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Valorization of Food Processing By-Products as Smart Food Packaging Materials and Its Application

Saroat Rawdkuen and Pimonpan Kaewprachu

Abstract

Traditional food packaging systems cannot provide any information related to the food quality during storage to consumers. Recently, the renewable resources have been considered as starting materials for making biodegradable packaging film. A variety of food processing by-products have been utilized, either alone or in mixtures, to produce packaging films with proper properties. It shows high possibility for smart biodegradable filmmaking as well as is applicable in the food industry. In order to monitor the food quality and to reduce the food loss and waste, a new packaging technology has been increasingly developed. Smart packaging refers to packaging systems which can monitor, detect, and inform about the qualities of food in real time. Indicator is the most commonly used device, which can communicate through direct visual change, especially in color. Natural extract and synthetic color are usually added into smart packaging films. However, synthetic dyes may be harmful to the consumers' health. Thus, the use of natural extract has been increased. Smart packaging films can be applied to various types of food products in order to monitor the food quality during transportation and storage. Thus, smart packaging could be used as a nondestructive tool to detect the food quality.

Keywords: biodegradable, by-products, food packaging, indicators, natural extract, smart films, valorization

1. Introduction

Nowadays, the food industry is continuously growing due to increasing levels of population and globalization and is providing a wide variety of food products to meet customer needs. The major food industries of the world include meat, poultry, fish and seafood, fruits and vegetables, dairy, and cereal. During food processing, a large number of by-products are thrown away into the environment, resulting in accumulation in the environment, and may also interfere with natural process of ecosystem. According to the FAO, 1.3 billion tons of foods are by-products. The global food losses and waste are strongly dependent on the kind of foods. The highest percentage of by-product is found in fruits and vegetables, plus roots and tubers (45%), followed by fish and seafood (35%), oilseed, meat, and dairy (20%),

respectively [1]. In developing countries, food waste is generally generated at post-harvest and processing levels at around 40%, while in industrialized countries, the sector contributing a huge by-product (more than 40%) is households [1]. These by-products still contain the organic matter that have a potential for making the packaging or developing new valuable products from them for commercial applications. Thus, valorization of food processing products is an interesting concept that offers sustainability rather than landfilling or disposal.

Traditional food packaging has a role to protect the packaged food from mechanical damage and physical stress, communicate with the consumers, provide convenience, and contain the product [2]. This packaging system cannot monitor the quality of packaged food products and inform the status of environment and food conditions to the consumers at any time and cannot meet the consumer preferences. Thus, a new packaging technology has been developed and also proposed with the aim to reduce the food loss and waste throughout the food supply chain. Thus, smart packaging or intelligent packaging concept has been raised, which is considered as innovative packaging. According to EC [3], intelligent materials and articles are materials and articles which monitor the condition of packaged food products or the surrounding environment of the food. Yam [4] defined intelligent packaging (also described as smart packaging) as a packaging system that used to monitor the condition that the packaged food product is exposed to and provide information about the packaged food quality to the consumers through the visual color changes.

Generally, there are three main types of smart packaging, which include sensors, indicators, and radio-frequency identification (RFID) devices [5]. Food spoilage or fermentation process is commonly accompanied with a pH change. Thus, pH indicator has gained much popularity, compared with others. Thus, pH indicator generally comprises a dye that changes in color in a function of pH. There are two main sources of indicators: chemical dye and natural extract. Chemical dyes include methyl red, methyl orange, bromothymol blue, bromocresol green, phenol red, and their combinations. A natural dye can be obtained from the root, flower, leaves of plants, and other parts of plant materials that contain a natural pigment, such as anthocyanins. The advantage of natural dye is safety and being eco-friendly to the consumers, compared to chemical dye. Thus, the natural dye has been increasingly used in smart packaging. These indicators can interact with the internal (metabolites in the head space and food components) and/or external factors (environmental surrounding). As a result of this reaction, they generate the response through the color changes or electrical signal and depend on types of intelligent packaging, which relate to the actual status of packaged food product. Thus, the use of smart packaging could help to decrease the number of food loss and waste by sensing or communicating the actual quality of the packaged food product to the consumer in real time through visible color changes.

Smart packaging has been widely applied on various types of food products [6–9]. The use of these packaging systems could facilitate the consumers to know the quality and the condition of packaged food (food spoilage, ripeness, or degree of fermentation) without damaging the package. From the food quality and safety point of view, smart packaging is very useful to the producers, sellers, and consumers to give information regarding the packaged food condition through a change of tools. In this context, the valorization of food processing by-products to produce packaging films is reviewed. The principle and types of smart/intelligent packaging are presented. The current researches on the applications of smart packaging on various kinds of food products are also discussed.

2. Valorization of food processing by-products

A large amount of food materials as by-products, which are generated along the chain of food production and transformation, are thrown into the environment. Food processing by-products generally include the residues or remain that were discarded after removing the desirable portion for further processing or direct consumption as food. According to Ezejiofor et al. [10], residues from food processing make up 30–60% of the product that is used for human consumption and animal feeding. Most of the by-products commonly contain an organic component, such as proteins, carbohydrates, and lipids, which are promising sources of value-added substances that can be extracted and utilized as a starting material for forming a smart packaging film. The different types of by-products produced by various food processing industries are listed in **Table 1**. Current trends in the world are to recover and utilize the food processing by-products into useful materials and to recycle by-products as a means of achieving goal of sustainable development. Hence, considerable efforts in the valorization of food processing by-products have been made with the purpose of minimizing the amounts of by-products, reducing the environment pollution, and increasing sustainability of these by-products. This section reviews by-products from various food processing industries that have a potential to be produced as smart packaging.

2.1 Meat and poultry processing by-products

The meat processing industry has emerged as a major food industry of the world. As per the FAO [11], the world pork meat production recorded 118 million tons, followed by poultry (117 million tons), sheep (9 million tons), and goat (5 million tons). A huge by-product is generated during the various stages of meat and poultry processing in industries. By-products generated during processing of large animals include the skin (6–10% of live weight), bones (15–20% of live weight), blood (3–9% of live weight), fat (3–4% of live weight), head (6–8% of live weight), and viscera (10–15% of live weight), while poultry by-products include the bones (8–10% of live weight), blood (3–5% of live weight), feather (5–7% of live weight), liver and heart (4–6% of live weight), and viscera (18–20% of live weight) [12]. Valorization of these by-products is a current trend concept for promoting the sustainable development. However, high value-added by-products from the meat and poultry industries are not exploited to its full potential when compared to the by-products from other industries such as fruits, vegetables, dairy, etc.

In general, humans will not consume bones; hence they are disposed into the environment. They are commonly used for animal feeding. However, bones and skin by-products obtained from meat and poultry are important source of proteins, which can be extracted as collagen and gelatin. Gelatin is a soluble protein obtained by degradation of collagen, which is commonly found in animal skin, bones, and connective tissue. Gelatin from poultry is an alternative source

Food processing industries	Generated by-products
Meat and poultry	Skin, bones, blood, head, feather, viscera
Fish and seafood	Skin, viscera, heads, backbones, blood, shells
Dairy product	Whey, lactose
Fruits and vegetables	Peels, pulp, seeds

Source: Ezejiofor et al. [10].

Table 1.
Various food processing industries and their by-products.

for the halal and kosher market. They have many applications in food industry and have been extensively used as a raw material in packaging. Feathers are by-products from industrial poultry production and are mostly discharged without any pretreatment, causing environmental problems. Chicken feather can be considered as attractive sources for the production of packaging films. Proteins (91%), lipids (1%), and water (8%) are the main components in chicken feathers [13]. Keratins are proteins found in chicken feathers. Thus, keratins from chicken feather protein are abundant and a cheap source, can be utilized as edible film material, and can reduce the environment pollution related to the by-products disposed by the processing industries.

2.2 By-products from fish and seafood processing

The industrial fish and seafood processing generates huge amounts of nonedible parts, which are discarded. According to Olsen et al. [14], fish and seafood by-products contribute to around 70% of the initial weight of the catch. Fish and seafood by-products generally contained valuable components such as oil, enzymes, collagen, gelatin, chitin, chitosan, and muscle protein. These valuable compounds have a high potential for using in food, packaging, pharmaceutical, medicine, and other industries. Some of these components from fish and seafood by-products have been isolated and currently sold commercially. The composition and the percentage of nonedible parts of fish and seafood processing are greatly dependent on fish and seafood types and processing methods. For example, by-products generated in finfish processing include the head (14–20%), gut (15–20%), skin (1–3%), bones (10–16%), and trimming (filets) (15–20%), while shrimp generates 65–85% (wet weight basis) of by-products, which is mainly obtained from the head and shell [15].

A high value-added compound can be isolated from fish and seafood processing by-product, which include collage, gelatin, chitin, chitosan, etc. Some of these valuable compounds are able to be formed as packaging films [16], which can reduce the environmental pollution at the same time (films are biodegradable). Collagen is the main structural component, which is widely found in the skin, bone, tendon, and cartilage. Collagen and gelatin have a wide range of applications in various industries. Nowadays, collagen and gelatin extracted from fish and seafood by-products have gained great attention due to the requirement of “halal and kosher” food products and the consumers’ concern about bovine spongiform encephalopathy in collagen and/or gelatin from mammals [17–19]. Thus, fish and seafood processing by-products are one of the most important sources of marine-derived collagen and gelatin.

The crustacean by-products contain chitin content between 2 and 75%, which are dependent on composition, processing methods, and their species [20]. Chitin possesses repeating units of poly- β -(1,4)-*N*-acetyl-D-glucosamine (**Figure 1**). It is the second most abundant natural polysaccharide on earth, after cellulose [21]. It is widely distributed in the nature as a structural polymer in the exoskeleton of insects and crustaceans, which is an important source for chitin industry. Chitin is insoluble in water and even in most organic solvents due to being highly hydrophobic [21]. Chitosan is the *N*-deacetylated derivative of chitin. On the other hand, chitosan is soluble in water, which makes it more convenient to use in different fields. Due to being biodegradable, nontoxic, and biocompatible, chitin and chitosan, which are an agent recovered from shellfish and/or crustacean processing by-products, have been recently found in various industrial applications such as antimicrobial agent, packaging films, pharmaceutical, cosmetic, etc.

2.3 Dairy processing by-products

Dairy processing generates enormous volume of by-products during the processing of milk and manufacture of varied products. These by-products are comprised of high amounts of lipids, proteins, vitamin, etc. The utilization of these by-products can hugely enhance dairy sector profitability. Whey, being a major by-product of the dairy industry, is generated during the manufacturing of cheese. It contains high amounts of proteins which can be converted into various whey protein products, including whey protein concentrates (WPC) and whey protein isolate (WPI). WPC contains 25–85% protein, while WPI contains >90% protein. The pretreatment prior to membrane separation and the membrane used typically affects the whey protein content [22]. Thus, the whey protein obtained has potential for use in the packaging and food industries such as protease inhibitor in surimi processing [23].

2.4 Fruit and vegetable by-products

The total worldwide annual production of fruit was reported to be more than 892 million metric tons in 2016 [11]. During fruit and vegetable processing, enormous numbers of peel, pulp, and seed are generated which are disposed into the environment. These by-products represent approximately 30–50% of the total weight, which can affect the environment and health sectors, due to their methane emissions and biodegradability [10]. These by-products are commonly utilized as animal feed; however, they still comprise of huge amounts of organic components such as polysaccharides, proteins, lipids, and other aliphatic and aromatic compounds. Thus, these by-products can be further utilized or extracted as high-value substance and can be used as alternative sources with a high potential for the development of new packaging technologies.

Banana (*Musa paradisiaca* L., Musaceae) is the most important fruit crop, with a global annual production of 113 million tons in 2016 [11]. Banana peels are usually discarded during the banana processing or used in the form of animal feed. Peels, by-products from banana, can represent 40% of the total weight [24]. However, this by-product is considered as a good and cheap source of valuable compounds. Moreover, a huge amount of banana that does not meet the standard exportation is obtained. The whole banana is a good source of polysaccharide, especially starch. Unripe banana generally contains starch more than 70% with other components (protein, fiber, and lipid) [25]. Banana starch and flour have been used as a starting material to form edible films [25, 26].

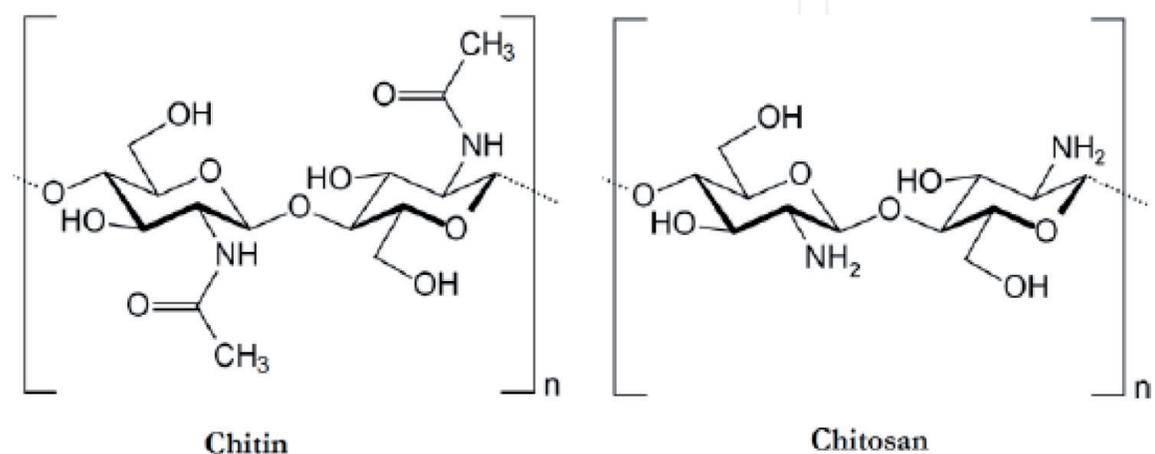


Figure 1.
Chemical structure of chitin and chitosan.

Mango (*Mangifera indica* L., Anacardiaceae) is the most important fruit in the tropical region, especially in Thailand. The major by-products of industrial mango processing are peels and kernels which represent about 35–60% of the total fruit weight [27]. Mango kernels are promising sources of edible oil, starch, flour, and essential amino acids [27–29]. Mango peels have been reported as a good source of pectin, which is considered a high-quality dietary fiber [27]. Thus, pectin extracted from mango peels has a potential for developing packaging films.

Rice (*Oryza sativa*) is one of the most common foods that people usually consume around the world. According to the FAOSTAT database, the worldwide annual production of rice was over 740 million tons in 2016 [11]. Rice milling process is required to remove unwanted material, prior to cooking. After rice milling process, 70% rice is obtained, while rice hull (20%), rice bran (8%), and rice germ (2%) are the by-products [24]. However, some amount of broken rice also occurred during milling process. Broken rice is a good source of starch and flour and can be used to produce as rice starch/flour films. Rice bran is a rich source of fat, protein, carbohydrate, vitamins, minerals, and antioxidants [30]. Rice bran has a potential to produce packaging film; however, it is very scarce to use rice bran as a starting material for film production.

According to the FAOSTAT database, the worldwide annual production of soybean was over 334 million tons in 2016 [11]. Soybean is the most important of legume crops. Soy protein resin is obtained from soybean harvesting and processing as by-products. Soy protein is typically categorized as soy flour (50–59% protein), soy protein concentrate (SPC) (65–72% protein), and soy protein isolate (SPI) (>90% protein) [31]. They are widely used as a starting material to form film due to their abundant, biodegradable, cheap, functional properties and high nutritional quality. Thus, the utilization of soy protein in packaging technology can provide a value to soybean by creating a new route for the marketing of soy protein materials.

3. Intelligent packaging

3.1 Principle

According to EC [3], intelligent materials and articles are materials and articles which monitor the condition of packaged food products or the surrounding environment of the food. Intelligent packaging refers to packaging materials that provide a total packaging solution and monitor changes in the quality of product or its environment [32]. Poças et al. [33] defined smart or intelligent packaging as a packaging system that is associated with communication. Intelligent packaging can also be defined as the packaging material that contains the communication functions for recording the internal and external environment changes and then inform the users about the packaged food product's status [4].

This packaging system utilizes a variety of sensors or indicators to detect the quality and safety of packaged food products and may inform the product's status and/or environment condition to the producers, retailer, or consumer at any given time. Food freshness, pH level, microbial growth, gas (carbon dioxide: CO₂ and oxygen: O₂) in the package headspace, time or temperature are an example of sensors or indicators. Smart packaging concepts and types including sensors, i.e., gas sensor and biosensor; indicators, i.e., freshness indicators, time-temperature indicators (TTI), and gas indicators; and radio-frequency identification devices are described in this chapter.

3.2 Sensors

Many smart or intelligent packaging concepts involve the use of sensors. This system is generally used in terms of a combination of sensors with packaging technique such as modified atmosphere (MAP) and vacuum packaging. Kerry [5] defined sensors as a small device that used to detect, locate or quantify energy or matter, giving a signal for the detection or measurement of a physical or chemical property to which the device responds. To qualify as a sensor, a device must provide continuous output of a signal. Most sensors commonly comprise of two basic functional parts, a receptor and a transducer. In the receptor, physical or chemical information is transformed into an energy form, which may be detected by a transducer. The transducer is a device capable of transforming the energy carrying the physical or chemical information about the sample into the signal [5].

3.2.1 Gas sensors

Gas sensors are devices that respond to the presence of a gaseous analyte by changing the physical parameters of the sensor and are monitored by external devices [34]. It includes organic conducting polymers, metal-oxide-semiconductor field-effect transistors, potentiometric carbon dioxide sensors, piezoelectric crystal sensors, and amperometric oxygen sensors [5]. Systems commonly contained a solid-state material, which operate on the principle of luminescence quenching or absorbance changes caused by direct contact with the gas analyte [34]. Optochemical sensor based on gas-phase protonated tetraphenylporphyrin (TPP) was developed by Tuerdi et al. [35]. This system was used for detecting volatile amines (ammonia, NH_3). The authors concluded that this sensor could detect NH_3 gas at very low levels (0.1 ppm). Thus, this system is a noninvasive technique, has high sensitivity and fast response-recovery times for gas analysis, and is potentially suitable for applications as smart packaging.

3.2.2 Biosensor

Yam et al. [2] defined biosensors as compact analytical devices that are used for detecting, recording, and transmitting information to biological reactions. Biosensors usually comprise of the bioreceptors and transducers [5]. The bioreceptors, including organic or biological materials (enzyme, hormone, microbes, etc.), recognize a target analyte. The transducers, such as electrochemical, acoustic, or optical, can convert biological signals into quantifiable electronic response [2]. Chemiluminescence biosensor for the detection of putrescine (biogenic amines) in meat product was developed by Omanovic-Miklicanin and Valzacchi [36] by covering the Co(II) and enzyme (putrescine oxidase or diamine oxidase) onto glass supports with hydroxyethyl cellulose membrane. Recently, biosensors based on heme entrapped in recombinant silk film on a glass carbon electrode modified with multiwalled carbon nanotubes for detecting nitric oxide (NO) at nanomolar levels in the presence and absence of oxygen were prepared by Musameh et al. [37]. This system should have high sensitivity, accuracy, precision, and stable detection of analyte.

3.3 Indicators

Indicators may also be defined as a substance that indicates the absence or presence of another substance or the degree of reaction between two or more substances by means of characteristic changes, especially in color [34]. The difference between

sensors and indicators is the components; indicators do not contain receptor and transducer components as the sensor, so it provides qualitative information through directly visible color change. According to Smolander [38], changes in color of pH dye indicator can be proposed to investigate acidic and/or basic volatile compounds and provide an irreversible color change in an appearance.

3.3.1 Freshness indicators

Freshness indicators are devices that are printed on the packaging film or in the form of a package label and then attached inside the packaging materials. Freshness indicators have been developed with the aim of investigating the food spoilage process, degree of fermentation, or ripening stage of fruits and vegetables and then informing the users about the freshness of packaged goods through color changes that can be directly detected by the naked eye. Freshness indicators provide direct information of the packaged food quality resulting from chemical changes or microbial growth within a food product. Indicator may react to metabolites that are generated in the package as a result of metabolism or the microbial growth [34]. Freshness indicator concepts based on pH sensing film, using CO₂ as the major target, have been reported [39]. Besides CO₂, also other metabolites like the volatile compounds trimethylamine (TMA), dimethylamine (DMA), and ammonia, collectively known as TVB-N; biogenic amines such as histamine, putrescine, tyramine, and cadaverine; ethanol; sulfuric compounds; and organic acids have been studied as suitable target molecules for these pH-sensing indicators [34]. It can sense and share information to the producer, retailer, or consumer at any time. Thus, the development of freshness indicator is based on a broad knowledge of quality-related metabolites closely related to product types, packaging material, the growth of microorganisms, and storage condition.

Various kinds of freshness indicators have been developed [40–43]. Freshness indicators are normally comprised of food spoilage and ripeness indicators [44]. Food spoilage indicators commonly investigate the spoilage of food due to the chemical changes, microbial growth, temperature abuse, or packaging leakage. The concepts for food spoilage indicators are based on changes in color indicator which response to microbial metabolites produced during spoilage process. Ripeness indicators are mostly used in fruit and some kind of vegetable product. However, this indicator is suitable for climacteric fruit rather than non-climacteric fruit because after harvest they still continue to ripen which is easily detected by indicators. During fruit ripening, there are many changes that occur such as loss of chlorophyll; ethylene production; conversion of starch to sugar; aroma development; changes in organic acids, proteins, and fats; etc. [33]. Changes in color indicator can help the consumers to decide the product when these fruits are fully mature, ripen, and ready to eat, e.g., durian, mango, and banana [7, 40]. However, freshness indicator has a disadvantage; it provides the broad-spectrum color change, which needs to be resolved before being used commercially.

Chen et al. [45] developed on-package indicator labels for the determination of lean pork freshness. The indicator label was based on methylcellulose immobilized with mixed dye (bromothymol blue/methyl red, 3:2). These labels were used to monitor freshness of pork at 5°C for 8 days. The indicator label presented visual color changes due to the presence of volatile compound (TVB-N) and aerobic plate counts. The authors reported that the colorimetric freshness indicator was able to recognize fresh (0–3 days) (red), medium fresh (4–5 days) (goldenrod), and spoiled stage (6–8 days) (green). Therefore, the indicator labels could be used

to monitor the real-time pork freshness as intelligent packaging tool and could enhance the guarantee of pork safety.

3.3.2 Time-temperature indicators

Meng et al. [6] defined time-temperature indicators (TTI) as a small device placed on a product or package that can be used to record, monitor, and indicate the time-temperature history on the quality of perishable product during the cold chain transportation and storage from the point of producer to the end consumer. Temperature is a critical factor influencing the food quality and safety during retail outlet or distribution and storage. The temperature history of packaged food is very difficult to control and monitor; thus, it is difficult to predict their shelf life. Thus, TTI can help to monitor and record the temperature condition during distribution and storage. The principle of TTI operation is based on chemical, enzymatic, mechanical, electrochemical, or microbiological change, generally expressed as a visible response in the form of a mechanical deformation, irreversible visible color development, or color change movement [46]. TTI should also be low-cost, reliable, with good stability, flexible to a wide range of temperatures, nontoxic, and easily integrated into a packaging material.

The TTI is useful because it can inform the consumers when the product has been temperature abused. If the temperature of the product is higher than the temperature recommended, the food quality can quickly deteriorate. TTI is mostly used in chilled or frozen foods, where the cold storage during distribution is important for food quality and safety. The role of developed system is to detect the quality changes by evaluating the pH alteration of the packaged foods during transportation and storage. Different kinds of food material will be stored at different temperatures; the system had the ability to detect temperature changes indirectly with the help of variation of pH of the food products, which occurred due to the improper temperature for the transportation and storage. Recently, a prototype diffusion-based TTI has been developed by Suppakul et al. [47]. A polydiacetylene (PDA)/SiO₂ nanocomposite was used as the color-developing substance and loaded on the diffusion path. Tween 20 was used as a moving substance. The authors said that when Tween 20 reached the test line of the PDA/SiO₂ nanocomposite, the color of the line changed from blue to red, indicating the TTI endpoint. Moreover, four TTIs were designed by matching the TTI endpoint with the deterioration time of food during storage at different temperatures (5, 10, 15, and 25°C). When diffusion-based TTI was applied on the tested food, the line of the PDA/SiO₂ nanocomposite was matched to the shelf life of the tested food during storage.

3.3.3 Gas indicators

It is very difficult to assess the packaged food quality because of many factors such as the production of gas by microorganisms within the package, changing concentration of gas and gas leakage from outside or inside of the packaging materials, or continuing fruit and vegetable respiration. Thus, gas indicators have been developed to solve these problems. Gas indicators, commonly produced in the form of label, are attached inside the food package for monitoring the changes of the level of gas, such as O₂ and CO₂, and then provide information through visual color changes [48]. A mixed pH dye-based indicator was developed for checking their ability of changes in color at different concentrations of CO₂ (10–80%) [48]. This indicator is relatively sensitive to the change of CO₂ level by changing the color from blue to green when the CO₂ level increased. The principle of gas indicators

(pH dye-based) was explained by Puligundla et al. [49]. CO_2 generally dissolves in an aqueous solution and then forms carbonic acid (H_2CO_3). H_2CO_3 dissociates into hydrogen ions (H^+) and bicarbonate ions (HCO_3^-). Then, H^+ , as a proton, combines with a water molecule to form a hydronium ion (H_3O^+). This H_3O^+ reacts with the basic (dissociated) form (In^-) of the pH dye indicator, resulting in an acid (protonated) form (HIn) which in turn develops a color change in the indicator containing label [49].

3.4 Radio-frequency identification devices

RFID, a food traceability system, is a small device that can be attached to the product, so that the product could be identified and tracked in real time. It is commonly grouped under the form wireless automatic identification together with biometrics, magnetic inks, QR codes, barcode, etc. [32]. RFID tags mainly comprise of three components including a tag produced from a microchip linked to a small aerial, a reader capable of discharging radio signals and also accepting answers from the tag in response to the sent signals, and a network system or web server that connects the company and the RFID equipment [50]. There are three categories of RFID, which based on the power supply include passive RFID tags, semi-passive RFID tags, and active RFID tags [32]. This tool is suitable for the large-scale production network, such as food supply chains, which can control material flow and/or give information. It is typically used in order to reduce uncertainties in the food purchasing process by giving information about the whole process in terms of quality and safety.

4. Sources of dye indicator

The inclusion of dye indicator into packaging materials is a promising method for the development of smart packaging. Dyes can interact with stimulus; then the color changes are obtained, which can be observed by the naked eye. Color change of dye is cause of the electron at double bonds in the dyes able to absorb the energy at visible range wavelengths (400–700 nm). Difference structures of dye will absorb different wavelengths, which are related to their colorations. It is an organic or inorganic substance. Generally, there are two main sources of indicator: chemical dye and natural extract (**Table 2**). These components are widely used as coloring agent because they are sensitive to the condition change and are able to inform the packaged food conditions to the manufacturers and the end consumers. Food deterioration or fermentation process is mostly correlated with pH change. Thus, pH indicator dye-based colorimetric is a type of dye indicator that is widely added into materials.

4.1 Chemical dye

Most of the chemical dyes are made up of synthetic material, which may be toxic to the consumer and not suitable for use in food application. The commercially available chemical pH dye is mostly based on halochromic compound and belonged to sulfonephthalein dye and azo dye groups. Sulfonephthalein dyes include phenol red, bromophenol blue, bromocresol green, and bromothymol blue, and azo dye includes methyl red (**Figure 2**). These chemical dyes are widely used as pH dye indicator because they exhibit a clear color according to pH. Change in color of pH dyes is due to a protonation/deprotonation reaction [64]. There are two forms of pH dye: HIn (acid form) and In^- (basic form). Furthermore, the chemical pH-sensitive dye is often limited in the pH range and showed very narrow color. For example,

bromophenol blue has only two color transitions: yellow at pH 3 and blue at pH 4.6, and phenol red also exhibits two color transitions, yellow at pH below 6.4 and pink at pH above 8.0, which is difficult to observe by the naked eye. Thus, it needs to combine with other chemical dyes to enable pH indication [65]. Baek et al. [48] studied the combination of two chemical dye indicators at different ratios (methyl red/bromothymol blue; 1:9, 3:7, 5:5, 7:3, and 9:1). The authors suggested that the mixing of two dyes exhibited various color changes in a narrow pH range, compared to the methyl red alone or bromothymol blue alone. Therefore, the combination of two dyes has been widely used as chemical indicator [7, 39, 48, 52].

4.2 Natural extract

Nowadays, the consumers concern about the use of the chemical compounds in foods and some chemical substances that are not generally recognized as safe compounds and may be harmful to the consumers' health. Thus, natural extract is an alternative source of indicator that can be used instead of chemical dye. Anthocyanins are natural colorants and water-soluble pigments belonging to the flavonoids family. These pigment compounds are responsible for the color (red, purple, and blue) of plant leaves, flowers, grains, fruits, and vegetables. Changes in color of anthocyanins are mostly due to the presence of phenolic or conjugated compound. The structure of anthocyanins changes when there is a difference in pH values [66]. Thus, anthocyanins are compounds that are mostly used as natural pH dye indicator because it shows a broad color change in function of pH. Color mechanism of anthocyanins can be described as illustrated in **Figure 3**, and the color chart of anthocyanins extracted from different sources of plants at different pH levels, compared to chemical dye (resazurin), is shown in **Figure 4**. Anthocyanins can be exhibited in different chemical structure forms depending on pH. In acidic condition (pH 1), intact structure forms of the flavylium cation (red color) are predominantly formed; at pH between

Sources	Indicators	References
Chemical dye	Bromophenol blue	Zaragoza et al. [51], Chen et al. [52], Dirpan et al. [40]
	Bromocresol green	Pirsa et al. [53]
	Methyl orange	Pirsa et al. [53]
	Methyl red	Niponsak et al. [7], Chen et al. [52]
	Phenol red	Kim et al. [54]
	Mixing methyl red and bromothymol blue	Rukchon et al. [39], Niponsak et al. [7], Baek et al. [48], Chen et al. [52]
Natural extract	Blueberry	Luchese et al. [55]
	Butterfly pea	Rawdkuen et al. [56]
	Grape	Ma and Wang [57]
	Mulberry	Ma et al. [58]
	Purple sweet potato	Liu et al. [59], Choi et al. [60], Yong et al. [61]
	Red cabbage	Pereira et al. [8], Silva-Pereira et al. [62], Pourjavaher et al. [63]
	Roselle	Zhai et al. [9]

Table 2.
 Example of dye indicator.

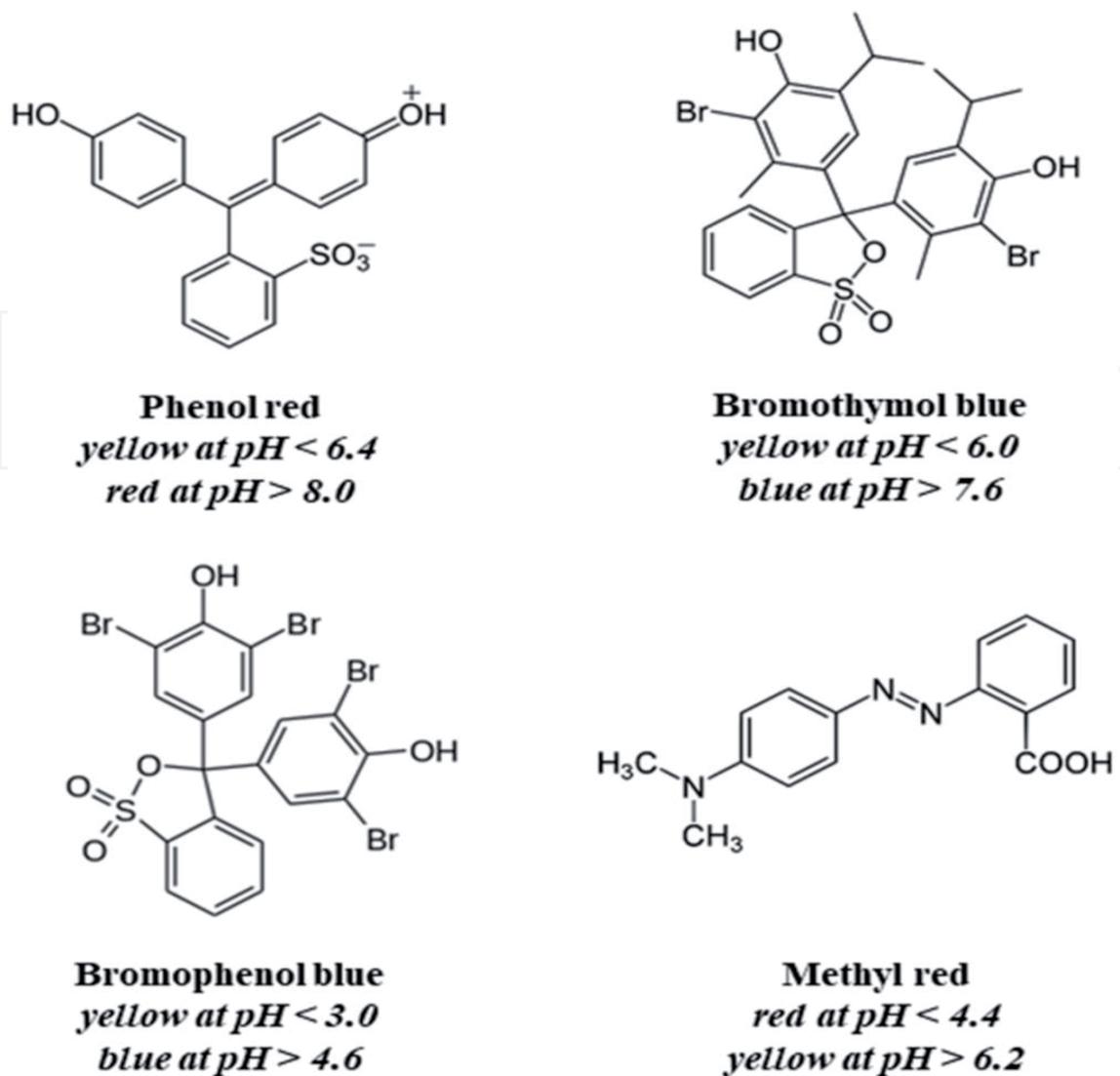


Figure 2.
 Structural formulas of some chemical dye indicators used in indicator labels.

2 and 4, the quinoidal blue species are formed; at pH value between 5 and 6, only two colorless structures (a chalone and a carbene pseudobase) are formed; and in basic conditions (pH > 7), the chemical structure forms are dependent on their substitution groups [67]. Therefore, anthocyanins can go through chemical changes due to pH variation. It can be successfully used as natural dye indicator for determining the food deterioration. Currently used extracted anthocyanins of some plants as natural indicator are shown in **Table 2**.

5. Applications of smart packaging films

The main purpose of packaging for fresh meat, poultry, fish, seafood, fruits and vegetables, etc. is to delay chemical, microbial, and biochemical changes, avoid contamination, reduce weight loss, and enhance overall product appearance to meet consumer expectations and preferences [34]. In general, food quality can be investigated through chemical analysis and microbiological evaluation. However, there are some quality attributes that need to alert the producers, retailers, or consumers during the whole supply chain. The qualities of the packaged food always change after processing or during distribution and

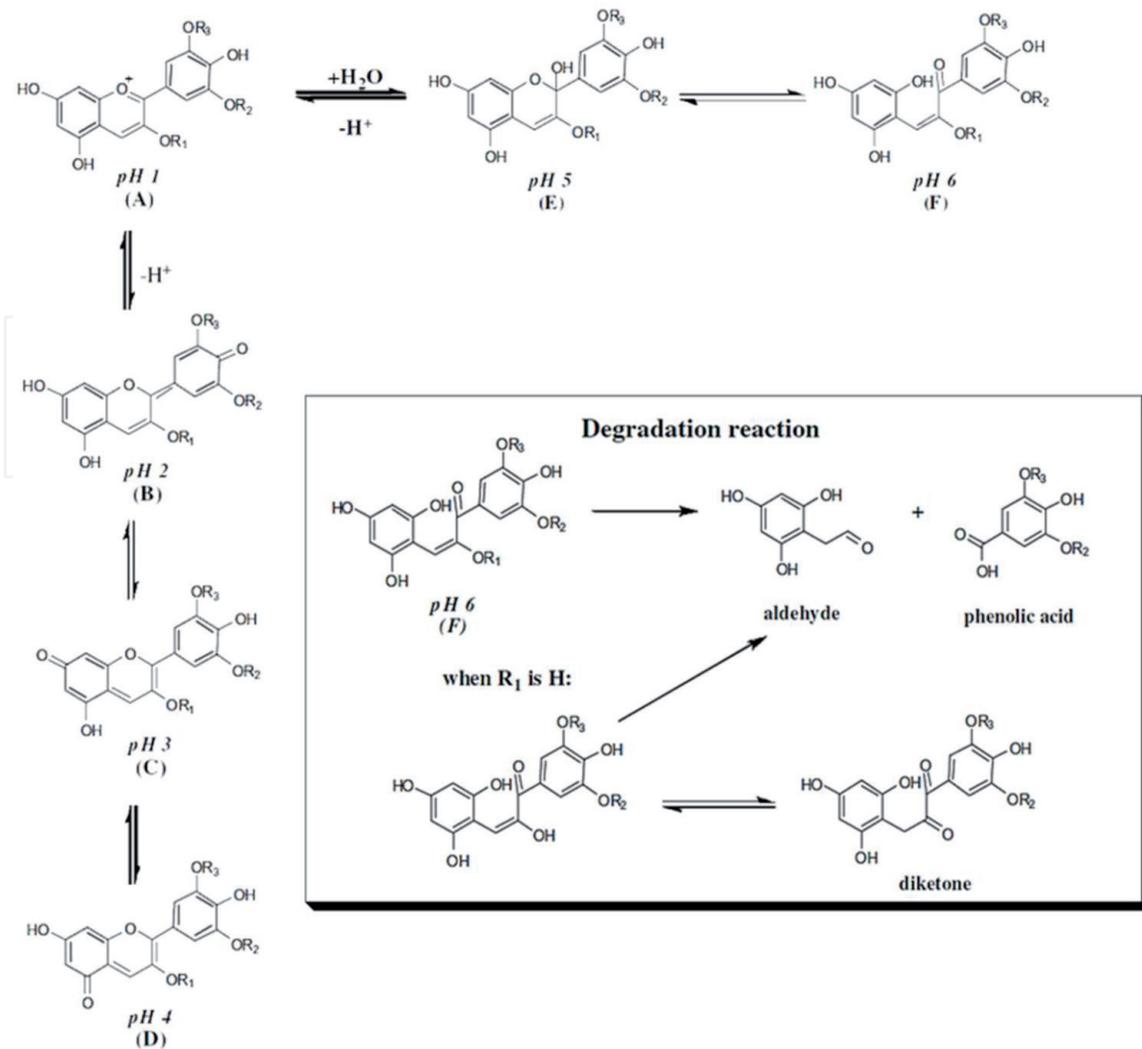


Figure 3. Anthocyanin chemical structure forms depending on pH. Source: Castaneda-Ovando et al. [67].

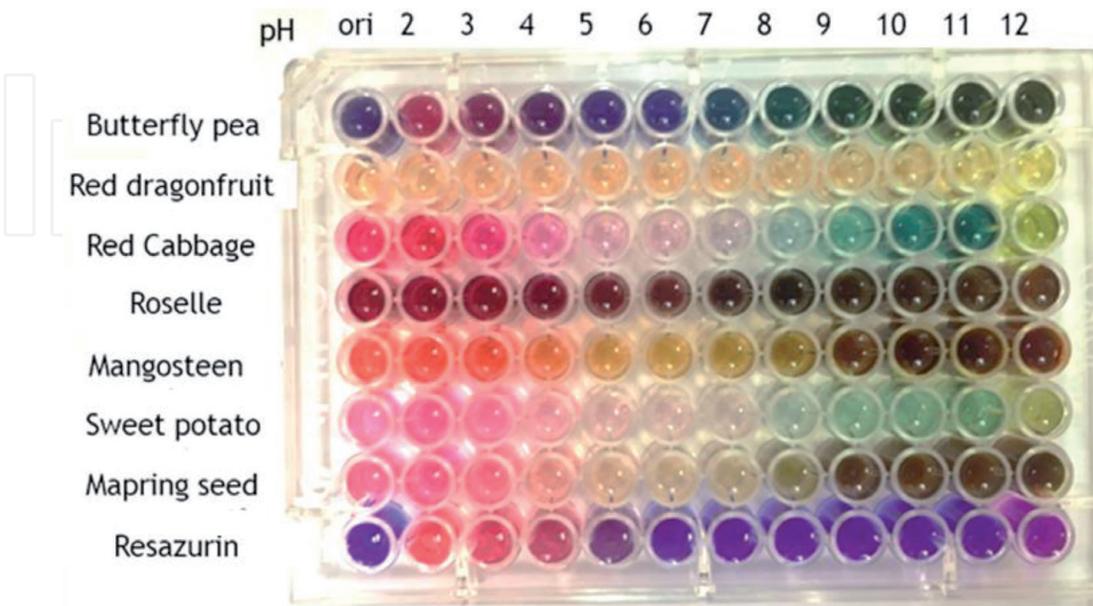


Figure 4. Color chart of anthocyanins extracted from different sources of plants at different pH levels, compared to chemical dye (resazurin). Source: Rawdkuen et al. [56].

storage. Thus, these quality changes of the packaged food are difficult to evaluate by the retailers or consumers. Moreover, different types of food require a specific condition and relative humidity (RH) for their transportation and preservation. With an advantage of smart packaging, it is possible to detect the quality of the food that is packed in the package. Smart packaging tools exhibit some changes in themselves (color development) which relate to the changes in the physicochemical and biological properties of the packaged food. They are mostly placed inside or outside of the package to observe the alteration in the quality of packaged product and to inform the users about the food safety and quality through visible color changes, as illustrated in **Figure 5**.

In the recent year, the applications of smart packaging tools or devices on food product have been described [48, 53, 58]. Indicators are the most frequently used tool due to their nondestructive method, simplicity, low cost, and easy detection by the naked eye. Chemical and biological changes in food product generally occur during handling, storage, and transportation. Contamination by microorganisms is also the main spoilage of most foods. Microbes can be generated the different chemical metabolites, such as CO₂, organic acids, alcohol (ethanol), and hydrogen sulfide (H₂S), and nitrogen containing molecules, which have been proposed as suitable target molecules for indicators [34]. When the levels of these volatile compounds increase, pH level will also increase. As a consequence, the freshness of packaged food is decreasing. The use of colorimetric indicator film as a smart packaging can inform the real conditions of the packaged food to consumer in real-time monitoring of food quality under certain condition. Smart packaging films are widely applied on high-value products, such as meat, fish, seafood, fruits, and vegetables, for monitoring package head-space and/or perishable food spoilage. Hence, smart packaging would be very helpful in ensuring the packaged food safety and quality throughout the whole distribution chain.



Figure 5. Example of freshness indicator for monitoring the degree of fermentation of plaa-som, a Thai fermented fish. The indicator produced from gelatin film incorporated with anthocyanins extracted from butterfly pea (left) and gelatin film containing rezasurin (right).

5.1 Application of smart packaging on meat products

Fresh meat is very perishable and susceptible for physicochemical changes. It generally deteriorates within 3–4 days of slaughter, even if it is kept in refrigeration. Meat products also represent excellent basic nutrients and are easily spoiled by microorganisms. The loss of meat freshness means meat has begun to spoil. The shelf life of meat is the storage time until spoilage, which are complex conditions because the combination of biological and physicochemical activities may interact and make the products unacceptable for human consumption [68]. An acceptable level of microbial, unacceptable appearance, and/or off-flavor is strictly depended on the degree of lipid oxidation, autolytic enzyme reaction, and the level of microbial contamination. Microorganism is one of the major meat spoilers. It can produce the compounds that correlate with spoilage, responsible for strong off-odors and discoloration. Slime production would constitute the major qualitative criteria for the rejection of meat. Meat itself comprises an essential nutrient (amino acids, nucleotides, and sugar). These nutrients are adequate for microbial growth, and the metabolism of these compounds leads to the formation of biogenic amines, H₂S, NH₃, indole, organic acids, and other substances characteristic of meat spoilage [68]. Meat industry requires rapid and simplest tool or device to determine the quality of packaged meat, to predict remaining shelf life of its product, and to provide information on the degree of spoilage. Consequently, smart packaging could be employed to detect microbial metabolites produced on packaged meat sample by reacting with volatile compounds and displaying an irreversible change in visual color, since it can be monitored directly by the naked eye. Applications of smart packaging on meat products are shown in **Table 3**.

Several freshness indicator concepts based on pH dyes, using CO₂ as the main target metabolite have been proposed. An alternative method for detection of chicken breast freshness was proposed by Rukchon et al. [39]. The authors used a

Food product	Material	Indicators	Type of intelligence	References
Chicken breast	Chitosan-corn starch	Hibiscus flower extract	Freshness indicator	Othman et al. [43]
Chicken	Cassava starch	Blueberry residue	Freshness indicator	Luchese et al. [55]
Pork	Chitosan	<i>Bauhinia blakeana</i> Dunn. flower extract	Freshness indicator	Zhang et al. [69]
Pork	Agar-potato starch	Purple sweet potato	Freshness indicator	Choi et al. [60]
Skinless chicken breast	Methylcellulose-hydroxypropyl methylcellulose	Bromothymol blue-methyl red (ratio of 2:3)	Gas indicator	Rukchon et al. [39]
Pork	A4 paper	Glucoamylase, amylose, and iodine	Time-temperature indicator	Meng et al. [6]
Pork	Reverse phase silica gel	Black rice extract	Gas sensor	Huang et al. [70]

Table 3.
 Application of smart packaging films on meat products.

colorimetric mixed pH dye indicator (bromothymol blue/methyl red, ratio of 2:3) based on a methylcellulose-hydroxypropyl methylcellulose film as a chemical barcode for real-time monitoring of skinless chicken breast spoilage. Changes in color indicator from green to orange-yellow were correlated with CO₂ level, microbial growth patterns, and the amount of volatile compounds of skinless chicken breast. The authors also concluded that the degree of chicken spoilage was related to CO₂ levels increased due to microbial growth. Zhang et al. [69] present a convenient, nondestructive, and visible method for pork freshness detection. The indicator response was correlated with pH changes in pork sample, thus enabling real-time monitoring of spoilage. The indicator was prepared by immobilizing natural pH-sensitive dye (*Bauhinia blakeana* Dunn. flower extract) in chitosan, which responded through visible color changes from red to green.

Recently, a system for monitoring chicken breast freshness was developed by Othman et al. [43] based on hibiscus flower extract. The extract was incorporated into chitosan-corn starch-based film, and changes from purplish-gray to darker gray with green color were observed when pH increased as a consequence of the spoilage of chicken breast due to the accumulation of amine and ammonia by mesophilic bacteria. Luchese et al. [55] applied the pH sensing film (anthocyanins extracted from blueberry residue added cassava starch film) on chicken meat indirectly and directly on meat surface for monitoring the spoilage of chicken during storage at 6°C for 10 days. The authors found color pigment migration from indicator to surface of chicken when the pH sensing film was placed directly on chicken meat. This phenomenon may cause the consumer to not accept the product due to changed appearance. Choi et al. [60] developed a pH-sensitive indicator for detecting pork spoilage. pH indicator was prepared by immobilizing a natural dye (anthocyanins extracted from purple sweet potato) into agar-potato starch film. Trials on pork have verified that the color indicator response (from red to green) correlates with pH values in pork sample, thus enabling the real-time detection of spoilage. Meng et al. [6] developed enzyme-based TTI for prediction of pork shelf life during cold storage. It is prepared from sodium alginate, amylose, iodine, and glucoamylase microcapsules and then was coated on A4 paper. After activation (cover with agar), PE film was used to seal the TTI. This type of indicator is based on the reaction between an enzyme and a substrate, which can cause a color change in the system to indicate the reaction degree. When this indicator was applied on pork sample, changed in color from mazarine to colorless was observed. The color change represents the spoilage of chilled pork. A gas sensor array has been proposed by Huang et al. [70] and was used for detecting the biogenic amines generated on packaged fresh pork during storage at 5°C for 7 days. The colorimetric sensor array was created by printing anthocyanins extracted from black rice extract on reverse phase silica gel plates (inorganic material). The authors found a good relationship between the gas sensor array results (i.e., color change) and biogenic amine content. The authors also demonstrated the reaction between indicator and generated volatile compound; the hydroxyl and carbonyl groups of anthocyanin molecules could be interacted with the amines generated during pork spoilage, resulting in changes in color.

5.2 Application of smart packaging on fish and seafood products

Fish and seafood products are classified as extremely perishable products. The decomposition of fish and seafood products can also be due to three mechanisms: enzymatic reactions, oxidative deterioration, and microbial spoilage [71]. Lipid oxidation is the main spoilage of chemical nature. When bacteria grow, they produce metabolic by-products, and their accumulations cause the organoleptic

rejection. However, microbial spoilage is the major mechanism that influences the quality deterioration of fresh fish and seafood and is responsible for shelf life duration. TVB-N, trimethylamines (TMA), sulfuric compounds, alcohols, aldehydes, ketones, and esters are the most common metabolites with characteristic odor that is produced by microorganisms during fish and seafood spoilage [71]. However, the produced compounds are dependent on the metabolic activity of selected specific spoilage organisms (SSO), which is directly influenced by storage conditions. It is difficult to determine the quality of packaged fish and seafood products; thus, a variety of approaches have been developed to monitor the freshness of fish and seafood products in real time. Indicators, one form of intelligent packaging, can react to changes—usually chemical or biological—occurring within the headspace of packaging as the fish and seafood spoil. Thus, indicator generally gives characteristic change: a highly visual color change of the indicator allows for a rapid assessment of the quality of the packed fish seafood products. Considering the food quality and safety, intelligent packaging is an alternative tool that can be used to assess the shelf life and freshness status of fish and seafood products during the supply chain. Applications of smart packaging on fish and seafood products are shown in **Table 4**.

A real-time technique for evaluating fish freshness was prepared by Silva-Pereira et al. [62]. Chitosan-corn starch was used as starting material for making freshness indicator, and red cabbage was used as a dye indicator. The indicator film was used to monitor the fish spoilage storage at room (25°C) and refrigeration temperature (4–7°C) during 7 days. The authors found that the color of the indicator film stored at room temperature was completely changed after 72 h (from blue to yellow), indicating fish spoilage. Ma et al. [41] developed a smart film as indicator for monitoring fish freshness. Smart film was prepared based on tara gum-cellulose nanocrystals containing *Vitis amurensis* husk extract (by-product from white wine processing). The

Food product	Material	Indicators	Type of intelligence	References
Fish	Chitosan-corn starch	Red cabbage	Freshness indicator	Silva-Pereira et al. [62]
Fish	Tara gum-cellulose nanocrystals	<i>Vitis amurensis</i> husk	Freshness indicator	Ma et al. [41]
Fish	PVA-chitosan nanoparticles	Mulberry extract	Freshness indicator	Ma et al. [58]
Silver carp	Starch-PVA	Roselle	Freshness indicator	Zhai et al. [9]
Squid	Aluminum oxide and silica gel	Thymol blue, bromothymol blue sodium salt, bromocresol purple, dinuclear rhodium complex, and bromophenol blue	Optoelectronic nose (sensor)	Zaragoza et al. [51]
Cooked crabs	Paper cellulose fibers	Anthraquinone and azo chromophores	Freshness	Zhang et al. [72]
Shrimp	Tara gum-PVA	Curcumin	Sensor	Ma et al. [41]

PVA: polyvinyl alcohol.

Table 4.
 Application of smart packaging films on fish and seafood products.

colorimetric film was adhered in a sealable bag to directly detect the color difference during fish spoilage. The color changed from pink to yellow-green due to fish spoilage. These color changes were related to the production of volatile basic nitrogen and the increase of pH. Ma et al. [58] developed a pH indicator which consisted of polyvinyl alcohol (PVA)-chitosan nanoparticles and mulberry extract. This indicator was tested for monitoring fish spoilage. The authors found that change in color indicator from red to green was correlated with the presence of volatile nitrogenous compounds, which is characteristic of fish spoilage. Anthocyanins that were extracted from roselle immobilized onto starch-PVA-based film were proposed by Zhai et al. [9] as visual colorimetric film for volatile nitrogenous compounds released in fish spoilage. The visual changes were monitored along 165 h refrigeration temperature (4°C), and the beginning purple color of colorimetric film changed over time to green (at 90 h) and finally yellow (after 135 h). These color changes indicated that the colorimetric films became more basic due to the increasing TVB-N.

The optoelectronic nose is based on the combination of pH indicator and selective chromogenic reagents supported on inorganic materials with diverse acidities and topologies [73]. A novel optoelectronic nose to monitor squid spoilage is proposed by Zaragozá et al. [51]. It is based on five pigments prepared by mixing the different corresponding dyes (thymol blue, bromothymol blue sodium salt, bromocresol purple, dinuclear rhodium complex, and bromophenol blue) with two inorganic supports (aluminum oxide and silica gel). Changes in color of sensor array were characteristic of packaged squid, which kept in cold storage for 12 days. The chromogenic array data were assessed with principal component analysis (PCA; qualitative) and partial least squares (PLS; quantitative) study tools. The PCA analysis carried out with CIE Lab showed that the colorimetric array was able to discriminate between fresh and spoiled squid. The statistical models obtained by PLS, with the optoelectronic nose, successfully predicted CO₂ and O₂ content in the headspace as well as microbial growth. The authors also suggested that this optoelectronic nose could help to evaluate cephalopod freshness, which is easy to use and rapid. Recently, a biosensor (tara gum-PVA-curcumin) was prepared by Ma et al. [41] for evaluation of shrimp spoilage at ambient temperature. The authors found a linear correlation between changes in total volatile basic nitrogen (TVB-N) and pH with the colorimetric response of sensor (slightly yellow to orange-red) within 1–3 min. The authors also suggested the possibility of using the developed film as a sensor for monitoring the spoilage of shrimp and other food products.

5.3 Application of smart packaging on fruits and vegetables

Fruits and vegetables are extremely perishable products and require appropriate handling and storage conditions over the distribution chain in order to preserve their quality and safety and to prolong the shelf life. These products are living products; it means they keep respiring, consuming O₂, and producing CO₂ after harvesting. Thus, the postharvest deterioration process is affected by intrinsic factors of the product. One of the major issues of fresh-cut products is the potential microbial spoilage. Smart packaging can be applied in fruit and vegetable product for facilitating the consumer to know the quality of packaged fruits and vegetables and to promote choosing the fruits and vegetables according to freshness or maturity without damaging the packaging. Generally, changes in color indicators represent the extent of ripening of packaged fruit and vegetable products, or in contrast the indicators represent the spoilage condition (microbial growth). According to Poças et al. [33], the packaging-specific requirements for fruits and vegetables are related to maintaining, controlling, and/or monitoring temperature, gas composition and humidity, and mechanical damage. Applications of smart packaging on fruit and vegetable products are shown in **Table 5**.

The use of freshness indicator on fruit and vegetable products has been proposed. Niponsak et al. [7] described a colorimetric mixed pH dye-based indicator that responds through visible color changes to sulfur-containing volatile compounds released in durian ripening. The strip was obtained by casting method using cassava starch-chitosan, as a based film, and mixing methyl red and bromothymol blue (ratio of 3:2), as indicators. Changes in color in the strip from red to orange and finally turned to yellow indicated the ripening stages of fresh-cut durian. The authors also explained that when the volatile compounds (sulfur; diethyl disulfide, diethyl trisulfide, 3,5-dimethyl-1,2,4-trithiolane) evaporated and reacted with the strip, its changed to the basic form. It is due to the presence of moisture from durian respiration resulting changes in color. Dirpan et al. [40] developed freshness indicator from cellulose membrane and soaked it in bromophenol blue solution and then applied on mango (*Mangifera indica* L. var. *Arummanisa*). The authors found that the color indicator changed from blue to green color (over-ripening) and also suggested that this indicator could be used to determine the freshness of mango.

Chen et al. [52] reported a system for freshness indicator based on methylcellulose and mixing of methyl red and bromothymol blue (ratio 3:2). The indicator was tested in the presence of fresh-cut green bell pepper sample during storage at $7 \pm 1^\circ\text{C}$ for 9 days. The visible change of freshness indicator was monitored for 9 days, and the color of indicator changed from yellow-green to orange when the sample was completely spoiled. The authors concluded that the deterioration of bell pepper could be detected in real time by freshness indicator. Lee et al. [74] developed a Maillard-type TTI for predicting the maturity of melon (*Cucumis melo* L.). Maillard-type TTI was based on a combination of d-xylose and glycine to create highly reactive Maillard reaction systems. The color of TTIs changed from colorless to yellow, brown, and finally dark brown. The authors concluded that

Food product	Material	Indicators	Type of intelligence	References
Fresh-cut durian	Cassava starch-chitosan	Mixing methyl red and bromothymol blue (ratio of 3:2)	Freshness indicator	Niponsak et al. [7]
Mango	Cellulose membrane	Bromophenol blue	Freshness indicator	Dirpan et al. [40]
Fresh-cut green bell pepper	Methylcellulose	Mixing methyl red and bromothymol blue (ratio of 3:2)	Freshness indicator	Chen et al. [52]
Kimchi	Cellulose fiber	Mixing chlorophenol red and bromothymol blue (ratio of 2:3)	Freshness indicator	Kim et al. [54]
Melon	Transparent pouch	D-xylose and glycine	Time-temperature indicator	Lee et al. [74]
Salad leaves	Plastic [poly(isobutyl methacrylate)]	PtTEPP and α -naphtholphthalein	Gas sensor	Borchert et al. [75]
Kimchi	Poly(ether-block-amide) and PET	Mixing methyl red and bromothymol blue (ratio of 3:7)	Gas indicator	Baek et al. [48]

PET: polyethylene terephthalate.

Table 5.
 Application of smart packaging films on fruits and vegetables.

Maillard-type TTI was successfully indicated a product's accumulated temperature as color changes during melon cultivation. The authors also suggested that Maillard reaction-based TTI is not only used for predicting melon maturity but also for monitoring the chilled food distribution and cooking time and temperature.

Some studies reported the combination of device and the packaging technique (MAP or vacuum packaging). Borchert et al. [75] developed an optochemical CO₂ sensor for measuring CO₂ in the headspace of the package under MAP storage. This sensor is composed of phosphorescent Pt-porphyrin (PtTEPP) reporter and a colorimetric pH indicator α -naphtholphthalein bounded in a plastic [poly(isobutyl methacrylate)] combined with tetraoctyl- or cetyltrimethylammonium hydroxide, as a phase transfer reagent. The authors used this sensor to measure the CO₂ levels in salad leaves stored under MAP (CO₂, 6.6%; O₂, 21.55%; humidity, 100%). It was demonstrated that the sensor could retain its color and sensitivity to CO₂ at 4°C for 21 days. Recently, an indicator for monitoring the fermentation stage of kimchi was developed by Baek et al. [48]. It was prepared by coating poly(ether-*block*-amide) that contained chemical dye (mixing of methyl red and bromothymol blue, ratio of 3:7) on polyethylene terephthalate (PET). The determination was worked on the changes in color produced by the levels of CO₂ gas in the packaged kimchi. The authors found that changes in color indicator were correlated with the CO₂ levels. Kim et al. [54] prepare an accurate indicator from mixing chlorophenol red and bromothymol blue in a ratio of 2:3 and combined it with cellulose fiber to work as labeling film for the aim to develop freshness indicator. The authors found that a total color difference of indicator was correlated well with pH and titratable acidity changes, indicating the possibility of using this indicator in commercial kimchi for indicating the degree of fermentation prior to the final purchase decision during distribution and retail sale.

5.4 Application of smart packaging on milk and dairy products

Milk and other dairy products are important sources of several essential nutrients. Liquid bovine milk generally comprises approximately 87% water, 4.9% carbohydrate, 3.9% protein, 3.5% fat, and 0.7% ash such as vitamins and minerals [76]. However, the milk compositions varied depending on the species and their immediate environment. For cheese, it can generally be classified as soft, semisoft, or hard, depending on their moisture content. Soft cheese contains high levels of moisture (50–80%), such as cottage cheese and mozzarella, while semisoft and hard cheeses, such as Gorgonzola and Parmesan, respectively, have lower moisture contents and more acidic pH levels [76]. Thus, a large variety of microorganisms quickly proliferate in soft cheese rather than hard cheese due to their moisture content and pH levels (5.0–6.5). Moreover, the presence of oxygen inside the package is also the factor to control. During milk and dairy product spoilage, a wide variety of metabolic by-products are exhibited and caused off-flavor and off-odor, in addition to visible change in color and texture. Most dairy products are distributed, handled, and stored in the cold chain (<7°C) to inhibit the microbial growth. However, temperatures should be kept as low as possible (<4°C) to prolong the storage life, and it has been reported that the shelf life of processed milk increases by decreasing temperatures during storage. To know the quality of milk and dairy products and the environment condition that packaged food product is exposed to, intelligent packaging can be used as a tool or device for detecting the spoilage process in real time and informing the real status of packaged product to the manufacturers and the end consumers by changes in color indicator without opening the packaging. The most recent studies focused on the use of intelligent packaging on milk and dairy products are presented in **Table 6**.

Pereira et al. [8] prepared a time-temperature indicator (TTI) based on a chitosan-PVA film containing anthocyanins from red cabbage, as a natural dye

Food product	Material	Indicators	Type of intelligence	References
Pasteurized milk	Chitosan-PVA	Red cabbage	Time-temperature indicator	Pereira et al. [8]
Milk	Tara gum-cellulose nanocrystal	Grape skin extract	Time-temperature indicator	Ma and Wang [57]
Pasteurized milk	Starch-PVA	Purple sweet potato	Time-temperature indicator	Liu et al. [59]
Milk	Starch-nanoclay	Methyl orange and bromocresol green	Freshness indicator	Pirsa et al. [53]
Italian cheese	RFID	—	RFID system	Papetti et al. [77]

PVA: polyvinyl alcohol.

Table 6.
 Application of smart packaging films on milk and dairy products.

indicator, for monitoring milk spoilage. During storage at a temperature above the cooling temperature, pH lowers from 6.7 (unspoiled) to 4.6 (spoiled). This value was correlated with the color values (CIELab) of the indicator. Color of the dye (red cabbage) turned from dark gray to dark pink. Thus, color changes of the indicator represented the milk spoilage. Ma and Wang [57] prepared pH sensing film from tara gum-cellulose nanocrystal incorporated with natural dye (grape skin extract) to evaluate the pH changes of the milk at ambient temperature for 48 h. During the test, the color of indicator clearly changed from bright red (acidic) to dark green (alkaline) which correlated with microbial contamination and pH decreased (from 6.48 to 2.94). The authors suggested that the developed pH sensing film could be used as a visible color indicator and changes in color of the film provide information to monitor the packaged food freshness.

Liu et al. [59] have reported the intelligent packaging made with starch-PVA added with anthocyanins extracted from purple sweet potato as an indicator for monitoring pH changes in pasteurized milk. The visual changes were monitored along 48 h storage at room temperature, and the original purple color of starch-PVA-anthocyanin film changed with time to red, which is related to milk spoilage. Recently, Pirsa et al. [53] developed a pH indicator for determination of milk spoilage. These indicators were based on starch-nanoclay incorporated with methyl orange and bromocresol green. For methyl orange indicator, the color changed from red (pH 3) to yellow (pH 4.5), while bromocresol green showed green (pH 3.8) to blue (pH 5.5). The authors suggested that the colorimetric pH indicator displayed color changes which correlated to pH changes. Thus, the use of these indicators is easily visible to the naked eye. Papetti et al. [77] developed an integrated electronic tracking system for analyzing the quality of Italian cheese. The system was able to identify the cheese products, and the consumer can know information with the help of the RFID code.

6. Conclusion

Food processing industry including meat, poultry, fish and seafood, fruits and vegetables, and dairy processing generates a large amount of by-products. However, these by-products still contain organic matters that can be recovered and exploited as high value-added compounds. They can be utilized in packaging applications as a starting material. Thus, it is a great potential of food processing by-products for

effective utilization in the field of food packaging applications that would be more cost-effective, efficient, and sustainable. Currently, low-cost sensing packaging technologies have become increasingly interesting because they can give the real status of packaged food quality through visual indication. Thus, the use of smart packaging can be applied on various types of food-based products for facilitating the consumer to know the quality of packaged food in real time and to promote choosing the products according to its freshness without damaging the packaging. These packaging technologies are a noninvasive method, cost-effective, and rapid and can also reduce food loss and waste at the same time. Thus, the use of smart packaging is an alternative way that could reduce the food loss and waste, which is one of the world's major issues.

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