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1 An overview of the intelligent packaging technologies
2 in the food sector

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7Structured Abstract

8Background

9Intelligent packaging is the newest technology within the food packaging field. Even though
10this technology is still growing and not fully commercially viable, it has enormous potential
11to improve the safety, quality, and traceability of food products, as well as its convenience for
12consumers.

13Scope and Approach

14This paper first describes both the technical aspects and commercial applications of the most
15representative intelligent technologies—indicators, data carriers, and sensors—with special
16focus on systems and devices that are directly integrated into the package. Secondly, to
17provide useful guidelines for future research in the field, the paper discusses some important
18aspects that still hinder the full exploitation of intelligent technology within the food
19packaging industry.

20Key Findings and Conclusions

21Future research needs to consider some important aspects in order to make intelligent systems
22commercially viable, such as cost, consumers' acceptance and confidence, regulatory aspects
23(e.g., labeling), and multifunctionality.

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

29

30Introduction

31 Packaging is one of the main processes to preserve the quality of food products for
32transportation, storage, and end use. It slows quality decay and makes distribution and
33marketing more efficient. Packaging has four basic functions: protection, communication,
34convenience, and containment (Han, 2005a). Packages protect products from the external
35environment; communicate with the customer through written texts, brand logo, and
36graphics; accommodate the lifestyle of the customer, for example saving time (ready-to-eat
37and heat-and-eat meals) or making the manipulation and handling of packaged food easier for
38the customer (examples of convenient features are easy opening, resealability, and
39microwavability); and act as containers for differently shaped and sized products, with the
40goal of optimizing logistic efficiency (Yam & Lee, 2012). Secure delivery and the
41preservation of packaged foods before consumption are the main goals throughout the food
42supply chain. However, loss of food quality attributes occurs during distribution and storage
43due to biological, chemical, and physical degradation (Han, 2013).

44 Food quality preservation is an important research target because it is intimately linked
45to the more global goal of enhancing the quality of our lives (Sandulescu, Cristea, Harceaga,
46& Bodoki, 2011). Food quality control is necessary, both to better protect consumers against
47foodborne illness and to maximize the efficiency of the food industries, e.g., by reducing
48losses due to the microbial spoilage of perishable foods. At the company level, food quality is
49usually assessed by periodic microbiological and chemical analyses underlying routine tests
50(Viswanathan & Radecki, 2008).

51 Of late, two new concepts have greatly contributed to achieving an advanced concept of
52packaging for safer and healthier food: the active packaging and intelligent packaging
53concepts. Active packaging materials are “materials and articles that are intended to extend
54the shelf-life or to maintain or improve the condition of packaged food. They are designed to

55deliberately incorporate components that would release or absorb substances into or from the
56packaged food or the environment surrounding the food” (European Commission, 2004). To
57improve the functionality of food packages and give them additional functions, different
58active substances can be incorporated into the packaging material (Singh, Abas Wani, &
59Saengerlaub, 2011). Several active packaging systems have been widely reported, such as O₂
60and ethylene scavengers, moisture regulators, CO₂ scavengers and emitters, antioxidant and
61antimicrobial controlled-release packages, and devices to control the release or adsorption of
62flavors and odors (Vermeiren, Devlieghere, van Beest, de Kruijf, & Debevere, 1999).

63 Intelligent packaging materials are “materials and articles that monitor the condition of
64packaged food or the environment surrounding the food” (European Commission, 2004). In a
65broader meaning, intelligent packaging is defined as science and technology that use the
66packaging system’s communication function to facilitate decision making by monitoring
67changes in the internal and external environments and communicating the conditions of the
68packaged food product (Yam, 2012). Differently from active packaging systems, intelligent
69packaging does not directly act to extend the shelf life of foods. Rather, intelligent packaging
70aims to convey information to the stakeholders of the food supply chains (e.g.,
71manufacturers, retailers, and consumers) related to the food’s quality (Restuccia et al., 2010).
72For example, an intelligent packaging system can show when the food product is fresh or
73whether its shelf life has expired; it can show the food’s temperature using thermochromic
74inks or microwave doneness indicators (MDIs); and it can display the food’s temperature
75history using time-temperature indicators (TTIs) (Robertson, 2012). Additionally, intelligent
76packaging can be used to check the effectiveness of active packaging systems (Kerry,
77O’Grady, & Hogan, 2006). In other words, active packaging is the component that takes some
78action, while intelligent packaging is the component that senses and shares the information
79(Yam, Takhistov, & Miltz, 2005). Intelligent packaging and active packaging can work

80synergistically to yield what is defined as “smart” packaging, i.e., a total packaging concept
81that combines the benefits arising from active and intelligent technology (Vanderroost,
82Ragaert, Devlieghere, & De Meulenaer, 2014).

83 Over the last decade, the research interest in intelligent packaging lagged far behind the
84interest in active packaging, as demonstrated by the number of related publications (Figure
851). An explanation for this trend can be twofold. On one hand, researchers first focused on
86new tools to improve the quality and safety of foods through new, “active” functions, in
87contrast to the original “passive” attributes, such as mechanical strength, barrier performance,
88and thermal stability. Intelligent packaging systems have come to the forefront because of the
89growing usage of active components in food packaging, which requires a means of
90monitoring both the active device’s performance and the overall packaging conditions. On the
91other hand, the higher complexity needed to achieve a sophisticated, intelligent system that is
92simultaneously reliable, efficient, and cost effective represented a hurdle to the development
93of intelligent devices. Moreover, the development of such devices requires different technical
94skills and backgrounds to be merged, e.g., food science, materials science, and chemical and
95electrochemical engineering, which makes the overall design and development process more
96complex.

97 Due to the increasing interest in intelligent developments, which have been forecast as
98the main food packaging innovations in the next years (Kuswandi, Wicaksono, Jayus,
99Abdullah, Heng, & Ahmad, 2011; Vanderroost et al., 2014), this paper summarizes the
100intelligent packaging applications in the food sector, with special reference to the applications
101designed to integrate intelligent devices into packages. Therefore, instruments developed
102based on an intelligent technology (e.g., nose and tongue systems), as well as Internet-based
103technologies (e.g., *Internet of everything*, IoE), are out of the scope of this review. The focus
104is on the technical aspects and market applications of three main categories of intelligent

105 systems, namely indicators, data carriers, and sensors. The last part of the work provides
106 guidance for tomorrow's research in the field, with the goal of covering some important
107 aspects that still hinder the full exploitation of intelligent systems for food quality and safety.

108

109 **Intelligent systems in food packaging**

110 The term "intelligent" involves an "ON/OFF" switching function on the package in
111 response to changing external/internal stimuli, in order to communicate the product's status to
112 its consumers or end users (Yam et al. 2005). In practice, an intelligent packaging system is
113 manufactured by incorporating an external, discrete component in the final package, e.g.,
114 two-dimensional films or three-dimensional objects. It is widely accepted that intelligent
115 packaging systems can be realized by three main technologies: (i) indicators, which aim to
116 provide more convenience and/or to inform consumers about the food quality; (ii) data
117 carriers, such as barcodes and radiofrequency identification tags (RFID), which are
118 specifically intended for storage, distribution, and traceability purposes; and (iii) sensors,
119 which allow for a rapid and definite quantification of the analytes in foods (Kerry et al.,
120 2006).

121

122 *Indicators*

123 Indicators convey information to the consumer that is linked to the presence or absence
124 of a substance, the extent of a reaction between two or more substances, or the concentration
125 of a specific substance or class of substances. Most often, such information is displayed by
126 immediate visual changes, e.g., different color intensities or the diffusion of a dye along the
127 indicator geometry (O'Grady & Kerry, 2008). A distinct feature of indicators is the type of
128 information involved, which is qualitative or semi-quantitative in nature. Despite the large
129 varieties of indicators, all of them can be reasonably included within three categories: time-

130temperature indicators, freshness indicators, and gas indicators (Hogan & Kerry, 2008). All of
131them fall within the main category of “product quality and value-improving systems,” which
132are undoubtedly the most widely used devices for food packaging applications (Robertson,
1332012).

134

135Temperature indicators

136 There are two types of temperature indicators: simple temperature indicators and time–
137temperature integrators (TTIs) (Ahvenainen & Hurme, 1997). Temperature indicators show
138whether products have been heated above or cooled below a reference (critical) temperature,
139warning consumers about the potential survival of micro-organisms and protein denaturation
140during, for example, freezing or defrosting processes (Pault, 1995). TTIs, sometimes also
141called integrators, are the first generation of indicators intended to monitor any detrimental
142change in temperature change (e.g., above or below a reference critical value) along the food
143supply chain, i.e., over time. The basic operating principle is based on mechanical, chemical,
144electrochemical, enzymatic or microbiological change, usually expressed as a visible
145response in the form of a mechanical deformation, colour development or colour movement
146(Taoukis & Labuza, 2003). Due to the pivotal role of both time and temperature in
147influencing the kinetics of physical and chemical deterioration, TTIs have gained increasing
148interest for acquiring information about the temperature history of a packaged food over time,
149thus preventing any sort of abuse and/or misuse. TTIs are recognized as user-friendly and
150readily usable devices, whose information is readily understood by consumers as being
151directly related to the quality of the food item at a certain temperature (Pereira Jr, de Arruda,
152& Stefani, 2015). Usually, they consist of small, self-adhesive labels attached to single
153packages or larger configurations (e.g., containers).

154 TTIs' market applications include Monitor Mark™ by 3M (USA), Fresh-Check® by
155Lifelines Technologies Inc. (USA), CoolVu™ and OnVu™ by Freshpoint (Switzerland),
156Checkpoint® by Vitsab International AB (Sweden), Tempix® by Tempix AB (Sweden),
157Timestrip® by Timestrip Plc (UK), Smartpak® by Trigon Smartpak Ltd (UK), and Insignia
158Cold Inspection Intelligent Labels™ by Insignia Technologies Ltd (UK) (Han, Ho, &
159Rodrigues, 2005b; Kuswandi et al., 2011) (Figure 2). Several other examples of TTIs are still
160in the laboratory stage (Table 1). These devices are especially suited for warning of
161temperature abuses of frozen or chilled food products (Yam, Takhistov, & Miltz, 2005).

162

163Freshness indicators

164 The development of freshness indicators over the last two decades stemmed from
165increasing consumer demand for healthy and fresh foods. Freshness indicators have to be
166intended as smart devices that enable the monitoring of the quality of food products
167throughout storage and transportation. Freshness decay may be due to both exposure to
168detrimental conditions and exceeded shelf life. Freshness indicators provide direct
169information on the product's quality regarding microbial growth or chemical changes (Siro,
1702012). For example, freshness indicators intended for seafood are based on the total volatile
171basic nitrogen content (TVB-N), i.e., volatile amines, which are formed as the food spoils and
172can be detected by different methods, such as conductometric (Heising, van Boekel, &
173Dekker, 2015) and pH variations (Kuswandi, Jayus, Oktaviana, Abdullah, & Heng, 2014).
174Hydrogen sulfide indicators can be used to determine the quality of meat products. Hydrogen
175sulfide, which is released by the meat matrix during aging, is correlated with the color of
176myoglobin, which is considered a quality attribute for meat products. Smolander et al.
177developed a freshness indicator based on this principle for modified-atmosphere packed
178poultry meat (Smolander et al., 2002). Other freshness indicators are based on sensitivity

179 toward other microbial metabolites, such as ethanol, diacetyl, and carbon dioxide (Pereira de
180 Abreu, Cruz, & Paseiro Losada, 2011). Commercial applications of freshness indicators
181 include Toxinguard[®] by Toxin Alert Inc., to monitor *Pseudomonas* sp. growth, and
182 SensorQ[™] by FQSI Inc., which senses spoilage in fresh meat and poultry products (O'Grady,
183 & Kerry, 2008). The ripeness indicator RipeSense[™] allows consumers to choose fruit that
184 best appeals to their tastes (Pocas, Delgado, & Oliveira, 2008) by detecting aroma
185 components or gases involved in the ripening process (e.g., ethylene) released by the fruit.

186

187 Gas indicators

188 Gas concentration indicators, in the form of labels, are placed inside the package to
189 monitor changes in the inside atmosphere due to permeation phenomena across the packaging
190 material, microorganisms metabolism, and enzymatic or chemical reactions on the food
191 matrix (Yam et al., 2005). Gas indicators are also used to either assess the efficacy of active
192 packaging components (e.g., O₂ and CO₂ scavengers) or to detect the occurrence of leakages.
193 Because the indicators are placed inside the package, some requirements must be met during
194 the design of these devices, such as being non-water soluble and non-toxic (these components
195 must have food contact approval) (Mills, 2005). The most widely known gas indicators are
196 used to check oxygen and carbon dioxide concentrations. Due to the importance of these
197 gases in food applications, much research has been devoted to the development of O₂ and
198 CO₂ indicators over the last decade and in more recent years (Roberts, Lines, Reddy, & Hay,
199 2011; Jung, Puligundla, & Ko, 2012; Lee & Ko, 2014; Vu & Won, 2014). Most devices are
200 based on redox dyes (e.g., methylene blue, 2,6-dichloroindophenol, or N,N,N',N'-tetramethyl-
201 *p*-phenylenediamine), a reducing compound (e.g., reducing sugars), and an alkaline
202 compound (e.g., sodium hydroxide) (Kuswandi et al., 2011). However, such indicators suffer
203 from dye leaching upon contact with the moisture in the package's headspace. The latest

204developments concern UV-activated colorimetric oxygen indicators (Lee, Mills, & Lepre,
2052004; Lee, Sheridan & Mills, 2005; Roberts et al., 2011) with very limited dye leaching due
206to either encapsulation or coating technologies (Mills, Hazafy, & Lawrie, 2011; Thai Vu &
207Won, 2013).

208 Trade names for several commercial applications include Ageless Eye™ by Mitsubishi
209Gas Chemical Co., Shelf Life Guard by UPM, Vitalon® by Toagosei Chemical Inc., Tufflex
210GS by Sealed Air Ltd., and Freshilizer by Toppan Printing Co (Rodrigues & Han, 2003;
211O'Grady & Kerry, 2008).

212

213*Data carriers*

214 Data carrier devices, also known as automatic identification devices, make the
215information flow within the food supply chain more efficient, to the advantage of food quality
216and safety. More specifically, data carrier devices do not provide any kind of information on
217the quality status of food but are rather intended for automatization, traceability, theft
218prevention, or counterfeit protection (McFarlane & Sheffi, 2003). Moreover, data carriers are
219more often placed onto tertiary packaging (e.g., multi-box containers, shipping crates, pallets,
220large paperboard packages). The most important data carrier devices in the food packaging
221industry are barcode labels and RFID tags, which belong to the main category of
222convenience-enhancing intelligent systems (Robertson, 2012).

223

224Barcodes

225 The first Universal Product Code (UPC) barcodes found market application in the
2261970s. Due to their low cost and ease of use, barcodes have been increasingly used in the
227large-scale retail trade and stores to facilitate inventory control, stock reordering, and
228checkout (Manthou & Vlachopoulou, 2001). A barcode is a pattern of parallel spaces and bars

229arranged to represent 12 digits of data. The encoded information is read by an optical barcode
230scanner that sends the information to a system where it is stored and processed (Han, 2013).
231One-dimensional (1-D) barcodes were developed first. The basic working principle is the
232same of a laser beam cutting a horizontal slice from the vertical code bars. As the beam
233moves over the symbol (see Figure 3a), it measures the relative time it spends scanning dark
234bars and light spaces. A lookup table is then used to decode individual characters from those
235times. Because of the line of the laser beam, these kinds of barcodes are referred to as being
2361-D. The storage capacity of the first-generation barcode labels was limited, such as to the
237manufacturer identification's number and the item number (Robertson, 2012; Drobnik, 2015).
238Reduced Space Symbology (RSS) barcodes were developed successively to encode more
239data in a smaller space. The most frequently used RSS symbologies are the RSS-14 stacked
240omnidirectional barcode and the RSS expanded barcode, the latter encoding up to 74
241alphanumeric characters (Yam, Takhistov, & Miltz, 2009).

242 Two-dimensional (2-D) barcodes (Figure 3b) allow a larger amount of information to
243be stored, compared to 1-D barcodes, by combining dots and spaces arranged in an array or a
244matrix, instead of bars and spaces. This allows for an increased density of data within a
245reduced space. For example, the Portable Data File (PDF) 417 is a 2-D symbol that carries up
246to 1.1 kB of data in the space of a UPC barcode (Yam et al., 2009). The more recent Quick
247Response (QR) 2-D barcode (Figure 3c) enables an even larger amount of data to be stored
248using four different encoding modes: numeric, alphanumeric, byte/binary, and kanji, the latter
249referring to logographic Chinese characters. Reading 2-D barcode symbologies requires a
250scanning device capable of simultaneous reading in two dimensions—vertically as well as
251horizontally (Kato, Tan, & Chai, 2010).

252

253

254 Radio-frequency identification systems

255 RFID tags are the most advanced example of a data carrier device. An RFID system
256 includes three main elements: a tag formed by a microchip connected to a tiny antenna; a
257 reader that emits radio signals and receives answers from the tag in return; and middleware (a
258 local network, web server, etc.) that bridges the RFID hardware and enterprise applications
259 (Kumar, Reinitz, Simunovic, Sandeep, & Franzon, 2009; Sarac, Absi, & Dautère-Pères,
260 2010). Two distinct features of RFID technology are the high number of various codes that
261 can be stored in the tag and the possibility of transferring and communicating information
262 even at a long distance, thus improving automatic product identification and traceability
263 operations (Plessky, 2009). Most advanced RFID systems (2.45 GHz—super high frequency
264 active tags) have a reading range of up to 100 meters, with up to 1 MB data in storage
265 capacity. Nowadays, RFID technology includes two types of tags: active and passive tags; the
266 main difference is that active tags rely on a battery while passive tags do not. Table 2
267 provides a full comparison between the two types of tags.

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

279Sensors

280 Sensors are considered the most promising and innovative technology for future
281intelligent packaging systems (Kuswandi et al., 2011; Bagchi, 2012). A sensor is a device or
282system with control and processing electronics, an interconnection network, and software
283(Patel & Beveridge, 2003). A sensor is used to detect, locate, or quantify energy or matter, by
284giving a signal for the detection or measurement of a physical or chemical property to which
285the device responds (Kress-Rogers, 1998). In practice, a sensor replies to a chemical or
286physical quantity to make a quantifiable output that is proportional to the measure. Most
287sensors are made up of four major components (Scheme 1). (i) The first is a receptor, i.e., the
288sensing part of the sensor, represented by a sampling area (generally a chemo-selective
289coating) where the surface chemistry occurs. Here, the analytical information is obtained
290from the adsorption of the target analyte on the recognition layer. The energy variation
291associated with detecting the analyte induces a change of a property of the receptor in terms
292of, for example, redox potential, pH, temperature, or light. (ii) The second is the transduction
293element, i.e., the measuring part of the sensor (e.g., an electrode), which is capable of
294transforming the energy variation and carrying the physical or chemical information into a
295useful analytical signal (e.g., electrical, optical, thermal, or chemical). Next are (iii) the signal
296processing electronics, and (iv) a signal display unit (Neethirajan, Jayas, & Sadistap, 2009).
297The ideal sensor should possess the following characteristics (Hanrahan, Patil, & Wang,
2982004): (i) specificity for the target species (i.e., selectivity); (ii) sensitivity to changes in
299target-species concentrations; (iii) fast response time; (iv) extended lifetime of at least several
300months; and (v) small size (miniaturization), with the possibility of low-cost manufacture.
301In recent years, different kinds of sensors intended for food applications have been
302developed, such as electrochemical sensors (Goulart, Cruz de Moraes, & Mascaro, 2016;
303Feng Gao et al., 2015; Liu, Xiao, Cui, & Wang, 2015; Pacheco et al., 2015; Nasirizadeh,

304Hajihosseini, Shekari, & Ghaani, 2015) and luminescence sensors (Fan, Shen, Wu, Wang, &
305Zhang, 2015; Pénicaud, Guilbert, Peyron, Gontard, & Guillard, 2010). Electrochemical
306sensors represent an important subclass of chemical sensors, in which an electrode is used as
307the transduction element. The working principle of electrochemical sensors is based on redox
308reactions that take place at the electrode/analyte interface upon applying a voltage by means
309of a potentiostat. The electrons transfer between electrode and electroactive species gives
310origin to a current that is proportional to the concentration of the analyte (Wang, 2006). In
311luminescence sensors the emitted fluorescence, phosphorescence or chemiluminescence
312signals are measured after the analyte is immobilized in a suitable solid support, giving origin
313to the expression solid-phase luminescence (SPL) or to its equivalent solid-matrix
314luminescence (SML). Under certain conditions, these analytical signals can be related to the
315concentration of analyte in the sample (Ibañez & Escandar, 2011).

316 However, most of these developments concern the detection of food components and
317contaminants in food matrices. Although flexible printed chemical sensors integrated into
318food packages have a promising future (Vanderroost et al., 2014), most advanced sensor
319technologies that can incorporate intelligent devices into packaging belong to two main
320groups: biosensors and gas sensors.

321

322Biosensors

323 The main difference between chemical sensors and biosensors lies in the recognition
324layer. While in chemical sensors the receptor is a chemical compound, the recognition layer
325of biosensors is made of biological materials, such as enzymes, antibodies, antigens, phages,
326and nucleic acids (Wang, 2006). Current uses of biosensor systems integrated into packaging
327are limited to a few examples. SIRA Technologies (USA) has developed the Food Sentinel
328System, a packaging barcode technology that can alert consumers and retailers when a

329product has been exposed to adverse conditions, thus affecting its safety. The technology is
330based on a biosensor carrying an antibody of a specific pathogen, in the form of a membrane
331attached to the barcode. In the presence of contaminating bacteria, an ink incorporated into
332the biosensor will turn red, and the barcode will be rendered incapable of transmitting data
333when scanned (Yam et al., 2005). Toxin Guard™ (Toxin Alert Inc., USA) is a visual diagnostic
334tool used to detect pathogens or other selected micro-organisms that may contaminate food,
335such as *Campylobacter* spp., *Escherichia coli* O157, *Listeria* spp., and *Salmonella* spp. The
336Toxin Guard™ immunoassay works based on antibody–antigen reactions on polymer
337packaging films: in the presence of pathogenic bacteria, the bacterial toxin is bound to the
338antibodies and immobilized on a thin layer of flexible polymer film (e.g., polyethylene, PE),
339yielding a clear change in the color of the smart device (Han, 2013). Bioett (Bioett AB,
340Sweden) is a system technology that combines biochemistry and electronics to monitor the
341temperature of foods during refrigerated transport. The system consists of a biosensor
342attached to the food package, a detector reading the data from the biosensor, and a database to
343store information about the goods. The main parts of the Bioett system are a chip-less RF
344circuit and a built-in biosensor. At different points in the supply chain, this sensor can be read
345using a handheld scanner (Hogan & Kerry, 2008). Flex Alert (Canada) developed
346commercially available flexible biosensors to detect toxins in packaged foods throughout the
347supply chain. Flex Alert biosensors have been specifically developed against *Escherichia coli*
348O157, *Listeria* spp., *Salmonella* spp., and aflatoxins (Vanderroost et al., 2014).

349

350Gas sensors

351 The development of sensors that can respond quantitatively and reversibly to gaseous
352analytes has been a fervid research field during the last two decades. Established systems for
353gas detection include metal oxide semiconductor field-effect transistors (MOSFETs), piezo-

354electric crystal sensors, amperometric oxygen sensors, organic conducting polymers, and
355potentiometric carbon dioxide sensors. However, these systems exhibit various limitations,
356such as cross-sensitivity to carbon dioxide and hydrogen sulfide, fouling of sensor
357membranes, and consumption of the analyte (e.g., oxygen), and these systems involve
358destructive analyses of packages in most cases (Kerry et al., 2006). More recent
359developments have especially focused on new O₂ and CO₂ sensors, with the aim to overcome
360these drawbacks.

361 The development of smart sensors to quantify oxygen permeating across the package
362has been a main research topic over the last two decades, as demonstrated by the number of
363works published in the literature and the instruments and devices in the market. Oxygen
364sensors are based on luminescence detection and represent an alternative approach to purely
365visual oxygen indicators, providing higher sensitivity and accuracy of the quantitative
366measurements, compared to systems based on absorption or reflectance (MacCraith et al.,
3671993). Distinct features of these systems include the possibility of carrying out the
368measurements on 3-D samples with non-destructive experiments. Moreover, they offer fast
369responses, do not consume any analyte, and lack electrical connections. Huber et al. proposed
370a new non-destructive and non-invasive fiber-optic oxygen meter to quantify the oxygen
371permeability of containers and plastic bottles (Huber, Nguyen, Krause, Humele, &
372Stangelmayer, 2006). The principle of the sensor's operation is based on the quenching of
373luminescence caused by the collision between molecular oxygen and luminescent dye
374molecules in the excited state. Oxygen determination (i.e., oxygen partial pressure) can take
375place in both solutions (dissolved oxygen) as well as in the gaseous phase. No cross-
376sensitivity exists for carbon dioxide, hydrogen sulfide, ammonia, pH, or any ionic species
377such as sulfide, sulfate, or chloride, and the measurement is not affected by salinity. Turbidity
378and changes in the stirring rate have no influence on the measurement. The sensors can also

379be used in methanol– and ethanol–water mixtures, as well as in pure methanol and ethanol. In
380another paper using an optical oxygen sensor, Fitzgerald et al. fabricated a phosphorescence
381lifetime-based oxygen sensor made of fluorescent complexes of ruthenium(II) and
382platinum(II)-octaethylporphyrine-ketone (PtOEPK) dye (Fitzgerald et al., 2001). The authors
383first demonstrated that the sensor allows for efficient and sensitive measurements of oxygen
384in food packages, besides being non-destructive. They also tested the sensor on real samples,
385such as packaged sliced ham, smoked fish, and raw and cooked meat, demonstrating that it
386provides accurate and reliable results even on real samples, which is a requirement for market
387applications of the sensor.

388 Baleizao et al. presented an optical dual sensor for oxygen and temperature (Baleizao,
389Nagl, Schaferling, Berberan-Santos, & Wolfbeis, 2008). The sensor is based on luminescence
390lifetime measurements and is highly sensitive to oxygen, while covering a very wide
391temperature range. The sensor contains two luminescent compounds incorporated into
392polymer films, one sensitive to temperature and the other sensitive to oxygen. Due to its
393highly temperature-dependent luminescence, Ruthenium tris-1,10-phenanthroline was used as
394the temperature-sensitive dye and is incorporated in poly(acrylonitrile) to avoid cross-
395sensitivity to oxygen. The oxygen-sensitive probe used is fullerene C₇₀, due to its strong,
396thermally activated, delayed fluorescence at high temperatures and its exceptional oxygen
397sensitivity. The dual sensor exhibits a temperature operation range between at least 0 and 120
398°C, as well as detection limits for oxygen in the ppbv range, operating for oxygen
399concentrations up to at least 50 ppmv.

400 The development of CO₂ sensors for food packaging applications has lagged behind
401that of O₂ sensors because of oxygen's role as a primary factor in the degradation of many
402foods. However, especially since the use of MAP packaging systems was established,
403controlling the amount of CO₂ in packages has become just as important for both shelf life

404and freshness studies (Fu, Molins, & Sebranek, 1992). Conventional techniques for a
405quantitative and qualitative analysis of CO₂ include Severinghaus-type electrodes, infrared
406(IR) spectroscopy, gas chromatography (GC), and mass spectroscopy (MS). However, these
407techniques suffer from a series of drawbacks: instruments are often expensive, bulky, and not
408particularly robust; require long pathlengths; are prone to interference; lack mechanical
409stability; and require rather sophisticated equipment (Mills & Eaton, 2000; Sipior, Randers-
410Eichhorn, Lakowicz, Carter, & Rao, 1996; Schulz, Jensen, Balsley, Davis, & Birks, 2004).
411For this reason, great effort has been made over the last 20 years to fabricate sensitive, robust,
412fast, cheap, flexible, and easily miniaturized sensors to detect CO₂. Von Bultzingslowen et al.
413developed an optical sensor to measure carbon dioxide in modified atmosphere packaging
414(MAP) applications (von Bultzingslowen et al., 2002). This sensor is based on the fluorescent
415pH indicator 1-hydroxypyrene-3,6,8-trisulfonate (HPTS) immobilized in a hydrophobic,
416organically modified silica (ormosil) matrix obtained by sol-gel chemistry. The authors
417showed that oxygen cross-sensitivity is minimized (0.6% quenching in air) by immobilizing
418the reference luminophore in polymer nano-beads. Moreover, cross-sensitivity toward
419chloride and pH was found to be negligible.

420 Borisov et al. developed optical carbon dioxide sensors based on an emulsion of room-
421temperature ionic liquids (RTILs)—1-butyl-3-methylimidazolium salts in a silicone matrix
422(Borisov, Waldhier, Klimant, & Wolfbeis, 2007). In particular, for the quantitative
423determination of CO₂, they used 8-hydroxypyrene-1,3,6-trisulfonate (HPTS) to prepare a
424fluorimetric sensor, which was claimed to have potential applications in several fields, such
425as food packaging technology. Borchert et al. developed an optochemical CO₂ sensor that
426includes a phosphorescent reporter dye PtTFPP and a colorimetric pH indicator α -
427naphtholphthalein incorporated in plastic matrix, together with a phase transfer agent—
428tetraoctyl- or cetyltrimethylammonium hydroxide (Borchert, Kerry, & Papkovsky, 2013).

429 Experiments were carried out to optimize the composition and working characteristics of
430 such a sensor in order to measure headspace CO₂ in foods packaged under a modified
431 atmosphere. The authors demonstrated that in food and modified atmosphere environments,
432 the sensor retained its sensitivity to CO₂ for 21 days at 4 °C, which is sufficient for many
433 packaged products.

434

435 *Other intelligent packaging systems*

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

453the form of holograms, thermochromic inks, micro-tags, tear labels, and tapes (Han, Ho, &
454Rodrigues, 2005c).

455

456**Market and legislative considerations**

457 Besides historical and technical factors, the commercial application of intelligent
458systems in the food packaging industry has had to face (and still does face) some important
459considerations. Consumers' perceptions and legislative aspects, in particular, are key factors.

460 One of the main issues that hinder the market penetration of intelligent devices in food
461packaging is consumers' acceptance of non-edible items separate from the package. Sachets,
462inserts, spots, and dots are sometimes thought to be unnecessary, i.e., the benefit of intelligent
463systems is still unclear. In other circumstances, consumers are worried that innovative
464packages might mislead them regarding the product's quality (Day, 2008; Vanderroost et al.,
4652014). In more recent years, retailers have reconsidered the use of intelligent systems for two
466main reasons: (i) alerts and messages provided by the intelligent devices (e.g., indicators) can
467push consumers to buy only newly displayed items, leading to an increased amount of unsold
468foodstuffs (Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback 2008);
469and (ii) some devices (e.g., TTIs) might display temperature abuses that occurred before the
470food reached the retailers' shelves. However, unambiguously identifying the failing step (and
471thus the responsibility for that abuse) in the supply chain might be difficult.

472 From a legislative point of view, the lack of an adequate regulatory framework in the
473EU for intelligent (and active) packaging systems until 2004 hindered the placement of new
474packaging solutions into the market, in contrast to the United States, Australia, and especially
475Japan, where intelligent packaging systems are widespread. In fact, the lack of a clear
476regulatory framework for many years led to reluctance by food packaging manufacturers to
477take on new concepts that are not fully covered by the legislation on food contact materials.

478The first EU legislative attempt to address the topic of active and intelligent materials was
479Framework Regulation EC 1935/2004 ([European Commission, 2004](#)), which describes the
480general requirements for all food contact materials. In particular, Article 3 covers stipulations
481on the packaging material containing the intelligent component, e.g., “the packaging material
482shall not transfer constituents to food in quantities that could endanger human health, bring
483about an unacceptable change in the composition of foods, or bring about deterioration in
484organoleptic characteristics thereof” ([European Commission, 2004](#)). Article 4 refers to the
485intelligent component, dealing with some issues in particular: “intelligent materials shall not
486give information about the food’s condition that could mislead the consumer, adequate
487labelling must allow non-edible parts to be identified, and adequate labelling must indicate
488that the materials are active and/or intelligent” ([European Commission, 2004](#)). Finally, Article
48915 clearly states that “consumers and food packaging companies must be informed on how to
490use the active and intelligent materials and articles safely and appropriately” ([European
491Commission, 2004](#)). Although useful, the Framework Regulation of 2004 can have multiple
492interpretation ([Dainelli et al., 2008](#)).

493 The Good Manufacturing Practice (GMP) represented a legislative implementation
494regarding materials and articles intended to come in contact with food (Regulation EC
4952023/2006). It aims to ensure that these materials do not transfer into foods (in a process
496called migration) in unacceptable quantities ([European Commission, 2006](#)).

497 However, the only specific regulation entirely devoted to intelligent (and active)
498materials intended for food packaging applications is Regulation 450/2009, which sets out
499specific requirements on the use and authorization of active and intelligent materials and
500articles intended to come into contact with food. The regulation also establishes an EU-wide
501list of substances that can be used in manufacturing these materials; substances may only be
502added to the list once their safety has been evaluated by the European Food Safety Authority

503(EFSA) (European Commission, 2009). In addition, this regulation introduces an
504authorization scheme for substances used for active and intelligent functions in food contact
505materials. Article 11 of the regulation also states that, in order the consumer to identify non-
506edible parts, active and intelligent materials and articles or parts thereof shall be labelled,
507whenever they are perceived as edible, (i) with the words “DO NOT EAT” and (ii) always,
508where technically possible, with a specific symbol.

509 Although the issue related to the potential migration concerns mainly active packaging
510systems, the risk associated to the unintended release/contact of certain substances/materials
511also includes intelligent packaging devices, especially when they are positioned inside the
512primary packaging. Consumers’ reluctance toward intelligent packaging devices is often
513associated to the potential risk of leaching of active components (e.g., inks) from the device,
514or swallowing of the sachet. This may be the case of intelligent systems including water
515soluble components that are susceptible to leaching upon direct contact with foods with a
516high moisture content (Mills, 2009). At least in Europe, the perception of this risk by
517consumers seem to be higher for separate non-edible objects (e.g., sachets and inserts)
518compared to structures that are incorporated/attached to the package (e.g., labels) (Han,
5192005c; Lee, Yam, & Piergiovanni, 2008). Therefore, preserving the integrity of intelligent
520components inside the package throughout the shelf life of the food plays a role to minimize
521any potential safety issue while increasing the consumers’ trust toward this technology.

522

523**Concluding remarks**

524 The interest in innovative packaging systems to achieve higher food quality and safety,
525consumer convenience, and management (i.e., storage, distribution, and traceability) along
526the food supply chain has boosted the development of intelligent devices, in the form of
527labels, tags, dots, and inks that perform different functions. Although the potential advantages

528 arising from such technologies have been widely explored and documented, there is still an
529 existing gap in market applications. For this reason, future research needs to consider some
530 important aspects in order to make intelligent systems commercially viable and, ultimately,
531 into everyday packaging commodities. For instance, the final cost of intelligent packaging
532 systems should account for a minimal part of the whole packaging cost. Due to the
533 technology involved, the cost attributed to intelligent devices is estimated to be ~ 50–100%
534 of the whole cost of the final package. However, for most food products the packaging cost
535 should not exceed 10% of the total cost of the goods placed on the shelves, provided that the
536 claimed benefits are unambiguously demonstrated to outweigh the possible extra expenses
537 arising from the new technology. This mismatch between the new technology and market
538 penetration eventually results into a negative cost/benefit analysis (Dainelli et al., 2008).

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

551 Another technical goal for the future is the integration of several functions within only
552 one device (multi-functional intelligent packaging), as well as the development of new

553functions, e.g., systems able to communicate the presence of potential allergens, warnings
554related to diet management, and error prevention alerts. In particular, advances in biosensors
555and biotechnology applied to food packaging systems are expected (Han, 2005c).

556 Equally important is to educate consumers on the extra benefits arising from intelligent
557systems. This can be achieved using clear information about the device, e.g., what purpose it
558serves, how it works, and how to use it.

559 Intelligent devices also need to be adequately labeled, in order to increase consumers'
560confidence in the safety of packaged food. Packaging manufacturers must also consider
561regulatory aspects , such as the potential effects on human health, changes in the composition
562and sensory profiles of foods, and the possible migration of contaminants, especially for
563devices intended to be placed inside the package. Finally, another aspect concerns the
564sustainability of the intelligent systems, according to a globally emerging concept of
565sustainable packaging. A first challenge in this direction could be to think of reusable,
566reversible, and long-lasting devices instead of the current single-use, irreversible, and
567disposable items.

568

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862

863 **Figure captions**

864

865 **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

869

870 **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>).

878

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

881

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

883

Figure 1

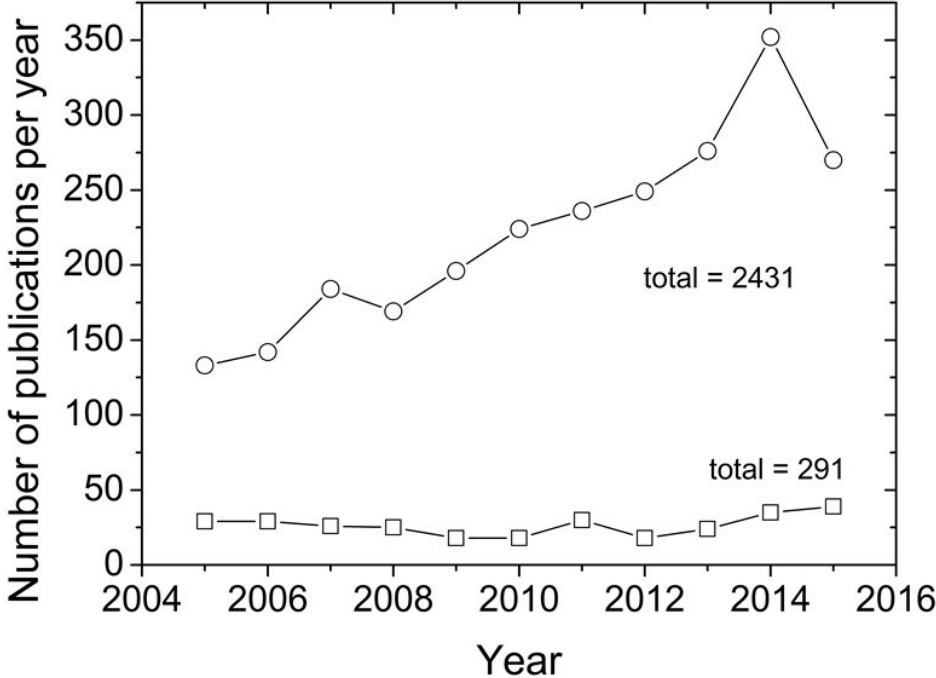
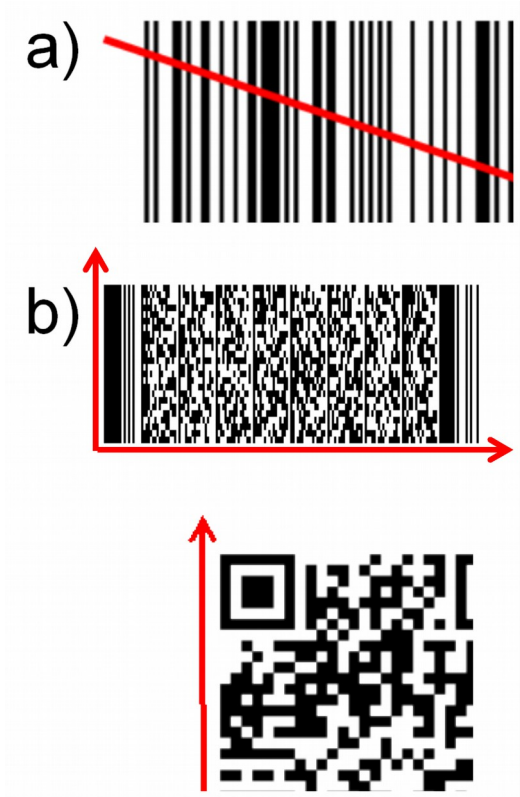


Figure 2



Figure 3



Scheme 1

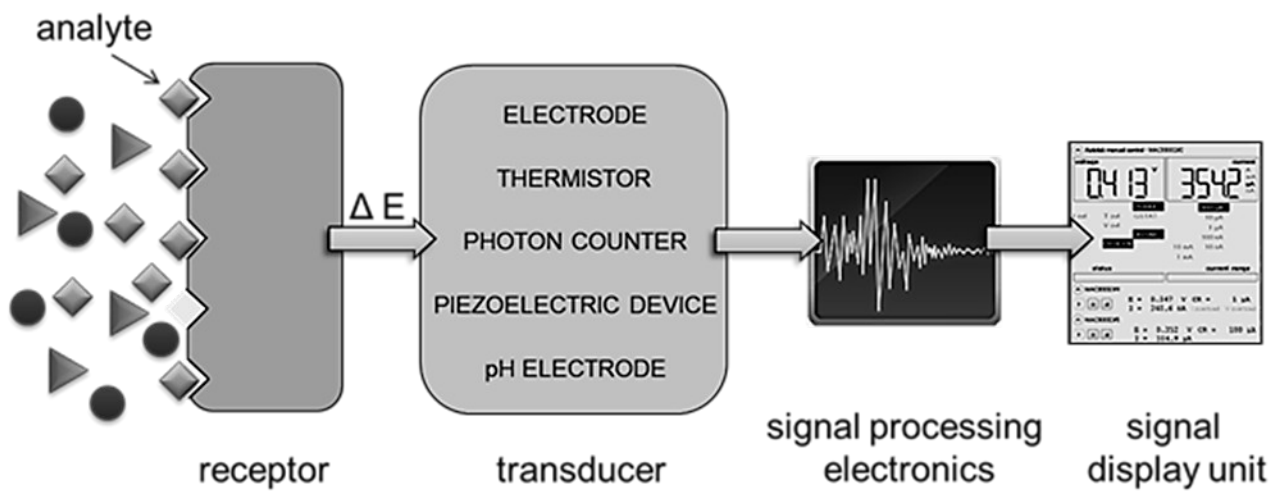


Table 1. List of recent works on the development of time-temperature indicators.

Sensing element/system	Application	Reference
Chitosan – PVA – Anthocyanin (Red Cabbage)	Milk	Pereira Jr et al., 2015
Glycerol tributyrate - <i>Aspergillus niger</i> 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
<i>Weissella cibaria</i> - 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
<i>Weissella cibaria</i>	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
<i>Bacillus subtilis</i> 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
<i>Aspergillus oryzae</i> 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