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Bio-Based Smart Materials for Food Packaging and Sensors – A Review

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Food industry must guarantee food safety and seek sustainable solutions for increasing shelf life and decreasing food waste. Bio-based smart packaging is a potential option, where sustainability and real-time monitoring of food quality are combined assuring

highlight the potential of natural compounds for sensing chemical and physical changes of the environment to monitor the food quality. Finally, different sustainability aspects of the bio-based materials are discussed.

Introduction

Busy lifestyles and growing urban populations mean an increasing demand for food that is fresh, healthy, convenient, and fast. One of the key drivers of this growth is the world's rising population which by the year 2050 will reach 9.7 billion people with increase of 26% (The United Nations, 2019). With the global population rising, wastage of food including 47% of all fruit and vegetables and 12% of meat and animal products, is one of the greatest challenges to achieve food security (Food and Agriculture Organization of the United Nations, 2019a). Although food is our basic necessity, its production, processing, transportation and storage are rather complex from many aspects and need to fulfill a number of criteria to ensure the health and environmental safety and economic feasibility.

Foods pose potential danger of diseases due to bacterial (*Salmonella, Campylobacter, Listeria*, and *Cholera*), viral (Norovirus, Hepatitis A), parasite (tapeworms, trematodes, *Ascaris, Cryptosporidium, Entamoeba histolytica*, and *Giardia*), fungal (*Aspergillus, Candida*, and *Fusarium*) and even prion infections as consequence of inappropriate handling and processing of the products causing foodborne diseases that affect ~10% of global population with a death toll of 420,000 deaths each year. Chemical contaminants, which may even accumulate in various food chains, represent further risks. These include phytochemical residues, mycotoxins, marine toxins from algae, cyanogenic glycosides from plants, and different metabolites from products aging and decaying (ethanol, putrescine, cadaverine, histamine, ethylene etc.) but also environmental and industrial pollutants, e.g., dioxins, polychlorinated biphenyls and heavy metals (Pb, Cd, and Hg) (World Health Organization, 2019). Another issue in the context of food safety is deliberate fraud to counterfeit the origin, content or quality (i.e., expiration dates) of products (Europol, 2015). Just diluting a high-quality wine with cheaper one mainly hurts the wallet and pride but more severe cases may endanger health permanently or cost lives (Branigan, 2008). Furthermore, the flip side of food safety is food waste as 1/3 of all produced food is lost or goes into waste (Food and Agriculture Organization of the United Nations, 2019a) meaning safe and edible food products are thrown away although the "best if used before" dates are only recommendations without information of the true status of the food. This is ethically and practically controversial as still today 820 billion people suffer from undernutrition and agricultural production would need to increase with 50% to feed the growing population by 2050 (Food and Agriculture Organization of the United Nations, 2019b).

As a partial solution to complex problems of food safety and decreasing unnecessary

packages, which include advanced packaging materials with improved properties, and sensors that can monitor food quality (Yam et al., 2005; Kuswandi et al., 2011). Over the years, several approaches toward smart packaging have been demonstrated including time-temperature indicators, modified atmosphere packaging sensors for CO_2 and O_2 monitoring, total volatile base nitrogen sensors to detect food decay, fruit ripeness indicators, pathogen sensors, and solutions for food tracking and authentication (RFID tags) (Fuertes et al., 2016; Ghaani et al., 2016; Ahmed et al., 2018; Badia-Melis et al., 2018; Galstyan et al., 2018; Mustafa and Andreescu, 2018; Yousefi et al., 2019).

As of today, plastics (rigid and flexible) have the largest shares of the market in food packaging (37% market), followed by paper and board (34%), glass (11%), and metal (9%) (Muncke, 2012). If we consider the properties of the given food packaging materials, the high market share of paper and board can be explained by the renewable source and recyclability, it is printable, wet and dry food can be stored in paper and board after laminating/covering process and in general paper and board are very suitable for mass production lowering the costs (Kirwan, 2011). Containers made of glass, on the other hand, are among the oldest materials man has used as it can be shaped to practically any form, has high chemical resistance, is impermeable to gasses, absorbs UV and even parts of visible spectrum (amber glass, green and glass partially), is hygienic and reusable, and the consumers associate it to high-quality products (Grayhurst and Girling, 2011). Metal is also an important food packaging material as it is durable withstanding packaging conditions in vacuum or under pressure and high temperature stabilization process for long shelf-life foods. Metal is reusable, UV-resistant and the food contact surface may be coated with different coatings in case the interaction between the product and the plain container would downgrade the shelf-life/quality of the product to an unacceptable level due to, e.g., metal surface corrosion or undesired food coloring as a consequence of combination of metal ions with the food components (Oldring and Nehring, 2007). Typically metal containers or cans are made of steel (tin-coated or tin-free) or aluminum (Kraus and Tarulis, 2009; Reingardt and Nieder, 2009; Robertson, 2012).

The most frequently used plastics in food packages in Europe are PP (19.3%), LDPE and LLDPE (17.5%), and PET (7.4%); and in fact, 39.9% of all produced plastics (61.8 million tons in Europe in 2018) goes to packaging in general (PlasticsEurope, 2019a). The success of plastic as packaging material can be explained by low cost, ease of modification from flexible films to rigid containers, strength, stability, light-weight, impermeability with gasses and many solvents, and enabled sterilization without affecting the food quality (PlasticsEurope, 2019b). Despite of numerous benefits, plastics are also problematic: annual global production of plastics is around 350 million tons of which only 1% is bio-based (European Bioplastics e.V., 2020) and the rest is fossil-derived with large carbon footprint [6% of all produced oil goes to plastics, having a carbon footprint equivalent to the aviation sector (World Economic Forum et al., 2016)]. In addition, plastic pollution is alarming, as polymers do not degrade but break down to smaller pieces ending up in the air, soil and water as microplastics, found even in deep-sea sediments (Jamieson et al.,

materials. The scope of this short review is to collect the contemporary literature on bio-based smart food packages covering not only bio-based food packaging materials but also bio-based sensors for monitoring various physical, chemical and biological conditions of foodstuff.

Bio-Based Plastic Packaging Materials

Production data for bio-based and biodegradable plastics are available to a limited extent only, although production capacity data are more readily accessible. Currently the production capacities of bio-based and biodegradable plastics are low (Figure 1), however, the market of some bio-based and/or biodegradable plastics are expected to grow significantly during the coming years (Bio-PET, PBS, and PLA) others are expected to consolidate (CA and Bio-PA) (van den Oever et al., 2017). Overall, it is expected that the global bioplastics production capacity is set to increase from around 2.11 million tons in 2019 to approximately 2.43 million tons in 2024 (Figure 2).

Figure 1

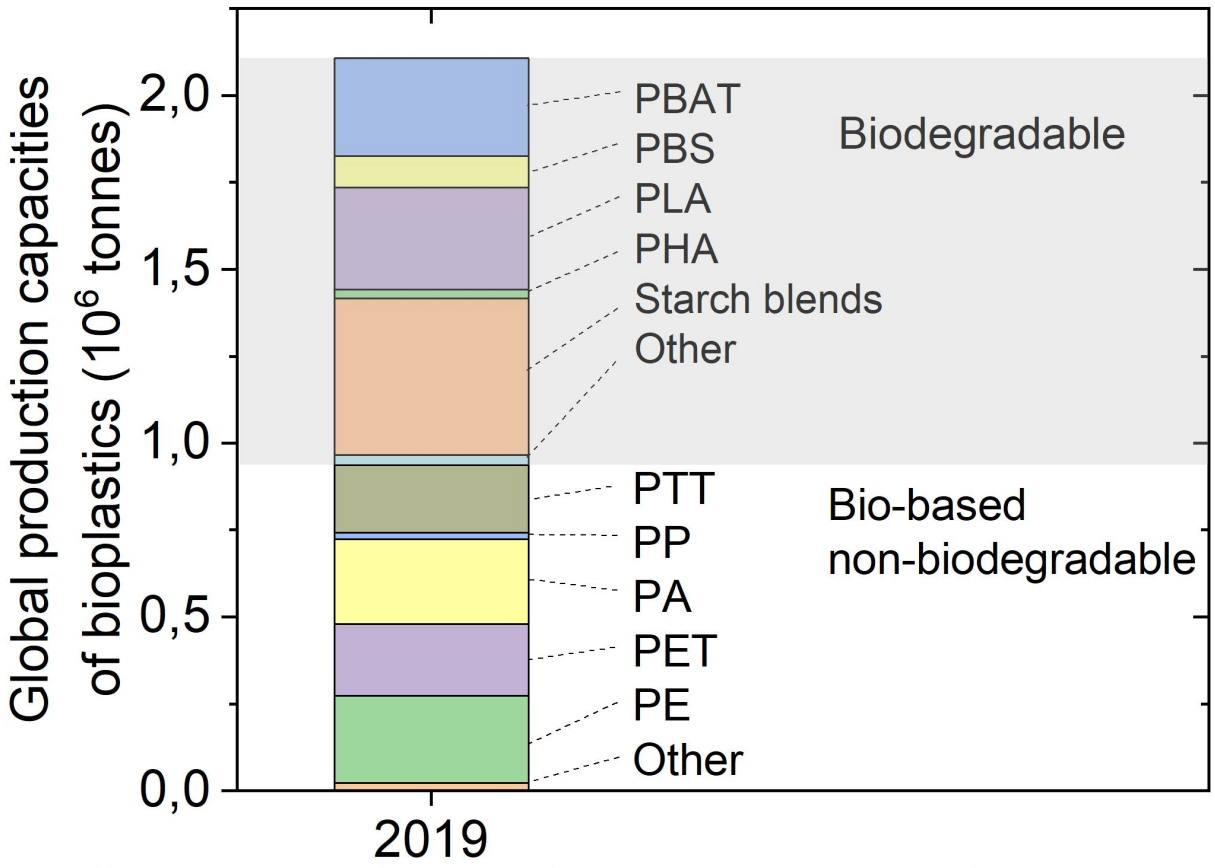

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Figure 2

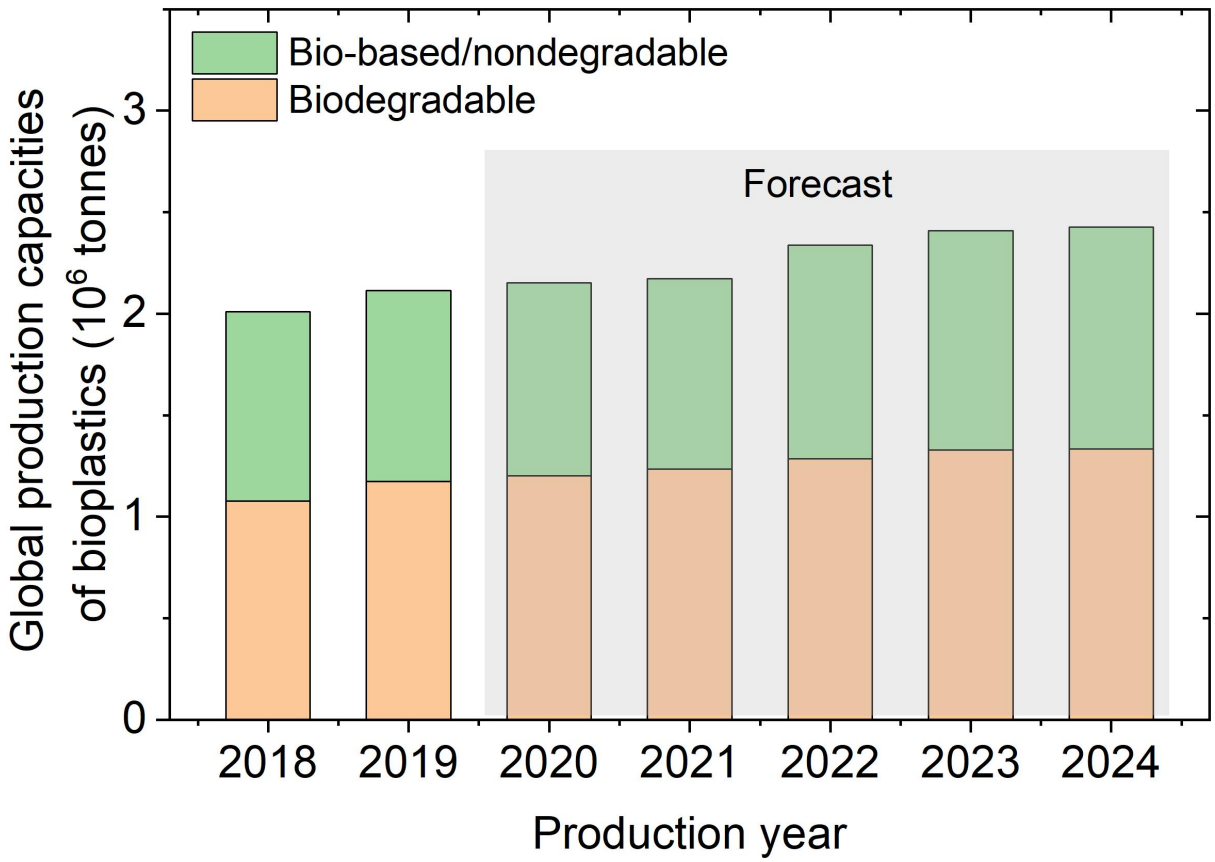

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FIGURE 2. Current and forecast global production capacities of bioplastics (European Bioplastics e.V., 2020).

A number of different routes have been developed during the past decade to produce bio-based materials [i.e., either made of bio-based source or it is biodegradable or contains both of these features (European Bioplastics e.V., 2019)] with a large variety of properties and applications areas (Table 1). Among these, only a few families are made of renewable biomass and are biodegradable [e.g., cellulose and starch thermoplastics, PHAs, PLA, polyester amides (Avérous, 2008)], viz. bio-based polyethylene, PP, polyamide and polyethylene terephthalate are non-biodegradable, and PCLs and PVAs are from non-renewable resources (Chen and Patel, 2012; Geueke, 2014; van Crevel, 2016). The main production routes are as follows:

1. Direct extraction of biopolymers such as starch and cellulose with subsequent

used as additives in polymers (Weinmann and Colton, 1950).

2. Hydrolysis to sugars followed by bacterial synthesis of polyesters, e.g., PHAs including PHB.
3. Conversion into sugars that are fermented to lactic acid followed by its direct polycondensation or by ring-opening condensation of lactide to PLA (Avérous, 2008).
4. Chemical conversion into monomers followed by polymerization, e.g., amino acids obtained by hydrolysis and separation are polymerized with esters of lactonized unsaturated fatty acids in PEA synthesis.

Table 1

	Type	Source and Properties	Applications	References
Bio-based - biodegradable	Starch-based polymers (polysaccharides)	Sourced from corn or potato Thermoplastic starch (TPS) alternative to PS Additive-free TPS has low water vapor barrier, poor mechanical and processing properties, brittle	Disposable tableware and cutlery, coffee machine capsules, bottles	Peelman et al., 2013; Niaounakis, 2013; Geueke, 2014
	Cellulose-based polymers (polysaccharides)	Sourced from de-lignified wood pulp or cotton linters Cellophane, cellulose acetates/ethers Additive-free cellulose derivatives have a low water vapor barrier and poor mechanical properties, brittle, expensive	Coated cellulose films are used for bread, fruit, meat, and dried product packaging	Geueke, 2014
	PLA (polyester)	Sourced from corn or other carbohydrate rich plants followed by conversion to dextrose and fermentation to lactic acid High tensile strength, transparent A potential alternative to LDPE, HDPE, PS, and PET	Cups, bowls, bottles, bags, jars, and films	Chen and Patel, 2012; Geueke, 2014
	PHA (polyester)	Produced by micro-organisms (such as *Cupriavidus necator*, *Methylobacterium rhodesianum*, or *Bacillus megaterium*) from glucose or starch Brittle, stiff, and thermally unstable	As composite can be tuned into different applications	Ackermann et al., 1995; Chen and Patel, 2012; Geueke, 2014
Bio-based – non-degradable	PP and PE (vinyl polymers)	Mainly based on sugar cane [sugar to ethanol (pentose fermentation), ethanol to propylene via metathesis of ethylene with 2-butene] Identical to fossil-based PP and PE	Similar to fossil-based PP and PE	Chen and Patel, 2012; Geueke, 2014; Machado et al., 2016
	PET (aromatic polyester)	Partially bio-based, synthesized from MEG and terephthalic acid (MEG from a renewable source, TA from fossil-based source)	Bottles	Chen and Patel, 2012; Geueke, 2014
	PEF (aromatic polyester)	Synthetized from FDCA and MEG Precursor can be found in human urine Better barrier properties than PET	Bottles, fibers, and films (bio-based alternative to PET)	Koopman et al., 2010a,b; Chen and Patel, 2012; Geueke, 2014
	PA	High-performance polymer Sourced from resin-rich wood or vegetable oils	Not common in food contact material	Geueke, 2014
Fossil-based - biodegradable	PVOH (vinyl polymer)	CO_2 barrier in PET	Coatings, a component of adhesives, paper and board	Chiellini et al., 2003; Geueke, 2014
	PCL (polyester)	Usually blended with biopolymers, eg., starch	Medical applications, food contact material as blends	Plackett et al., 2006; Geueke, 2014
	PBS, PES, and PBSA [aliphatic (co)polyesters]	Monomers are fossil-based aliphatic dicarboxylic acids and diols, bio-based monomers start emerging	Disposable cutlery	Chen and Patel, 2012; Geueke, 2014
	PBAT, PBST [aliphatic-aromatic (co)polymers]	Condensation of aliphatic diols and dicarboxylic acids and aromatic dicarboxylic esters/acids	Fast food disposable packaging, films	Geueke, 2014

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TABLE 1. Bio-based plastics in food packaging.

Based on the technical report published by Wageningen Food & Biobased Research in 2017 (van den Oever et al., 2017), it has been shown that the bio-based and biodegradable plastics are currently more expensive than fossil-based plastics on weight basis (Tables 2, 3). However, specific material properties can allow costs reductions in the use or end-of-life phase. Further, the price of fossil-based plastics is dependent on oil

plastics will come down.

Table 2

Plastic	Price level (euro/kg)	Density (kg/m^3)
CA	5	1,200–1,300
Bio-PA	+10–20%	1,040–1,190
Bio-PE	+20–40%	910–970
Bio-PET	No information	1,370–1,390
Bio-PP	+80–100%	900–920
PP (certified bio)	+40–50%	900–920
PBAT	3.5	1,250
Bio-PBS	4	1,260
PHA	5	1,200–1,250
PLA	2	1,250
PTT	4	1,320
Starch blends	2–4	1,250–1,350

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TABLE 2. Price level for bio-based and/or biodegradable plastics.

Table 3

Plastic	Price level (euro/1,000 kg)	Density (kg/m^3)
LDPE	1,250–1,450	910–940
HDPE	1,200–1,500	930–970
HIPS	1,350–1,525	1,080
PET	850–1,050	1,370–1,390
PP	1,000–1,200	900–920
PS	1,250–1,430	1,040

TABLE 3. Price level for fossil-based plastics.

In addition, since most bio-based plastics have a higher density, this directly contributes to their higher price. But there are exceptions when prices are compared on a product level. By selecting specific material properties and redesigning can allow material savings. For example, a traditional HIPS-based cup of 0.89 mm wall thickness could be down-gauged using impact modified PLA to 0.66 mm thickness (Schut, 2016).

Bio-Based Smart Food Packages

Many new concepts in food packaging, like the smart functionalities, have been introduced during the last years in response to the increasing demand of ready-to-eat and higher quality foods (Vanderroost et al., 2014).

Smart functionalities of food packages refer to active coatings and physical/chemical sensors combined with the packaging materials. The purpose of smart antimicrobial coatings is to mitigate the proliferation of various microbes thus prolonging the shelf-life of products, whereas sensors play role in monitoring physical and chemical conditions that influence or reflect the quality of the food products. These add-ons have inevitable positive health, environmental and socio-economic effects, which may be amplified even further by accomplishing the smart functions using renewable natural materials and robust technologies (Arroyo et al., 2019).

Antimicrobial Films

As mentioned, active food packaging involves the use of polymeric films that act as a support for various active compounds such as natural extracts that can be incorporated during the manufacturing process of the packaging itself (Kuorwel et al., 2015; Bassani et al., 2019). Antimicrobial incorporation may result in a material with antibacterial activity which can suppress the growth of bacteria on the material surface (according to the international norm ISO 22196:2011 – Measurement of antibacterial activity on plastics and other non-porous surfaces). In the food sector, a greater interest is toward materials enriched with antimicrobials so that the direct use of food additives in products is limited.

Antimicrobial materials act in two different ways. Antimicrobials can be incorporated into the film or coated either on the surface of the film or on the surface of the food (in the form of edible film). In both cases, the substance may migrate partially or completely through gradual diffusion into the food or headspace (which is typical for essential oils, for example) where it exerts its protective action, or it may not migrate, acting only when the food is in contact with the surface of the film and the target microorganism comes

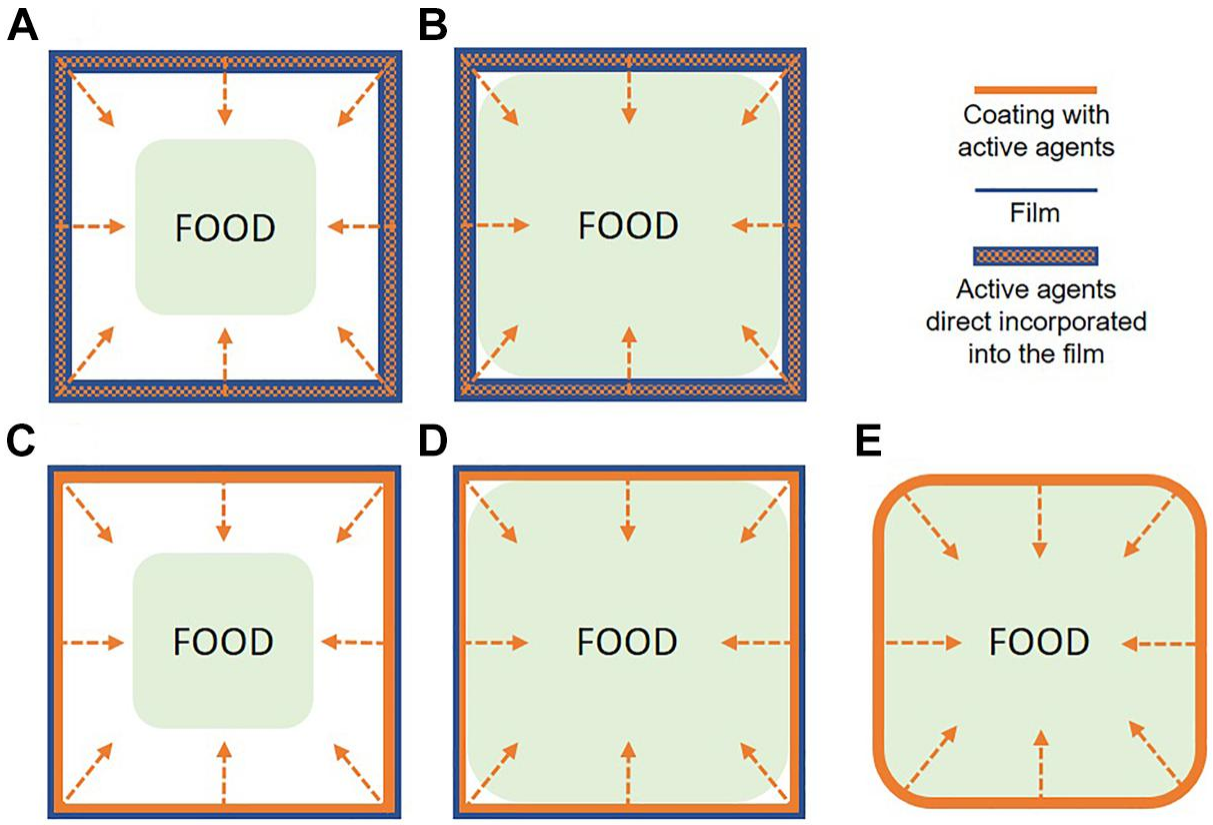

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FIGURE 3. Different ways of incorporation and release of antimicrobial agents into food: **(A)** direct incorporation into the film before extrusion and migration via gradual diffusion from the material into the headspace; **(B)** direct incorporation into the film before extrusion and migration via gradual diffusion from the material into the food through direct contact; **(C)** surface coating on the film and migration via gradual diffusion from the material into the headspace; **(D)** surface coating on the film and migration via gradual diffusion from the material into the food through direct contact; **(E)** edible film and migration via gradual diffusion from the material into the food through direct contact.

In both the cases, this kind of packaging is called active packaging [Regulation (EC) No 450/2009 — active and intelligent materials and articles intended to come into contact with food].

Antimicrobial agents used for the preservation of foods are either chemically synthesized or extracted from biomass of plants, animals and microorganisms. Conventional chemical preservatives, including ethanol and other alcohols, organic acids, and their salts (benzoates, propionates, and sorbates) are the predominant food preservatives thanks to their low price and facility to use. However, research has been focusing on replacing them with natural antimicrobial agents such as enzymes, bacteriocins, chitin

natural substances into films have involved grapefruit seed and green tea extracts, which have shown to be active as antioxidants and against different pathogens (e.g., *Escherichia coli* and *Listeria* spp.) (Wang and Rhim, 2016; Wrona et al., 2017). Cinnamaldehyde, derived from cinnamon, was also studied for its bioactivity against *E. coli* and *Salmonella* spp. (Ma Y. et al., 2018). Moreover, cinnamon oil in the PVA matrix showed repellent effect toward *Plodia interpunctella larvae* (Jo et al., 2015) and in PP film inhibited the formation of molds (Manso et al., 2015). However, clove and cinnamon in cassava starch films failed to show clear antimicrobial effect even though they reduced the water vapor transmission (Kechichian et al., 2010). Another example was provided by Seydim and Sarikus (2006) who tested edible films made of whey protein isolate loaded with rosemary, oregano and garlic essential oils against *E. coli, Staphylococcus aureus, Salmonella enteritidis, Listeria monocytogenes*, and *Lactobacillus plantarum*. Oregano proved to be the most effective against bacteria, while rosemary showed no effect.

The most popular technique to include natural extracts into the final film formulation is the extrusion (Gómez-Estaca et al., 2014). This technique involves the incorporation of the bioactive compounds before extrusion so that the high temperatures of extrusion (the exact values depend on the melting temperature of the processed polymer) allow their effective and homogeneous distribution in the film. However, this technique can often result in thermal degradation of the bioactive compounds and decrease in their activity. For instance, Ha et al. (2001) used high-temperature profile 160–190°C to extrude an antimicrobial LLDPE-based film resulting in high loss of grapefruit seed extract functionality up to complete loss of antimicrobial activity. For this reason, heat-sensitive bioactive agents (i.e., natural extracts) are preferably produced by non-heating method (e.g., electrospinning and surface coating). Among these methods, surface coating is a simple process based on low temperatures. However, this technique may suffer from poor adhesion to plastics and, if applied to make an active packaging, needs to be designed to be in direct contact with the food. Examples of antimicrobial-coated films include chitosan/essential oil-coated PP film (Torlak and Nizamlioğlu, 2011), cinnamaldehyde, garlic oil and rosemary oil-coated PP/LDPE film (Gamage et al., 2009), oregano essential oil and citral-coated PP/EVOH film (Muriel-Galet et al., 2013), chitosan-coated plastic film (Ye et al., 2008a, b), and thyme and oregano-coated LDPE. Interestingly, as reported by Valderrama Solano and de Rojas Gante (2012), antimicrobial films produced by elevated temperature processes showed better microbial inhibition compared to the ones obtained by the coating method. In particular, they found that antimicrobial films produced by extrusion method are more effective against *E. coli, Salmonella typhimurium*, and *L. monocytogenes* compared to ionizing-coated antimicrobial films with the identical amount of agent incorporated antimicrobial. The results suggest that the extrusion method allows a better incorporation of the active compounds on the polymer. Given the number of pros and cons highlighted by the literature for both the techniques, more studies comparing the efficacy of two methodologies will be needed in order to address future researches in this field. Indeed, there isn't any large scale industrial production of active bio-based films yet. For this

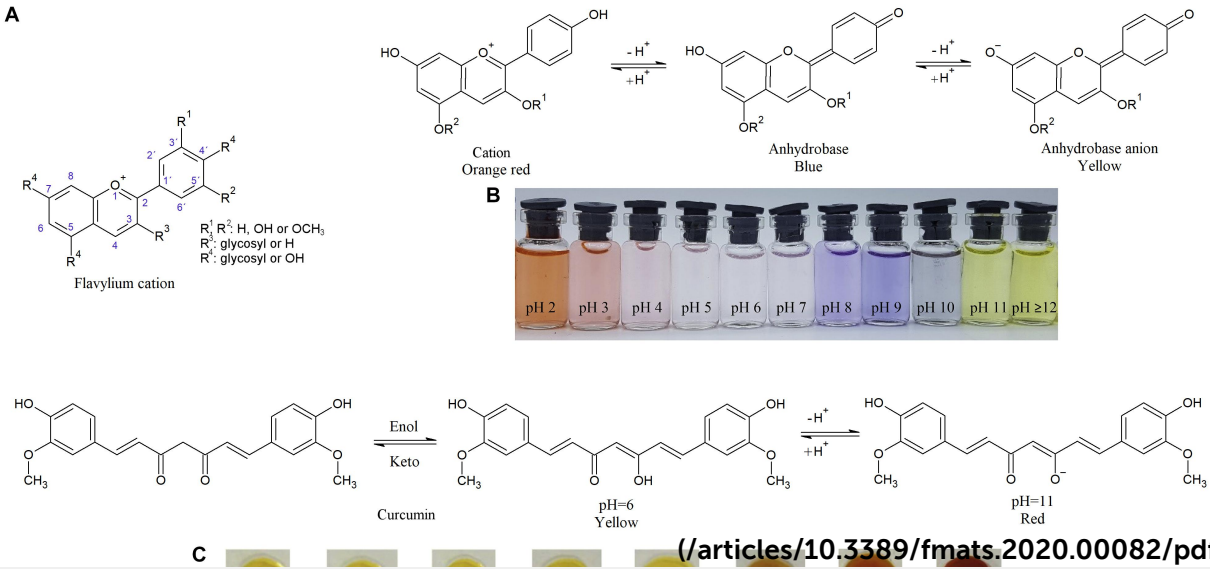
cyclodextrins (370 €/kg), an estimation of the final price of active films was done resulting in about 6.4 €/kg. It is useful to point out that this evaluation was made considering an addition of encapsulated extract equal to 2% wt as maximum (Bassani et al., 2019) and that this estimation already includes the costs necessary to encapsulate the extract by spray-drying technique.

Bio-Based Sensors

Bio-based sensors have at least one component from bio-based source which may either be the substrate (i.e., the plastics listed in the previous section) or the sensing element. Most of the sensors related to bio-based materials in food packaging are based on colorimetric detection of analytes.

Many fruits, berries, vegetables and flowers with colors covering practically the entire visible spectrum are dyed by natural compounds such as anthocyanins and curcumin known as natural pH indicators (Yoshida et al., 2009; Silva-Pereira et al., 2015; Choi et al., 2017; Dudnyk et al., 2018; Majdinasab et al., 2018; Saliu and Pergola, 2018; Zhai et al., 2018; Kurek et al., 2019). Upon protonation/deprotonation of these molecules, their delocalized electronic structure rearranges and the change of the total number of resonant electrons as well as their confinement result in a change of their color (Figure 4). For instance, Choi et al. (2017) demonstrated a pH sensor made of agar and potato starch with anthocyanin extracts from purple sweet potato that showed color variations at pH 2.0–10.0. Zhai et al. (2018) used a gelatin-gellan gum matrix with red radish anthocyanin having a slightly broader pH range from 2.0 to 12.0. In Table 4 more examples of bio-based sensors developed for food quality monitoring in recent years are listed.

Figure 4



chemical groups (–H, –OH, and $–OCH_3$) in the 3, 5, 6, 7 and the 3′, 4′, and 5′ positions determine the original color of the molecule. pH-sensitivity and color change of **(B)** pelargonidin and **(C)** curcumin. [**(A)** Reprinted from *Phytochemistry* 64(5), Kong et al. (2003). Copyright (2020) with permission from Elsevier. **(B)** Reprinted with permission from Zhai et al. (2018). Copyright (2020) American Chemical society. Reprinted from *Spectrochim. Acta A* 226, Chayavanich et al. (2020) Copyright (2020), with permission from Elsevier. **(C)** Reprinted from *Food Hydrocolloid.* 83, Liu et al. (2018) Copyright (2020), with permission from Elsevier].

Table 4

	Function	Matrix	Sensing material (source)	Response range/limit	References
Starch matrix	Colorimetric sensing, antibacterial	Starch – PVA	Anthocyanin and limonene	pH 1 – 14	Liu et al., 2017
	Colorimetric sensing	Corn starch – glycerol	Anthocyanin (blueberry powder)	pH 1–14	Luchese et al., 2017
	Colorimetric sensing, antioxidant	Cassava starch – glycerol	Chlorophyll and carotenoids (green tea and basil)	pH 3 and pH 12	Medina-Jaramillo et al., 2017
	Colorimetric sensing	Corn starch	Anthocyanin (black bean seed coat or red cabbage)	pH 1 – 10	Prietto et al., 2017
	Colorimetric sensing, NH_3 sensing	Starch – PVA	Anthocyanin (roselle)	pH 2 – 12	Zhai et al., 2017
	Colorimetric sensing	Starch – cellulose	Alizarin	pH 1 – 11	Ezati et al., 2019
	Colorimetric sensing	Corn starch – glycerol	Anthocyanin (blueberry and jaboticaba powder)	pH 2 – 12	Luchese et al., 2019
	Colorimetric sensing	Starch – gelatin	Anthocyanin (red radish)	pH 2 – 12	Chayavanich et al., 2020
Chitosan matrix	Colorimetric sensing	Chitosan	Anthocyanin (grapes)	pH 2 – 13	Yoshida et al., 2014
	Colorimetric sensing	Chitosan – microcrystalline cellulose – plasticizer	Curcumin	pH 8 – 14	Pereira and Andrade, 2017
	Colorimetric sensing, antioxidant	Chitosan	Anthocyanin (blueberry and blackberry pomace)	pH 2,4,5,6,7,10,12	Kurek et al., 2018
Cellulose matrix	Resistive sensing of H_2S	Cellulose (paper)	Copper acetate	10 ppm of H_2S	Sarfraz et al., 2012
	Colorimetric sensing	Cellulose (filter paper)	Anthocyanin (rose and red cabbage)	N/A	Shukla et al., 2016
	Colorimetric sensing	Bacterial cellulose nanofibers	Anthocyanin (red cabbage)	pH 2 – 10	Pourjahaver et al., 2017
	Resistive sensing of NH_3 at 25°C	Cellulose nanofibrils	Hydroxyapatite	5 ppm (NH_3)	Narwade et al., 2019
Gum matrix	Colorimetric sensing, NH_3 sensing	Tara gum – PVA	Curcumin	N/A	Ma et al., 2017a
	Colorimetric sensing	Tara gum – cellulose	Anthocyanin (*Vitis amurensis* husk)	pH 1,2,4,6,8,10	Ma et al., 2017b
	Colorimetric sensing, NH_3 sensing	*Artemisia sphaerocephala* Krasch. gum – carboxymethyl cellulose sodium	Anthocyanin (red cabbage)	pH 3 – 10	Liang et al., 2019
	Colorimetric sensing, H_2S sensing	Gellan gum	Ag nanoparticles	0.81 μM (H_2S)	Zhai et al., 2019
Carrageenan matrix	Colorimetric sensing	ι-Carrageenan	Anthocyanin (butterfly pea and red cabbage)	pH 1 – 11	Ahmad et al., 2019
	Colorimetric sensing	κ-Carrageenan	Curcumin	pH 3 – 10	Liu et al., 2018
Other matrices	Resistive sensing of methylamine	TiO_2 thin film	Anthocyanin sensitizer (spinach, red radish, winter jasmine, black rise)	2 ppm of methylamine	Yanxiao et al., 2015
	Colorimetric sensing	Hydrophobic nanoporous films	Anthocyanin (*rosa chinensis, roselle, camellia japonica, rose, carnation, myosotis sylvatica, zhaoshui plum blossom, Ive plum blossom, red plum blossom*)	10–30 ppm of biogenic amines	Xiaowei et al., 2015
	Colorimetric sensing	Agarose – epichlorohydrin	Anthocyanin (red grapes)	pH 1 – 10	Abolghasemi et al., 2016
	CO_2 sensing by dielectric permittivity and loss	Wheat gluten	Amino groups of wheat gluten	40% CO_2	Bibi et al., 2017
	Colorimetric sensing	Tamarin seed polysaccharide	Litmus lichen	pH 4 – 10	Liang and Wang, 2018
	Colorimetric sensing	PVA – Chitosan nanoparticles	Anthocyanin (mulberry)	pH 1 – 13	Ma Q. et al., 2018
	Colorimetric sensing	Polycaprolactone – polyethylene oxide fibers (electrospun)	Anthocyanin (acai)	pH 1 – 10	da Silva et al., 2019
	Colorimetric sensing	PVA mat (electrospun)	Anthocyanin (red cabbage)	pH 2 – 12	Maftoonazad and Ramaswamy, 2019
	Colorimetric sensing, antioxidant	Gelatin	Anthocyanin (red cabbage)	pH < 4, pH > 11	Musso et al., 2019

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TABLE 4. Bio-based sensors developed for food monitoring.

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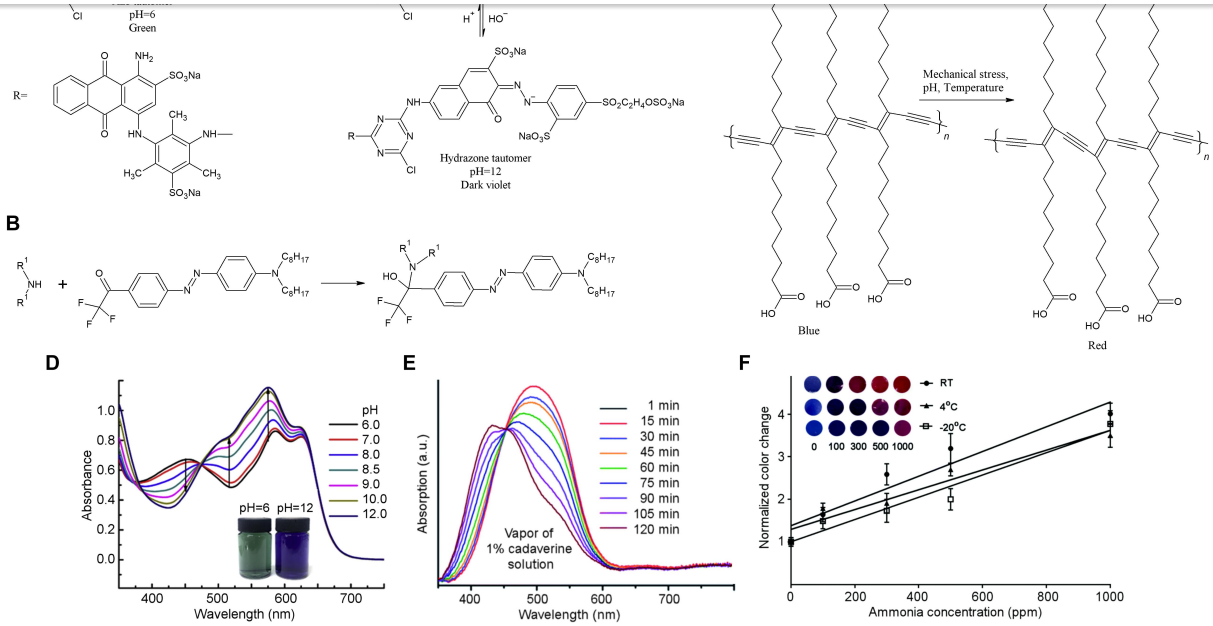
2019) and bacterial cellulose membranes (Ruswandi et al., 2012), blueberry and red grape skin pomace in chitosan and carboxymethyl cellulose matrix (Kurek et al., 2019), chitosan-corn starch film with red cabbage extract (Silva-Pereira et al., 2015), alginate beads with red cabbage extract (Majdinasab et al., 2018) as well as red cabbage extract in pectin films (Dudnyk et al., 2018) have been shown as feasible indicators of meat, shrimp and fish spoilage by detecting amines and cyclic N-containing compounds. In a similar way, acidic CO_2 evolves during the metabolism of pathogens in the food thus lowering pH, which may be detected, e.g., by anthocyanin/polylysine in cellulose matrix in a reversible manner as demonstrated by Saliu and Pergola (2018).

Other natural dyes such as chlorophyll and β-carotene might be also relevant for sensing since both structures are highly sensitive to oxidative species. Silva et al. (2017) showed that replacing the coordinated Mg^{2+} with Zn^{2+} in chlorophyll A, the fluorescence of the complex is faded when increasing the concentration of dissolved oxygen in the medium. The mechanism of luminescence suppression is suggested to be caused by an energy transfer to oxygen molecules that collide with the excited molecule. In the case of carotene, one may exploit several mechanisms for sensing. β-carotene is prone to oxidation and subsequent decomposition to shorter cleavage products leading to a gradual disappearance of the orange color (Pénicaud et al., 2011).

Synthetic dyes based on various azo-compounds and polydiacetylenes also hold promise for chemical sensing in food packages. Azo-anthraquinone based dyes immobilized on paper (cellulose) as pH sensors working either in acidic or alkaline conditions, depending on the selected pigment, were shown by Zhang et al. (2019). Selective amine sensing colorimetric indicators utilizing trifluoroacetyl azobenzene dyes developed by Mohr (2004a; 2004b), Reinert and Mohr (2008); Kirchner et al. (2006) have been exploited in colorimetric and electrochemical detection of ammonia, ethylamine, cadaverine and putrescine (Lin et al., 2015). The carbonyl carbon of the trifluoroacetyl group is highly electron-deficient thus readily reacts with electron donors such as amines or alcohols. In the presence of amines (primary, secondary, or tertiary) it forms a hemiaminal group, i.e., the number of delocalized electrons in the diazobenzene backbone is decreased (so as the confinement length of electrons) resulting in a blue shift of optical absorption. Sensors printed on paper could detect vapors of the analytes having a concentration of 1.0–0.1 vol.%. Furthermore, highly sensitive ammonia sensors that operate even at very low temperatures (down to –20°C) were demonstrated by using polydiacetylenes that were polymerized in self-assembled vesicles stabilized with cellulose nanocrystals in the chitosan matrix. The sensing mechanism is based on a conformal change of the polydiacetylene backbone (from planar to non-planar) upon external stimuli such as pH, mechanical stress or temperature. Films of the sensors could detect 100 ppm ammonia (Nquyen et al., 2019) (Figure 5).

Figure 5 **(/articles/10.3389/fmats.2020.00082/pdf)**



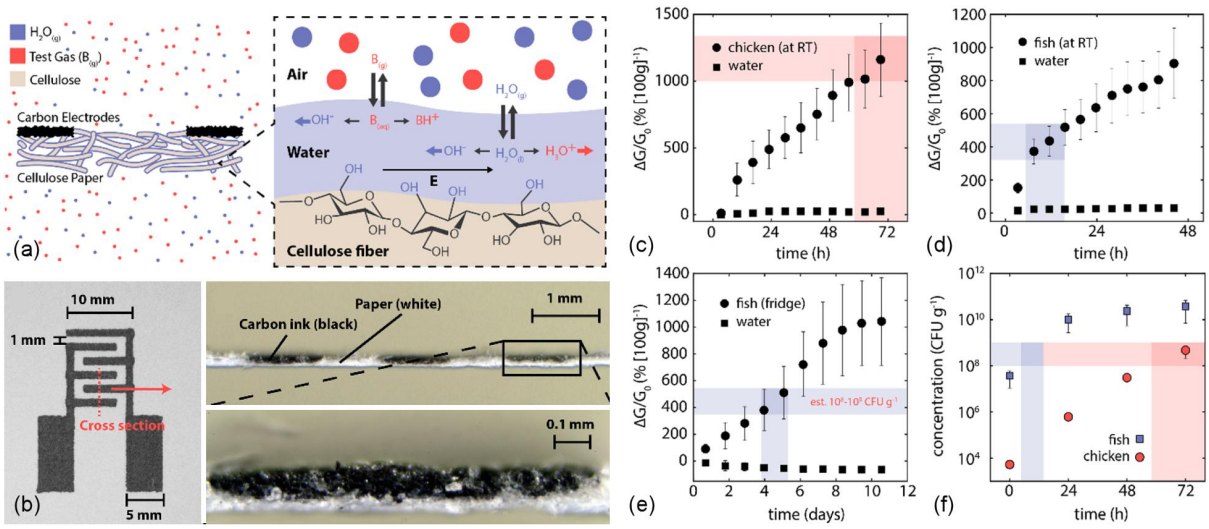
(https://www.frontiersin.org/files/Articles/521914/fmats-07-00082-HTML/image_m/fmats-07-00082-g005.jpg)

FIGURE 5. Reaction mechanisms of **(A)** anthraquinone and **(B)** trifluoroacetyl azo dyes and **(C)** polymerized self-assembled polydiacetylene vesicles and the corresponding color change during analyte sensing as displayed in panels **(D–F)**, respectively. Note, that the normalized color change in panel **(F)** is the ratio of red and blue components in the RGB coordinates in reference to the original values. [**(A,D)** Reprinted from *Sens. Actuat. B* 286, Zhang et al. (2019) Copyright (2020), with permission from Elsevier. **(B,E)** Reproduced from Lin et al. (2015). **(C,F)** Reproduced from Nquyen et al. (2019) with permission from The Royal Society of Chemistry.].

Colorimetric sensors/indicators may be also accomplished by using enzymatic processes, in which the color change is typically a function of temperature and time. Capitalizing on these, Yan et al. (2008) developed a temperature indicator that combines the coloration of iodine-starch clathrates and the influence of temperature on the hydrolysis of starch in the presence of amylase enzyme. From the kinetic reaction rates of hydrolysis (and the corresponding coloration of the clathrates), one may assess the time-temperature history of cooled products. Another example of time-temperature indicator based on enzymatic oxidation of ABTS [2,20-azino bis-(3-ethyl benzthiazoline-6-sulphonic acid] substrate resulting in green color was demonstrated by Rani and Abraham (2006). By applying a fuse-type melting medium between peroxide and the mixture of the enzyme and substrate, the reaction starts only when the medium is warm enough to melt the separator thus enabling the mixing of the reactants. A similar melting fuse type of color indicator was proposed by Lorite et al. (2017), in which erythrosine B food dye printed on PLA was applied in a microfluidic device in conjunction with a frozen solvent. As soon as the temperature exceeds the melting point, the transparent solvent flows and dissolves the dye producing red staining of the device. **(/articles/10.3389/fmats.2020.00082/pdf)**

temperature sensor by using an electrically conductive film of carbon nanotubes being a part of an RFID tag. When the temperature reached the melting point of the solvent, it flowed through a capillary toward the nanotube film, soaked it and increased its resistance detected by the RFID reader. Very recently, an interesting and simple resistive chemical sensor for water-soluble gasses was proposed by Barandun et al. (2019) on cellulose substrates. As water is always present on the surface of the hygroscopic cellulose, when it is exposed to water-soluble gas analytes, the surface conductivity increases depending on the chemistry and concentration of the interacting moiety, which can be monitored by electrical measurements using carbon electrodes printed on the surface. The devices were highly sensitive to ammonia (down to 200 ppb) among the gasses tested (TMA, H_2S, CO_2, and CO) and were feasible for monitoring the headspace of meat and fish food packages. The sensors could be integrated into RFID tags and read by an NFC enabled smartphone (Figure 6).

Figure 6



(https://www.frontiersin.org/files/Articles/521914/fmats-07-00082-HTML/image_m/fmats-07-00082-g006.jpg)

FIGURE 6. **(A)** Schematic diagram displaying the equilibrium of surface adsorbed water and vapor in the gas phase. Absorbed and then dissociated base increases the concentration of mobile ions in the liquid. **(B)** Optical images of the printed carbon electrodes on paper (top and cross-sectional views). Relative change of sensor conductance measured for decaying **(C)** meat and **(D)** fish at room temperature, and **(E)** fish at 4°C. The red and gray bands in the graphs correspond to the healthy limit of microbial contamination determined by microbial cultures displayed in panel **(F)**. [Reproduced from Barandun et al. (2019) with permission from American Chemical Society].

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the price of curcumin is around 500 €/kg (February 2020) or for the wild blueberries the purchase price is typically between 2.5 and 4 €/kg (in Finland) but may rise up to 6–8 €/kg if the availability is limited due to dry summer, for example like in Finland in 2019. However, as pointed out earlier, edible food should not be used as raw material, but rather side streams should be valorized. Luckily the anthocyanin content is reasonable high in pomaces and seeds, so in principle agro-food waste can be used as source for sensor material lowering the price. If we compare the prices of food to prices of typical gas sensor materials, such as titanium dioxide with bulk price around 2–3 €/kg (2016–2017, Industrial Minerals), the anthocyanin source are competitive, especially because colorimetric sensors do not require complex electronics for the output and the waste management can be expected to be cheaper.

Conclusion

Bio-based smart food packaging will be one answer to the global challenges related to the desperate quest for carbon neutrality, food saving and safety, as well as for renewable materials and technologies. In this review, we have collected the contemporary literature on three key components of bio-based smart materials including (i) the packaging materials themselves responsible for providing a safe envelope for the products, (ii) advanced coatings and additives to help preserving foodstuff as well as (iii) renewable sensor materials with enabling technologies that can detect the quality of foods and are potentially feasible for industrial scale-up. Although the corresponding fields of scientific research on bio-based and renewable materials with robust production technologies are becoming more and more relevant today, it is clear that careful life cycle, economic and even user perception analyses have to be made to assess the real environmental and socioeconomic impact of each potentially viable solution.

Author Contributions

NH and KK contributed to the conception and design of the review. NH wrote the first draft of the manuscript. PP contributed with the literature search, illustrations, and writing of the manuscript. AB, CF, RN, and GS wrote sections of the manuscript. All authors contributed to manuscript revision, read and approved the submitted version.

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Abbreviations

Bio-PA, bio-polyamide; Bio-PBS, bio-polybutylene succinate; Bio-PE, bio-polyethylene; Bio-PET, polyethylene terephthalate containing bio-based materials; Bio-PP, bio-polypropylene; CA, cellulose acetate; FDCA, 2,5-furandicarboxylic acid; HDPE, high-density polyethylene; HIPS, high impact polystyrene; LDPE, low-density polyethylene; LLDPE, linear low-density polyethylene; MEG, mono-ethylene glycol; NFC, near-field communication; PA, polyamide; PBAT, poly(butylene adipate-co-terephthalate); PBS, polybutylene succinate; PBSA, polybutylene succinate adipate; PBST, poly(butylene succinate-co-terephthalate); PCL, polycaprolactone; PE, polyethylene; PEA, polyesteramide; PEF, polyethylene furanoate; PES, polyethersulfone; PET, polyethylene terephthalate; PHA, polyhydroxyalkanoate; PHB, polyhydroxybutyrate; PHBV, poly(hydroxybutyrate-co-valerate); PLA, polylactic acid; PP, polypropylene; PS, polystyrene; PTT, polytrimethylene terephthalate; PVA, polyvinyl alcohol; PVC, polyvinyl chloride; PVdC, polyvinylidene chloride; PVOH, polyvinyl alcohol; RFID, radio frequency identification; TA, terephthalic acid; TPS, thermoplastic starch; UV, ultra violet.

Footnotes

1. ^ Amazon.co.uk (http://Amazon.co.uk)

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