Ashutosh Kumar Shukla *Editor*

Food Packaging: The Smarter Way

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To my parents.

Preface

Food packaging is of interest to different stakeholders including producers, retailers and consumers in view of societal and environmental concerns. Right choice of packaging materials to improve packaging experience and shelf-life are therefore the areas that have attracted the scientific community. This volume is a collection of different chapters dealing with novel packaging materials and trends from pre- to post-packaging of food items. Initial chapters have been planned to acquaint the audience with food packaging industry and provide a link to latest research on smarter ways of food packaging including artificial intelligence applications. Biodegradable packaging solutions for fruits, vegetables and animal-based food products including dairy and fishery products have been covered in the following chapters. Edible films and coatings have been presented in a separate chapter highlighting the challenges and applications. Role of sensors to improve food packaging has also been presented in a separate chapter as well. Similarly, another chapter presents the applications of functional nanomaterials for food packaging. Rules and regulations related to the packaging have also been included to bring in the sense of completeness.

While going through the chapters, I learnt many things and expect the same for the intended audience including novice researchers. I thank the expert contributors from different laboratories/countries/disciplines making this volume truly interdisciplinary. Some of the authors accepted my request to review the individual chapters and provided their valuable comments. This helped me a lot to present the quality content before the readers.

I sincerely thank Dr. Naren Aggarwal, Editorial Director—Books, Asia, Medicine & Life Sciences, Springer for giving me the opportunity to present this book to the readers. I also thank Madhurima Kahali, Editor Books—Medicine & Life Sciences, Springer and Mr. Suraj Kumar, Production Editor (Books), Springer for their support during different stages of publication.

Prayagraj, Uttar Pradesh, India Ashutosh Kumar Shukla

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Ashutosh Kumar Shukla is an Associate Professor of Physics at Ewing Christian College, Prayagraj, a constituent college of the University of Allahabad, India. He has also served as Associate Professor in the Department of Pure and Applied Physics, Guru Ghasidas Vishwavidyalaya, Bilaspur, Chhattisgarh (2011–2013), on lien from his parent organization. His research interest is primarily based on the applications of Electron Spin Resonance Spectroscopy technique in different fields of societal interest including Condensed Matter Physics. He has been awarded the Bilateral Exchange fellowship and Visiting fellowship of Indian National Science Academy, New Delhi, in addition to research fellowships of Council of Science and Technology, Lucknow, and Council of Scientific and Industrial Research, New Delhi. Dr. Shukla is on the reviewer panel and editorial boards for international journals. With 20 years of experience of Physics teaching, he has to his credit journal publications, textbooks, and many edited volumes in different areas such as medicine, food science, and crude oil in collaboration with experts globally. Dr. Shukla has successfully completed research projects and presented his research in different international events. He is a member of the International EPR Society (IES) and web member of the International Society of Magnetic Resonance (ISMAR).

Food Packaging Industry: An Introduction

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Vilásia Guimarães Martins and Viviane Patrícia Romani

1.1 Introduction

The basic materials used in the food packaging industry are glass, metal, paper and paperboard, plastics, or their combinations in the same material. Innovations in the sector of food packaging are adding many different compounds and using various technologies to produce intelligent, active, sustainable, and biodegradable packaging.

There is a growing demand from consumers who desire safer, high-quality, extended shelf life, and less processed food products. In this case, the innovations in terms of packaging are remarkably interesting. Active compounds with antimicrobial and antioxidant properties, for example, can be incorporated in the packaging material. These compounds are released throughout the product's shelf life, and then, it is not necessary to add preservatives in the food product formulation. This type of material is called active packaging. Another technology highlighted in recent years is the intelligent packaging, which also contributes for food safety. Intelligent packaging informs the consumer through indicators and/or labels if the food product is appropriated to be consumed, even if it is within the expiration date described on the product packaging. In some cases, food products are not stored, transported, or even handled at the appropriate temperature, reducing their shelf life. Through intelligent packaging, consumers will know if the product was appropriately transported and handled, and thus, they will be sure about the quality of the product.

The world produces about 300 million tons of plastic waste each year, and so far, only 9% of this waste has been recycled (ONU 2019a). Also, each year at least eight

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million tons of plastic go to sea and oceans, often decomposing into small microplastics that end up in our food chain (ONU 2019b). Durability, resistance, lightness, flexibility, and low cost are some of the characteristics that make the plastic unique and indispensable in daily life (Heidbreder et al. 2019). However, the qualities that make the plastic a good material for consumer products can also make it extremely resistant to biodegradation and can remain in the environment for decades.

Given the above, a sector of the packaging industry that is expanding is the sustainable and biodegradable packaging. Research has been carried out all over the world trying to find solutions to make possible to replace mainly the synthetic plastic packaging by biodegradable packaging. The main raw materials used to produce biodegradable packaging are proteins, lipids, and carbohydrates, usually extracted from agroindustrial sources, and waste from food industries. Important findings are reported in the production of biodegradable materials for food packaging; however, their use is still limited due to the mechanical and barrier properties, which are inferior to those of synthetic polymers. Several techniques have been used by researchers to improve these properties, including the use of blends, chemical modifications of polymers, addition of reinforcement materials (e.g., nanoparticles and fibers), application of plasma, and UV light treatments.

In addition to the sustainable, renewable, and biodegradable materials being used in the development of new plastic packaging, it is also possible to observe this environmental concern for other materials as well. For example, the immense exchange that is taking place from glass, metal, and plastic packaging by cellulosic packaging also has this environmental appeal. Today is possible to find whiskey “bottles” in cardboard on the market, which would have been unthinkable a few years ago.

This chapter will briefly discuss information about the global food packaging market, innovations in the food packaging sector, and what are the challenges and perspectives for the packaging produced in research laboratories to reach the market.

1.2 Global Food Packaging Market and Sustainability

According to the World Packaging Organization (2020), the revenue of the global packaging industry is over USD 500 billion. Sustainability, convenience, efficiency, protection, and flexibility are the parameters associated with packaging solutions. For consumers, easy-to-use and sustainable packaging is becoming a priority now than ever before.

There are a lot of market analysis reports about the global food packaging market size at the Internet. Most of them project the future market of food packaging based on type of material, application, outlook, region, among others. The growth of the food packaging market is estimated based on some factors such as convenience, increase in the population, improvement of the shelf life, single serve packs, food delivery, and high-performance materials; all these parameters have a positive impact on the market. In the industries, the consumers’ desire is what drives the vector of changes, and in the packaging industry, it would be no different. In addition to the improvement of the consumer experience when unpacking a product, its design and how it relates to sustainability also matter.

About 40% of all plastic produced in the world is used for food packaging, followed by building and construction industry (20%) (PlasticsEurope and EuPR 2015). Governments have been acting to reduce environmental impacts, establishing agreements with brands to reduce or change the use of raw materials, such as plastic. In the United Kingdom, for example, the government will introduce a tax on plastic packaging from 2022. Besides that, financial sanctions will be established to encourage the incorporation of recycled plastic in the production chain of these companies, creating an interesting precedent for their adhesion.

As mentioned by the Grand View Research (2020), the paper and paper-based material segment accounted for a revenue share of 31.9% of the total food packaging market in 2019. Growth of this segment is driven by high product adoption to substitute nonbiodegradable packing solutions. Innovations in design, ease of printability, and sustainability give paper packaging a competitive advantage over plastic and metal packaging solutions.

Regarding bioplastics, which represents a crucial way to reduce the environmental burden, European Bioplastics, Nova-Institute (2020), mentioned that the global production capacities of bioplastics including bio-based and biodegradable materials are 2.11 million tons in 2020. Bio-based/nonbiodegradable materials (PE, PET, PA, PP, PTT, and other) represent 41.9% of the bioplastics produced, and biodegradable materials (PBAT, PBS, PLA, PHA, starch blends, and other) represent 58.1% of the total. Going forward, the conscious consumption will be the meeting point for transformations in the global food industry. Implementing sustainable policies and philosophies in the manufacturing lines, from the product itself, through packaging and logistics, will be a necessity to survive in a modern market that is concerned with the planet.

1.3 Sustainable Food Packaging

Plastics are the most used materials in packaging applications in different fields. Commonly used polymers, such as polyvinylchloride (PVC), polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), polyamide (PA), polystyrene (PS), and ethylene vinyl alcohol (EVOH), are cost-effective and have enough properties to protect packaged products since production to consumption (Geyer et al. 2017; Luzi et al. 2019). However, these fossil-based polymers are obtained from finite sources. In addition, the incorrect plastic disposal, which has a nonrecyclable or nonbiodegradable nature, is driving the growing need of sustainable alternatives, due to the environmental burden resulting from the high amount discarded (Ahmed et al. 2018).

Sustainable materials protect the environment since the raw material extraction to the final disposal, meaning that there is no damage to the nature. The food industry is one of the largest users of packaging (All4Pack 2018) and thus has a valuable responsibility in using sustainable packaging as well as motivating industries and consumers to adhere to the use of such materials. Then, the idea is to use renewable and abundant resources to produce biodegradable/compostable alternatives. In this context, it is important to consider that plastics produced by renewable resources are not certainly biodegradable or compostable. Likewise, biodegradable materials are not necessarily produced with renewable resources, as biodegradation is related to the chemical structure in the matrix instead of its origin (Lambert and Wagner 2017; Asgher et al. 2020). In summary, a sustainable packaging is a material obtained from renewable sources that in the end of its cycle is biodegraded or composted.

Bioplastics are a potential alternative that can contribute to the sustainable development in food packaging. Biopolymers or natural polymers can be generated by plants or microorganisms and/or extracted from food industry by-products. Some examples include carbohydrates (e.g., chitosan, starch, cellulose), proteins (e.g., keratin, gluten, collagen), polylactic acid (PLA) polyhydroxyalkanoates (PHAs), and exopolysaccharides (EPSs) (Benbettaïeb et al. 2016; Asgher et al. 2020), which are being widely explored in the development of films, wraps, and laminates for food packaging. Some examples of recent developments include an edible film packaging made from a milk protein for dairy, a box for pasta packaging prepared with wasted vegetables and fruits, and a plant-based paper cup (TrendHunter 2017).

The production of bio-based materials for food packaging is a multistep process. Generally, it starts with the breakage of the intermolecular linkages of the biopolymer, followed by the synthesis of a new molecular structure and finally the development of the three-dimensional network from the new linkages formed (Galić et al. 2011). The resulting material characteristics depend on the raw polymer structure and the processing conditions. Bioplastics can be produced by wet or dry processing (Blanco-Pascual et al. 2013). Wet processing, known as casting technique or continuous spreading, is based on the solubilization of the biopolymer in the solvent with the posterior solvent evaporation. The final polymer is influenced by the pH and temperature of the suspension, and the type of solvent (Khwaldia et al. 2004; Mellinas et al. 2016). Differently, the dry processing, including extrusion, or thermal processing, depends on the thermoplastic properties of the polymers, based on the theory of glass transition. This method consists of the conversion of the glassy structure into a semi-solid condition at a specific temperature, where the molecules are broken, and new linkages and bonds are formed (Khwaldia et al. 2004; Hernandez-Izquierdo and Krochta 2008).

Bioplastics can also be incorporated with various additives, for example, antioxidant and antimicrobial compounds, nutrients, and colorants. Agricultural by-products are a potential and cheap source of renewable additives. These additives reduce the possibility of microbial growth and lipid oxidation, and also have the capacity to increase the shelf life of products (Jafarzadeh et al. 2020). Studies reported the incorporation of various additives from renewable sources in different matrices, such as durian leaf waste to enhance antioxidant activity in gelatin films (Joanne Kam et al. 2018), jambolão skin extract in methylcellulose films to provide antioxidant and color-changing properties (da Silva Filipini et al. 2020), grape seed extract—carvacrol microcapsules in chitosan films to increase the shelf life of refrigerated Salmon (antimicrobial properties) (Alves et al. 2018), chitosan nanoparticles in starch films to inhibit bacteria in wrapped cherry tomatoes (Shapi'i et al. 2020), pomegranate peel particles into starch-based films as antimicrobial and reinforcing agent (Ali et al. 2019), kombucha tea in chitosan films to retard lipid oxidation and microbial growth in minced beef (Ashrafi et al. 2018), and pink pepper phenolic compound incorporation in starch/protein blends to inhibit apple browning (Romani et al. 2018). Bioplastics have strong potential as sustainable alternatives to synthetic materials because they are produced using renewable sources, a prerequisite as previously mentioned, and despite some challenges that their use still faces (discussed in the section ahead), generally they present fast biodegradation in soil and water.

1.4 Technology Developed in the Laboratories X Scale Up to Industries

Sustainable materials have been broadly studied and interesting results are being obtained, but the large-scale production for commercial uses is still limited and the commercial use is insignificant compared to the conventional plastics. There are different reasons that prevent the wide use of sustainable packaging, including (1) the material properties, which are not enough to protect the products and can compromise the food shelf life; (2) scale-up, since the process of production, development of the new technology, and financial capacity; (3) production and logistic for feedstocks and composting infrastructure; (4) regulation policies; and (5) consumer behavior (Rydz et al. 2018; Steenis et al. 2018; de la Caba et al. 2019).

New sustainable materials for packaging are generally produced using hydrophilic biopolymers, which result in the low physicochemical properties (such as mechanical and barrier performance) due to sensitivity to humidity. To overcome these limitations, researchers are searching for strategies for material improvement. Examples of strategies are the incorporation of reinforcing agents, blending different raw materials, and chemical, enzymatic, and physical methods to alter polymer chains (Bourtoom 2009). Promising results are being obtained; however, the challenge is the improvement of the overall performance of the polymer. Usually, the increase in the mechanical resistance does not come with the increase in barrier properties and vice versa. Indeed, the protection of foods requires the equilibrium of different material's characteristics to prevent the component oxidation and microbial spoilage, and thus, attention is necessary at the minimum performance necessary in the packaging to keep the quality of products (Martins et al. 2019). Additionally, the change in the properties of bioplastics during the interaction with the food material needs to be considered. Either, the compatibility of the polymers with the products can affect the food quality and gained minimal attention (Asgher et al. 2020).

Another important hurdle that affects the wide adoption of biopolymers use is the difficult scale-up of the production process. The production of biopolymers in laboratories is widely performed through the casting technique. It consists of pouring a suspension on plates (e.g., Petri dishes) controlling the mass of suspension to generate an uniform thickness, but variations are difficult to avoid. This technique is suitable to produce films up to 30 cm, not larger, and takes long time in the drying step making it useless at industrial scale. The tape casting, which consists of the spreading of suspension in larger supports or on continuous carrier tapes, is useful to produce larger films, but industries in general have well-established extrusion processes (De Moraes et al. 2013; Werner et al. 2017). To advance in the large-scale production of biopolymers, the proof of concept needs to be effectively produced in industries. Even though some biopolymers are not suitable for extrusion processes, as in the case of proteins due to their molecular architecture and spatial arrangement (Mensitieri et al. 2011), it is important to focus on strategies to adequate such polymers in a way to facilitate their processing by the industry technologies. Generally, industries adopt and license unique raw materials to produce plastics, and then, an industry adjustment would be necessary for the production of the biopolymers. In addition, besides the fact that academic research mostly stops at the proof-of-concept stage, patented technologies are frequently incomplete in scope to be adopted by industry (Nerkar and Shane 2007; Tolfree and Jackson 2008; Inns 2012), impeding the advance of such technologies. That is why, it is also important the pilot scale fabrication and characterization, otherwise it is possible to be financially inviable for industries to take high risks to adopt the sustainable technologies for packaging (Werner et al. 2017).

1.5 Final Remarks

To increase the use of biopolymers, besides the mentioned aspects, the resource efficiency and composting infrastructure need to be considered, as well as the regulation policies. Currently, raw materials used to produce biopolymers often compete with requirements for food-based products. The expansion of the first-generation bio-based polymer production can generate unsustainable demands (Babu et al. 2013). Furthermore, a composition structure is necessary for the suitable biopolymer disposal. As the definition of ASTM-D6400, a compostable material is a material that biologically degrades; therefore, just substances that can degrade biologically in a composting environment can be labeled as “compostable” (ASTM D 2004). In this sense, besides the processing of biopolymers, it is fundamental to the planning of resource efficiency, raw materials obtaining, and composting infrastructure in the end of the biopolymer life.

Also, the consumer purchase probability and willingness to pay for sustainable materials have a critical influence in the advance of new biopolymer technologies. The consumer acceptance of these new materials has also an important role. Firstly, the higher costs of biopolymer production result in superior market prices and consumers usually do not pay the price for sustainability. Also, traditional consumers can question the quality and/or safety of the new material due to the limited information about the advantages and the low familiarity with biopolymers (De Marchi et al. 2020). In addition, consumer insights related to sustainable designs help designers to develop coherent strategies to adopt new initiatives. The lack of these consumer insights also complicate the adoption of new sustainable alternatives (Steenis et al. 2018). Still regarding the attitudes to adopt sustainable materials, industries have an essential role as well. Some manufacturers tend to support the use of old technologies and prevent the adoption of more sustainable solutions due to the efforts needed in adjusting their processes (Keränen et al. 2020). Therefore, reorientation of existing industries toward sustainability demands more attention and consumers need to be informed regarding the importance of sustainable packaging and motivated to consume such products.

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World Packaging Organization (2020)

Toward Smarter Food Packaging

2

Bambang Kuswandi

Abstract

Many food producers regularly know about new food packaging options, i.e., smart packaging, active packaging, intelligent packaging, and connected packaging. This chapter review shows that these types of packaging benefit specific types of food products, so producers can decide which ones most help them. Smart packaging is an umbrella term used to describe food packaging with enhanced functionality through technology. However, intelligent packaging that will be focused on in this chapter contains sensors to determine the condition (e.g., freshness or ripeness) of the food products. Another term used in smart packaging is active packaging, where it is used to interact with the packaged foods to enhance their condition, significantly to extend freshness or shelf life. In comparison, connected packaging allows consumers to interact with a food product through a label or code on the package that can be activated with a mobile device. Finally, the role of I.T. in smart packaging applications for food quality and safety monitoring is discussed.

Keywords

Smart packaging · Intelligent packaging · Sensors · Perishable foods · I.T

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2.1 Introduction

There will be many food products that lose their nutritional values and properties fastly if they are not packaging properly. Food packaging plays a very important role in the food supply chain management system. Since it can be employed as a protective layer to avoid food contamination and maintain food quality and safety (Bambang Kuswandi et al. 2011), however, the classical food packaging system can only protect or isolate food product from the external condition and environment without informing food quality, such as freshness and safety, for users (Kuswandi and Moradi 2019). Physical, chemical, and microbial properties and sensory evaluation are traditional methods for the determination of food quality and safety. The drawbacks of these methods are laborious works, long procedure, high cost, requiring sample pretreatment, and destructions. Based on these methods, the detection of food quality and safety is difficult to be performed in fast and nondestructive methods. Therefore, smart food packaging that informs the consumer regarding the food quality and safety evaluation is needed. It drives by the recent trends and consumer needs as well as regulatory requirements on food products.

Food packaging also plays a crucial role in reducing waste and carbon dioxide (CO_2) caused by used packaging and their disposal (Kuswandi 2017). This is due to the fact that our society has produced 8.3 billion metric tons of waste, where it contains 76% of plastic, which is mainly not recycled (90.5%) for over the last 60 years (UN Environment Programme 2018). Furthermore, the current situation due to the pandemic of COVID-19 caused food scarcity, which has to pay further attention to the food waste problem. Even though all in the food sectors have a role play in avoiding and decreasing food waste, starting from food producers to consumers, the significance of domestic waste recommended that innovative packaging can be an important tool for preventing and reducing food waste. In this regard, packaging has become an important technology to assure quality and safety in the food supply chain, enhance shelf life of foods, fulfill consumer satisfaction, and prevent undesired consumer complaints (Kuswandi 2020). Since consumers are more concerned about food quality and safety and require more accurate food information on quality and safety, in this case, it needs to provide a simple and real-time monitoring method for food product quality and safety during food supply, distribution, storage, and display according to the consumer interests and rights. Although there are many major innovations in food packaging systems and materials, the basic food packaging principles are still the same. Among them, smart packaging systems offer a great approach in reducing or even preventing food wastes.

Smart packaging, especially intelligent, active, and connected packaging, could support the optimization of the food supply management system, such as enhancing food shelf life and food quality monitoring, and reducing food loss and waste. Active packaging system helps in increasing the food product shelf life, enhancing their condition, and significantly extending food quality by “interacting” with the food product, while an intelligent packaging system helps in informing the food product quality and safety during transportation and storage as well as a display by "monitoring" the food product condition (Kuswandi et al. 2011; Kuswandi 2017; Rai et al. 2019). Furthermore, the connected packaging allows consumers to interact with a food product via package label or code that can be accessed using a mobile device. This chapter discussed smart packaging that will be more focused on intelligent packaging development over active packaging since its "monitoring" function could enhance to be smarter with the help of ICT via Apps, artificial intelligence, or IoT (Internet of Things). In this case, connected packaging concepts are also discussed that allow consumers to interact with a food product through a label or code on the package that can be activated with a mobile device.

2.2 Intelligent Packaging

Based on the Commission of the European Communities, it is stated that intelligent food contact materials are defined as materials that allow monitoring the packaged food condition or the environment surrounding the food (Communities 2004). Hence, intelligent food packaging could be defined as a new packaging technology that integrates intelligent functions with food packaging systems. Intelligent packaging can detect, sense, trace, record, and communicate internal or external changes related to food products that associate with the formation of food quality and safety, and extended the packaging information functions during transport, storage, and display that promotes and enhances safety and quality of food producers or consumer (Kuswandi et al. 2011; Salinas et al. 2014a, b). The intelligent packaging functions are given in Fig. 2.1. Intelligent packaging performs the communication functions on the packaged food condition during distribution, storage, sales, and packaging waste disposal to food producers and consumers (Yam et al. 2005).

In order to allow real-time or online monitoring of a food product during distribution, storage, and display, numerous smart devices are employed, such as indicators (for monitoring food freshness, pH, leakage, integrity, time, and temperature), data carriers (bar codes, IoT), and sensors (gas chemical sensors and biosensor) (Ghaani et al. 2016; Kuswandi 2018; Müller and Schmid 2019). Intelligent packaging allows many applications in food freshness detection, spoilage, chemical, and microbial contaminant detection, traceability, authentication, etc. (Majid et al. 2018; Kalpana et al. 2019; Vanderroost et al. 2014; Popa et al. 2019). Some current example of various smart devices used and integrated into intelligent food packaging is shown in Table 2.1.

2.2.1 Freshness Indicator

A freshness indicator is a smart colorimetric device integrated into intelligent food packaging. This type of colorimetric indicator is divided into two types: direct colorimetric indicator and indirect colorimetric indicator as given in Fig. 2.2 (Kuswandi et al. 2011). The direct colorimetric indicator commonly works based on color development of indicator caused by the volatile gas released by food that

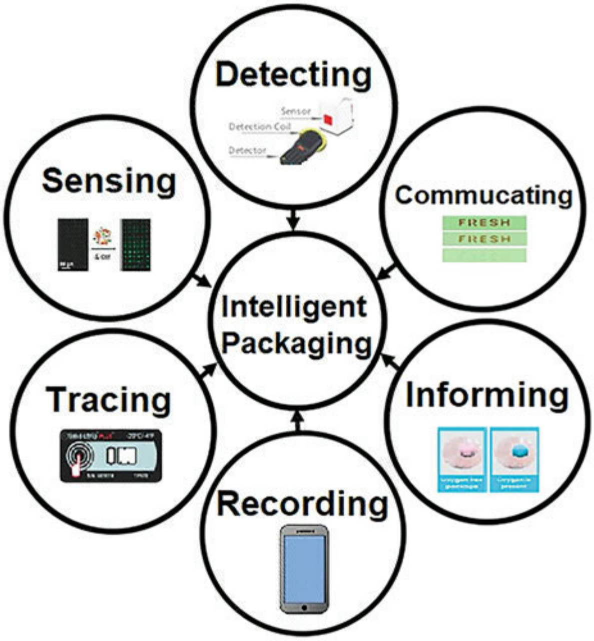

Fig. 2.1 Intelligent packaging functions in food packaging

Table 2.1 Some current examples of various smart devices integrated into intelligent food packaging

Type of smart device	Sensitive material	Target analyte	Material used	Type of food	References
Indicators	Black carrot	pH	Bacterial cellulose	Pasteurized milk	Ebrahimi Tirtashi et al. (2019)
	Red cabbage	pH	Bacterial cellulose	Pasteurized milk	Kuswandi et al. (2020)
	Alizarin	pH	Chitosan	Fish	Ezati and Rhim (2020)
	Shikonin	pH	CMC CNF	Fish fillet (mackerel)	Ezati et al. (2021)
Gas sensor	Oxygen sensor	O_2	A blending of polyethylene and ethylene-vinyl acetate	Beef	Kelly et al. (2020)
	RFID	O_2 and CO_2	Wheat gluten	Cheese	Saggin et al. (2019)
	RFID	O_2 and CO_2	Cellulose	Vegetables	Eom et al. (2012)

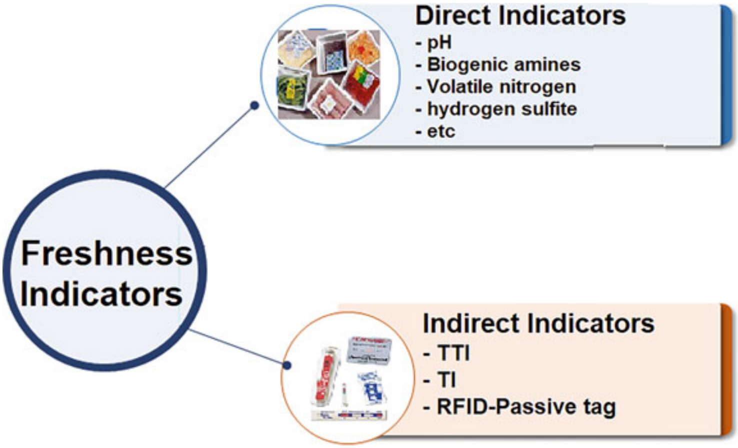

Fig. 2.2 Types of freshness indicators

can be directly related to the food freshness. This colorimetric indicator allows for qualitative or semi-quantitative detection of food quality and safety as a result of physiological changes or microbial growth during food storage, which directly integrated into food packaging, where it can help consumers to judge food quality easily by the naked eye (Ghaani et al. 2016; Kuswandi 2018), while indirect colorimetric indicator works based on imitating or mimicking degradation of targeted food to be monitored, for example, TTI (time–temperature indicator) and T.I. (temperature indicator) (Kuswandi 2018; Kuswandi 2020). In other perspectives, these freshness indicators could also be classified into two types, i.e., the first one is an internal indicator, and the second, external indicator. The internal freshness indicators are placed inside the food package, and the external freshness indicators are physically placed outside the food package (Pavelková 2013). Both types of freshness indicators can inform consumers related to the presence or absence of specific compounds, or the concentration of a specific substance, related to the level of food freshness. Thus, a distinct characteristic of freshness indicator is the type of information they provide, either qualitative or semi-quantitative toward the user for freshness level.

Freshness indicators employed in food packaging inform a consumer on the issue of food quality and safety related to food degradation due to microbial activity or other food properties. Mostly, the information is presented by immediate color change variations, such as different color intensities or the dye diffusion inside the indicator area (Kalpana et al. 2019; Kuswandi 2020). Even though freshness indicators are simple devices but crucial in assuring food quality and safety, they allow a reduction in the food loss and the cost loss due to the food damage. Therefore, it is very useful for nondestructive testing of food freshness, especially for perishable foods and dairy products, such as fish, meat, milk, fruits, and vegetables. Therefore, in other senses, it is also called a spoilage indicator since it is also used to indicate food deterioration or spoilage. Even though intelligent food packaging has many benefits for perishable foods, however, its application in the food industry is still limited, and it needs further exploration and commercialization.

Commonly, freshness indicators are monitoring the packaged food quality by detecting metabolite compounds produced associated with the food freshness causing by natural metabolism or microbial growth (Kuswandi et al. 2011; Realini and Marcos 2014). Physicochemical changes during food distribution and storage are indicators for food freshness. Food metabolites change during storage, such as pH, lactic acid, carbon dioxide, ethanol, volatile nitrogen, biogenic amines, or hydrogen sulfite, are an indication of microbial activities inside foods, which, in turn, can be employed as indicators of food freshness (Arvanitoyannis and Stratakos 2012; Kuswandi et al. 2011). Freshness indicator monitors food freshness via this metabolite detection that has been reported in the literature (Kuswandi 2018), even though, their successful commercialization is still limited. For instance, FreshTag® and Toxin Guard™ are commercial freshness indicators that have been discontinued. The former was a colorimetric freshness indicator that detects the volatile amines produced in fish, based on the immobilization of antibodies into plastic films to detect food pathogens (Kuswandi et al. 2011).

2.2.2 Ripeness Indicators

Freshness indicators are mainly employed for freshness monitoring of protein-based food products, e.g., meat and seafood. Besides these, particularly, fruits and vegetables produce a large number of volatile compounds during ripeness and maturation that can be used to indicate ripeness and maturity. For the first time, in 2004, P-P Enterprise's supermarket in New Zealand released a new ripeness indicator, namely ripeSence (T.M.) for pear, as a new intelligent packaging system. It is the first intelligent label in the world that can indicate the fruit ripeness or maturity or even rotten. The ripeness indicator could detect fruit ripeness by determining the fruit aroma compounds released during fruit ripening. Volatile aldehydes, such as acetaldehyde produced during stone fruit maturation, such as apple, could be used to detect their maturity. An indicator label based on methyl red can detect the release of aldehyde from an apple. The ripeness indicator is fabricated by special ink printed directly on a paper medium. Under mature conditions, the indicator color developed from yellow to orange and then to red (Kim et al. 2018).

Ethylene produce can directly relate to fruit ripeness or vegetable maturity. A colorimetric ripeness indicator for color change upon reaction with ethylene has been developed. The indicator can be used to indicate the ripeness or maturity via the ethylene release during ripening of fruits and vegetables, such as apple (Lang and Hübert 2012) and kiwifruit (Hu et al. 2016). With the development of ripeness indicators, a variety release of aromatic substances during fruit and vegetable maturation or rotten can be employed as releases indicating fruit and vegetable ripeness.

pH could also be used as an indicator for fruit freshness since many metabolite changes during fruit ripeness or vegetable maturity relate to the pH change inside

Fig. 2.3 Ripeness indicators were used for monitoring fruit ripeness (freshness) for strawberries (**a**) and grapes (**b**)

food packaging (Kuswandi 2018). For example, a ripeness indicator was developed for monitoring guava (*Psidium guajava* L.) freshness based on immobilized bromophenol blue onto a cellulose membrane by absorption (Kuswandi et al. 2013). The ripeness color indicator developed from blue to green as the pH declined as a result of the guava over-ripening, where at this stage, the volatile organic compounds were released. This indicator was successfully applied to detect the guava ripeness at ambient temperature (28–30 °C). Furthermore, fruit ripeness indicators have been developed by the same group for strawberries (Kuswandi 2013) and grape (Kuswandi and Murdyaningsih 2017) using a similar principle based on pH change in fruits monitored as given in Fig. 2.3. Another indicator for vegetable maturity was developed based on a silver nanoparticle that has yellow color as a colorimetric sensor for the detection of organosulfur compounds produced when onion spoilage. The color developments were observed during 10 days, where the color developed with time, from yellow to orange, then pink, and lastly colorless when the onion spoilage.

2.2.3 Other Indicators and Sensors

Other indicators or often called colorimetric sensors since these smart devices can be classified as colorimetric chemical sensors or biosensors depend on the sensitive membrane or film used. One example of this type of indicator is integrity indicator, where it is informing regarding how long a packaged food has been opened or inform the gases presence inside the food package. Usually, this integrity indicator is employed with MAP (modified atmospheres in packaging), where the oxygen gas replaces other gases, such as nitrogen or carbon dioxide, which, in turn, enhances the shelf life of foods.

Ageless Eye, Tell-Tab, and O_2 Sense are examples of commercial integrity indicators that provide color changes upon oxygen exposure so that suitable for the naked eye detection in terms of oxygen concentrations at low concentration (0.5%). Another example is Food Sentinel System™ that has been discontinued already. It is a biosensor system for the food pathogen detection using a specific pathogen antibody immobilized onto the membrane creating a barcode part (Yam et al. 2005). Here, when the contaminating bacteria are present, it will produce a localized dark bar, which, in turn, causes the error or unreadable barcode while scanning when it is used in the supermarket.

2.3 Smart Indicator for Food Freshness

Based on the type of design construction, the food freshness indicator could be grouped into (1) single indicator; (2) dual indicator; and (3) indicator array (Kuswandi 2020). The single indicator is a freshness indicator that only used one indicator to detect, sense, trace, and inform the food freshness quality and safety inside the package. The dial indicator is the freshness indicator that used two indicators in monitoring and informing the food freshness quality and safety. At the same time, an indicator array is the multiple indicators that used several indicators in an array to create a pattern in detecting or sensing, and informing the food freshness quality and safety. Mostly, commercially available freshness indicators are a single indicator type, while dual indicator and indicator array are undergoing laboratory-scale developments. Figure 2.4 shows the ripeness indicator or sensors based on design, type, and application for food freshness.

The smart indicators integrated into food packaging can act as an important tool in sensing, detecting, monitoring, and tracing food freshness quality and safety.

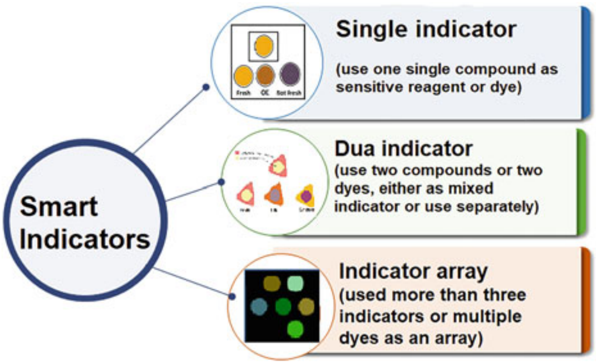

Fig. 2.4 Types of smart indicators used in food packaging for freshness monitoring

These smart indicators or sensors are critically important when foods are stored outside their desired conditions, e.g., extreme hot or cold. Furthermore, in the case of processed foods, where they should not be frozen, a smart indicator could be necessary to monitor whether they had been stored in freezing conditions improperly or not. On the other hand, a smart indicator could also inform if the processed food is sensitive to heat where it had been exposed to hot conditions and the duration involved. Moreover, the detailed description of the smart indicators for food freshness quality and safety is described in the following subsections.

2.3.1 Single Indicator

Commonly, a single indicator or sensor is used in a single color indicator construction, and the choice of reagent dye has a great impact on its colorimetric indicator performance. In order to construct high sensitivity of the colorimetric indicator or sensor, the reagent dye should have high sensitivity as well. For instance, if a single indicator works based on pH change inside the headspace of food packaging, therefore, a large pH range of the reagent dye is needed, or it should be sensitive enough to pH change and easy to distinguish its color change related to the food freshness during storage (Kuswandi 2018). By using a single indicator, simple, fast, and sensitive detection of food freshness can be performed by using a noninvasive colorimetric method detected by the naked eye. For example, it is used for fish freshness detection, where it was observed to relate well with bacterial growth trends in whiting and codfish samples, which in turn allowing real-time monitoring of fish spoilage (Pacquit et al. 2007; Kuswandi et al. 2012a, b). This single indicator allows the high potential for creating "best-before" dates where it could make improvements in the food quality evaluation of food freshness active label.

Polyaniline (PANI) could make a distinctive color change with total volatile base nitrogen (TVBN), so it is often used as a single indicator in an intelligent packaging system. A single indicator was developed based on a similar working principle based on PANI film for milkfish sample (*Chanos chanos*) freshness monitoring in the fish package headspace (Kuswandi et al. 2012a, b). This single indicator showed color changes toward a variety of TVBN produced during fish deterioration as shown in Fig. 2.5. Here, the PANI film response presented as a color change is also related to microbial activities in fish samples (*Pseudomonas spp*. and TVC (total viable count)). The single indicator or sensor allows for the real-time spoilage detection of fish either at stable or fluctuating temperatures. Furthermore, a single biosensor was developed for xanthine (adenine nucleotide degradation product in animal tissue) detection (Arvanitoyannis and Stratakos 2012). In this work, the xanthine oxide was attached to the electrodes, e.g., silver, platinum, and pencil graphite electrode (Devi et al. 2013; Dolmacı et al. 2012; Realini and Marcos 2014). Another novel single indicator was developed based on curcumin for volatile amine (TVBN) detection in shrimp (Kuswandi et al. 2012a, b). The curcumin was absorbed onto the bacterial cellulose membrane to create a sensitive indicator and edible membrane for food applications. The indicator develops color from yellow to orange and finally to

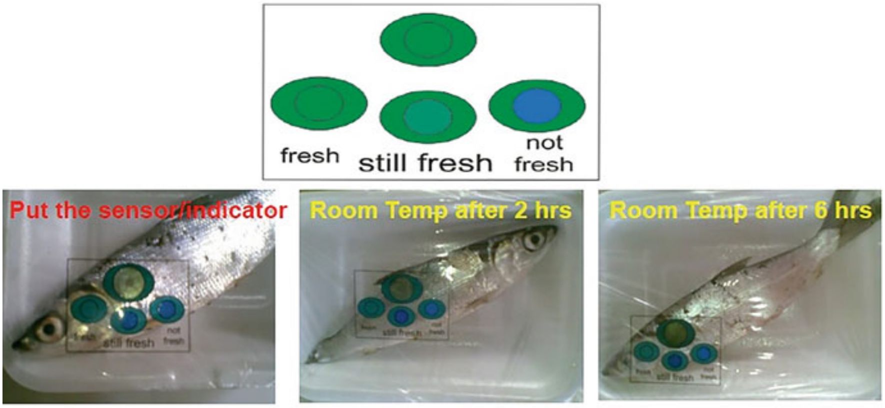

Fig. 2.5 Example of a single indicator used for fish freshness monitoring based on PANI

reddish orange for the indication of shrimp spoilage. Moreover, the indicator color change was correlated with bacterial activities in shrimp in ambient and chillier conditions.

A single indicator was constructed to monitor the guava ripeness. The single indicator was immobilized bromocresol blue on the cellulose membrane for acidic volatile compound detection. Here, when the indicator color changes from blue to green, the maturity of guava changes from ripeness to rotten (Kuswandi et al. 2013). It is often that ripeness and rotten fruit foods can be distinguished by only two color changes, since secondary ripeness fruits cannot be identified, such as medium condition. Therefore, the same group constructed a single indicator based on chlorophenol red as a ripeness indicator for the detection of grape maturity. Here, acid volatile organic compounds released during grape maturation create the indicator color development from white to beige and lastly to yellow that obviously show the ripeness, medium, and rotten grade of grape (Kuswandi and Nurfawaidi 2017).

2.3.2 Dual Indicator

Sometimes, a single indicator is difficult to distinguish the color transition in the process of color change, and therefore, mixture of two indicators could be employed to increase the sensitivity of color development end point. Dual indicator can be divided into three classes: First is to add an inert dye to the acid–base indicator, where the color does not change at the pH less than 7.0, and second is to mix the two acid–base indicators to accurately create the indicator color change in order to have a narrower range of color change by using the complementary effects of each indicator, and the last is using dual indicators simultaneously that used two indicators that referencing each other in detecting, monitoring, and informing the food freshness level, since the amount of TVBN and CO_2 in meat products is the key maker for food deterioration. Therefore, the mixed indicator color change was more obvious in the detection of TVBN at various levels of food freshness. A freshness indicator was developed based on bromothymol blue phenol red for skate fish (*Raja kenojei*) (Lee et al. 2016). Here, TVBN produced during the fish storage was increased in the fish package, causing the headspace pH to increase. As a result, the indicator changed from yellow to purple color when the fish product deteriorated.

The mixed dual indicator could also increase their sensitivity toward CO_2. The mixed dual indicator label of bromothymol blue and methyl red could accurately evaluate the skinless chicken freshness. The label works based on CO_2 detection as the main gas released by the microbial metabolism of skinless chicken. Here, when the mixed indicator shows green and orange color, it means freshness and spoilage, separately (Rukchon et al. 2014). A similar principle was developed using a mixed dye-based indicator in food spoilage for an effective shelf life detection by allowing dynamic freshness to be detected visually alongside the best-before date, which, in turn, reduces margins of error (Nopwinyuwong et al. 2010). Further applications of mixed dual indicator for food freshness indicator to other perishable food products are open up for future area of developments and commercialization.

The last type of dual indicator is dual indicators that used two indicators simultaneously as a label for food freshness monitoring. It was developed as a novel approach for food freshness monitoring, i.e., beef meat, and proposed to prevent the problem using a single indicator (Kuswandi and Nurfawaidi 2017). This is due to the fact that a single indicator is similar to traditional acid–base titration using a pH indicator dye, where it is often difficult to detect the onset of spoilage threshold, as it could be too late or too early if it is related to microbial activities on food products (Kuswandi et al. 2015). The dual indicator has several benefits as smart label, such as suitable for naked detection for the spoilage onset threshold, easily to be displayed and distinguished for each level of the food freshness as two color displays, more accurate food freshness determination, preventing false negative and positive for the level of freshness, more attractive due to two color tone as well as they referenced each other, as one indicator develops color a long side with other indicator color change. It was developed by using two pH dyes (methyl red and Bromocresol purple) to fabricate an on-package dual indicator label for the real-time detection of beef freshness (Kuswandi and Nurfawaidi 2017) (Fig. 2.6).

2.3.3 Indicator Array

Apart from single or dual indicator or colorimetric sensor, indicator or sensor array based on imaging approaches offers many benefits in nondestructive determination of food quality and safety. This imaging technique is using an indicator array or sensor array to capture a pattern of an “odor” fingerprint related to the food quality and safety (Chen et al. 2016; Morsy et al. 2016). Most of the developed indicator or sensor arrays are employed for monitoring chemical species in the food packaging headspace that are related to food freshness or spoilage of perishable foods, e.g., meat, seafood, fruits, and vegetables, which have high value. Since the cost of the sensor array is the most expensive and not simple compared to other smart indicators