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Review

Advanced technologies in biodegradable packaging using intelligent sensing to fight food waste

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ABSTRACT

The limitation of conventional packaging in demonstrating accurate and real-time food expiration dates leads to food waste and foodborne diseases. Real-time food quality monitoring via intelligent packaging could be an effective solution to reduce food waste and foodborne illnesses. This review focuses on recent technological advances incorporated into food packaging for monitoring food spoilage, with a major focus on paper-based sensors and their combination with smartphone. This review paper offers a comprehensive exploration of advanced macromolecular technologies in biodegradable packaging, a general overview of paper-based probes and their incorporation into food packaging coupled with intelligent sensing mechanisms for monitoring food freshness. Given the escalating global concerns surrounding food waste, our manuscript serves as a pivotal resource, consolidating current research findings and highlighting the transformative potential of these innovative packaging solutions. We also highlight the current intelligent paper-based food freshness sensors and their various advantages and limitations. Examples of implementation of paper-based sensors/probes for food storage and their accuracy are presented. Finally, we examined how intelligent packaging can be an alternative to reduce food waste. Several technologies discussed here have good potential to be used in food packaging for real-time food monitoring, especially when combined with smartphone diagnosis.

1. Introduction

Food packaging plays a vital role in its safety and shelf life [1]. The limitation of conventional plastic packaging in demonstrating accurate and real-time food expiration dates leads to food waste and foodborne diseases [2,3]. Most foods' quality and shelf life are strongly affected by the packaging, storage temperature and shipping conditions [4]. As such, it is difficult to accurately estimate the food expiry date, resulting in food spoiling before its printed expiry date and causing food poisoning [5,6].

According to the 2017 report by the World Health Organization (WHO), approximately 600 million cases of illnesses were attributed to the consumption of spoiled foods, resulting in 420,000 annual deaths due to food poisoning caused by bacteria, fungi, and mycotoxins in spoiled food [7,8]. On the other hand, the expiry date and the best before date are not accurate indicators of food spoilage, and customers may throw away foods that are not spoiled. Globally, approximately one-third of food produced goes to waste and never reaches a person's plate [9]. As the world's population continues to grow, the issue of food wastage is becoming increasingly apparent. This large amount of food waste is mainly due to the absence of real-time quality and freshness monitoring. Intelligent packaging is able to monitor the conditions of packaged foods, provide real-time information about the quality of foods, and offer dynamic expiry dates. The transition from fixed shelf life to dynamic shelf life reduces food waste and protects consumers from foodborne illnesses [10,11]. Over the past two centuries, the role of food packaging has transcended its basic function as a mere container. Fig. 1 illustrated evolution of food packaging. Today, it stands as a pivotal element in the branding and marketing strategies for food products. The design intricacies ranging from the package's shape and color to the

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specific details and claims inscribed on its label play a crucial role in enhancing product recognition and driving sales. Concurrently, advancements in food packaging technology have significantly bolstered both food safety and quality assurance measures in recent decades [12].

The cause of food spoilage may be due to lipid oxidation, protein oxidation, microbial activity, enzyme degradation, or changes in pH. Smart packaging, which combines intelligent and active systems, is able to overcome these causes of spoilage and wastage [13]. Intelligent packing provides real-time and dynamic information about food quality through the use of indicators and sensors. Active packaging extends food shelf-life and improves quality by controlling the level of oxygen and minimizing bacterial proliferation. Accordingly, smart packaging can reduce food waste and health problems by providing dynamic information about food edibility and delaying food spoilage [14]. Smart packaging is characterized as a packaging system capable of performing intelligent functions, including recording, detection, communication, sensing, and tracking [15]. There are different smart packaging techniques for assessing the freshness of food products. These smart packages comprise indicator-based (freshness, gases, time-temperature), sensor-based (biosensors and chemical sensors), and paper-based/arrays [16].

Although there is a considerable amount of research on intelligent packaging/sensors, there have been no comprehensive reviews focused on paper-based and smart phone diagnostics. Papers based sensors can be an economically viable option due to using a low-cost base material and their combination with smartphone facilities their communication with smartphones which can be a promising route for expanding application of intelligent packaging in food industry. The present review paper provides a general overview about intelligent films and the unique opportunity that nanomaterials provide in development of intelligent sensors. Colorimetric indicators-based nanomaterials and natural sources, their preparation, characterization, and mechanism are addressed. Paper-based sensors are deeply discussed and their combination with phone diagnostics and their pros and cons are explained. In addition, an overview of existing knowledge on application of intelligent probes on different food products in the form of food packaging is addressed. Finally, the review looks at how intelligent food packaging can help in reducing food waste, food poisoning, and contamination issues that affect the environment and health.

2. Intelligent plastic films

Microorganisms, enzymes, temperature, and other elements diminish the freshness and quality of food throughout storage, transport, and sale. Consequently, individuals place a larger emphasis on the quality and safety of food and need accurate information about food quality. A realistic and real-time method for monitoring the quality and safety of the food supply chain is necessary for standardizing the food market and safeguarding consumer rights and interests [17]. Active and intelligent packaging, on the other hand, is a revolutionary concept aimed to improve the safety and management of food products [18–20]. Intelligent packaging emphasizes the capacity to detect or quantify a property of the packed food item, the atmosphere inside the box, or the transportation environment. Active packaging often refers to a package that includes active capabilities beyond passive protection and confinement of the food product. The information gathered by intelligent packaging may be sent to consumers or utilized to activate the features of active packaging [21]. Intelligent packaging and active packaging may collaborate to create "smart" packaging, which combines the benefits of active and intelligent packaging systems [22]. Additional components in intelligent packaging interact with the packaging environment and track the condition (storage duration, temperature, etc.) of the packaged food products. Several intelligent packaging technologies may provide clients with real-time quality information for packaged food items through quality indicators [23]. Indicators are a simple but crucial tool for guaranteeing food safety, since they limit the risk of loss and the expenses associated with replacing or repairing defective products [24].

These are the indicators that function based on the temperature of both the product and its container. These indicators or processors monitor and display temperature and are often affordable and eco-friendly. They are further categorized depending on their functions as time-temperature indicators, essential temperature/time integrators, and critical temperature indicators [25].

Freshness indicator (FI) is a crucial feature of intelligent packaging and is usually accessible in two kinds: indicator card (label) and indicator film. The color change of FI generated by a food's specific volatile identify the food's freshness quickly [26]. FI has potential to offer qualitative or semi-quantitative information on food quality changes caused by physiological or microbiological growth without damaging food packaging, allowing consumers to intuitively and scientifically evaluate food quality [27].

Time and temperature indicators (TTIs) are easy-to-use and cost-effective tools for keeping track of a food package's temperature data from the moment it is packed through the packaging, storage, distribution, and retail stages. Several studies have shown that TTIs are quite

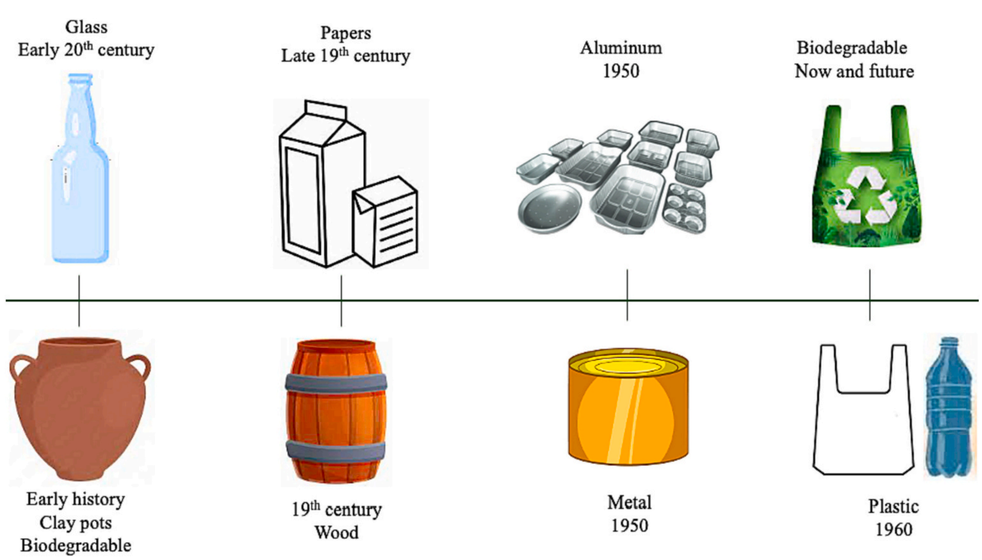

Fig. 1. A brief evolution of food packaging.

useful for monitoring changes in dietary characteristics. While numerous Time-Temperature Indicators (TTIs) have been designed and utilized to monitor shelf-life and storage properties in dairy products, other seafood, frozen foods, fish products and chilled meat, with the goal of enhancing intelligent packaging methods, there remains a lack of research dedicated to liquid food intelligent packaging [28].

As a result of a variety of factors, maintaining the quality of food materials within a packaging system is a challenge. These include the respiration rate of fresh fruits and vegetables, the alteration of gas concentrations, gas leaks into or out of packaging materials, and the gas produced by microbes. For the resolution of such issues, gas indicators have been developed. By changing color in response to a specified chemical or enzymatic reaction, these indicators give information on the oxygen or carbon dioxide gas concentration inside the packing material. These indicators may give information on the presence or absence of a gas by direct interaction with the foods. Typical forms of gas indicators include labels, tablets, printed layers, and laminated polymer films [29].

pH indicators may exhibit color changes based on chemical reactions or microbial growth. pH alterations are directly affected by the spoilage of food products. Foods with high water activity, such as meat and seafood-based products, are more susceptible to microbial biodegradation [30]. The pH within the package varies due to the accumulation of volatile nitrogen compounds (containing ammonia and amines, such as total volatile nitrogen compounds (TVB-N)) caused by the pH variations as a result of bacterial spoilage [31,32], which is visible in the color change of the pH-responsive color indicators. Moreover, the difference in pH can occur in packaged foods when they tend to decompose, which results in an alkaline pH environment, or when fermentation happens because of microbial activity. The growing demand for consuming natural/organic products and using natural pigments to produce indicators is a healthier and safer replacement for synthetic colorants. Indicators of food freshness based on natural dyes, such as curcumin, grape peel, and beetroot extracts, have also been investigated for detecting the deterioration of cod meat. These natural pigments are mostly containing anthocyanins molecules which have a pH-dependent color. The color changes are primarily due to structural changes in the anthocyanin molecule. Anthocyanins are composed of a chromophore, which is responsible for absorbing light and giving color. The chromophore undergoes changes in its conjugation and structure in response to pH changes, resulting in different colors. It has been shown that natural bio-based extracts may be employed efficiently as dyes in pH-based food quality indicators in intelligent packaging technologies since they can readily operate based on overall color difference values [33]. Previous investigations have highlighted the potential of natural dyes, pigments, and food colorings as intelligent packaging system indicators [24,34] and there is still a need to compile information on plant- and food waste-based pigments used in biodegradable material and their impact on food overall quality.

3. Natural-based pH sensitive indicators

Due to the fact that packaging should not negatively affect the environment, biodegradable and conveniently recyclable materials are becoming more popular [35,36]. Special standards are imposed on food packaging to ensure that it does not compromise the safety and health of customers and does not leach dangerous compounds from the plastics manufacturing into the packed food. Several substances may migrate from food packaging [37,38]. Such additives include plasticizers, antioxidants, heat and light stabilizers, slide agents, antistatic agents, lubricants, and nanoparticles. Some chemical substances (such as A diglycidyl ether, bisphenol A, primary aromatic amines, bisphenol, and phthalate) detected in food because of packing migration are harmful to human health [39]. The migration of dangerous compounds into food is strictly regulated and therefore cannot exceed permissible levels. Consequently, there is a growing interest in natural chemicals that may be employed as process additives in polymers [34]. There are numerous studies on the creation of pH indicator with anthocyanins derived from various natural sources into polymer coatings. A pH-sensitive film was developed and characterized using sago starch and anthocyanins from torch ginger by Mei et al. [16]. Elongation at break, moisture content and water solubility were improved with addition of torch ginger compared to control. A pH sensitivity analysis showed that the color of films containing TGE extract changed from pink to slightly green as the pH increased from pH 4 to 9. Therefore, the authors stated that the developed pH-sensitive film with torch ginger extract can be used to detect food freshness or spoilage to ensure their safety and quality.

The addition of grape anthocyanin (1.0 g/100 g, as natural pH indicator) to chitosan films was investigated. In this research, the incorporation of the indicator had no impact on the mechanical properties of the films, including tensile strength and stiffness. However, it resulted in a reduction of 47 % in elongation and a 48 % decrease in water vapor permeability (WVP) compared to films without the indicator. In another study, the casting method was used to develop an intelligent pH-sensing indicator made of gelatin film and anthocyanin extracted from dragon fruit skin. As a result of adding anthocyanin to the films, the moisture content, thickness, and water solubility increased, but the WVP and light transmittance decreased. Films exposed to different pH buffer solutions displayed significant differences in color. Gelatin film incorporated with anthocyanin was found to be a useful visual indicator of pH variations during food storage [40].

Natural dyes and polymer-carriers recovered from waste peels, seeds, tubers, and even pulps are the optimal option to produce natural pH-sensitive indicator films, one of the most crucial components of intelligent packaging [41].

Accordingly, natural pigments are increasingly replacing chemically manufactured pigments with pH-sensitive properties due to concerns about the side effects of the synthetic pigments, including toxicity and environmental impact [42]. Typically, natural dyes like anthocyanins, carotenoids, and chlorophylls serve as food indicators. These not only detect food quality changes but also offer additional benefits, such as antibacterial and antioxidant activity [43]. To meet pH-responsive food packaging requirements, enhancements are needed in the color rendering range, color sensitivity, and dye penetration of natural pigments.

Natural dyes are often utilized for intelligent packaging, particularly food freshness tracking, because to their abundant supply, ease of access, safety, broad signaling range, excellent pH-responsiveness, and other characteristics. Natural pigments' poor stability, varied solubility, and distinctive pH sensitivity are thought to be their key limiting factors. In general, natural pigments are categorized as anthocyanins, carotenoids, betalains, chlorophyll, and curcumin [44].

Anthocyanins are found in many plants, including berries and purple sweet potatoes. High color sensitivity and outstanding antibacterial and antioxidant properties made them ideal for use in smart food packaging [45]. Anthocyanin typically exhibits red under acidic conditions, pink under neutral situations, and blue under basic ones [46]. The most prevalent usage of anthocyanins' unique characteristic has been in the creation of smart packaging sheets that employ pH indicators to track the freshness of foods.

More recent research found that pH sensitive smart films made of chitosan, methylcellulose, and chitin nanoparticles combined with anthocyanin extract had improved mechanical characteristics and might be used as quality sensors in food containers [47].

Anthocyanins may affect the performance of the indicator film. For instance, the indicator film's mechanical and thermal resilience were improved by anthocyanins due to hydrogen bonds and intermolecular cross-linking [48]. Instead, because of their hydrophilicity, anthocyanins decreased the colorimetric film's water vapor barrier characteristics [49], and interfered by their own color due to excessive addition. In a research investigation, a pH indicator was synthesized using a combination of corn starch, polyvinyl alcohol (PVA), and natural anthocyanin sources, namely purple sweet potato extracts (PSPE) and red cabbage extracts (RCE). As the concentration of the extracts increased, notable enhancements in film thickness, mechanical strength, and thermal properties were observed, along with a significant reduction in light transmittance. Furthermore, at lower extract concentrations, films made with purple sweet potato extracts (PS-PSPE) displayed more intense colors, improved mechanical characteristics, and lower light transmittance than films containing red cabbage extracts (PS-RCE) [50].

As explained in the previous section, anthocyanin-rich films can also be produced by casting [51] extrusion [52] or electro-spinning. Depending on the kind of polymer, biopolymers are typically dissolved in suitable solvents with or without heating prior to casting. After extracts containing anthocyanins have cooled to room temperature, they are combined with biopolymers to create film-forming solutions. Frequently, plasticizers such glycerol [50,53], sorbitol [54], and polyethylene glycol [55] are added to film-forming mixtures. Plasticizers may enhance the films' mechanical and water vapor/gas barrier characteristics [56]. Fig. 2 illustrates the preparation process for natural pH-responsive colorimetric indicator films.

4. Nanomaterials enabled pH-sensitive indicators

Food pH levels serve as freshness indicators of chemical changes and the impact of irreversible chemical and biological interactions. The ability to detect the pH change of the product or the environment non-invasively by color change of the pH-sensitive freshness indicator during preservation provides possible real-time monitoring for food quality, safety, and shelf-life [57]. In the last decade, nanotechnology has been utilized in food packaging systems as pH indicators to control food freshness since the performance of pH-sensitive indicators can be enhanced by incorporating various types of nanomaterials/nanostructures [58,59]. The main advantages of the pH-responsive indicators combined with nanoparticles/nanomaterials in food research are antibacterial activity, stability, sensitivity, oxidation resistance, UV blocking ability, improvement of tensile strength (TS), water resistance, mechanical characteristics, and so forth. The following advantages of pH-sensitive freshness indicators are discussed in the following sections and explained in more detail using examples.

In the literature, cellulose nanocrystals have been incorporated in chitosan composite films to improve their TS, water barrier, and UV barrier properties [47,60]. Also, zinc oxide nanoparticles (ZnO) have been employed in konjac glucomannan/chitosan-based [61] and gelatin/agar active packaging systems [62] because of their effective antibacterial capabilities. Additionally, since silver nanoparticles (AgNPs) can increase the stability of the films, they have been utilized in anthocyanin-based films [63–65]. Moreover, using natural pigments and titanium dioxide nanoparticles (TiO_2 NPs) has been reported to improve the films' mechanical properties and moisture resistance. Most of nanomaterial-based pH indicators consist of following various nanomaterials: nano-biopolymers, carbon-based nanostructures, functional metallic nanoparticles, nano-metal oxides incorporated polymers, and biopolymers to improve for food monitoring quality [66]. Chitosan, cellulose, gelatin, starch, and polyvinyl alcohol (PVA) are typical examples of biopolymers/polymers that are often utilized [67].

4.1. Biopolymer-based nanostructures

Recently, Ezati et al. [68] established intelligent pH-responsive color indicator films based on shikonin-containing cellulose nanofibers (CNF) to monitor the freshness of fish. CNF was used in their research since it possesses more mechanical strength and flexibility. The colorimetric pH-sensitive indicator film changes depending on the pH from 2 to 12. When the fish sample was fresh, the color of the pH-sensitive indicator film was reddish-pink, and the pH was 5.7. After 36 h of storage, the color indicator film displayed significant color variations, and the color changed to bluish-violet, indicating the spoilage of fish with pH 6.9.

It is well known that chitosan nanofibers are a sustainable natural

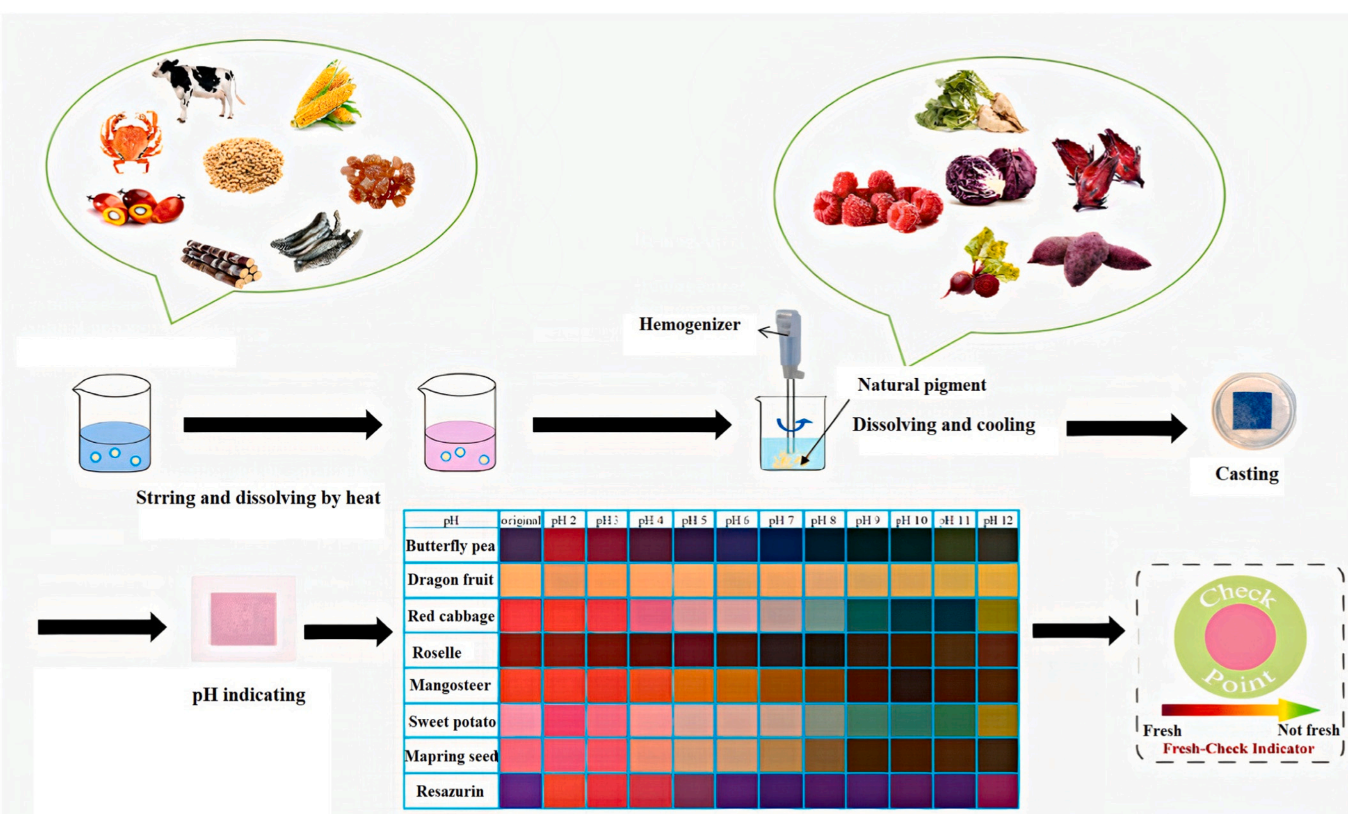

Fig. 2. Natural pH-responsive colorimetric indicator films preparation process.

polymer that has gained popularity for their antibacterial qualities [38,69]. Incorporating the chitosan nanofibers into the indicators enhances the mechanical and water barrier capabilities while decreasing the indicators' ability to transmit UV light. Moreover, in a recent study, Alizadeh-Sani and colleagues made pH-responsive color indicator films using methylcellulose, chitosan nanofibers, and anthocyanin to detect the quality of lamb meat for intelligent packaging technologies in real-time. Since the color of the indicator film varied from reddish/crimson to yellow when the pH ranged from 1 to 14, the pH significantly influenced the color. After storage at room temperature for 72 h, the pH of lamb meat considerably changed from 5.8 to 7.5. The color indicator film, which was used to identify spoilage of lamb visually and color change in response to an increase in pH values, turned from crimson to light peach during preservation [70].

4.2. Carbon-based nanostructures

Carbon dots (C-dots) can act as sensors in intelligent food packaging due to the fact that they are fluorescent and possess unique properties. In acidic conditions (low pH), carbon dots exhibit protonation of functional groups, causing changes in their electronic structure. This results in a shift in absorption and emission wavelengths, leading to a color change (e.g., from blue to green or red). Conversely, at high pH, functional groups deprotonate, leading to an altered electronic structure as well as color changes.

One of the most promising carbon-based nanomaterials is graphene oxide (GO) nanoparticle/nanosheets. GO exhibits outstanding bio-safety performances, especially at low concentrations [71,72], in addition to their unique mechanical and thermal properties [72,73]. Therefore, it appears that the use of GO as reinforcement or filling agent helps to improve the aforementioned properties of pH-sensitive colorimetric indicators. In a recent study, the color response of a colorimetric fresh indicator film to real-time pH variations in lamb was investigated to determine whether or not food spoilage was indicated. It was found that the addition of GO to the prepared freshness indicator film not only enhanced the mechanical properties and thermal stability, but also significantly improved the moisture content, water solubility, surface hydrophilicity and biosafety. The color changes of the freshness indicator were observed from red/pink to yellow with the increase of pH from 2 to 13. After 96 h of storage at 4 °C, the lamb samples began to decay, which was due to the formation of volatile nitrogen compounds and a rise in the pH of the meat sample. The pH of the lamb was 5.8 (beige) when the meat sample was fresh, and the sample's pH increased to 7.8 (light indigo) at the end, indicating their deterioration/spoilage [73].

In another work, Koshy and colleagues [74] have developed an intelligent starch-based biopolymer film containing carbon dots (CDs) and anthocyanins that can detect food spoilage. Because the starch film containing anthocyanin has high hydrophilicity and poor mechanical characteristics, anthocyanin extracted from *Clitoria ternatea* flower (CTE) is combined with CDs. The starch/CD/CTE film (SED) was prepared by solution casting technique and characterized as novel, high sensitive, pH indicator film having the potential to monitor the freshness of packed pork. Visual color alterations of the developed film from red to yellow were observed, with pH changes in the range of 1–12. In this study, the freshness of pork meat at room temperature was monitored using colorimetric pH-sensitive films. When the pork sample was fresh and kept at a temperature of 25 °C, the pH of the sample was slightly acidic and was 5.8 (pink/purple). However, the freshness/quality of the pork tended to decrease after 30 h of storage, causing the pH to become basic and increase to 7. When the pH reached 8 (green color) after 48 h of storage, the meat was completely decomposed.

4.3. Functional metallic nanoparticles

Sulfur nanoparticles (SNPs) exhibit a variety of biological functions, including anti-inflammatory, antibacterial, and antitumor properties [75]. According to a recent study, SNPs have been employed in developing antimicrobial films demonstrating considerable antimicrobial activities against bacteria that lead to foodborne disease [75]. A smart pH-responsive packaging film indicator was made by utilizing SNPs, curcumin, and pectin to detect the freshness of shrimp samples. SNPs improved the indicator film's UV barrier characteristics, thermal stability, antibacterial activity, antioxidant activity, and sensitivity to ammonia vapor. As the pH of the composite film increased from 2 to 12, its perceived appearance changed from pale yellow to dark red. The pH-sensitive indicator film showed a noticeable color shift from fresh to the spoiling stage after 36 h of storage at 25 °C. The pH of the shrimp increased from 6.3 to 7.1 during storage, changing the original appearance of the pH-responsive film from yellow to orange [76].

AgNPs are among the most extensively employed metal nanoparticles. They have antimicrobial properties [63], excellent thermal stability [77], and UV-blocking properties [78]. AgNPs have been shown to significantly increase the mechanical strength and stability of biopolymer films in earlier studies [63,64,79]. Recently, You and coworkers [80] established a pH-sensitive freshness indicator based on κ-carrageenan polymer filled with AgNPs and anthocyanin to monitor marine fish's freshness in real-time. AgNPs were also included in the composite film, much like SNPs, to enhance the developed film's antibacterial characteristics. The developed indicator film has a pH range of 3-10 and is colored red in acidic medium and dark green in alkaline medium. It was examined to determine the freshness of the seabass sample after being preserved at 4 °C. The color of the film was purple (around pH 5–6) at the beginning of the experiment. After four days, the color changed to blackish-purple (pH 8), and on the last day, it was dark green (pH 10), meaning that the seabass sample was spoiled.

Copper nanoparticles (CuNPs), which are noble metals, are less expensive than other metals. CuNPs, unlike other nanoparticles, are used to detect volatile sulfur compounds due to their strong interaction with sulfur ions. Since fish degradation releases hydrogen sulfide, a volatile sulfur gas, food freshness indicators with CuNPs additions are used to determine the freshness or spoilage of fish. Recently, Teymouri and Shekarchizadeh [81] synthesized and characterized CuNPs for developing an intelligent fish packaging system that utilizes a colorimetric pH-responsive indicator of volatile sulfur compounds (Fig. 3a–d). CuNPs-based colorimetric indicators changed color during fish storage to assess the degradation of fish at room temperature and 4 °C. Fresh fish samples stored at 4 °C had a pH of 6.23, and a CuNPs-based colorimetric indicator showed the first white color. Fish samples kept in the refrigerator for six days with a pH of 6.75 were spoiled, and the indicator turned light-brown color. Similarly, fresh fish samples preserved at 25 °C had a pH of 6.03, and the colorimetric indicator based on CuNPs exhibited white color at the beginning. Fish samples were maintained at room temperature for 30 h with a pH of 6.68 degraded, and the indicator displayed a light-brown color.

4.4. Metal oxide nanoparticles

ZnO are used as a nanofiller in numerous studies to enhance/improve the stability [82–86], the mechanical and moisture barrier qualities of certain biopolymer-based food grade films/coatings [87,88], absorb UV radiation, and, most significantly, for their excellent antibacterial effect [89]. Liu and colleagues [90] recently developed a pH-sensitive colorimetric film made of ZnO that contained chitosan, PVA, and anthocyanins to monitor the progress of shrimp decomposition in real-time. ZnO was employed in the films as an antibacterial agent, and the prepared films had noticeable color changes from pH 2 to pH 12 at 4 °C. The pH value of the shrimp samples at the beginning of the experiment was 7.41, and the indicator film showed a purple hue. After four or more days of storage, the pH of the spoiled shrimp samples increased to about 8, shifted to the alkaline range, and the color of the indicator turned to light green.

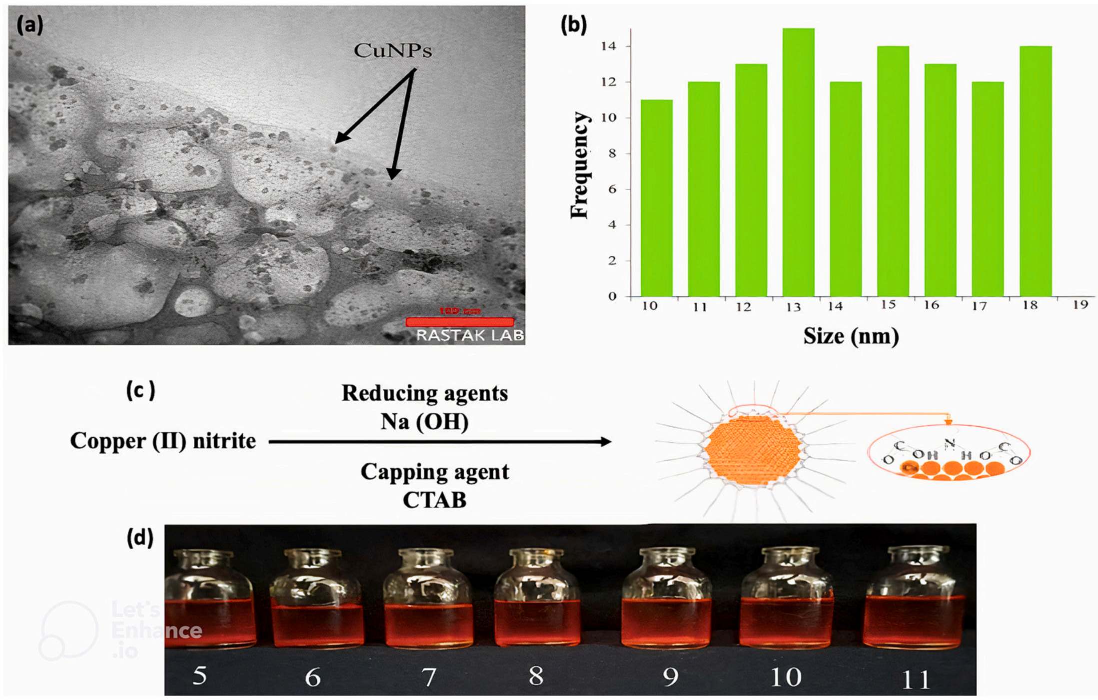

Fig. 3. (a) TEM image, (b) size distribution diagram of CuNPs, (c) schematic of the formation of nanoparticles by the chemical reduction method and (d) color of CuNPs at different pHs. Reprinted with permission from Elsevier [81].

TiO_2 NPs are broadly used as reinforcement agents in food packaging applications [91]. They are mainly used as UV light absorbers [92] and also have antibacterial and antimicrobial functions [93], which helps to prevent the growth of dangerous microorganisms. Moreover, incorporating TiO_2 NPs advanced the water vapor and oxygen permeability of food packaging films [94]. According to a study, Mary and co-workers successfully designed a starch-based food packaging film integrating anthocyanin and TiO_2 NPs as the pH-sensitive indicators to determine the freshness of prawn samples. The film containing TiO_2 NPs had superior physical characteristics to neat film, including color, transparency, thickness, moisture content, water vapor permeability, and thermal stability. In the pH range of 1 to 12, the color of the fabricated freshness indicator changed from reddish pink in acidic media to green in alkaline media. The fresh prawn samples had an initial pH of 6.58 and depict pale pink color. The color of the prawn samples changed to vivid green and indicates spoilage, and the sample's pH raised during

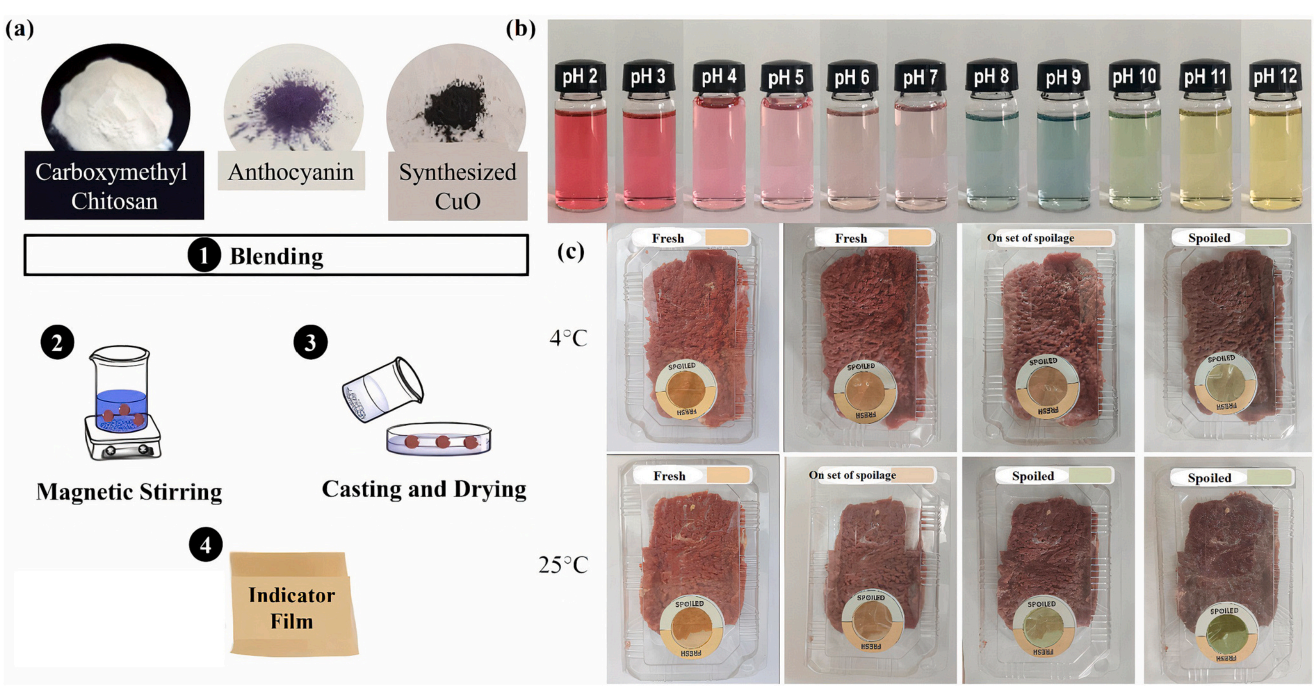

Fig. 4. (a) Preparation of pH-sensitive indicator film containing carboxymethyl chitosan, anthocyanin dye and CuO NPs using solvent casting method; (b) Color change profile and pH range of the prepared freshness film by using different pH buffer solutions; (c) Freshness test of meat samples with pH-sensitive bio-based film at refrigerator (4 °C), and room temperature (25 °C). Reprinted with permission from Springer Nature [96].

deterioration due to the hydrolysis of proteins [95].

The incorporation of copper oxide nanoparticles (CuO) into pH-sensitive colorimetric indicators improves their mechanical properties, UV absorption, barrier, and thermal stability. A bio-based nanocomposite film based on carboxymethyl chitosan (CMCS) containing CuO and anthocyanin (Fig. 4a), was designed by Fathi et al. [96] to monitor lamb meat freshness. The pH-sensitive indicator film, ranging from pH 2 to 12, depicted in Fig. 4b, clearly shows color changes from pink to green in acidic (pH 3 and 5) and alkaline (pH 9 and 11) media, respectively. They evaluated the freshness/spoilage of lamb meat at 4 °C and 25 °C, and it accurately determined the freshness of lamb meat during preservation which is depicted in Fig. 4c. As can be seen, due to the formation of volatile compounds and increased pH, the meat samples degraded after 72 h at 4 °C and 12 h at 25 °C storage. When the meat sample was fresh, the pH-sensitive film had a light brown color. While it started to degrade, the color changed to a pastel gray color, eventually turning green, indicating the complete degradation.

5. Paper-based food freshness sensors

Several determination methods are used for food freshness monitoring. Among them, paper-based sensors are greatly interesting due to their high specific strength and stiffness, lightweight, nontoxicity, abundance, recyclability, biodegradability, and low cost [97]. Paper-based sensors offer consumers and scientists a fast and easy-to-use detection technique for food freshness. Therefore, through integrating effective or required chemicals and polymers into the cellulose network, cost-effective paper-based technologies for filtering, printing, packaging, and sensing have developed quickly in recent years. In Table 1, some of the recent paper-based food spoilage sensors along with their properties, sensing elements, detection approaches, and performances are summarized.

In order to broaden the application limits, and increase the efficiency of detection methods, paper-based sensors have been started to be utilized. Porphyrins are frequently utilized in pesticide detection, medicine, and energy sectors because of their excellent biocompatibility and distinctive photophysical characteristics [98–100]. A relative study in which a fluorescence-visualized paper-based sensor for organophosphorus pesticides (OPPs) detection using filter paper as the sensor material was developed by Wang et al. [101]. In their paper-based sensor, dimethoate from apple and red cabbage samples was detected and analyzed with a fluorescence visualization established by double quantum dots (QDs) combined with a high-activity nanoporphyrin, Zn-nanoporphyrin (nano-ZnTPyP). CdTe and ZnCdSe QDs were selected to be used as a combination in order to produce additional color regions for more accurate measurement of various dimethoate concentrations, considerably increasing probe sensitivity. Paper chips were loaded with nano-ZnTPyP at the initial state. With the addition of QDs to paper chips, the sensing mechanism of the "Turn-off-on" fluorescence probe starts with the "off" state, quenching of the QDs and change in the color of the paper chip from red to purple or dark purple. With the addition of dimethoate to paper chips, onto the quenched QDs, color of the paper chips changed again and turned back to red. Fig. 5 represents the loading and "Turn-off-on" states of the developed sensor. Data analysis was performed by chemometrics using *Matlab* software.

A smartphone-assisted hydrogel-based "*optoelectronic nose*" was designed in 2022 by Das et al. [102]. This device has taken the name CN-2 and was synthesized via multiple inter/intramolecular C—N fusion

Table 1
Some examples of paper-based sensors for the detection of food samples.

Food sample	Paper type	Target/marker	Reagent	Detection	Properties	LOD/LOQ/DR	Reference
Salmon, chicken, beef, and pork	The Whatman 42 filter paper (ZIF-8 coating on AuNP impregnated)	Biogenic amines (Putrescine, cadaverine)	4-Mercapto-benzaldehyde	SERS	Accumulation/ enrichment of gaseous molecules, additional enhanced Raman signal of molecules	76.99 ppb for putrescine, 115.88 ppb for cadaverine	[107]
Chicken, beef, pork and fish	Commercial paper sheets "Colour Catcher®"	Volatile byproducts other than biogenic amines	Five sensitive acid-base indicators (m-cresol purple, o-cresol red, bromothymol blue, thymol blue, chlorophenol red)	Colorimetric, PCA	Able to follow the entire spoilage process	–	[108]
Pork meat	Filter paper	Biogenic amines (Putrescine, cadaverine)	Porous poly(lactic) acid film with GO coating onto paper	Colorimetric, LDI-MS	Dual and detection platform, Disposable sensor, Qualitative and quantitative detection	0.07 pM for putrescine, 0.02 pM for cadaverine	[109]
No specific food sample	Office wastepaper	Biogenic amines (Putrescine, cadaverine, histamine, tyramine)	Methyl red and chitosan double layer	Colorimetric	Roughness of the surface: 10–12 μm, Thermally stable up to 300 °C	–	[110]
Yellow croaker	Filter paper	Hydrogen sulfide (H_2S)	Naphthalimide-fluorophore bearing a H_2S-cleavable moiety modified with a long hydrophilic PEG chain	Fluorescence, Colorimetric	Naked-eye detection, A specific nucleophilic substitution reaction	9.95 nM	[111]
Beef and milk	Whatman Grade 4 filter paper	Bacterial Contamination - Quorum sensing molecules	Modified filter paper	Colorimetric	Early detection, 3 h of assay time, $0.15 of cost	1.0 nM for beef and 0.1 nM for milk	[112]
Salmon fish	Whatman glass microfibre filter paper	Amine vapor	Dihydroquinoxaline derivative (H + DQ2)	Photoluminescence	A solid-state fluorescence sensor Providing a baseline information	1 mg/L	[113]
Shrimp, chicken, and weever	Qualitative filter paper	Biogenic amines	(ASQ) coating on paper	Fluorescence, pH	Bimodal real-time monitoring, Protonation/ deprotonation on the H^+ASQ label	17.67 mM	[114]

ZIF-8: Zeolite imidazole framework-8; SERS: Surface enhanced Raman spectroscopy; PCA: Principal component analysis; LDI-MS: Laser desorption-ionization mass spectrometry; ASQ: 4-(dimethylamino)styryl quinoxalin-2(1*H*)-one; LOD: Limit of detection; LOQ: Limit of quantification; DR: Dynamic range.

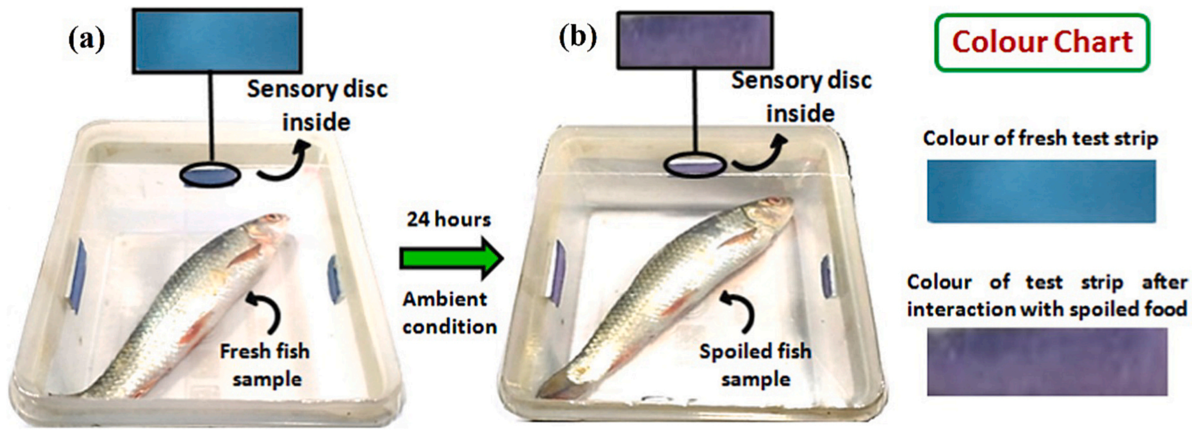

Fig. 5. Testing the freshness of fish using a CN-2 coated test strip (a) before and (b) after the fish has spoiled after being kept for almost 24 h at room temperature. Reprinted with permission from Elsevier [102].

reactions. Aliphatic amines were selected as the biomarker, and the synthesized monoprotonated CN-2 could overcome chromo-fluorogenic detection of primary, secondary, and tertiary aliphatic amines and ammonia selectively with low detection limits. For NH_3, hydrazine (primary amine), diethanolamine (secondary amine), and triethylamine (tertiary amine), the detection values were 27.2 ppb, 0.7 ppm, 5.4 ppm, and 1.7 ppm for UV–Vis and 42.5 ppb, 1.61 ppm, 5.5 ppm, and 6.14 ppm for fluorescence spectral data, respectively. When exposed to ammonia and aliphatic amine, a naked eye-observable color change occurred with the selective deprotonation of monoprotonated CN-2. Color of the test

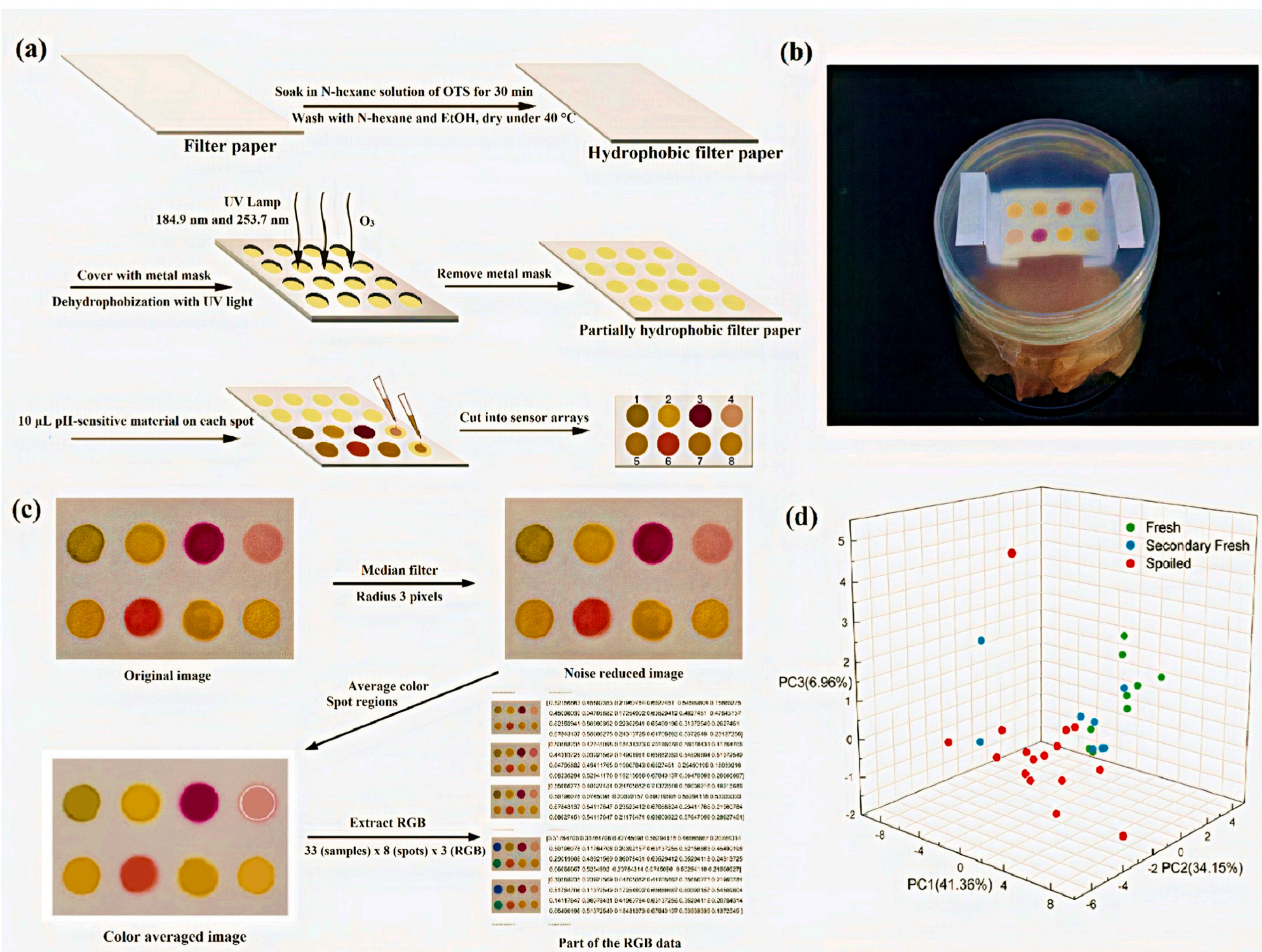

Fig. 6. (a) Fabrication process of the sensor array; (b) Packed fish sample with a colorimetric label; (c) Image processing progress of the sensor array; (d) PCA score plot using the first three principal components. Reprinted with permission from John Wiley and Sons [104].

strip changed upon the interaction of test strip with amine groups from blue to distinguishable purple (Fig. 5). The retrieved RGB (Red, Green, Blue) values from the smartphone application strongly correlate with the observed chromogenic signals. Also, a specific protonated antenna center was present in the CN-2 with an antioxidant activity which makes it possible to detect aliphatic biogenic amines released from spoiled food, such as spermidine, triethylamine, and putrescine [103]. A free Android app called Color Grab 3.9.2 was used to track the changes in RGB values in order to get quantitative data regarding the CN-2 "naked eye" recognition phenomena in the presence of ammonia vapor.

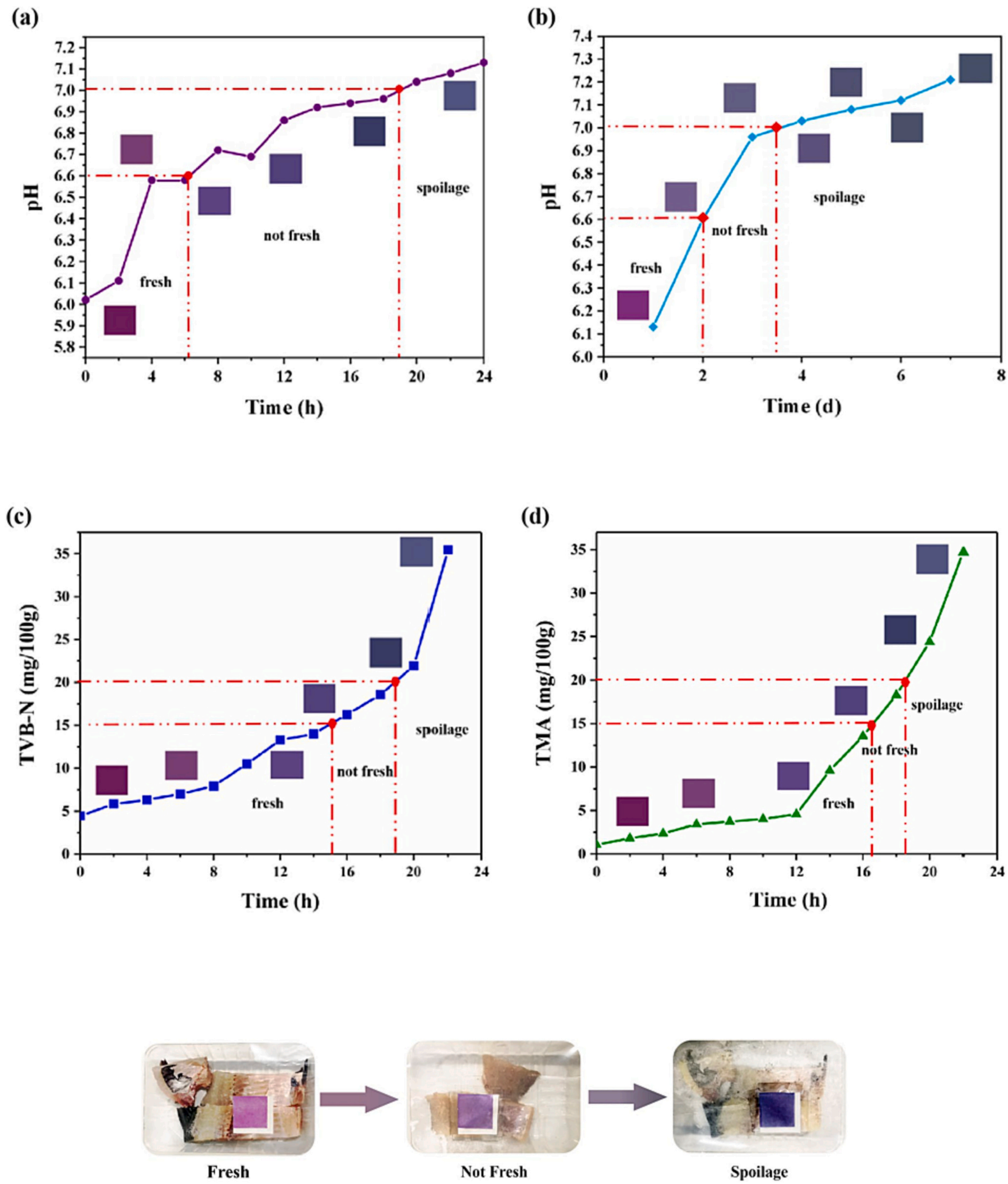

Fig. 7. (a) pH variations in grass carp stored at 25 °C within 24 h. (b) pH variability in grass carp stored at 4 °C over 7 days. (c) Changes in TVB-N values of grass carp kept at 25 °C within 24 h. (d) Shifts in TMA values of grass carp kept at 25 °C within 24 h. (e) Alterations in paper sensors' color while monitoring grass carp freshness within 24 h at 25 °C. Reprinted with permission from Springer [106]

In another study by Wang et al. [104], a colorimetric paper-based sensor array was fabricated using pH indicators. The sensor was prepared by dropping eight different pH indicators (bromophenol blue (BPB), bromocresol purple (BCP), neutral red (NR), purple sweet potato anthocyanidin (PSPA), bromocresol green (BCG), methyl red (MR), bromothymol blue (BTB) and curcumin (CUR)) onto UV lithography filter paper. Preparation step of the colorimetric sensor array can be seen in Fig. 6a. The sensor array was placed 1 cm below the jar's cap full of Grass carp samples. Placement of the sensor into the jar is shown in Fig. 6b. Through pattern recognition, the sensor array can separate the Grass carp into three stages of degradation or spoilage and provide stable color change over the storage time. The People's Republic of China's national standard GB2733-2015 states that the spoil TVB-N threshold for freshwater fish is 20 mg/100 g. As a result, grass carp samples were categorized into three freshness stages: primary fresh, defined as TVB-N content under 10 mg/100 g; secondary fresh, defined as TVB-N content between 10 mg/100 g and 20 mg/100 g; and spoiled, defined as TVB-N content exceeding 20 mg/100 g. After the color change on the paper sensor, image processing was performed, and RGB data belonging to the color were obtained, as seen in Fig. 6c. Then, principal component analysis (PCA) was performed in order to pattern recognition and to form groups depending on the Grass carp's spoilage stage. Fig. 6d represents the PCA results of the spoilage stages.

A colorimetric paper-based sensor was fabricated by Alamdari et al. [105] using a simple and scalable coating method. The sensor detector was prepared by coating a mixture of soybean hulls (SBHs), bentonite, and bromocresol purple onto the filter paper. This matrix resulted in an improved gas adsorption capacity and a useful dye-immobilization. By adjusting the detector's pH, it was possible to assess the freshness and spoilage of fish meat with different weights. It was demonstrated that the color of the detectors changes at different times during storage by fabricating them with a wide range of starting pH values (0.25–5.20), and exposing them to catfish fillets that had been kept at 4 °C. The correlation between the activation time and daily quantifiable microbiological growth (TVC) enables the development of a set of detectors that operate within the ideal pH range for the detection of spoilage and freshness levels. The activated detectors' capacity to give information on the history of the food product at various points in the food supply chain can be possible by the stability of color change and irreversibility of the activated detectors.

A non-destructive and paper-based sensor for trimethylamine (TMA) and TVB-N determination upon spoilage of Grass carp samples was recently developed by Sun et al. [106]. In this study, a mixture of carboxymethyl cellulose, glycerin, and natural purple cabbage pigment was used to produce a printable ink, and this ink was printed on the filter paper by the screen-printing method. The resulting printout was then applied to modified atmosphere packaging (MAP). Fig. 7a and b represent the change in pH values with the increasing amount of TVB-N and TMA, as well as the amount of TVB-N with increasing time at room temperature, respectively. On the other hand, Fig. 7c and d display the increasing amount of TVB-N and TMA contents with color changes over time. At 25 °C, the sensor's color changed from purplish red to blueish violet in 18 h, and at 4 °C, it turned to blueish violet in 3 days. For the color stability of the paper sensor, the total color difference (ΔE) values of the paper sensors were calculated, and the ΔE values changed from 17.19 ± 0.912 to 22.75 ± 0.215 for 25 °C and from 23.34 ± 0.211 to 26.16 ± 0.182 for 4 °C. The color change of the paper-based sensor is given in performed as seen in Fig. 7e. High ΔE values during the storage of grass carp show that the paper sensors have the necessary visual discriminating capabilities. Moreover, using the screen-printing method resulted in a low-cost, massive, and efficient fabrication of paper sensors.

5.1. Lab-on-paper optical sensors

Optical sensors are analytical tools that have drawn the interest of researchers, particularly in the analytical sector. They are regarded as chemical or bio-based sensors since they combine optical transducers with chemical recognition phases (sensing receptors) [115]. Typically, optical sensors are tools that can identify and measure a range of lighting characteristics, including frequency, wavelength, polarization, and intensity [116]. This type of sensors is frequently employed in the food industry to evaluate food quality parameters including pH, temperature, and spoiling [115].

Research has explored the development of optical sensors for detecting food freshness across various samples, including fish, shrimp, fruit, and milk. Weston et al. developed the creation of a ZnO solution with the incorporation of conjugated polymers and polydiacetylene (PDA) to assess cream milk freshness [117]. A disc sensor was created by immobilizing the PDA/ZnO nanocomposite in agarose after it was made using the thin film hydration process. PDAs are appealing starting points for colorimetric sensors thanks to their distinct colorimetric characteristics. In this work, PDA/ZnO nanocomposite's structure has been adjusted to obtain the ideal pH value that was recorded from the spoiled milk. As the pH increased, the PDA/ZnO/agarose film's color changed from blue to red. The quantity of lactic acid created by the milk allowed this sensor to distinguish between fresh, spoilt, and ruined milk. Because it changes color when forces inside the PDA/ZnO nanocomposite are disrupted by acid, this material is an excellent indicator. Moreover, PDA/ZnO demonstrated more excellent stability in a variety of solvents than pure PDA. As a result, it can be cast into many different kinds of materials, making it perfect for use in food packaging systems.

Hasanah et al. [118], have successfully fabricated an optical pH sensor to determine the freshness of fish by employing chromoionophore as the reagent. Chromoionophore was bound in a pectin hydrogel membrane, and the protonation and deprotonation of the functional groups of the chromoionophore allowed for the detection of pH changes in fish samples. Additionally, the good quality of the pectin hydrogel membrane makes it an ideal candidate to be used as a sensor matrix. Pectin, being a hydrophilic polymer, exhibits superior permeability compared to synthetic hydrophobic polymers. Its hydrophilic nature results in the efficient adsorption of the chromoionophore into the polymer, allowing for a rapid response to pH changes. Protonation under acidic conditions and deprotonation under neutral conditions were assessed using a UV–Vis spectrophotometer at 615 nm and 535 nm respectively. An isosbestic point, which shows the optimal absorbance of the optical pH sensor in both situations, was created by changes in the protonation and deprotonation processes. The sensor was able to respond quickly and generate satisfactory linearity and repeatability based on the analytical performance tests.

Moreover, in a recent research a colorimetric or optical sensor was developed to measure the TVB-N emitted during the rotting of tilapia fish using polyaniline (PANI), a conductive polymer that is frequently used for electrochemical sensors. In this work, doped-PANI was created using hydrochloric acid. The indication based on doped-PANI shifted from green to blue when there was spoiled tilapia present. The created sensor could be used at least three more times and had benefits, including being simple to use and affordable for mass manufacture [119].

Additionally, a lab-on-paper colorimetric sensor for bacterial spoilage in packaged meat was developed [120]. Two pH-indicator dyes, bromocresol purple (BCP) and bromothymol blue (BTB), were utilized to produce the on-package sensor, which was then bonded to the inside of the packing film before the storage process.

Circular filter-paper pieces, measuring 15 and 35 mm diameter, were used as sensory material. These sensors were applied to the inner side of chicken fillet packages, allowing direct contact with the package atmosphere. The samples were then stored at 25 °C and 4 °C, separately, and they were periodically tested for microbiological population, pH, and TVB-N content. In the presence of humidity, the dye in the sensor can interact with the emerging gaseous by products of bacterial decomposition to produce the required color shift, which can then be recognized by an Android Smartphone. A smartphone camera was used to take sensor photos, which were then edited with an Android program that comes pre-installed to read color changes and offers qualitative and quantitative data regarding the freshness of the meat. For BTB and BCP, the linearity ranges were determined to be 11.2×10^3 to 1.12×10^6 and 38.0×10^3 to 1.12×10^6 CFU/g, respectively. According to the obtained calibration plots, the correlation coefficients (r) for BTB and BCP were 0.9998 (slope: 2.48 g/CFU) and 0.9999 (slope: 1.95 g/CFU), respectively. According to the obtained calibration plots, the correlation coefficients (r) for BTB and BCP were 0.9998 (slope: 2.48 g/CFU) and

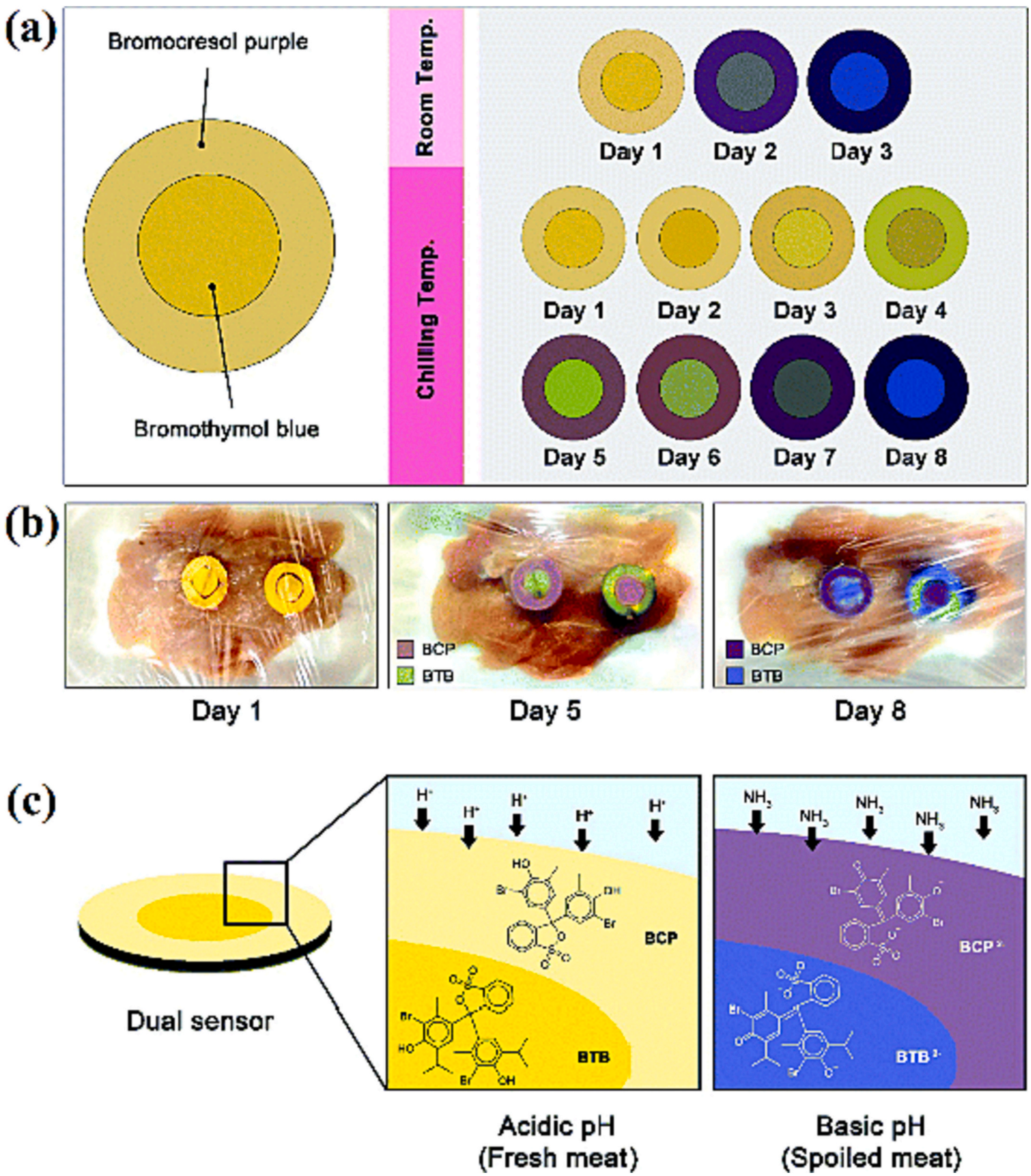

Fig. 8. (a) The lab-on-paper colorimetric sensor structure and the color changes in room and cooler temperatures; (b) Images of one of the test samples of chicken meat at 4 °C chiller temperatures on days 1, 5, and 8 were taken using a smartphone; (c) Color responses of BCP and BTB indicators in fresh and spoiled meat samples. Reprinted with permission from The Royal Society of Chemistry [120].

0.9999 (slope: 1.95 g/CFU), respectively. The as-prepared sensor was used to assess the freshness of chicken meat products for 8 days in the refrigerator and 3 days at room temperature. Based on the microbiological amount of the standard samples determined by the APC technique, the Android application may build a standard curve using the R and G values (for BTB and BCP, respectively) from which the application can read the level of freshness. Fig. 8 represents the structure and working principles of the as-prepared lab-on-paper optical sensor. The suggested sensors have several benefits, including ease of use via the first Android application to be created, cost-effective and simple preparation thanks to the use of ordinary filter papers and pH indicator dyes, and easy detection by the human eye due to the high contrast between the sensor colors under various conditions of microbial spoilage at chiller and room temperatures during the course of storage.

6. Smartphone diagnostics

Given the growing need for technology and the most recent technical developments, cell phones have become an inevitable part of our daily life. They are highly functional and easily transportable, providing enhanced communication capabilities due to their integrated sensors. By connecting the biosensors to cell phones, food assessment may become simple and broadly accessible. Toxins, allergies, pollutants, and viruses may all be detected or determined with these convenient and portable detectors. Throughout the whole procedure of production, sale, storage, transportation and consumption, food items might get contaminated [121,122]. The traditional detection techniques have been duplicated by smartphone-based biosensors, but they are more effective [123,124]. Table 2 summarizes recently developed smartphone-based sensors for food spoilage applications. The potential for biosensors to identify pathogens in food samples is quite significant. Biosensors are also compatible with portable devices since they can be readily integrated into them for the detection of food pollutants [121,124–126], pathogens and shelf life. Smartphones are now great data processors thanks to the most recent operating systems, sensors, transducers, and data processors [124,127,128]. Smartphones have also been used to monitor the environment and assess food quality [129,130]. The advent of smartphones has significantly altered on-site sensing equipment and systems.

Recently, fluorescent-based colorimetric technologies have been employed on smartphones to gather color information, making them more precise, sensitive, and portable. In particular, the RGB analysis is used for digitizing the color information so that the three-channel ratio may derive the fluorescence intensity fluctuation [131]. Consequently, a definitive assessment can be achieved by using smartphone-based fluorescence sensors in food safety issues. In the study conducted by Zhang et al. in 2022 [132], it was found that amine vapor can mediate β-ᴅ-glucose pentaacetate (β-D-GP) to produce photoluminescent polymer-carbon nanodots (PCNDs) with good optical properties. One of the most commonly utilized biomarkers is volatile amines, which are typically produced during the breakdown of food high in protein [133]. Trimethylamine (TMA) was used as a volatile amine in this study and β-D-GP was solidified in agarose hydrogels to produce a practical hydrogel detection platform. The β-D-GP in the hydrogel was instantly mediated by amines to generate PCNDs when exposed to volatile amines released by the decomposition of shrimps, leading to a noticeable fluorescence-based color shift of the functional hydrogel. For on-site quantitative investigation, digital images of hydrogels and RGB data were collected using a smartphone. The hydrogel's G/(R + B) gray value exhibited good linearity with TMA vapor concentrations between 0 and 59.49×10^{-9} mol/dm^3 [132].

Another smartphone-based colorimetric detection platform for meat spoilage was established [134]. In this detection platform, the CO_2 level associated with bacterial growth was tracked. Using the acidity of CO_2, the sensor was developed using three supports which are Byodine B, Nytran N, and Nytran SPC. Supports were used to immobilize the CO_2 sensing membrane. The best results were obtained by using the Nytran SPC support. First, sensors were attached onto the inner part of the meat packages; then, the pork loin fillets were sealed. Only photos of the freshness sensor were taken using these two control samples. The bacterial growth and CO_2 gas emitted by packed meat were well associated with the color information evaluated as a gray scale. As a result, the designed mobile phone application was able to signal that the meat was

Table 2
Some studies performed regarding Smartphone-assisted detection of food spoilage.

Food sample	Target	Reagent	Detection tool	LOD/LOQ/DR	Reference
Fish	Biogenic amine (Cadaverine)	Synthesized PTCN	Fluorescence	46 nM	[137]
Fish	Histamine	CDs	Visual monitoring	36 nM	[138]
Beef, fish, chicken, and pork	TVB-N	Poly(vinyl alcohol)/sodium alginate (PVA/SA) hydrogel	pH, RGB analysis	–	[139]
Fish	Biogenic amines	NH_2-rich lanthanide MOF coupled fluorescein 5-isothiocyanate	Fluorescence	2.17 mg/L	[140]
Beef meat and salmon fish	Biogenic amines	Hydrolysis-induced silver metallization of Au nanorods	Colorimetric	8.6×10^{-9} mol.dm^{-3}	[141]
Beef meat	Microbial contamination	–	Mie scatter angle analysis	10 CFU/mL	[142]
Beef, fish, chicken, and pork	Biogenic amines (NH_3, putrescine, cadaverine)	p-Toluene sulfonate hexahydrate doped nanostructured polyaniline (PTS-PANI)	NFC tag, Smartphone readout	5 ppm	[121]
No specific food sample	Microbial contamination (*S. enteritidis* and *E. coli*)	Mesoporous Pd@Pt nanoparticles	Colorimetric	20 CFU/mL for *S. enteritidis* and 34 CFU/mL for *E. coli*	[128]
Milk, cheese, and water	Microbial contamination (*S. enteritidis*)	The combination of the magnetic-antibody and the HRP - Antibody - nanoflower based ELISA	Colorimetric	1.0 CFU/mL	[143
Yoghurt and egg	Microbial contamination (*E. coli*)	Compact laser-diode-based photosource, a long-pass thin-film interference filter and a high-quality insert lenses on the Smartphone	Fluorescence	1.0 CFU/mL and 10.0 CFU/g	[143]
No specific food sample	LPS	A grating coupled SPR Smartphone spectrometer	SPR	32.5 ng/mL	[144
Fruit juice	Malathion	A 3D printed chip	Colorimetric	51.9 ng/mL	[145
Fruits and vegetables (Apple, tomato, grape, green pepper)	OPPs	OPH-based biosensor system on the index finger of a flexible lab glove	Enzyme-immobilized biosensing	–	[146]

NFC: Near-field communication; SPR: Surface Plasmon Resonance; LPS: Lipopolysaccharides; OPH: Organophosphorus hydrolase; MOF: Metal-organic framework, PTCN: A ratiometric fluorescence probe.

already spoiled if the value of the gray scale exceeded the established threshold of 98.5 (Fig. 9).

In another study performed by Kilic et al. [135], anthocyanins-doped fish gelatin (FG) films were developed for food freshness monitoring. CDs were used as potential crosslinkers in producing FG films. UV emission of the CDs improved the optical, surface, structural, barrier, and mechanical properties of FG films. Ammonia vapor was the biomarker interacting with CDs-incorporated FG films. Sensor films with different CD concentrations were prepared, and their color changes upon interaction with ammonia vapor were utilized to develop the mobile application monitoring food spoilage. The most sensitive results to bacterial growth and TVB-N were obtained with 100 mg/L CD incorporation into FG films. Accordingly, it was predicted that FG-UVCD100 films would exhibit a colorimetric reaction to TVB-N as soon as spoiling started. A custom-designed smartphone app (Smart Food) with picture processing capabilities was created for the quantitative study of food deterioration. The Smart Food's embedded analysis tool tracks and displays the level of spoilage and notifies the user of the food's current freshness state.

To achieve the quantitative detection of NH3 and enable intelligent real-time monitoring of seafood spoilage using standard smartphones, a fly antennae-inspired fluorescence platform (PAA-FP) was recently created [136]. Filter paper and self-aggregated NH_3 sensitive fluorescent probe were combined, and the resulting PAA-FP with anticipative "fly antennae-like" convex microstructure showed good solid-state fluorescence characteristics and selective detection of NH_3 by naked eyes in daylight (Fig. 10a). The visible color of the shrimps was changed from cyan to orange upon storage at room temperature. Accordingly, the color of the PAA-FP was changed from light yellow to dark orange. The G value determined by color recognition had a good linear connection with storage time. PAA-FP was also used to track the degradation of fish meat samples, revealing a solid linear connection between G value and storage duration. Initial tests of the study determined a correlation between the NH_3 concentration and the RGB values. Also, a simple AcOH fumigation procedure was employed to reuse PAA-FP. As the concentration of NH_3 rose, the color of PAA-FP and the pertinent values of RGB also altered simultaneously (Fig. 10b, c and e). The G value and the concentration of NH_3 showed a good linear relationship which can be seen in Fig. 10d. Therefore, it was concluded that the developed PAA-FP could detect NH_3 quantitatively and visually by using a smartphone.

7. Application of intelligent probes on food products

Nowadays, the idea of intelligent food sensors for food quality and freshness monitoring has been found as a very interesting area of exploration in food science to improve food shelf life and reduce food waste [147. This part of the present review is focused on the review of the most recent literature related to the application of intelligent food packaging with an emphasis on their functional characterization in food freshness control. Term food freshness indicator refers to the probes/ sensors with the ability to communication the product spoilage. In some cases, sensors reveal just binary signal fresh-spoilage, but solutions with higher resolution are also reported. These more accurate probes can indicate also intermediate states such as: *less fresh*, still edible, spoiling which makes them more effective and attractive.

In some cases, presented intelligent probes are given in the form of some pattern which increases its attractivity and usability for a customer such as QR-code like sensor [148], happy/sad face pattern [159] or imprinted information being visible when the sensor recognizes a product as spoiled [150]. Some the sensors can be applied as inkjet-printed bars [151]. These less typical and more complex but much more attractive solutions from the commercial perspective are presented in Fig. 11. Table 3 presents various sensors tested in the real sample applications by pointing their composition, however the consideration of the functional principles is the best way to find the common points between different sensors. Based on the information in Table 3 it is seen that meat is the general type of product targeted in the considered research. It is because meat (next to dairy) has been definitely recognized as a very perishable food, what is more, wasting of this kind of product is not only related to hunger and money – it is also about the social aspects of using animal products. Since the food pH changes are directly connected with its quality loss and spoilage, one of the main functional principles used for the development of freshness sensor action is among others pH-sensitivity. In this kind of intelligent probes, the chromatic/fluorescent answer relates to the presence of pH responsive compound or compounds mixture in the overall sensor composition. Anthocyanins are a group of natural pigments commonly applied in sensors as pH-responsive agent [152–155]. The colorimetric anthocyanins-based sensors for freshness monitoring of chicken, shrimp and pork meat have been successfully applied by immobilization of various sources derived anthocyanins in different matrices and carriers. For example, Franco et al. reported a happy/sad face indicator of chicken freshness [149] based on anthocyanins obtained from black carrot immobilized in cellulose acetate film. The adhesive paper with printed circular Emoji - happy and sad faces playing a role of pH inactive reference layer whereas the eyes and mouths were filled with pH sensitive anthocyanin/cellulose acetate composite. As prepared, the label was attached to the fresh chicken meat and as long as the pH of the environment did not reach the values above 6.2 the sensor color stayed pink and the visible Emoji face was smile. Spoiling of the meat resulted in pH increasing of the label surroundings and subsequent color change to purple was reflected by the change of the Emoji face to the sad one. In another effort, the impregnation of filter paper with anthocyanin solution resulted in an indicator card for monitoring of shrimp freshness

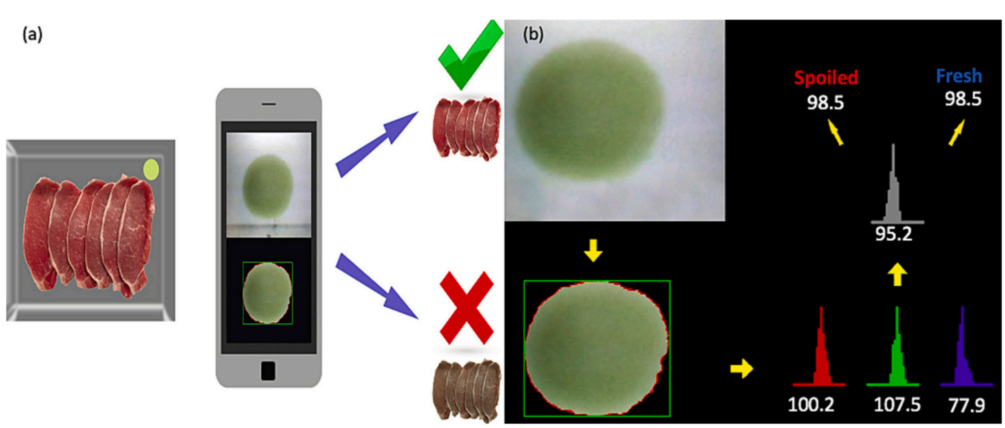

Fig. 9. (a) Schematic representation of colorimetric Smartphone-based meat freshness sensor; (b) Evaluation of the threshold value through the signal. Reprinted with permission from Elsevier [134.

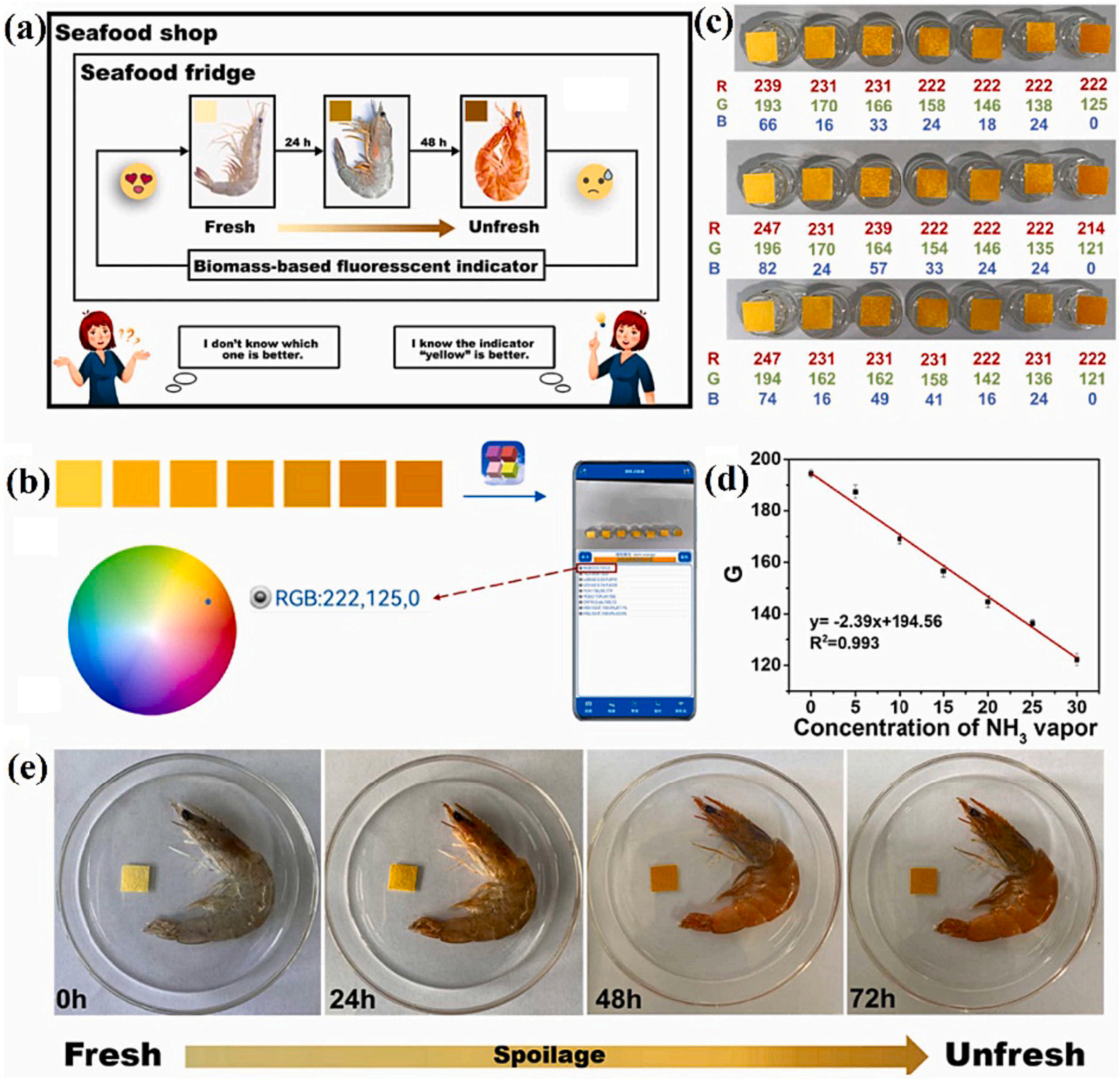

Fig. 10. (a) Schematic diagram of the application of the PAA-FP; (b) Quantitative detection of NH_3 through a Smartphone; (c) RGB values of PAA-FP after exposure to different concentrations of NH_3 (0 ppm, 5 ppm, 10 ppm, 15 ppm, 20 ppm, 25 ppm, and 30 ppm from left to right); (d) graphical relationship between NH_3 concentrations and G values of PAA-FP; (e) monitoring of shrimp spoilage with time. Reprinted with permission from Elsevier [136].

[150]. In this case, the expected color change varied from pink at pH around 4.5 through purple for pH 6 to green at pH higher than 9. The probe was placed at the inner side of the container contained shrimp and its color changes were monitored. Finally, nineteen days storage of the shrimps at 4 °C resulted in spoiling the product which was clearly announced by green color of the indicator. Authors ascribed the pH changes to ammonia and volatile amines generation during the shrimp spoilage process that was confirmed via model experiments with ammonia solution. The applied filter paper was imprinted with pink word "BAD". The shade of the printed mark was similar to the anthocyanin at pH below 5 which made it invisible in contact with fresh products, hence the pink-green color change resulted in the appearance of "BAD" on the indicator. Chumee et al. [156] developed a pork freshness indicator by coating of a chromatographic paper with a biofilm containing anthocyanins extracted from a red cabbage. Similar to the above-described cases, the pH increases related to the spoiling of the meat resulted in the color change from initial pink to orange after 24 h of storage at 25 °C. Ammonia level depended pH changes were also used as a base for the preparation of agar/sugarcane wax film modified with anthocyanin extracted from butterfly pea flower [157]. The film was applied as the freshness indicator of shrimp via its placing on the cover of the container without direct contact with the product. The samples were kept at room temperature for 24 h and the colorimetric response of the probe was monitored. Nitrogen contained volatile compounds generated during the *spoiling process* were recognized as responsible for pH changes that was confirmed with digital pH measurements and determination of total volatile basic nitrogen in the samples. The pH during the storage time changed from 6.7 in the beginning to 8.7 after 24 h which was continuously revealing by the colorimetric sensor.

As mentioned earlier, pH variation related to negative changes in the food has been one of the main bases for designing new freshness sensor. Commonly, anthocyanins play a role of sensing agent, however the probes based on other pH indicators are also known. Chen et al. [148] reported a QR-code like food freshness sensor based on nile red, zinc tetraphenylporphyrin and methyl red as pH sensitive dyes. Anion exchange resin was saturated with a solution of nile red and methyl red, cation exchange resin was, in turn, a matrix for Tetraphenylporphyrin. The idea of giving a predefined shape and form of the sensor was realized by using a laser cut polymer template and PDMS mold. Finally, one sensor comprised of three different probes (differentiated by shape/pattern) deposited on a filter paper. The sensor was applied on chicken meat in a way allowing for direct contact between the meat and the filter paper and the samples were stored under different temperature conditions. The color variations assigned to the product spoilage were

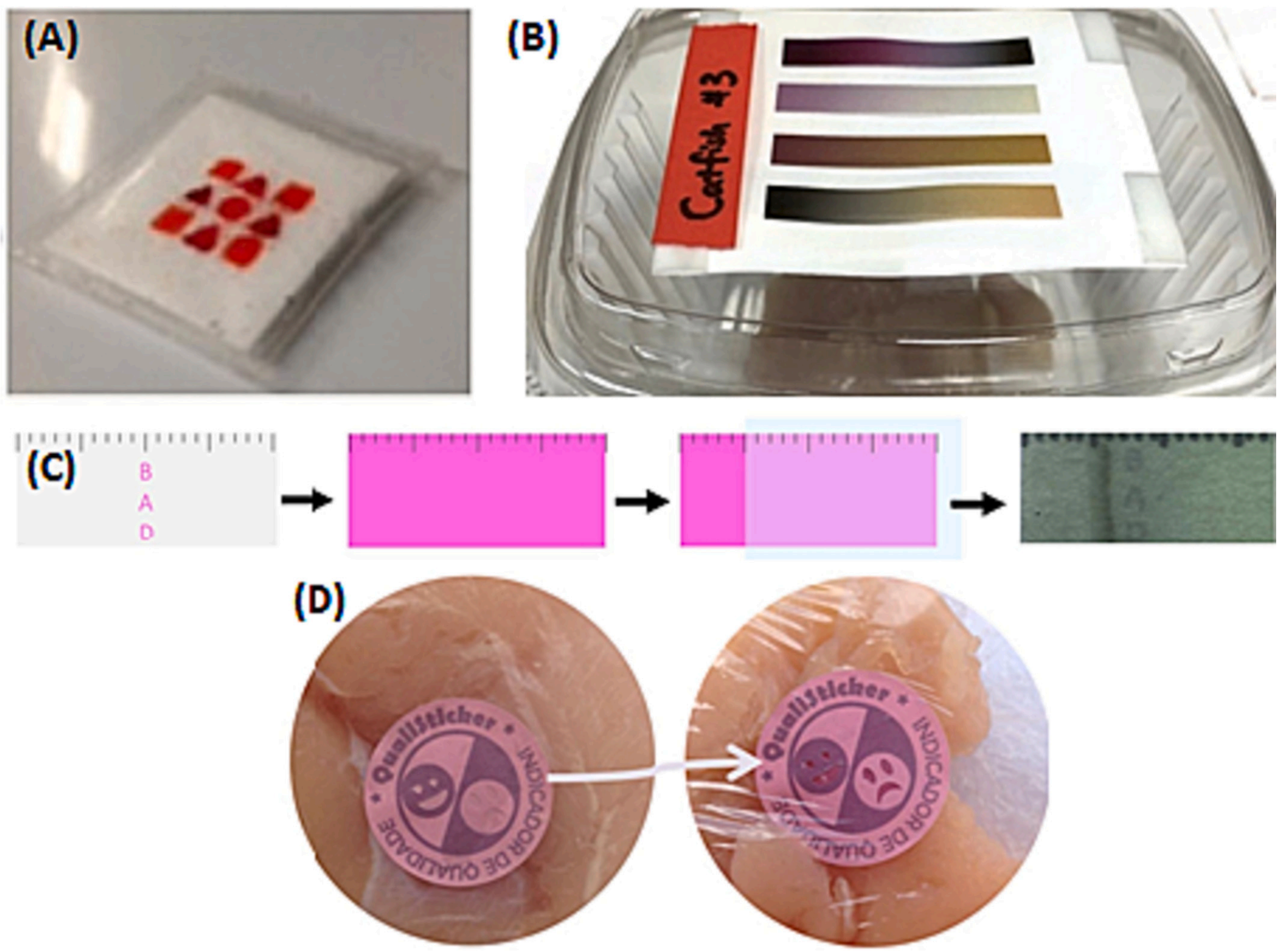

Fig. 11. Different forms of food freshness/spoilage indicator with increased customer attractivity: (A) QR-code like [148], (B) progress bar [151], (C) imprinted information [150], (D) happy/sad face [159]. Reprinted with permission from Elsevier [148–151].

monitored using a smartphone camera. The chromatic response of the three chosen probes made the overall sensor promising as the indicator of pH changes connected with spoiling of the examined product. Later, a printing paste contained pH sensitive dye for application in screen printing using cellulose paper was described by Zhang et al. [48]. The authors describe detailed synthesis and characterization of two dyes (anthraquinone and azo chromophores based), whereas both revealed activity under different pH ranges. One of the prepared dyes, called D-1 was placed in a zip-lock bag containing a cooked crab and left at 37 °C. A clear color change (dark violet to black) was noted which indicated the spoilage of the product after 4 h of storage. The authors emphasized that the application of cellulose paper and the printing technique guarantees a stable and irreversible fusion of the dye and carrier thanks to covalent bonds creation. An array based on 18 various pH indicators, 5 porphyrins and phthalocyanine deposited as separate circles on a filter paper played the role of the multipurpose sensor for freshwater bass meat freshens monitoring [172]. The colorimetric probes array was responsive to pH changes and/or generation of various gaseous products such as trimethylamine, aldehydes and sulfides. The continuous monitoring of the color change of particular probes allowed authors to determine a color difference map which made the sensor a promising tool for more accurate and selective food freshness control.

Among colorimetric freshness probes applied in real food samples one can also find various solutions using fluorescent responses of the sensor active part. Template synthesis of a sensor containing SNARF-1 fluorescent agent was reported by Kiryukhin et al. [158]. Successful synthesis of the sensor comprising encapsulated SNARF-1 (SNARF-1 Dextran) in microchambers well-distributed on a polyethylene film was confirmed by SEM and fluorescent microscopy. Fish juice prepared by homogenization of golden pomfret meat in deionized water was used to assess the performance of the prepared sensor as a freshness controller in semi-real food sample. Fluorescent pH response of the sensor during the storage time agreed with bacteriological analysis which allowed to consider the described approach as a promising indirect method of monitoring microbial spoilage of the fish. The challenge with such sensors is that fluorescent methods usually require additional tools like UV lamp, fluorescence microscope or spectrometer for reading the results. However, in the modern world of miniaturization this kind of solutions should be also taking into account which can make the final industrial application of the sensor more difficult.

As mentioned above, pH changes identified by sensors are frequently assigned to gaseous compounds generation during the food spoilage process. Gaseous products such as different amines, ammonia or hydrogen sulfide arise as a result of protein or more accurately amino acids decomposition during the food spoilage process. Generated gases influence the pH of fluids presented in the product which can be indicated by the probe. The second scenario is a chromatic response caused by pH changes of the water/humidity reaching the sensor in the case of food-probe contactless solutions. The specific process responsible for NH_3 creation is the deamination of amino acids. One example of such

Table 3
An overview of the compositions, applications and functional principles of various real-time food freshness/spoilage sensors applied on different types of food products.

Probe description	Probe composition	Food application (freshness control)	Functional principle	Ref.
Paper-based colorimetric sensor (smartphone detection)	pH responsive agent: nile red/zinc tetraphenylporphyrin/methyl red Polymeric matrix: anion/cation exchange resins Carrier of the sensor: filter paper	Chicken breast fillet	pH monitoring	[148
An active two-faced indicator label	Film creating agent: cellulose acetate pH responsive sensor: black carrot anthocyanin	Chicken meat		[149]
An acid-base indicator card	pH responsive agent: purple sweet potato solution - Carrier of the sensor: filter paper	Shrimp		[150]
Colorimetric biofilm sensor	Film creating agents: agar, sodium alginate, polyvinyl alcohol pH responsive sensor: red cabbage anthocyanin	Pork		[156]
Smart color-changing paper packaging sensors	pH responsive agent: anthraquinone and azo chromophores Carrier of the sensor: cellulose paper	Cooked crabs		[48]
Membrane film sensor	Fluorescent sensing dye: SNARF-1 Array for the dye immobilization: polyelectrolyte multilayer film Support of the sensor: polyethylene film	Golden pomfret fillet		[158]
Colorimetric pH sensoring packaging films	Film creating agent: agar Film modifier (mechanical and optical properties): sugarcane wax pH responsive sensor: butterfly pea flower extract	Shrimp		[157]
Progress bar colorimetric strip sensor	Colorimetric sensors: bromophenol blue and bromocresol green	Cod fillet	NH_3 level monitoring	[159]
Ionic-liquid doped polymeric composite colorimetric sensor	NH_3 responsive agent: 1,2-naphthoquinone-4-sulfonic acid sodium salt Polymeric matrix: polydimethylsiloxane Matrix additives: tetraethylorthosilicate and SiO_2 NPs 3 alternative ionic liquids (promotors of porosity and permeability of the matrix): 1-methyl-3-octylimidazolium hexafluorophosphate; 1-butyl-3-methylimi-dazolium-octyl sulfate; 1-butyl-4-methylpyridiniumhexafluorophosphat	Chicken meat		[160]
Extruded LDPE -curcumin film	NH_3 responsive agent: curcumin Sensor matrix: LDPE	Silver carp and beef		[161]
Colorimetric sensor array with classification algorithm	Responsive agents: 3 pH indicators and 9 metalloporphyrins Support of the sensor: C2 reverse phase silica-gel plates	Chicken breast fillet	Volatile compounds detection	[162
Colorimetric indicator grafted on MOF carrier	Responsive agent: bromothymol blue Film creating agents: cellulose acetate/polyethylene glycol 4000	Grass carp		[163]
Reversible AIE-active fluorescent probe	Fluorescent probe: CLBZ Carrier of the sensor: filter paper	Beef and shrimp	H_2S level monitoring	[164]
Water based-ionic liquid carbon dioxide sensor	CO_2 selectivity promoter: 1-ethyl-3-methyl-imidazolium chloride Responsive agent: meta cresol purple sodium salt Film forming agent: ethyl cellulose	Pork	CO_2 level monitoring	[165]
Colorimetric gas-sensitive array sensor	Responsive agents: various pH indicators, five porphyrins and one phthalocyanine Carrier of the sensor: filter paper	Freshwater bass meat	Trimethylamine, aldehydes and sulfides levels monitoring	[166]
Sensor array with portable electronic analyzer	Colorimetric sensors: various metals (Zn, Cu, Ni, Co, Al, Sr, Cd, Fe, Cr, Li, Pb) complexes of single azophenol dyes	Spiked meat and cottage cheese enriched with spermine and tryptamine	Biogenic amines determination	[167]
Sensitive ratiometric fluorescent sensor	pH responsive sensor: CdTe quantum dots	Shrimp		[168]
Fast visual fluorescent sensor	Fluorescence sensor: dicyanovinyl coumarin	Beef	Cadaverine determination	[169]
Enzymatic time-temperature colorimetric Indicator	Bioactive compound: laccase Electrospun film: chitosan/polyvinyl alcohol/tetraethylorthosilicate	Milk	Enzymatic time–temperature indicator	[170]
PDA/ZnO colorimetric sensor	Colorimetric responsive agent: polydiacetylene/zinc oxide Matrix film creating agent: agarose	Milk	pH and lactic acid level monitoring	[171]

LDPE: low density polyethylene; CLBZ: 4-chlorosalicylaldehyde and 2-benzothiazo-leacetonitrile probe; PDA: polydiacetylene.

sensors is NH_3 responsive food freshness probes. BPB and BCG were used as pH sensitive dyes for the preparation of progress bar for monitoring of cod fillet freshness [163]. The sensor was successfully applied in the freshness monitoring of cod fillets. Fig. 12a presents the gradual color changes of the progress bar in relation to a different stage of the fish freshness/spoilage level. The authors determined the total volatile basic nitrogen (TVB-N) generated during 30 h storage of the fish fillets at 25 °C which allowed to calibrate the bar and regulate three freshness states of the product according to the official standards in China. The reported progress stripe could help to differentiate the fish as fresh, less fresh and spoiled via naked eye and simple readings. Ballester-Caude et al. [160] proposed a chicken freshness colorimetric sensor based on 1,2-naphthoquinone-4-sulfonic acid sodium salt as a NH_3 responsive agent with the aid of an ionic liquid which positively influences the effectivity of the sensor. A commercially available Silicone Elastomer Kit was the base of the sensor matrix. The prepared probe was calibrated with ammonia standard and tested with chicken meat. The calibration experiments proved that the colorimetric response of the studied sensor showed a clear shift in its color from orange in the case of fresh meat (low ammonia content) to dark brown for the spoiled product. Moreover, the sensor color changed gradually which allowed separating of three stages of the chicken meat quality-fresh, sub-fresh and putrid. Fig. 12b shows the colorimetric response of the probe (red half circle) in comparison to reference probes (orange half circles). The LDPE extrusion method was used to develop a NH_3 sensitive probe containing curcumin as an active agent [161]. Application of LDPE film as a sensor

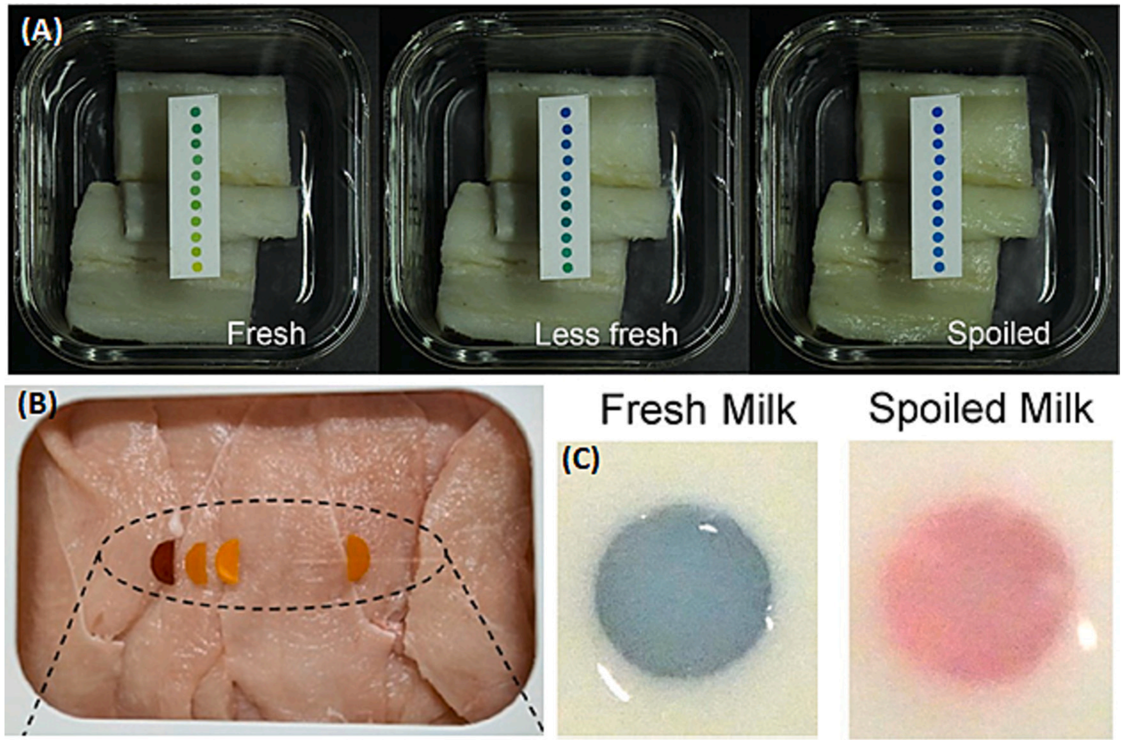

Fig. 12. Practical application of different indicators in relation to: (A) fish filet [163], (B) chicken [160], (C) milk [171]. Reprinted with permission from Elsevier [160,163,171].

matrix guaranteed perfect mechanical and chemical stability of the sensor or packaging materials allowing to use the sensor in the storage of foods with very high humidity which is common for meat products. Silver carp fillet and beef samples were used as example food products for freshness monitoring during storage. The meat spoilage was reflected by the sensor color change; however, the color difference was not as well-defined as in the case of previously described cases. The color variation related to the NH_3 release was slightly visible with naked eye, but the accurate quantitative differences were delivered by colorimetric analysis.

TVB-N is a frequently used parameter in meat spoilage studies which gives a quantitative view on nitrogen contained volatile compounds generated during meat storage, namely NH_2, trimethylamine and dimethylamine. This section presents a brief description of the sensor assigned by authors as volatile compounds sensitive. For example, the sensor containing 12 responsive agents placed on C2 reverse phase silica-gel was tested as volatile compounds responsive array for chicken spoilage control [162]. Moreover, the authors developed a classification algorithm that helped with the freshness level evaluation. Application of the sensor on real chicken samples, followed by the digitalization of the results using a computer scanner in combination with the developed classification algorithm resulted in a promising and selective indicator of the product freshness. Another probe based on volatile compounds level monitoring was reported by Wang et al. [163]. Chemical and mechanical stability of the sensor was guaranteed by grafting the bromothymol blue on a Metal-organic framework (MOF) with its subsequent immobilization in the structure of cellulose acetate film Grass carp spoilage control tests were conducted in order to evaluate a practical implementation of the sensor. A slight color change was observed by a change in the freshness level of the product. This chromatic response was in good correlation with the TVB-N content of the fish samples.

Hydrogen sulfur (H_2S) can be also commonly found among the gases in food headspace generated by a wide group of bacteria called H_2S producing bacteria during the meat and fish spoilage process. Therefore, H_2S has been also targeted as a compound for monitoring food bacterial spoilage using intelligent sensors. For example, a fluorescent active compound called CLBZ (synthesized by authors using 4-chlorosalicylaldehyde and 2-benzothiazoleacetonitrile) was deposited on a filter paper and used as a beef and shrimp freshness sensor [164]. The chromatic response of a sensor was presented both under UV lamp and sunlight to reveal its practical usefulness in everyday application. The main difference between the observed color changes under UV and sunlight was better resolution in the presence of UV radiation. The color changes of the sensor applied in the food containers both under UV and sunlight were sharp enough to communicate the total spoilage of the product but also gradual changes indicating an intermediate state between fresh and spoiled products were observed. The presence of visible light allowed the sensor to be practically applied in the monitoring of beef and shrimp freshness which was clearly described and documented in the article (Fig. 13). Moreover, the CLBZ-based sensor was highly selective in relation to H_2S.

The next gaseous product of meat spoilage is CO_2 which changes the pH of fluids by its dissolving followed by the creation of unstable carbonic acid. Meta cresol purple sodium immobilized in 2-hydroxyethyl cellulose film was practically applied as a sensor for pork freshness control [165]. Additionally, the sensor was enriched with an ionic liquid which promotes its selectivity by enhancing CO_2 solubility. The authors conducted systematic tests of the sensor colorimetric response to CO_2 levels in model systems followed by microbiological test during pork storage which allowed to calibrate the sensor and determine the critical CO_2 level indicating the product spoilage.

Biogenic amines generated in the atmosphere of spoiling food as a result of protein and amino acid degradation have also found some dedicated probes and sensors for application in intelligent food packaging. Singh et al. [167] developed multipurpose sensors array for the determination of different amines, namely: spermine, histamine, spermidine, tryptamine, creatine, histidine and cysteamine. Every single probe contained a metal complex of azophenol dye. Application of different metal complexes guaranteed a selective chromatic response in the case of contact with a particular biogenic amine. In addition, the

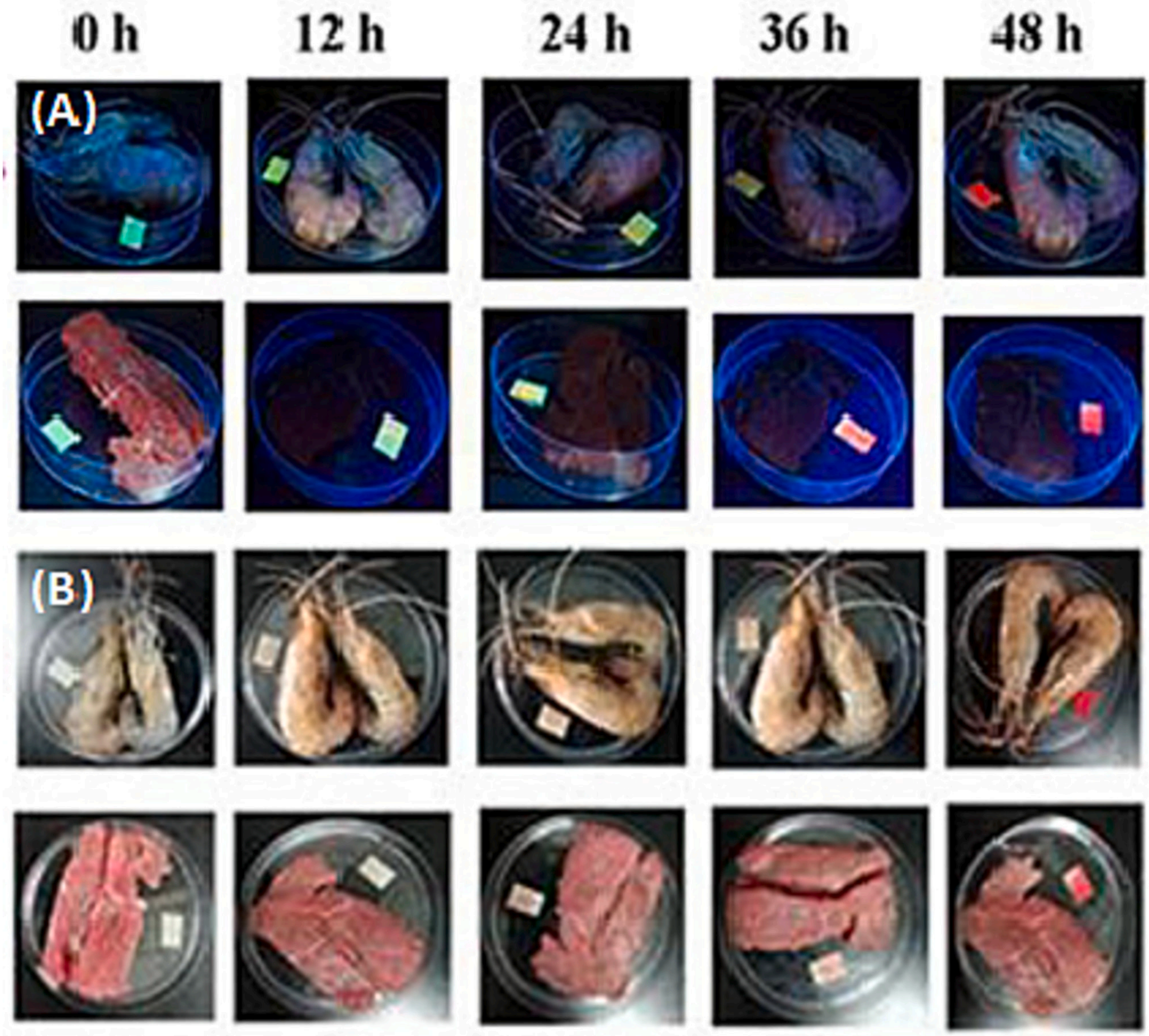

Fig. 13. Practical application of fluorescent indicator in relation to shrimps and beef under (A) UV lamp and (B) sunlight [164]. Reprinted with permission from Elsevier [164].

authors used a portable plate reader for the automatization of the analysis. The practical application experiments were conducted on spermine and tryptamine enriched ethanol extracts of meat and cottage cheese. The successful semi-real application of the sensor resulted in the effective detection of two examined amines in the relatively wide concentration range between 0 and 40 μM with 100 % accuracy. Biogenic amines determination was also a working principle of the sensor based on blue and yellow fluorescent quantum dots deposited on a filter paper [168]. The amine sensitivity of the sensor was successfully confirmed with individual experiments for eight standard solutions, whereas the practical application test included shrimp spoilage monitoring. In this case, the fluorescent response could be noted under the presence of UV radiation by 365 nm UV light. A color change from blue to bright green after 24 h of storage indicated spoilage of the shrimp which was photographically documented. Cadaverine is a well-known biomarker of food spoilage, hence, the fluorescent sensor of this biogenic amine was developed [169]. The test strips were prepared by immersion of a filter paper in an ethanol solution of CMCD (dicyanovinyl coumarin) and the reading was conducted under UV radiation. Firstly, authors conducted competitivity tests with different chemical species which can be present in a packed food environment, namely: ethylamine, triethylamine (TEA), N, N-diisopropy-lethylamine (DIPEA), diethylamine (DEA), ammonia, S^{2-}, SO_3^{2-}, H_2O_2, NO_2 and glutathione (GSH). No significant influence on cadaverine sensing was observed with the presence of the competing compounds. Additionally, experiments for different spoilage biomarkers including histamine, tyramine, putrescine, and cadaverine were conducted, and the sensor was found selective towards cadaverine and putrescine. The practical application test for beef freshness monitoring was conducted by placing the sensor on the inner wall of a Petri dish lid while there was no direct contact between food sample and the sensor. Three temperature modes were chosen for the storage test, namely 25, 0 and −20 °C and during this time the fluorescence images of the sensors were recorded after pre-defined time intervals. The readings were correlated with the results of the total volatile base nitrogen analysis which allowed to assign the color change of the sensor to the beef spoilage level. TVB-N content in beef on the levels of 15, 20 and 25 mg·100 g^{-1} refer to the fresh, slightly spoiled and totally spoiled meat, respectively which was clearly reflected in the fluorescent signal of the sensor under 365 nm UV light. The results for different storage temperatures presented in a clear and elegant manner how the storage conditions influenced the shelf life of the product. In this view, the product was recognized as not fresh after 16 and 60 h of storage at 25 and 0 °C, respectively, but it was still fresh after 25 days in the frozen form (−20 °C).

Generally, meat products are recognized as perishable, thus the research on real time freshness control is mainly focused on this kind of products. However, dairy products also deserve attention in this field. In another study an enzymatic milk quality indicator containing laccase immobilized on a chitosan/PVA/tetraethylorthosilicate electrospun film was developed [170]. The chromatic response of the sensor was consistent with the lactic acid bacteria growth in the product. Another milk freshness colorimetric indicator with a well-pronounced chromatic response was synthesized [171]. In the presented case, agarose film containing ZnO trapped in polydiacetylene liposome indicated the pH changes in spoiling milk. Observable pH changes are related to the bacterial conversion of lactose to lactic acid. The prepared probe was applied in the real sample tests which brought a positive result-clear change of the color from pink (fresh milk, pH about 6.8) to blue for the spoiled product with pH about 4.0 which is presented in the Fig. 12c.

Food freshness sensor is a strongly explored area in material and food science. In accordance with the presented example studies, it is clearly seen that the majority of current solutions are based on continuous monitoring of pH changes in the product environment. Observable pH variations can be related to different chemical changes of the product during the storage time – among others: generation of ammonia, hydrogen sulfur, CO_2 or biogenic amines. In many cases, the colorimetric response of the sensors allows to differentiate the spoilage-related chemicals and sometimes their quantitative or semi-quantitative analysis is possible. Numerous forms of sensors have been proposed: progress bars, set of the sensors or one, single color change sensor and sometimes more complex and commercially attractive forms and patterns. The recent literature review allows to conclude that the filter paper can be successfully used as a sensor matrix or carrier which makes its practical application easier. Noteworthy, bio-based and biodegradable polymers based on PVA, cellulose acetate or agar were also profitably applied as a sensor matrix which is very important in today's material science.

8. Intelligent packaging in facing food waste

Food waste and hunger have been recognized as the global problems of today's world while the global food system is facing serious environmental sustainability issues too. According to the available data, the mass percentage of wasted food can reach up to 33 % [173,174] of the global food production wherein 8.9 % world's population suffers from hunger [175]. Irresponsible food management finds the reflection not only in a problem of undernourishment - it relates also to many financial, social and environmental worldwide issues. Food is wasted in different ways, from the initial production stages (usually called food loss), through transport and sale to our households. Noteworthy, according to the report of Stenmarck et al. [174], responsibility for more than half of lost food should be assigned to private households, which is essential information from the perspective of this review. Large-scale food production is fraught with a high risk of food material loses. In this case, the responsibility for the reduction of food waste lies with the relatively narrow group of people such as factory management and employees. From the perspective of large-scale production plants, introducing of well-organized actions targeted on mitigation of food wasting can be more effective in comparison to actions by billions of single households. In this view, there is an urgent need to provide some novel effective solutions dedicated to retail consumers Billions of people over the world having different economic statuses feel different responsibilities for social and environmental live aspects including food management in their own households. Among many reasons and mechanisms, one of the possible answer to the question why such a large amount of food is wasted in households is the imperfect communication of the time for safe product consumption called shelf life date. Firstly, there is an existing problem of misunderstandings of different ways of the product freshness predictions placed on the packaging such as: “best by”, “best before” or “expiration date” which results in wasting of still fresh and edible food [176,177]. Frequently, the role of the part of these dates is to give a tip related to the freshness of the product, but its final evaluation belongs to the consumer. In fact, the final quality of the bought food in some particular day depends on its delivery chain history. In the case of perfect production and storage conditions over the course of the whole product lifecycle its final shelf life can be much longer than suggested by “best before” date. On the other hand, if the condition on some production or delivery stage differed from optimal the product spoiling processes can be accelerated.

Social campaigns are one of the non-invasive methods of influencing people in order to raise their awareness of various social aspects or problems, in other words, it is a way of making society realized about some problem without introducing legal duties, prohibitions and orders [178–180]. In the view of well-developed social consumers-directed actions both on a local or wider scale one can say that the growing awareness of the food waste issue should meet novel tools and solutions which could be used against it. At this moment intelligent/smart food packaging beside active food packaging which represents the separate technology in the food industry can respond to the expectations. This promising technology based on real-time freshness/spoilage control can effectively replace or complement the current printed date approach. Moreover, intelligent packaging brings double benefits since it indicates both early spoilage of the products related to inappropriate storage as well as prolonged freshness caused by careful storage. Some of the reported solutions provides also information about gradual freshness losing what can help consumer to take the action to use it before spoiling.

Traditional and basic roles of the packaging include (i) mechanical and biological protection, (ii) conveying practical and marketing information, (iii) enabling long distance transport. The above review article presented a wide range of solutions related to the novel, environmentally-oriented role of the packaging – real time indication of the product freshness/quality. This relatively young area of material and food science is a real challenge for the scientific world, but in this case, there is an additional social responsibility to provide effective and commercially applicable solutions and it is obvious that mitigation of food wasting is the primary objective of the research. Such indicators placed into the packaging allow customers for continuous control of the freshness and quality of their products which can actively and effectively contribute to food waste limitation. Moreover, as it was mentioned in the previous paragraph, the current stage of research on this technology allows to design and produce of sensors that can actively communicate with the customer by nice/funny form or shape. The rapid development of intelligent packaging together with worldwide social campaigns will hopefully bring a synergistic effect with real, visible results.

9. Conclusion

Growing concerns for environmental conservation and food safety will inevitably lead to the replacement of packaging materials derived from petroleum and synthetic dyes with safe, biodegradable natural components. Innovative intelligent packaging technologies are beneficial for addressing rapidly changing consumer and industry expectations and developments. Several recent studies have proved the practical potential of intelligent probes for assessing food freshness utilizing naturally sourced pigments for real-time quality monitoring of perishable foods.

Incorporating colorimetric and paper-based indicators based on natural dyes into food packaging has multiple advantages, including the timely disclosure of real-time quality details, the facilitation of decision-making, the long-term reduction of food waste, the decrease of foodborne diseases, spoilage, and deterioration of food products, and the enhancement of the packaged product's appeal to retailers and consumers. In contrast, the colorimetric sensor system technology currently in use still relies on synthetic dyes, offering certain advantages to both the industry and food quality. However, using synthetic dyes have several disadvantages such as the toxicity, carcinogenic effects, and allergenicity of synthetic dyes compared to natural colorants, and the use of toxic solvents that are unacceptable for the food industry. This study reviewed the ideas of different types of intelligent packaging based on natural dyes that could help to tackle the current issues. Presently, pH-responsive colorimetric indicator films based on natural ingredients are utilized in the laboratory, but further research is required to boost their wide production and commercialization. In conclusion, the study emphasizes the potential impacts of intelligent food packaging on reducing food waste, food poisoning, and contamination. Intelligent packaging offers significant implications for public health and environmental sustainability by addressing these issues. Therefore, as scientific research and related technologies develop, colourimetric and paper-based probes that monitor food quality and safety will become one of the future research trends.

CRediT authorship contribution statement

Shima Jafarzadeh: Conceptualization, Investigation, Project administration, Validation, Visualization, Writing – original draft, Writing – review & editing. **Zeynep Yildiz:** Investigation, Writing – original draft. **Pelin Yildiz:** Investigation, Methodology, Writing – original draft. **Przemyslaw Strachowski:** Methodology, Visualization, Writing – original draft. **Mehrdad Forough:** Investigation, Visualization, Writing – review & editing. **Yasaman Esmaeili:** Methodology, Writing – original draft. **Minoo Naebe:** Supervision, Visualization, Writing – review & editing. **Mehdi Abdollahi:** Investigation, Validation, Visualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No research data were shared. There was no need for ethics approval for the present research.

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