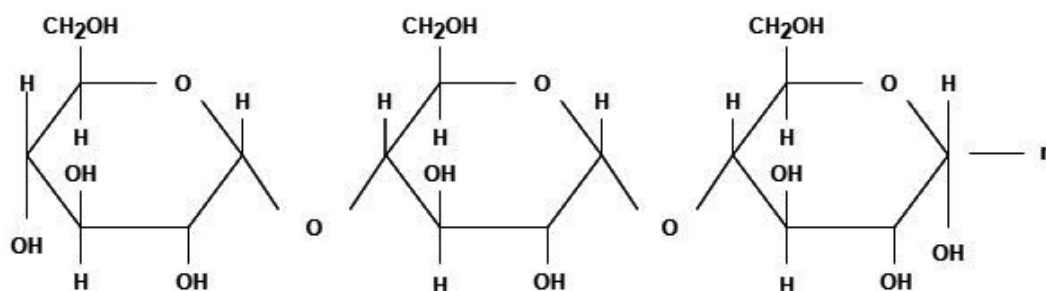


Figure 9. Chemical structure of polymeric starch.

Carotenoids are a part of the lipophilic pigment class. The most well-known sources are red, yellow, and orange fruits and vegetables, especially carrots, tomato, watermelon, and some species of fish such as salmon and crustaceans, principally cooked lobster and crab [61,116]. In addition to other pigments such as chlorophyll, β -carotene, anthocyanins, and lycopene, which are presented in Figure 10, β -carotene is added in the composition of biofilms used to make food packaging. The great attention of the researchers received the importance of β -carotene for its proprieties of excellent natural antioxidant and colorant products [117]. The encapsulation of β -carotene is used for obtaining active biodegradable packaging films. The presence of β -carotene has proved better thermal protection for the packaging films and greater protection against the oxidation process [61]. A recent study has developed a poly-lactic acid (PLA) film with three different compositions, using lycopene, β -carotene, and bixin. The standard curves of the three compounds were made in the following concentrations: β -carotene ($\lambda_{\max} = 449 \text{ nm}$), from 2.2 to 105.4 $\mu\text{g mL}^{-1}$ ($R^2 = 0.999$); lycopene ($\lambda_{\max} = 480 \text{ nm}$), from 0.3 to 13.9 $\mu\text{g mL}^{-1}$ ($R^2 = 0.999$); bixin ($\lambda_{\max} = 457 \text{ nm}$), from 0.3 to 5.3 $\mu\text{g mL}^{-1}$ ($R^2 = 0.996$). All types were destined for sunflower oil oxidation protection [118]. The carotenoids were progressively released to the food simulant, the attainment of approximately 45% release of β -carotene and lycopene, and approximately 55% release of bixin. The PVA films improved with β -carotene and lycopene have proved a good barrier against light and oxygen, and PVA films with bixin presented the best antioxidant activity for sunflower oil [118].

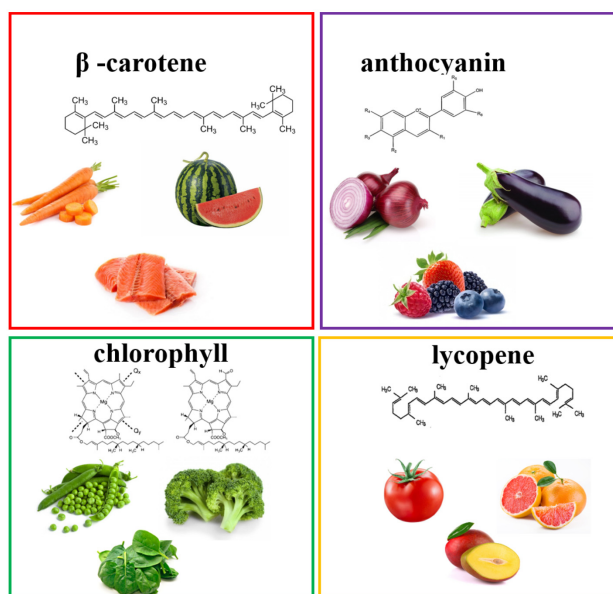


Figure 10. Bioactive pigments and their chemical structure.

Intelligent and active packaging has been obtained by poly(vinyl alcohol) (PVA) films mixed with chitosan, itaconic acid, and tomato by-products extract (TBE) [117]. TBE was used in another research study as a natural pigment due to its carotenoid content [119]. TBE is rich in carotenoids (lutein, lycopene, and β -carotene), an content of 3.273 mg/100 g DW, and phenolic compounds (e.g., caffeic acid–glucoside isomer, 5-caffeoyl quinic acid, quercetin-diglucoside), the total content of 80.596 mg/100 g DW [117]. This active film has proven to have increased physical properties (diameter, thickness, density, weight), inhibition effects against bacterias such as *S. aureus* and *P. aeruginosa*, and antimicrobial activity to *S. aureus*, *E. coli*, *P. aeruginosa*, *S. enterica* Enteritidis, and *S. enterica* Typhimurium [117].

The literature shows that chlorophyll is recommended to be used as a colorimetric temperature indicator for 50 to 70 degrees in chitosan films, along with anthocyanin, which is capable of detecting a temperature variation in intelligent packaging due to the property of changing its color depending on the temperature [34,120].

Another bioactive compound from the pigment class with high antioxidant activity that can be incorporated in active packaging is lycopene [116]. The most known natural sources are tomatoes, especially the peels [121], but it can be found also in guava or papaya [122]. An important aspect is that lycopene can be extracted from the tomato product waste [123]. A study shows that tomato processing by-products contain up to 0.1% lycopene and other bioactive compounds such as tocopherols and other carotenoids [124]. A research paper presented a method to replace synthetic oil-based stabilizers with natural antioxidants compounds obtained from tomato and grape seeds recovered from the tomato and wine industries [125].

By-products can be recovered from food waste, which represents a high-value source of functional components such as proteins, fibers, polysaccharides, phytochemicals, lipids, and fatty acids [126]. One of the best-known by-products that is very frequently used in the world is bran resulting from the grain industry. Bran has a low price, great availability, and multiple functional properties (antioxidant and anti-inflammatory activity) due to the high content of phenolic compounds, fibers, and minerals [127].

A by-product that can be used from the winery waste is grape marc. [128]. Globally, the production of grapes, achieved in 2017 an amount of more than 77,000 tons and only the pulp is used for the wine production. The rest (seeds and peels) forms the grape marc abundant in epicatechin, catechin, gallic acid, procyanidins, and phenolic acids, which makes it have good antioxidant and antimicrobial activity [129].

Globally, banana production is registering an amount of 102 tons annually, of which 35% is only the peel, which is considered waste. One study developed an antioxidant chitosan biofilm improved with banana peels extract, which is fraught in bioactive compounds. The active biofilms were particularly made for maintaining the postharvest quality of apple during storage [109]. The by-product of mango is another example of a fruit that is used for developing active films for the food packaging industry. The seeds of mango had a large concentration of bioactive compounds, while the peel is rich in polysaccharides. The film created present good properties of permeability, color stability, antioxidant activity, and greater hydrophobicity [112].

7. Future Perspectives

Intelligent food packaging is an area with great potential for increasing the food packaging sector, providing fast, inexpensive, and efficient ways to monitor the environmental conditions of food in the supply chain. The next generation of food packaging developments and food packaging materials must be more environmentally friendly, and more importantly, they should be reusable, easy to use, and communicative with the consumers, to avoid the specific problems related to food waste, food quality managing, or foodborne diseases. For all this to become possible, supplementary attention should be driven to the innovative packaging and materials. Furthermore, recent strategies involving agro-industrial by-products showed that numerous bioactive compounds can be recovered and integrated into functional food packaging. Overall, future studies will be made on this topic with the purpose of continuing evolution in the food packaging industry.

8. Conclusions

This review paper presents the most important aspects associated with active and intelligent packaging and highlights many bioactive compounds recovered from agro-industrial by-products that may be useful in active or intelligent packaging. Smart packaging has a strong impact on food quality and control by allowing consumers to directly interpret the freshness and safety of the food inside. Intelligent packaging is an improved version of active packaging, simply by including devices from indicators and sensors classes for increased accuracy. The next future research can be made for by-product compounds integration in the elements of smart packaging (e.g., indicators, sensors) or for the design of new concepts of smart packaging that includes recovered bioactive compounds. Nevertheless, the trend of zero waste and the efforts made in the world of science

to find environmentally friendly solutions to combat food waste and to capitalize raw materials at high capacity is constantly growing; accordingly to this tendency, shortly, more compounds will be integrated into food coatings.

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