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An overview of the intelligent packaging technologies in the food sector

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Structured Abstract

Background

Intelligent packaging is the newest technology within the food packaging field. Even though this technology is still growing and not fully commercially viable, it has enormous potential to improve the safety, quality, and traceability of food products, as well as its convenience for consumers.

Scope and Approach

This paper first describes both the technical aspects and commercial applications of the most representative intelligent technologies—indicators, data carriers, and sensors—with special focus on systems and devices that are directly integrated into the package. Secondly, to provide useful guidelines for future research in the field, the paper discusses some important aspects that still hinder the full exploitation of intelligent technology within the food packaging industry.

Key Findings and Conclusions

Future research needs to consider some important aspects in order to make intelligent systems commercially viable, such as cost, consumers' acceptance and confidence, regulatory aspects (e.g., labeling), and multifunctionality.

Keywords: barcode; consumer perception; indicator; legislation; sensor; RFID

Introduction

Packaging is one of the main processes to preserve the quality of food products for transportation, storage, and end use. It slows quality decay and makes distribution and marketing more efficient. Packaging has four basic functions: protection, communication, convenience, and containment (Han, 2005a). Packages protect products from the external environment; communicate with the customer through written texts, brand logo, and graphics; accommodate the lifestyle of the customer, for example saving time (ready-to-eat and heat-and-eat meals) or making the manipulation and handling of packaged food easier for the customer (examples of convenient features are easy opening, resealability, and microwavability); and act as containers for differently shaped and sized products, with the goal of optimizing logistic efficiency (Yam & Lee, 2012). Secure delivery and the preservation of packaged foods before consumption are the main goals throughout the food supply chain. However, loss of food quality attributes occurs during distribution and storage due to biological, chemical, and physical degradation (Han, 2013).

Food quality preservation is an important research target because it is intimately linked to the more global goal of enhancing the quality of our lives (Sandulescu, Cristea, Harceaga, & Bodoki, 2011). Food quality control is necessary, both to better protect consumers against foodborne illness and to maximize the efficiency of the food industries, e.g., by reducing losses due to the microbial spoilage of perishable foods. At the company level, food quality is usually assessed by periodic microbiological and chemical analyses underlying routine tests (Viswanathan & Radecki, 2008).

Of late, two new concepts have greatly contributed to achieving an advanced concept of packaging for safer and healthier food: the active packaging and intelligent packaging concepts. Active packaging materials are “materials and articles that are intended to extend the shelf-life or to maintain or improve the condition of packaged food. They are designed to deliberately incorporate components that would release or absorb substances into or from the packaged food or the environment surrounding the food" (European Commission, 2004). To improve the functionality of food packages and give them additional functions, different active substances can be incorporated into the packaging material (Singh, Abas Wani, & Saengerlaub, 2011). Several active packaging systems have been widely reported, such as O_2 and ethylene scavengers, moisture regulators, CO_2 scavengers and emitters, antioxidant and antimicrobial controlled-release packages, and devices to control the release or adsorption of flavors and odors (Vermeiren, Devlieghere, van Beest, de Kruijf, & Debevere, 1999).

Intelligent packaging materials are "materials and articles that monitor the condition of packaged food or the environment surrounding the food" (European Commission, 2004). In a broader meaning, intelligent packaging is defined as science and technology that use the packaging system's communication function to facilitate decision making by monitoring changes in the internal and external environments and communicating the conditions of the packaged food product (Yam, 2012). Differently from active packaging systems, intelligent packaging does not directly act to extend the shelf life of foods. Rather, intelligent packaging aims to convey information to the stakeholders of the food supply chains (e.g., manufacturers, retailers, and consumers) related to the food's quality (Restuccia et al., 2010). For example, an intelligent packaging system can show when the food product is fresh or whether its shelf life has expired; it can show the food's temperature using thermochromic inks or microwave doneness indicators (MDIs); and it can display the food's temperature history using time-temperature indicators (TTIs) (Robertson, 2012). Additionally, intelligent packaging can be used to check the effectiveness of active packaging systems (Kerry, O'Grady, & Hogan, 2006). In other words, active packaging is the component that takes some action, while intelligent packaging is the component that senses and shares the information (Yam, Takhistov, & Miltz, 2005). Intelligent packaging and active packaging can work synergistically to yield what is defined as "smart" packaging, i.e., a total packaging concept that combines the benefits arising from active and intelligent technology (Vanderroost, Ragaert, Devlieghere, & De Meulenaer, 2014).

Over the last decade, the research interest in intelligent packaging lagged far behind the interest in active packaging, as demonstrated by the number of related publications (Figure 1). An explanation for this trend can be twofold. On one hand, researchers first focused on new tools to improve the quality and safety of foods through new, "active" functions, in contrast to the original "passive" attributes, such as mechanical strength, barrier performance, and thermal stability. Intelligent packaging systems have come to the forefront because of the growing usage of active components in food packaging, which requires a means of monitoring both the active device's performance and the overall packaging conditions. On the other hand, the higher complexity needed to achieve a sophisticated, intelligent system that is simultaneously reliable, efficient, and cost effective represented a hurdle to the development of intelligent devices. Moreover, the development of such devices requires different technical skills and backgrounds to be merged, e.g., food science, materials science, and chemical and electrochemical engineering, which makes the overall design and development process more complex.

Due to the increasing interest in intelligent developments, which have been forecast as the main food packaging innovations in the next years (Kuswandi, Wicaksono, Jayus, Abdullah, Heng, & Ahmad, 2011; Vanderroost et al., 2014), this paper summarizes the intelligent packaging applications in the food sector, with special reference to the applications designed to integrate intelligent devices into packages. Therefore, instruments developed based on an intelligent technology (e.g., nose and tongue systems), as well as Internet-based technologies (e.g., *Internet of everything*, IoE), are out of the scope of this review. The focus is on the technical aspects and market applications of three main categories of intelligent systems, namely indicators, data carriers, and sensors. The last part of the work provides guidance for tomorrow's research in the field, with the goal of covering some important aspects that still hinder the full exploitation of intelligent systems for food quality and safety.

Intelligent systems in food packaging

The term "intelligent" involves an "ON/OFF" switching function on the package in response to changing external/internal stimuli, in order to communicate the product's status to its consumers or end users (Yam et al. 2005). In practice, an intelligent packaging system is manufactured by incorporating an external, discrete component in the final package, e.g., two-dimensional films or three-dimensional objects. It is widely accepted that intelligent packaging systems can be realized by three main technologies: (i) indicators, which aim to provide more convenience and/or to inform consumers about the food quality; (ii) data carriers, such as barcodes and radiofrequency identification tags (RFID), which are specifically intended for storage, distribution, and traceability purposes; and (iii) sensors, which allow for a rapid and definite quantification of the analytes in foods (Kerry et al., 2006).

Indicators

Indicators convey information to the consumer that is linked to the presence or absence of a substance, the extent of a reaction between two or more substances, or the concentration of a specific substance or class of substances. Most often, such information is displayed by immediate visual changes, e.g., different color intensities or the diffusion of a dye along the indicator geometry (O'Grady & Kerry, 2008). A distinct feature of indicators is the type of information involved, which is qualitative or semi-quantitative in nature. Despite the large varieties of indicators, all of them can be reasonably included within three categories: time-temperature indicators, freshness indicators, and gas indicators (Hogan & Kerry, 2008). All of them fall within the main category of "product quality and value-improving systems," which are undoubtedly the most widely used devices for food packaging applications (Robertson, 2012).

Temperature indicators

There are two types of temperature indicators: simple temperature indicators and time–temperature integrators (TTIs) (Ahvenainen & Hurme, 1997). Temperature indicators show whether products have been heated above or cooled below a reference (critical) temperature, warning consumers about the potential survival of micro-organisms and protein denaturation during, for example, freezing or defrosting processes (Pault, 1995). TTIs, sometimes also called integrators, are the first generation of indicators intended to monitor any detrimental change in temperature change (e.g., above or below a reference critical value) along the food supply chain, i.e., over time. The basic operating principle is based on mechanical, chemical, electrochemical, enzymatic or microbiological change, usually expressed as a visible response in the form of a mechanical deformation, colour development or colour movement (Taoukis & Labuza, 2003). Due to the pivotal role of both time and temperature in influencing the kinetics of physical and chemical deterioration, TTIs have gained increasing interest for acquiring information about the temperature history of a packaged food over time, thus preventing any sort of abuse and/or misuse. TTIs are recognized as user-friendly and readily usable devices, whose information is readily understood by consumers as being directly related to the quality of the food item at a certain temperature (Pereira Jr, de Arruda, & Stefani, 2015). Usually, they consist of small, self-adhesive labels attached to single packages or larger configurations (e.g., containers).

TTIs' market applications include Monitor Mark™ by 3M (USA), Fresh-Check® by Lifelines Technologies Inc. (USA), CoolVu™ and OnVu™ by Freshpoint (Switzerland), Checkpoint® by Vitsab International AB (Sweden), Tempix® by Tempix AB (Sweden), Timestrip® by Timestrip Plc (UK), Smartpak® by Trigon Smartpak Ltd (UK), and Insignia Cold Inspection Intelligent Labels™ by Insignia Technologies Ltd (UK) (Han, Ho, & Rodrigues, 2005b; Kuswandi et al., 2011) (Figure 2). Several other examples of TTIs are still in the laboratory stage (Table 1). These devices are especially suited for warning of temperature abuses of frozen or chilled food products (Yam, Takhistov, & Miltz, 2005).

Freshness indicators

The development of freshness indicators over the last two decades stemmed from increasing consumer demand for healthy and fresh foods. Freshness indicators have to be intended as smart devices that enable the monitoring of the quality of food products throughout storage and transportation. Freshness decay may be due to both exposure to detrimental conditions and exceeded shelf life. Freshness indicators provide direct information on the product's quality regarding microbial growth or chemical changes (Siro, 2012). For example, freshness indicators intended for seafood are based on the total volatile basic nitrogen content (TVB-N), i.e., volatile amines, which are formed as the food spoils and can be detected by different methods, such as conductometric (Heising, van Boekel, & Dekker, 2015) and pH variations (Kuswandi, Jayus, Oktaviana, Abdullah, & Heng, 2014). Hydrogen sulfide indicators can be used to determine the quality of meat products. Hydrogen sulfide, which is released by the meat matrix during aging, is correlated with the color of myoglobin, which is considered a quality attribute for meat products. Smolander et al. developed a freshness indicator based on this principle for modified-atmosphere packed poultry meat (Smolander et al., 2002). Other freshness indicators are based on sensitivity toward other microbial metabolites, such as ethanol, diacetyl, and carbon dioxide (Pereira de Abreu, Cruz, & Paseiro Losada, 2011). Commercial applications of freshness indicators include Toxinguard® by Toxin Alert Inc., to monitor *Pseudomonas* sp. growth, and SensorQ™ by FQSI Inc., which senses spoilage in fresh meat and poultry products (O'Grady, & Kerry, 2008). The ripeness indicator RipeSense™ allows consumers to choose fruit that best appeals to their tastes (Pocas, Delgado, & Oliveira, 2008) by detecting aroma components or gases involved in the ripening process (e.g., ethylene) released by the fruit.

Gas indicators

Gas concentration indicators, in the form of labels, are placed inside the package to monitor changes in the inside atmosphere due to permeation phenomena across the packaging material, microorganisms metabolism, and enzymatic or chemical reactions on the food matrix (Yam et al., 2005). Gas indicators are also used to either assess the efficacy of active packaging components (e.g., O_2 and CO_2 scavengers) or to detect the occurrence of leakages. Because the indicators are placed inside the package, some requirements must be met during the design of these devices, such as being non-water soluble and non-toxic (these components must have food contact approval) (Mills, 2005). The most widely known gas indicators are used to check oxygen and carbon dioxide concentrations. Due to the importance of these gases in food applications, much research has been devoted to the development of O_2 and CO_2 indicators over the last decade and in more recent years (Roberts, Lines, Reddy, & Hay, 2011; Jung, Puligundla, & Ko, 2012; Lee & Ko, 2014; Vu & Won, 2014). Most devices are based on redox dyes (e.g., methylene blue, 2,6-dichloroindophenol, or N,N,N',N'-tetramethyl-*p*-phenylenediamine), a reducing compound (e.g., reducing sugars), and an alkaline compound (e.g., sodium hydroxide) (Kuswandi et al., 2011). However, such indicators suffer from dye leaching upon contact with the moisture in the package's headspace. The latest developments concern UV-activated colorimetric oxygen indicators (Lee, Mills, & Lepre, 2004; Lee, Sheridan & Mills, 2005; Roberts et al., 2011) with very limited dye leaching due to either encapsulation or coating technologies (Mills, Hazafy, & Lawrie, 2011; Thai Vu & Won, 2013).

Trade names for several commercial applications include Ageless Eye™ by Mitsubishi Gas Chemical Co., Shelf Life Guard by UPM, Vitalon® by Toagosei Chemical Inc., Tufflex GS by Sealed Air Ltd., and Freshilizer by Toppan Printing Co (Rodrigues & Han, 2003; O'Grady & Kerry, 2008).

Data carriers

Data carrier devices, also known as automatic identification devices, make the information flow within the food supply chain more efficient, to the advantage of food quality and safety. More specifically, data carrier devices do not provide any kind of information on the quality status of food but are rather intended for automatization, traceability, theft prevention, or counterfeit protection (McFarlane & Sheffi, 2003). Moreover, data carriers are more often placed onto tertiary packaging (e.g., multi-box containers, shipping crates, pallets, large paperboard packages). The most important data carrier devices in the food packaging industry are barcode labels and RFID tags, which belong to the main category of convenience-enhancing intelligent systems (Robertson, 2012).

Barcodes

The first Universal Product Code (UPC) barcodes found market application in the 1970s. Due to their low cost and ease of use, barcodes have been increasingly used in the large-scale retail trade and stores to facilitate inventory control, stock reordering, and checkout (Manthou & Vlachopoulou, 2001). A barcode is a pattern of parallel spaces and bars arranged to represent 12 digits of data. The encoded information is read by an optical barcode scanner that sends the information to a system where it is stored and processed (Han, 2013). One-dimensional (1-D) barcodes were developed first. The basic working principle is the same of a laser beam cutting a horizontal slice from the vertical code bars. As the beam moves over the symbol (see Figure 3a), it measures the relative time it spends scanning dark bars and light spaces. A lookup table is then used to decode individual characters from those times. Because of the line of the laser beam, these kinds of barcodes are referred to as being 1-D. The storage capacity of the first-generation barcode labels was limited, such as to the manufacturer identification's number and the item number (Robertson, 2012; Drobnik, 2015). Reduced Space Symbology (RSS) barcodes were developed successively to encode more data in a smaller space. The most frequently used RSS symbologies are the RSS-14 stacked omnidirectional barcode and the RSS expanded barcode, the latter encoding up to 74 alphanumeric characters (Yam, Takhistov, & Miltz, 2009).

Two-dimensional (2-D) barcodes (Figure 3b) allow a larger amount of information to be stored, compared to 1-D barcodes, by combining dots and spaces arranged in an array or a matrix, instead of bars and spaces. This allows for an increased density of data within a reduced space. For example, the Portable Data File (PDF) 417 is a 2-D symbol that carries up to 1.1 kB of data in the space of a UPC barcode (Yam et al., 2009). The more recent Quick Response (QR) 2-D barcode (Figure 3c) enables an even larger amount of data to be stored using four different encoding modes: numeric, alphanumeric, byte/binary, and kanji, the latter referring to logographic Chinese characters. Reading 2-D barcode symbologies requires a scanning device capable of simultaneous reading in two dimensions—vertically as well as horizontally (Kato, Tan, & Chai, 2010).

Radio-frequency identification systems

RFID tags are the most advanced example of a data carrier device. An RFID system includes three main elements: a tag formed by a microchip connected to a tiny antenna; a reader that emits radio signals and receives answers from the tag in return; and middleware (a local network, web server, etc.) that bridges the RFID hardware and enterprise applications (Kumar, Reinitz, Simunovic, Sandeep, & Franzon, 2009; Sarac, Absi, & Dauzère-Pérès, 2010). Two distinct features of RFID technology are the high number of various codes that can be stored in the tag and the possibility of transferring and communicating information even at a long distance, thus improving automatic product identification and traceability operations (Plessky, 2009). Most advanced RFID systems (2.45 GHz—super high frequency active tags) have a reading range of up to 100 meters, with up to 1 MB data in storage capacity. Nowadays, RFID technology includes two types of tags: active and passive tags; the main difference is that active tags rely on a battery while passive tags do not. Table 2 provides a full comparison between the two types of tags.

While RFID technology was well known for a long time, the market penetration of these devices has lagged behind barcodes, mainly due to cost reasons (Preradovic & Karmakar, 2012). However, RFID technology should not be considered as a replacement for barcodes. Because of important differences between the two systems (Table 3), which are ultimately reflected in advantages and disadvantages that depend on the application, they will continue to be used, either alone or in combination. Current applications of RFID tags, aside from traffic control, pallet identification, building security, parking guidance, and the tracing and identification of animals, also have different applications in the food industry, such as product identification and traceability (Hwang, Moon, & Yoo, 2015), cold chain monitoring (Badia-Melis, Ruiz-Garcia, Garcia-Hierro, & Villalba, 2015), livestock management (Ariff, Ismarani, & Shamsuddin, 2014), and shelf life prediction (Uysal, Emond, & Bennett, 2011).

Sensors

Sensors are considered the most promising and innovative technology for future intelligent packaging systems (Kuswandi et al., 2011; Bagchi, 2012). A sensor is a device or system with control and processing electronics, an interconnection network, and software (Patel & Beveridge, 2003). A sensor is used to detect, locate, or quantify energy or matter, by giving a signal for the detection or measurement of a physical or chemical property to which the device responds (Kress-Rogers, 1998). In practice, a sensor replies to a chemical or physical quantity to make a quantifiable output that is proportional to the measure. Most sensors are made up of four major components (Scheme 1). (i) The first is a receptor, i.e., the sensing part of the sensor, represented by a sampling area (generally a chemo-selective coating) where the surface chemistry occurs. Here, the analytical information is obtained from the adsorption of the target analyte on the recognition layer. The energy variation associated with detecting the analyte induces a change of a property of the receptor in terms of, for example, redox potential, pH, temperature, or light. (ii) The second is the transduction element, i.e., the measuring part of the sensor (e.g., an electrode), which is capable of transforming the energy variation and carrying the physical or chemical information into a useful analytical signal (e.g., electrical, optical, thermal, or chemical). Next are (iii) the signal processing electronics, and (iv) a signal display unit (Neethirajan, Jayas, & Sadistap, 2009). The ideal sensor should possess the following characteristics (Hanrahan, Patil, & Wang, 2004): (i) specificity for the target species (i.e., selectivity); (ii) sensitivity to changes in target-species concentrations; (iii) fast response time; (iv) extended lifetime of at least several months; and (v) small size (miniaturization), with the possibility of low-cost manufacture.

In recent years, different kinds of sensors intended for food applications have been developed, such as electrochemical sensors (Goulart, Cruz de Moraes, & Mascaro, 2016; Feng Gao et al., 2015; Liu, Xiao, Cui, & Wang, 2015; Pacheco et al., 2015; Nasirizadeh, Hajihosseini, Shekari, & Ghaani, 2015) and luminescence sensors (Fan, Shen, Wu, Wang, & Zhang, 2015; Pénicaud, Guilbert, Peyron, Gontard, & Guillard, 2010). Electrochemical sensors represent an important subclass of chemical sensors, in which an electrode is used as the transduction element. The working principle of electrochemical sensors is based on redox reactions that take place at the electrode/analyte interface upon applying a voltage by means of a potentiostat. The electrons transfer between electrode and electroactive species gives origin to a current that is proportional to the concentration of the analyte (Wang, 2006). In luminescence sensors the emitted fluorescence, phosphorescence or chemiluminescence signals are measured after the analyte is immobilized in a suitable solid support, giving origin to the expression solid-phase luminescence (SPL) or to its equivalent solid-matrix luminescence (SML). Under certain conditions, these analytical signals can be related to the concentration of analyte in the sample (Ibañez & Escandar, 2011).

However, most of these developments concern the detection of food components and contaminants in food matrices. Although flexible printed chemical sensors integrated into food packages have a promising future (Vanderroost et al., 2014), most advanced sensor technologies that can incorporate intelligent devices into packaging belong to two main groups: biosensors and gas sensors.

Biosensors

The main difference between chemical sensors and biosensors lies in the recognition layer. While in chemical sensors the receptor is a chemical compound, the recognition layer of biosensors is made of biological materials, such as enzymes, antibodies, antigens, phages, and nucleic acids (Wang, 2006). Current uses of biosensor systems integrated into packaging are limited to a few examples. SIRA Technologies (USA) has developed the Food Sentinel System, a packaging barcode technology that can alert consumers and retailers when a product has been exposed to adverse conditions, thus affecting its safety. The technology is based on a biosensor carrying an antibody of a specific pathogen, in the form of a membrane attached to the barcode. In the presence of contaminating bacteria, an ink incorporated into the biosensor will turn red, and the barcode will be rendered incapable of transmitting data when scanned (Yam et al., 2005). Toxin Guard™ (Toxin Alert Inc., USA) is a visual diagnostic tool used to detect pathogens or other selected micro-organisms that may contaminate food, such as *Campylobacter* spp., *Escherichia coli* O157, *Listeria* spp., and *Salmonella* spp. The Toxin Guard™ immunoassay works based on antibody–antigen reactions on polymer packaging films: in the presence of pathogenic bacteria, the bacterial toxin is bound to the antibodies and immobilized on a thin layer of flexible polymer film (e.g., polyethylene, PE), yielding a clear change in the color of the smart device (Han, 2013). Bioett (Bioett AB, Sweden) is a system technology that combines biochemistry and electronics to monitor the temperature of foods during refrigerated transport. The system consists of a biosensor attached to the food package, a detector reading the data from the biosensor, and a database to store information about the goods. The main parts of the Bioett system are a chip-less RF circuit and a built-in biosensor. At different points in the supply chain, this sensor can be read using a handheld scanner (Hogan & Kerry, 2008). Flex Alert (Canada) developed commercially available flexible biosensors to detect toxins in packaged foods throughout the supply chain. Flex Alert biosensors have been specifically developed against *Escherichia coli* O157, *Listeria* spp., *Salmonella* spp., and aflatoxins (Vanderroost et al., 2014).

Gas sensors

The development of sensors that can respond quantitatively and reversibly to gaseous analytes has been a fervid research field during the last two decades. Established systems for gas detection include metal oxide semiconductor field-effect transistors (MOSFETs), piezo-electric crystal sensors, amperometric oxygen sensors, organic conducting polymers, and potentiometric carbon dioxide sensors. However, these systems exhibit various limitations, such as cross-sensitivity to carbon dioxide and hydrogen sulfide, fouling of sensor membranes, and consumption of the analyte (e.g., oxygen), and these systems involve destructive analyses of packages in most cases (Kerry et al., 2006). More recent developments have especially focused on new O_2 and CO_2 sensors, with the aim to overcome these drawbacks.

The development of smart sensors to quantify oxygen permeating across the package has been a main research topic over the last two decades, as demonstrated by the number of works published in the literature and the instruments and devices in the market. Oxygen sensors are based on luminescence detection and represent an alternative approach to purely visual oxygen indicators, providing higher sensitivity and accuracy of the quantitative measurements, compared to systems based on absorption or reflectance (MacCraith et al., 1993). Distinct features of these systems include the possibility of carrying out the measurements on 3-D samples with non-destructive experiments. Moreover, they offer fast responses, do not consume any analyte, and lack electrical connections. Huber et al. proposed a new non-destructive and non-invasive fiber-optic oxygen meter to quantify the oxygen permeability of containers and plastic bottles (Huber, Nguyen, Krause, Humele, & Stangelmayer, 2006). The principle of the sensor's operation is based on the quenching of luminescence caused by the collision between molecular oxygen and luminescent dye molecules in the excited state. Oxygen determination (i.e., oxygen partial pressure) can take place in both solutions (dissolved oxygen) as well as in the gaseous phase. No cross-sensitivity exists for carbon dioxide, hydrogen sulfide, ammonia, pH, or any ionic species such as sulfide, sulfate, or chloride, and the measurement is not affected by salinity. Turbidity and changes in the stirring rate have no influence on the measurement. The sensors can also be used in methanol– and ethanol–water mixtures, as well as in pure methanol and ethanol. In another paper using an optical oxygen sensor, Fitzgerald et al. fabricated a phosphorescence lifetime-based oxygen sensor made of fluorescent complexes of ruthenium(II) and platinum(II)-octaethylporphiryne-ketone (PtOEPK) dye (Fitzgerald et al., 2001). The authors first demonstrated that the sensor allows for efficient and sensitive measurements of oxygen in food packages, besides being non-destructive. They also tested the sensor on real samples, such as packaged sliced ham, smoked fish, and raw and cooked meat, demonstrating that it provides accurate and reliable results even on real samples, which is a requirement for market applications of the sensor.

Baleizao et al. presented an optical dual sensor for oxygen and temperature (Baleizao, Nagl, Schaferling, Berberan-Santos, & Wolfbeis, 2008). The sensor is based on luminescence lifetime measurements and is highly sensitive to oxygen, while covering a very wide temperature range. The sensor contains two luminescent compounds incorporated into polymer films, one sensitive to temperature and the other sensitive to oxygen. Due to its highly temperature-dependent luminescence, Ruthenium tris-1,10-phenanthroline was used as the temperature-sensitive dye and is incorporated in poly(acrylonitrile) to avoid cross-sensitivity to oxygen. The oxygen-sensitive probe used is fullerene C_{70}, due to its strong, thermally activated, delayed fluorescence at high temperatures and its exceptional oxygen sensitivity. The dual sensor exhibits a temperature operation range between at least 0 and 120 °C, as well as detection limits for oxygen in the ppbv range, operating for oxygen concentrations up to at least 50 ppmv.

The development of CO_2 sensors for food packaging applications has lagged behind that of O_2 sensors because of oxygen's role as a primary factor in the degradation of many foods. However, especially since the use of MAP packaging systems was established, controlling the amount of CO_2 in packages has become just as important for both shelf life and freshness studies (Fu, Molins, & Sebranek, 1992). Conventional techniques for a quantitative and qualitative analysis of CO_2 include Severinghaus-type electrodes, infrared (IR) spectroscopy, gas chromatography (GC), and mass spectroscopy (MS). However, these techniques suffer from a series of drawbacks: instruments are often expensive, bulky, and not particularly robust; require long pathlengths; are prone to interference; lack mechanical stability; and require rather sophisticated equipment (Mills & Eaton, 2000; Sipior, Randers-Eichhorn, Lakowicz, Carter, & Rao, 1996; Schulz, Jensen, Balsley, Davis, & Birks, 2004). For this reason, great effort has been made over the last 20 years to fabricate sensitive, robust, fast, cheap, flexible, and easily miniaturized sensors to detect CO_2. Von Bultzingslowen et al. developed an optical sensor to measure carbon dioxide in modified atmosphere packaging (MAP) applications (von Bultzingslowen et al., 2002). This sensor is based on the fluorescent pH indicator 1-hydroxypyrene-3,6,8-trisulfonate (HPTS) immobilized in a hydrophobic, organically modified silica (ormosil) matrix obtained by sol-gel chemistry. The authors showed that oxygen cross-sensitivity is minimized (0.6% quenching in air) by immobilizing the reference luminophore in polymer nano-beads. Moreover, cross-sensitivity toward chloride and pH was found to be negligible.

Borisov et al. developed optical carbon dioxide sensors based on an emulsion of room-temperature ionic liquids (RTILs)–1-butyl-3-methylimidazolium salts in a silicone matrix (Borisov, Waldhier, Klimant, & Wolfbeis, 2007). In particular, for the quantitative determination of CO_2, they used 8-hydroxypyrene-1,3,6-trisulfonate (HPTS) to prepare a fluorimetric sensor, which was claimed to have potential applications in several fields, such as food packaging technology. Borchert et al. developed an optochemical CO_2 sensor that includes a phosphorescent reporter dye PtTFPP and a colorimetric pH indicator α-naphtholphthalein incorporated in plastic matrix, together with a phase transfer agent—

tetraoctyl- or cetyltrimethylammonium hydroxide (Borchert, Kerry, & Papkovsky, 2013). Experiments were carried out to optimize the composition and working characteristics of such a sensor in order to measure headspace CO_2 in foods packaged under a modified atmosphere. The authors demonstrated that in food and modified atmosphere environments, the sensor retained its sensitivity to CO_2 for 21 days at 4 °C, which is sufficient for many packaged products.

Other intelligent packaging systems

Additional intelligent devices that have found fewer applications compared to the aforementioned systems include doneness indicators and thermochromic ink convenience-enhancing–type systems (Robertson, 2012). Thermochromic inks are based on thermosensitive inks printed on the package, e.g., onto shrink sleeves of beverage cans. The color of the ink changes when the temperature is within a specific pre-set range that is best for food consumption. In some cases, the color change is accompanied by a simultaneous display of a short message, such as “ready to serve.” Thermochromic inks are produced by several companies, such as LCR Hallcrest (U.S.A.), CTI Inks (USA), QCR Solutions Corp. (USA), Siltech Ltd. (UK), and B&H Colour Change (UK). Based on the same principle, doneness indicators inform the consumer when heated food is ready. One of the main drawbacks of doneness indicators is the difficulty of observing the color change distinctly, especially when the oven is still closed (Robertson, 2012). Another type of intelligent device is represented by systems intended to tackle theft, counterfeiting, and tampering. Although not very common in the food industry, these systems are drawing increasing interest, especially as a means of containing the economic burden posed by the aforementioned threats. Electronic article surveillance (EAS), in the form of electronic tagging systems, is an example of systems against theft, whereas anti-counterfeiting and anti-tampering devices take the form of holograms, thermochromic inks, micro-tags, tear labels, and tapes (Han, Ho, & Rodrigues, 2005c).

Market and legislative considerations

Besides historical and technical factors, the commercial application of intelligent systems in the food packaging industry has had to face (and still does face) some important considerations. Consumers' perceptions and legislative aspects, in particular, are key factors.

One of the main issues that hinder the market penetration of intelligent devices in food packaging is consumers' acceptance of non-edible items separate from the package. Sachets, inserts, spots, and dots are sometimes thought to be unnecessary, i.e., the benefit of intelligent systems is still unclear. In other circumstances, consumers are worried that innovative packages might mislead them regarding the product's quality (Day, 2008; Vanderroost et al., 2014). In more recent years, retailers have reconsidered the use of intelligent systems for two main reasons: (i) alerts and messages provided by the intelligent devices (e.g., indicators) can push consumers to buy only newly displayed items, leading to an increased amount of unsold foodstuffs (Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback 2008); and (ii) some devices (e.g., TTIs) might display temperature abuses that occurred before the food reached the retailers' shelves. However, unambiguously identifying the failing step (and thus the responsibility for that abuse) in the supply chain might be difficult.

From a legislative point of view, the lack of an adequate regulatory framework in the EU for intelligent (and active) packaging systems until 2004 hindered the placement of new packaging solutions into the market, in contrast to the United States, Australia, and especially Japan, where intelligent packaging systems are widespread. In fact, the lack of a clear regulatory framework for many years led to reluctance by food packaging manufacturers to take on new concepts that are not fully covered by the legislation on food contact materials.

The first EU legislative attempt to address the topic of active and intelligent materials was Framework Regulation EC 1935/2004 (European Commission, 2004), which describes the general requirements for all food contact materials. In particular, Article 3 covers stipulations on the packaging material containing the intelligent component, e.g., “the packaging material shall not transfer constituents to food in quantities that could endanger human health, bring about an unacceptable change in the composition of foods, or bring about deterioration in organoleptic characteristics thereof” (European Commission, 2004). Article 4 refers to the intelligent component, dealing with some issues in particular: “intelligent materials shall not give information about the food’s condition that could mislead the consumer, adequate labelling must allow non-edible parts to be identified, and adequate labelling must indicate that the materials are active and/or intelligent” (European Commission, 2004). Finally, Article 15 clearly states that “consumers and food packaging companies must be informed on how to use the active and intelligent materials and articles safely and appropriately” (European Commission, 2004). Although useful, the Framework Regulation of 2004 can have multiple interpretation (Dainelli et al., 2008).

The Good Manufacturing Practice (GMP) represented a legislative implementation regarding materials and articles intended to come in contact with food (Regulation EC 2023/2006). It aims to ensure that these materials do not transfer into foods (in a process called migration) in unacceptable quantities (European Commission, 2006).

However, the only specific regulation entirely devoted to intelligent (and active) materials intended for food packaging applications is Regulation 450/2009, which sets out specific requirements on the use and authorization of active and intelligent materials and articles intended to come into contact with food. The regulation also establishes an EU-wide list of substances that can be used in manufacturing these materials; substances may only be added to the list once their safety has been evaluated by the European Food Safety Authority

(EFSA) (European Commission, 2009). In addition, this regulation introduces an authorization scheme for substances used for active and intelligent functions in food contact materials. Article 11 of the regulation also states that, in order the consumer to identify non-edible parts, active and intelligent materials and articles or parts thereof shall be labelled, whenever they are perceived as edible, (i) with the words “DO NOT EAT” and (ii) always, where technically possible, with a specific symbol.

Although the issue related to the potential migration concerns mainly active packaging systems, the risk associated to the unintended release/contact of certain substances/materials also includes intelligent packaging devices, especially when they are positioned inside the primary packaging. Consumers’ reluctance toward intelligent packaging devices is often associated to the potential risk of leaching of active components (e.g., inks) from the device, or swallowing of the sachet. This may be the case of intelligent systems including water soluble components that are susceptible to leaching upon direct contact with foods with a high moisture content (Mills, 2009). At least in Europe, the perception of this risk by consumers seem to be higher for separate non-edible objects (e.g., sachets and inserts) compared to structures that are incorporated/attached to the package (e.g., labels) (Han, 2005c; Lee, Yam, & Piergiovanni, 2008). Therefore, preserving the integrity of intelligent components inside the package throughout the shelf life of the food plays a role to minimize any potential safety issue while increasing the consumers’ trust toward this technology.

Concluding remarks

The interest in innovative packaging systems to achieve higher food quality and safety, consumer convenience, and management (i.e., storage, distribution, and traceability) along the food supply chain has boosted the development of intelligent devices, in the form of labels, tags, dots, and inks that perform different functions. Although the potential advantages arising from such technologies have been widely explored and documented, there is still an existing gap in market applications. For this reason, future research needs to consider some important aspects in order to make intelligent systems commercially viable and, ultimately, into everyday packaging commodities. For instance, the final cost of intelligent packaging systems should account for a minimal part of the whole packaging cost. Due to the technology involved, the cost attributed to intelligent devices is estimated to be ~ 50–100% of the whole cost of the final package. However, for most food products the packaging cost should not exceed 10% of the total cost of the goods placed on the shelves, provided that the claimed benefits are unambiguously demonstrated to outweigh the possible extra expenses arising from the new technology. This mismatch between the new technology and market penetration eventually results into a negative cost/benefit analysis (Dainelli et al., 2008).

Concurrently, technological advancement is requested. For example, especially the companies providing the technology leading to these materials claim improvements in efficiency and performance of the intelligent materials. The main criticism arises from the discrepancy between the results obtained within model tests and real foods. The complexity of real food systems (e.g., different quantity of foodstuffs packed, ratio and distribution of fat and non-fat parts, fluctuation and variability of physical and chemical parameters such as water activity, pH etc.) has been indicated has the main reason for the decrease in activity of the intelligent materials compared to *in vitro*/lab scale trials (Dainelli et al., 2008). However, intelligent materials may need a demonstration of the reliability of the information provided, especially to avoid misleading the consumer (Rijk, 2008). As an example, the use of a freshness indicator that has lower capacity to monitor and alert about a certain microbial growth may mislead and even endanger consumers' health.

Another technical goal for the future is the integration of several functions within only one device (multi-functional intelligent packaging), as well as the development of new functions, e.g., systems able to communicate the presence of potential allergens, warnings related to diet management, and error prevention alerts. In particular, advances in biosensors and biotechnology applied to food packaging systems are expected (Han, 2005c).

Equally important is to educate consumers on the extra benefits arising from intelligent systems. This can be achieved using clear information about the device, e.g., what purpose it serves, how it works, and how to use it.

Intelligent devices also need to be adequately labeled, in order to increase consumers' confidence in the safety of packaged food. Packaging manufacturers must also consider regulatory aspects , such as the potential effects on human health, changes in the composition and sensory profiles of foods, and the possible migration of contaminants, especially for devices intended to be placed inside the package. Finally, another aspect concerns the sustainability of the intelligent systems, according to a globally emerging concept of sustainable packaging. A first challenge in this direction could be to think of reusable, reversible, and long-lasting devices instead of the current single-use, irreversible, and disposable items.

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Figure captions

Figure 1. Publication trends (research articles and review papers) on active packaging (−○−) and intelligent packaging (−□−) in the period 2005–2015. The total number is the cumulative sum of publications at the date of the last access to the web (December 2015). Source: www.scopus.com

Figure 2. Examples of time-temperature indicators: a) Monitor Mark™ by 3M (USA) (http://3m.com); b) Fresh-Check® by Lifelines Technologies Inc. (USA) (http://fresh-check.com/); c) CoolVu™ by Freshpoint (Switzerland) (http://www.freshpoint-tti.com/product/coolvu.aspx); d) Checkpoint® by Vitsab International AB (Sweden) (http://vitsab.com/index.php/tti-label/); e) OnVu™ by Freshpoint (Switzerland) (http://www.freshpoint-tti.com/links/default.aspx); f) Tempix ® by Tempix AB (Sweden) (http://tempix.com/the-indicator/); and g) Timestrip® by Timestrip Plc (UK) (http://timestrip.com).

Figure 3. Example of: a) **a** 1-D barcode; b) a PDF 417 2-D barcode; and c) a QR 2-D barcode.

Scheme 1. Representation of the working principle and components of a sensor.

Table 1. List of recent works on the development of time-temperature indicators.

Sensing element/system	Application	Reference
Chitosan – PVA – Anthocyanin (Red Cabbage)	Milk	Pereira Jr et al., 2015
Glycerol tributyrate - *Aspergillus niger* lipase	Some fruits and vegetables Some fish and shell fish	Wu et al., 2015
Tyrosinase	-	Kocak & Soysal, 2014
Lactic acid bacteria loaded Ca-alginate microparticles	Beef products	Choi, Jung, Lee, & Lee, 2014
PEGylated laccase - 2,2′-azino-bis	Kimchi	Kang et al., 2014
Polydiacetylene - SiO_2 - surfactant	-	Nopwinyuwong, Kitaoka, Boonsupthip, Pechyen, & Suppakul, 2014
Weissella cibaria - Man-Rogosa-Sharpe broth	Chicken breast meat	Park, Kim, Jung, Kim, & Lee, 2013
Ag shell - Au nanorod	-	Zhang et al., 2013
Alkaline lipase - PVA	Milk	Lu, Zheng, Lv, & Tang, 2013
Phenol red – Carbamide - Urease	-	Wu et al., 2013
PDA – Silica nanocomposites	-	Nopwinyuwong, Boonsupthip, Pechyen, & Suppakul, 2012
Weissella cibaria	Ground beef	Kim, Jung, Park, Chung, & Lee, 2012
TOPAS 5013 - BBS chromophores	High temperature processed food products	Lee & Shin, 2012
Burkholderia cepacia lipase	Ground beef	Kim, Choe, & Hong, 2012
Gelatin-Templated Gold Nanoparticles	Frozen foods	Lim, Gunasekaran, & Imm, 2012

Lactic acid bacteria	-	Kim, Jung, Park, & Lee, 2012
PDA - Pluronic F127	-	Nopwinyuwong, Boonsupthip, Pechyen, & Suppakul, 2012
Ag nanoplates	-	Zeng, Roberts, & Xia, 2010
Bacillus subtilis α-amylase	-	Grauwet, Plancken, Vervoort, Hendrickx, & Loey, 2009
α-Amylase	Bogue fish	Yan, et al., 2008
Anionic peroxidase	-	Rani & Abraham, 2006
Bromothymol blue - methyl red - lactic acid	Apple - Carrot - Cake	Wanihsuksombat, Hongtrakul, & Suppakul, 2010
Aspergillus oryzae α-Amylase	-	Raviyan, Tang, Orellana, & Rasco, 2003
Malachite green leuco	-	Bhattacharjee, 1988

Table 2. Comparison between active and passive RFID tags.

Attribute/Feature	Active	Passive
Power source	Have their own power supply (battery)	Acquire the power from the external radio frequency communication.
Cost	$20 to $100	10 cents per tag (for large quantities)
Typical capability	Read/Write	Read-only
Transmission distance	20 to 100m	A few centimeters to 10m
Lifespan	Depends on battery duration and on use	Depends only on use
Communicate with the reader	Can communicate with the reader at any time	Activated when they come within the range of a RFID reader
Size	> Passive	< Active
Frequencies	433 MHz, 2.45 GHz or 5.8 GHz	128 KHz, 13.6 MHz, 915 MHz or 2.45 GHz

Table 3. Comparison between barcode and RFID

Attribute/Feature	Barcode	RFID
Technology	Optical (Laser)	RF (Radio Frequency)
Environment condition	Sensitive to environment, dirt, scratches and temperature	Customized to resist environmental stress and severe processes
Read/Write	Cannot be updated	New information can be over-written
Price	Cheap	Expensive
Identification	Most barcodes only identify the type of item (UPC Code) but not uniquely	Can uniquely identify each item/asset tagged
Read Range	Several inches up to several feet	Passive UHF RFID: - Up to 40 feet (fixed readers) - Up to 20 feet (handheld readers) Active RFID: - Up to 100 feet or more
Data Storage	Barcode is the representative of numbers and cannot store any data	RFID tags contain chips which can store data around 32-128 Bit
Type of tracking	Require manual tracking and therefore are susceptible to human error	Can be automatically tracked removing human error
Integrability	Not integrable	Integrable with sensors

Figure 1

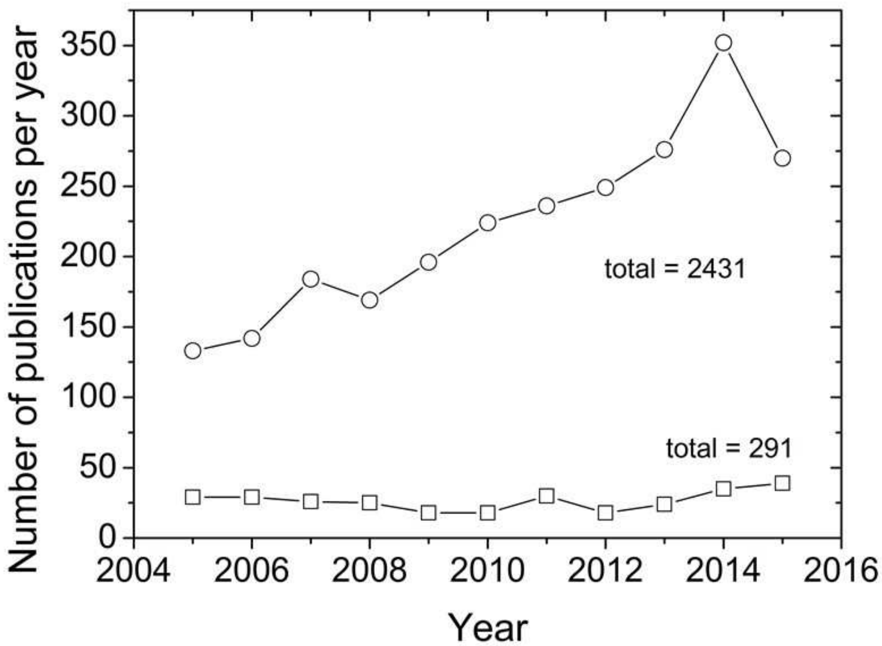

Number of publications per year
350
300
250
200
150
100
50
0
2004
2006
2008
2010
2012
2014
2016
Year
total = 2431
total = 291

Figure 2

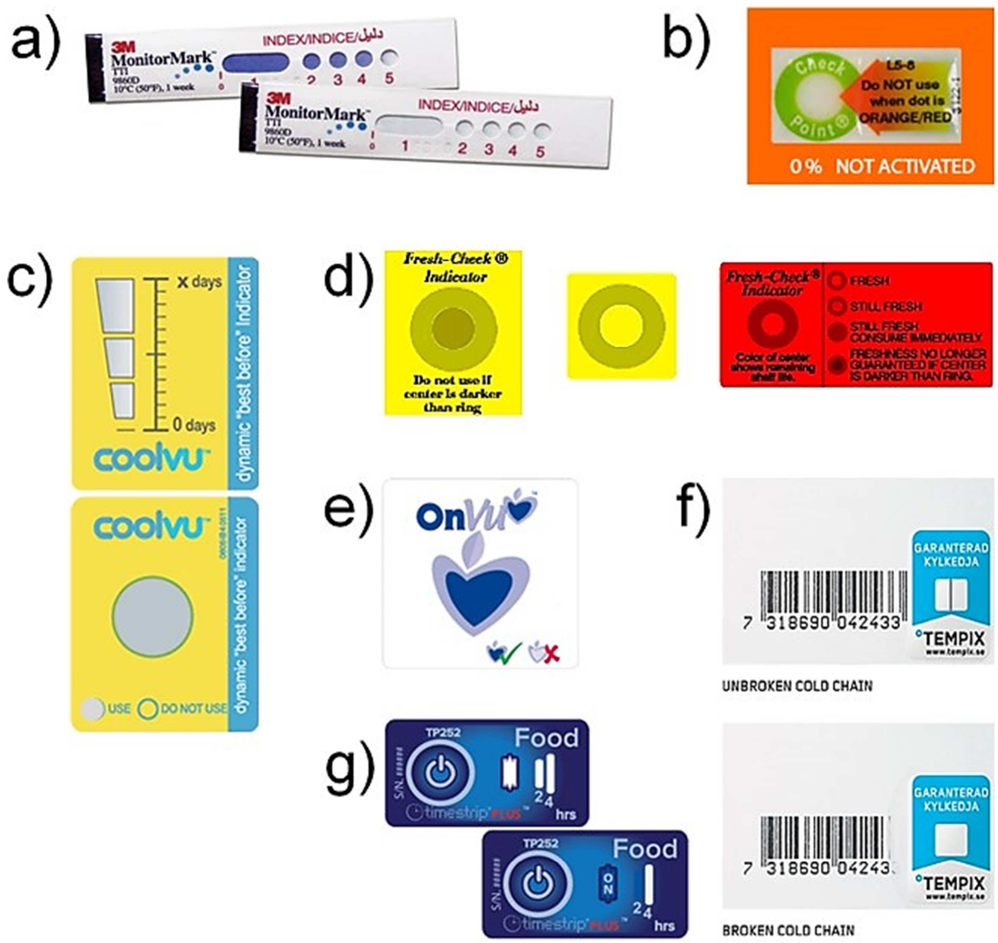
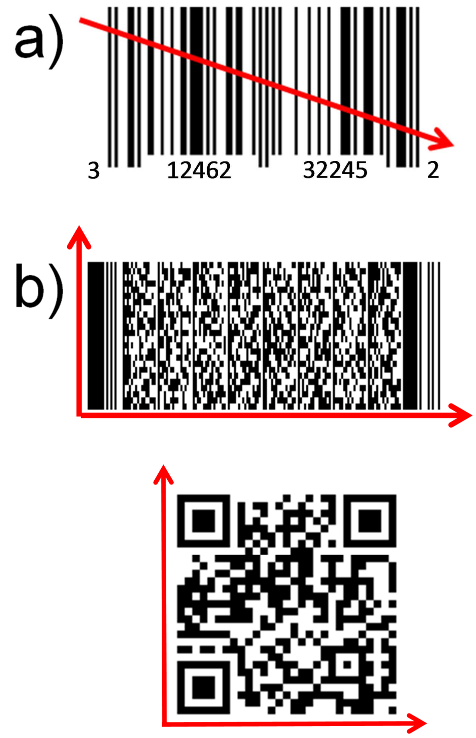
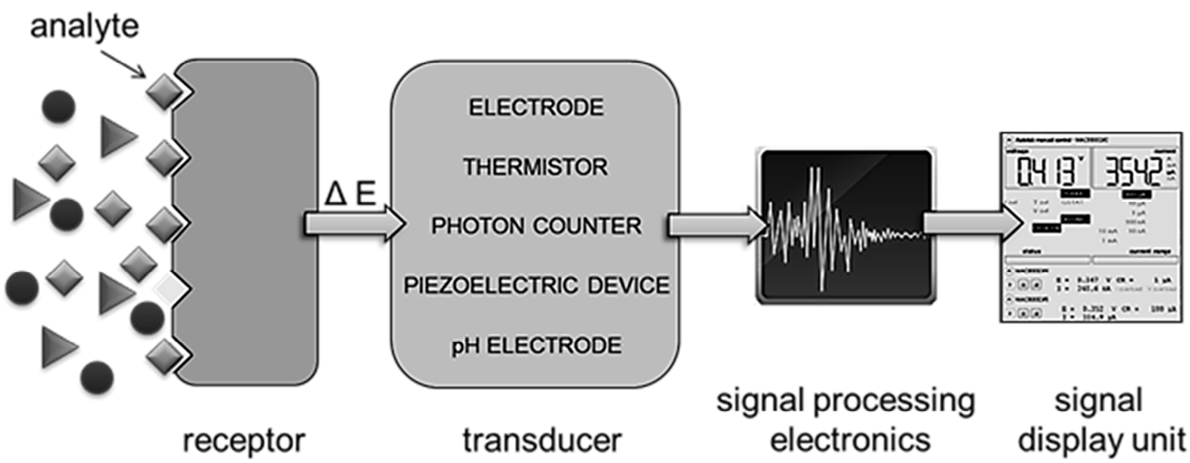

a)
3M
MonitorMark
INDEX/INDICE/دليل
1 2 3 4 5
b)
Check Point
L5-8
Do NOT use when dot is ORANGE/RED
0% NOT ACTIVATED
c)
x days
0 days
coolvu
dynamic "best before" indicator
USE
DO NOT USE
d)
Fresh-Check® Indicator
Do not use if center is darker than ring
FRESH
STILL FRESH
STILL FRESH CONSUME IMMEDIATELY
FRESHNESS NO LONGER GUARANTEED IF CENTER IS DARKER THAN RING.
e)
OnVu
f)
GARANTERAD KYLKEDJA
TEMPIX
UNBROKEN COLD CHAIN
BROKEN COLD CHAIN
g)
TP252
Food
timestrip PLUS
hrs

Figure 3



Scheme 1


analyte
Δ E
ELECTRODE
THERMISTOR
PHOTON COUNTER
PIEZOELECTRIC DEVICE
pH ELECTRODE
receptor
transducer
signal processing
electronics
signal
display unit

Highlights

- Intelligent packaging refers to systems that monitor the condition of packaged foods
- These systems provide information on the food quality during transport and storage
- We overview the basic principles and market applications of intelligent systems
- Aspects concerning legal, economic, and consumers' behavior have been covered
- Guidelines for tomorrow's research in intelligent packaging are provided