Journal of Pharmacognosy and Phytochemistry

Available online at www.phytojournal.com

E-ISSN: 2278-4136
P-ISSN: 2349-8234
JPP 2018; 7(4): 809-812
Received: 11-05-2018
Accepted: 15-06-2018

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Indicator sensors for monitoring meat quality: A review

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Abstract
Modern storage facilities have made it possible to store meat for and transport meat for long distances. No doubt these practices are helping to tackle food security, but cold chain breakdown conditions open gates for abuse and supply of spoiled meat to consumers. Even though we can develop very good manufacturing and packaging facilities, a minute abuse at any stage of transport or in the retail shops will be sufficient enough to spoil the whole effort and providing unsafe meat to consumers. A large number of cases of food poisoning are seen due to consumption of unsafe packed food products including meat, making it necessary to track the freshness in supply chain. Estimation of freshness is of utmost importance from the point of food safety especially due to changing food habits when the consumer demands minimally processed products. These cases of food poisoning not only question the reputation of meat industry but also compromise consumer's health. Most of the times these are related to abuse during supply chain which gives an opportunity for microbes to flourish. In recent years, intelligent packaging is emerging as a new concept for food safety. Intelligent packaging systems are those that monitor the condition of packaged foods to give information regarding quality of the packaged food during transport and storage. This information will be available to consumer as readymade through colour change indicators in packed meat upon spoilage. So if an intelligent packaging system is developed, it will be of immense use in meat sector for monitoring safety throughout supply chain benefitting both manufacturers and consumers. By this manufacturers can track quality and withhold unsatisfactory meat before they find their place in the display cabinets for sale. Whereas, consumers will confidently purchase meat without any doubt over quality simply by observing the colour change in the indicator placed along with packaged meat. Several indicator sensors have been developed using various dyes, indicators carrier and techniques. This review focuses on newly developed indicator sensors developed for real time monitoring of meat and meat products.

Keywords: indicator sensors, time temperature indicators, gas sensors

Introduction

Food safety and control requires the application of analytical techniques capable of producing reliable data in order to establish if food complies with the regulatory organization criteria. As a society is becoming more complex, users (food producers, food processors, logistic operators, retailers and consumers) continuously demand innovative and creative food packaging to guarantee food safety, quality, and traceability. This requires appropriate technologies that can be integrated in food packaging. As a result, over the last decades, researches have shown a growing interest in developing different food safety measures to ensure security and quality of food. The food industry is constantly trying to enhance the safety of products by acquiring new technologies (Duncan 2011) [1].

Indicators are devices that give some information about the presence or absence of a substance or the degree of interaction between two substances by changing in characteristics, like color. The difference between sensors and indicators is that the latter do not have receptor and transducer components and instead communicate information through direct visual changes (Kuswandi *et al.*, 2011) [2]. Three categories of well known indicators are time temperature, gas and dye based freshness indicators.

Time temperature indicator

Time–temperature indicator (TTI) is a simple quality recording device that is able to show irreversible visual response to monitor and record critical parameters throughout the entire supply chain from production to the storage and distribution including domestic storage (Nuin *et al*., 2008) [3]. Consumers can easily check the quality of food using TTIs, expressing a visible response of colour development that correlates to the shelf life of a food stuff at a target temperature (Kerry *et al*., 2006) [4].

TTIs are applied to reflect the time-temperature history of the chilled and frozen food such as marine food products (Tsironi *et al*., 2008) [5], meat and poultry products (Ellouze and Augustin, 2010) [6]. TTIs have also been applied to assess the pasteurization and sterilization process (Mehauden *et al.*, 2007) [7], to estimate the remaining shelf life of food products (Taoukis, 2001) [8].

TTIs can be sub divided into three types based on the working principle: (1) physical systems, (2) chemical systems, and (3) biological systems. Commercially available TTIs include a number of diffusion, enzymatic, polymer based, solid state reaction and microbiological systems. Diffusion-based TTIs such as Monitor Mark TTI, commercialized by the 3 M Company are based on temperature-dependent diffusion reaction of a coloured fatty acid ester along a porous wick made of high quality blotting paper. Its measurable response is the distance of the advancing diffusion front from the origin.

Polymer-based systems such as the Fresh Check TTI produced by the company Temp Time, are based on solid state polymerization of a thinly coated colourless acetylenic monomer that changes to a highly coloured opaque polymer at a temperature-dependent rate (Nuin *et al.,* 2008) [10]. Solid state reaction systems represented by On Vu™ TTI produced by the Ciba company are based on photosensitive compounds such as benzylpyridines. Once exposed to allow wavelength light, they become coloured and this coloured state reverses to the initial colourless state according to temperature (Tsironi *et al*., 2008) [10].

Enzymatic systems such as the VITSAB Check Point TTI are based on a colour change in the TTI induced by a pH drop resulting from controlled enzymatic hydrolysis of a lipid substrate which changes colour of the chromatic indicator from green over yellow to orange red (Kerry *et al*., 2006; Tsironi *et al*., 2008) [11, 12]. Active Chitosan/PVA Films with anthocyanins from *Brassica oleraceae* (Red Cabbage) was prepared which acted as time-temperature indicators in intelligent food packaging applications (Pareira *et al*. 2014) [13]. Microbiological TTIs are proposed by the French company CRYOLOG. TRACEO and eO are microbiological TTIs made of selected strains of lactic acid bacteria. Prior to utilization, these TTIs are stored in a frozen state (-18°C) to prevent the bacterial growth in the TTI medium. As they are very thin, their activation is obtained simply by defrosting them for a few minutes at the room temperature. Once they are put on the food, and in case of temperature abuse, or when the product reaches its use by date, the temperature-dependent growth of the TTI microorganisms causes a pH drop in the tags leading to an irreversible colour change of the medium chromatic indicator which becomes red (Ellouze *et al*., 2008) [14].

Dye based Indicator/Sensors

Chemical Indicator dyes

Indicators use an indicator dye and a carrier for indicator dye which will hold and carry it. Intelligent packaging applications have been used for monitoring spoilage of meat and to predict its remaining shelf life. A chromogenic sensor array was prepared for monitoring boiled marinated turkey meat freshness using 13 indicators (including pH indicators, nucleophilic sensing dyes, etc.) and three different inorganic supports i.e. UVM-7, alumina and silica gel (Vivancos *et al*., 2014) [15].

Several attempts have also been made for utilizing different chemical dyes used as an indicator solution for the development of an indicator sensor. Mixture of bromothymol blue and phenol red was proposed for fabrication of the indicator sensor responsive to TVBN released from chicken meat during storage (Rukchon *et al*. 2011) [16]. Polyaniline (PANI) film-based chemical sensor was developed for real-time monitoring of microbial breakdown products in the headspace of packaged fish that responded through visible colour change to a variety of basic volatile amines specifically to TVBN released during fish spoilage (Kuswandi *et al*. 2012a) [17]. Bromocresol green was used to monitor the production of TVBN and prediction of fish freshness (Pacquit *et al*. 2007) [18]. A novel on-package sticker sensor based on methyl red as an indicator solution was developed to monitor broiler chicken freshness (Kuswandi *et al*. 2014) [19]. Shukla *et al.,* (2015) [20] used bromophenol blue to develop an indicator sensor based on TVBN for monitoring buffalo meat quality. Kim *et al.,* (2017) [21] developed a colorimetric bromocresol purple dye-based pH-responsive indicator to monitor the quality of chicken breast meat by direct surface contact. Kuswandi and Nurfawaidi (2017) [22] used bromocresol purple and methyl red to construct an on-package dual sensor label for monitoring the beef freshness. Boscher *et al*. (2014) [23] described the detection of volatile amines such as trimethylamine (TMA), triethylamine (TEA) and dimethylamine (DMA) using a novel metalloporphyrin-based coating applied onto PET films.

Natural indicator dyes

Chemical indicator dyes are synthetic chemicals which harmful to the consumers if leaked and are difficult to handle also. Moreover chemical dyes are difficult incorporate in indicator carrier. In recent years different natural dyes have been used for development of indicator sensors. There are many reports of the use of dyes contained in plant tissues for colourimetric determination of pH (Brotto *et al*., 2002; Chigurupati *et al*., 2002; Mohd *et al*., 2011) [24, 25, 26]. It has been found that colour changes in such dyes are due to the presence of phenolic or conjugated substances, such as anthocyanins, which are subjected to structural changes when there is a variation in pH (Shahid & Mohammad, 2013) [27]. Zhang *et al.,* (2014) [28] developed visual pH sensing film using natural dyes from Bauhinia blakeana Dunn to monitor pork and fish freshness.

Some natural pigments from fruits and vegetable sources, anthocyanins for example, have great potential as indicators in intelligent packaging systems because the color expression of anthocyanins is strongly influenced by its structural conformation which is highly influenced by its pH. This color instability of anthocyanins makes these pigments especially useful as a tool to monitor food quality and therefore can be used as an indicator of food spoilage in intelligent packaging systems. Anthocyanin extracted from Ripen black mulberry (*Morus nigra*) was used for developing quality indicator for Monitoring Quality of fresh chicken meat during storage at room temperature (Talukder *et al*., 2017) [29]. Shukla *et al*., (2016) [30] used natural dyes extracted from the flower of rose and red cabbage to develop an indicator sensor based on anthocyanin for monitoring buffalo meat quality. The sensor changes its colour from yellow to orange and then to reddish orange during spoilage. Chitosan based intelligent film mixed with anthocyanin was developed as fast pH–colorimetric device for spoilage detection of food based on pH variation). Curcumin [(1E, 6E)-1,7-bis-(4- hydroxy-3-methoxyphenyl)-hepta-1,6-3dione]-based indicator sensor which showed colour response in reaction to TVBN was developed to monitor the spoilage level in shrimp (Kuswandi *et al*. 2012b) [31]. Active Chitosan/PVA Films with Anthocyanins from *Brassica oleraceae* (Red Cabbage) was developed as Time-Temperature Indicators for Application in Intelligent Food Packaging. A colorimetric pH indicator film was developed using natural dye anthocyanin extracted from purple sweet potato, *Ipomoea batatas* (Choi *et al*. 2017) [32].

Carrier for dye based indicator/sensor

Agarose was used as carrier for myoglobin to detect hydrogen sulphide production during spoilage of unmarinated broiler cuts (Smolander *et al*. 2002) [33]. Different types of papers (untreated cellulose), polyamides, cellulose acetate, gel, foam and resins were used as carriers with most preferred thickness of about 1 mm (Wallach and Hollis 2002) [34]. Polytetrafluoroethylene (PTFE) solid substrate was used to immobilize ammonia-sensitive indicator dye which showed change in spectral characteristics on exposure to volatile acidic and basic compounds (Khalil *et al*. 2010) [35]. Rukchon *et al*. (2011) [36] developed an indicator carrier consisting of methylcellulose as a binder and polyethylene glycol as plasticizers. Bacterial cellulose membrane made from Acetobacter xylinum culture was used as an indicator carrier for curcumin (Kuswandi *et al*. 2012a) [37]. Shukla *et al*. 2015 [38] used filter paper for development of on-package indicator sensor for real-time monitoring of buffalo meat quality during refrigeration storage.

Janjarasskul and Krochta, (2010) [39] stated that biodegradable materials derived from natural resources can be used as interesting potential substitutes for traditional non–biodegradable plastic polymers due to their low cost, easy availability from reproducible resources and biodegradability. As biodegradable films acts as barriers to control the transfer of moisture, oxygen, lipids and flavour, they can be used as self–standing films for indicators.

Kim *et al.,* (2017) [40] used filter paper as indicator coating materials for bromophenol blue and bromocresol purple respectively. Kuswandi *et al*. (2014) [41] developed methyl red/cellulose membrane for determination of broiler chicken. Methyl red was immobilized onto a bacterial cellulose membrane via absorption method. The methyl red/cellulose membrane as a freshness sensor worked based on pH increase as the basic spoilage volatile amines produced gradually in the package headspace, and subsequently, the colour of the sensor changed from red to yellow for spoilage indication, which was visible to the naked eye. Potato starch and agar was used as solid matrices to immobilize natural dyes. Chitosan and anthocyanin based pH-colourimetric indicator was prepared for monitoring pH variations (Yoshida *et al*. 2014) [42].

Gas Sensors

Gas sensors are devices that respond reversibly and quantitatively to the presence of a gaseous analyte by changing the physical parameters of the sensor, and are monitored by an external device (Kerry *et al*. 2006) [43]. An optochemical CO_2 sensor (Borchert *et al*. 2013) [44] which uses a phosphorescentreporter dye and a colourimetric pH indicator showed robust optical responses to CO2. The sensor is designed as film coatings to be applicable in meat packaging. In recent years, a number of instruments and materials for optical oxygen sensing have been described. Such sensors are usually comprised of a solid-state material, which operate on the principle of luminescence quenching or absorbance changes caused by direct contact with the analyte. These systems provide a non-invasive technique for gas analysis through translucent materials and as such are potentially suitable for intelligent packaging applications. The solid-state sensor is inert and does not consume analyte or undergo other chemical reactions. Optochemical sensors have the potential to enhance quality control systems through detection of product deterioration or microbial contamination by sensing gas analytes such as hydrogen sulphide, carbon dioxide and amines.

Conclusion

In the food sector, one of the most important problems is the time-consuming and laborious process of food quality-control analysis. Innovative devices and techniques are being developed that can facilitate the preparation of food samples and their precise and inexpensive analysis. From this point of view, the development of colorimetric sensors to detect spoilage is a particularly promising application of food microbiology.

Intelligent packaging applications in the meat industry are still limited. It could be expected that the continuous advances in biotechnology, analytical chemistry, microelectronics, and materials science will contribute to the development of new intelligent packaging solutions. The growing need for information on packaging points to a step change in the way this information is provided, driving the need for smart packaging use, particularly for food products. Consumers increasingly need to know what ingredients or components are in the products and how they should be stored and used. Another important need is consumer safety assurance, particularly for perishable food products. The question is whether, for instance, a chilled ready-meal is safe to use or consume. Currently the answer to this question is the 'best by' date stamping. However, this does not take into account whether the product has inadvertently been exposed to elevated temperatures during storage or transportation. In the future, the use of smart packaging in food products will give a clear, accurate, and unambiguous indication of product quality, safety, and shelf-life condition.

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