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Responsive Food Packaging: Recent Progress and Technological Prospects

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Abstract: Responsive food packaging is an emerging field in food packaging research and the food industry. Unlike active packaging, responsive packaging systems react to stimuli in the food or the environment to enable real time food quality and food safety monitoring or remediation. This review attempts to define and clarify the different classes of food packaging technologies. Special emphasis is given to the description of responsive food packaging including its technical requirements, the state of the art in research and the current expanding market. The development and promises of stimuli responsive materials in responsive food packaging are addressed, along with current challenges and future directions to help translate research developments into commercial products.

Keywords: active packaging, food packaging, intelligent packaging, responsive packaging, stimuli responsive materials

Introduction

Historically, food packaging has been developed to contain food products, maintain food quality, and inform consumers about the properties of the enclosed product. These primary functions are often stated as containment, protection, and communication (Coles and others 2003). A commonplace example is a milk carton. A typical carton of milk consists of plastic-lined paperboard that provides an effective barrier that both contains the milk and protects it from the outside environment. The carton then communicates with the consumer with printed branding, nutritional information. Innovations in the food packaging industry involve contributions from engineers, microbiologists, food scientists, chemists, regulators, and other professionals. As a result, a variety of terminologies have been used to describe similar concepts and the same terms are sometimes used to describe different ideas. The terms "smart" and "intelligent" packaging have been widely used by industry professionals and the media to refer to different types of functional packaging systems. There is no official definition of smart packaging. For our purposes, we will use the widely cited definition from "the A to Z of Materials," an international network of engineers and researchers dedicated to advancement of materials science research and applications. By this definition, smart packaging is any type of packaging that provides specific functionality beyond the role of physical barrier between the food product and the surrounding environment (Butler 2001). Put simply, smart packaging can be viewed as an enhancement of the 3 primary packaging functions. Within the greater realm of smart packaging the term intelligent packaging is currently used to describe any packaging system that conveys information to the consumer about the enclosed product (Yam and others 2005). However, the 2 terms are often used interchangeably and their definitions are inconsistent both within food packaging research and in the media (Singh and Heldman 2001; Vanderroost and others 2014). This inconsistency leads to confusion about the designed purpose of the package and hinders communication between researchers, the media, and the general public. In order to alleviate confusion surrounding intelligent and smart packaging, we consider that "smart" and "intelligent" are synonyms that refer to any packaging that enhances the primary functions of the package or adds new functionalities. This definition is also shared by other authors (Yam and others 2005). Such clarification is necessary because from an engineering standpoint the synonyms "smart" and "intelligent" are generic descriptions that do not provide information on the designed functionality of a particular package. Smart/intelligent packaging systems can be classified based on the engineered functionality of the package. Here, we propose the classification of food packaging into 4 main categories based on functionality: ergonomic, informative, active, and responsive (or reactive) (Figure 1). Ergonomics is the practice of performing tasks with minimal physical effort and discomfort while maximizing efficiency. This definition stems from guidelines outlined by the Natl. Inst. for Occupational Safety and Health (NIOSH) concerning handling materials in the workplace (Cal/OSHA Consultation Service Div. of Occupational Safety and Health 2007). Ergonomic packaging enhances the containment aspect of packaging and make the package easier to transport, store, use, and discard (Azzi and others 2012). Ergonomic packages aid with package handling, sealing, dispensing. Other functionalities that enhance convenience are also included in ergonomic packaging. These functionalities include easy-to-open bottles for arthritis patients, microwaveable pouches, edible packaging, and bottles with unique handle shapes (Singh and Heldman 2001). Informative, active, and responsive packaging serve to enhance both protection and communication. We present the term

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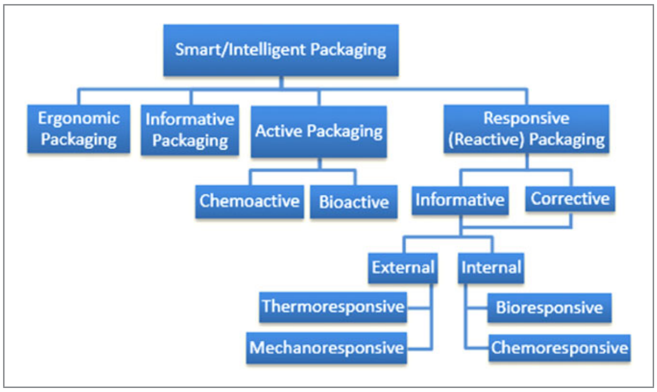

Figure 1–Diagram showing the classification of smart packaging from the functionality or engineering standpoint.

informative food packaging to describe packaging that enhances the way the information about the food product is stored, disclosed or transferred without interacting with the food. This enhancement is particularly important because without it nearly every food package would be considered informative due to the product information printed on the package. This type of information transfer is possible due to recent technological advancements, namely advances in radio-frequency identification (RFID) tags. This technology is considered smart/intelligent packaging as it has drastically improved the communication aspect of packaging with streamlined information transfer between food producers and consumers. Briefly, RFID tags are enhanced bar codes that can be read via frequency scanners or smart phones. The distinct advantage that they have over traditional barcodes is that information can be uploaded or altered electronically on these tags. These technologies will be described further in this review. However it must be noted that food packaging sensors do not fall into this packaging class as they provide dynamic information in response to a change in the food quality and safety. Food packaging sensors fall under the class of informative responsive packaging and are discussed later in this review.

Active packaging is a widely accepted term for packaging systems that directly interact with the enclosed food and affect its quality (Yam and others 2005). These systems include moisture scavengers, antimicrobial composites, and modified atmosphere packaging (MAP). Unlike responsive packaging, these systems work constantly and do not respond to a specific trigger. More information on this distinction can be found later in this review. The fourth and much less known category we present is responsive (or reactive) packaging technology.

We define "responsive packaging" as any package that elicits a curative or informative response as a result of a specific trigger or change occurring in the food product, food package headspace, or the outside environment. This triggering is important because current active packaging technology is primarily based on passive diffusion or initial package modification (Kruijf and others 2002). Responsive packaging systems work differently by only reacting to a stimulus present in food or the environment. Stimuli can be a variety of foodborne threats, substances or organisms that negatively affect food quality or safety, like molds, bacteria, and contaminants. Stimuli can also be benign factors like moisture, pH, or gas levels in the package headspace. Response systems can be informative, corrective, or both. An informative responsive package is one that incorporates a sensor that can detect specific target compounds and responds with a measurable signal, thus providing information to the user. Corrective responsive packaging systems perform a curative action that can either maintain food safety (release of antimicrobials) or promote food quality (release of color compounds, and flavoring agents) when triggered by a foodborne or environmental threat or other change within the food package. In other words, these responses can be triggered by internal chemical/biological stimuli or external temperature changes and mechanical stresses (Ulijn and others 2007).

In order for a food packaging system to be considered responsive, it must incorporate a material that responds to internal or external stressors. These materials are referred to as "stimuli responsive," and research in this area has grown significantly in the past 30 y. However, the majority of the work on stimuli responsive materials has been concentrated in the engineering, chemistry, materials science, and medical fields, with limited implementation in food packaging (Figure 2) (Takae and others 2008; Schneider and others 2012; Lau and Wang 2013). Incorporating responsive material technology into food packaging could have enormous impacts in food safety. The U.S. Centers for Disease Control and Prevention (CDC) estimates that 1 in 6 Americans gets sick from foodborne illness each year (2014). Responsive food packaging can help alleviate these issues as foodborne pathogens are identified in real time thus preventing harmful consumption. Additionally, Responsive food packaging can prevent food waste as viable food is more easily identified by producers and consumers, thus saving the United States an estimated $165 billion annually (Gunders 2012). This review starts with a short description and update on active food packaging, followed by an overview of responsive materials and technologies specifically targeted to the food industry. Recent developments, perspectives and challenges in this field will be discussed. Additionally, this review will provide a framework for communication between food industry professionals, chemical engineers, and materials scientists.

Active Food Packaging Technologies

Before responsive packaging can be discussed, current research in active packaging must be addressed to highlight the differences between the 2 technologies and the major characteristics of each. Research in packaging that directly interacts with and affects the quality of food, also known as active packaging, has gained popularity due to recent advances in materials science and nanotechnology. The largest sectors of technological development include active packaging systems that release antimicrobials or antifungal compounds into food during storage to increase shelf-life and combat foodborne illnesses (Kruijf and others 2002; Duncan 2011). Unlike responsive packaging, active packaging operates without specific trigger mechanisms. This distinction is important as the design and goal of active packaging is inherently different from responsive packaging, and active packaging systems will operate whether or not a change is present in the food. For example, antimicrobial silver particles embedded in packaging films and gas releasing systems like equilibrium modified atmosphere packaging (EMAP) that utilize micro-perforated materials are considered active packaging due to the fact that they are not triggered by any change within the food product or package environment (Jacxsens and others 1999). However, biodegradable gels loaded with antimicrobials would be considered responsive due to the fact that they must be broken due to the biological activity of select organisms within the food product (bioresponsive). The same logic applies to compounds released by water-soluble polymers embedded in packaging films. These systems would be

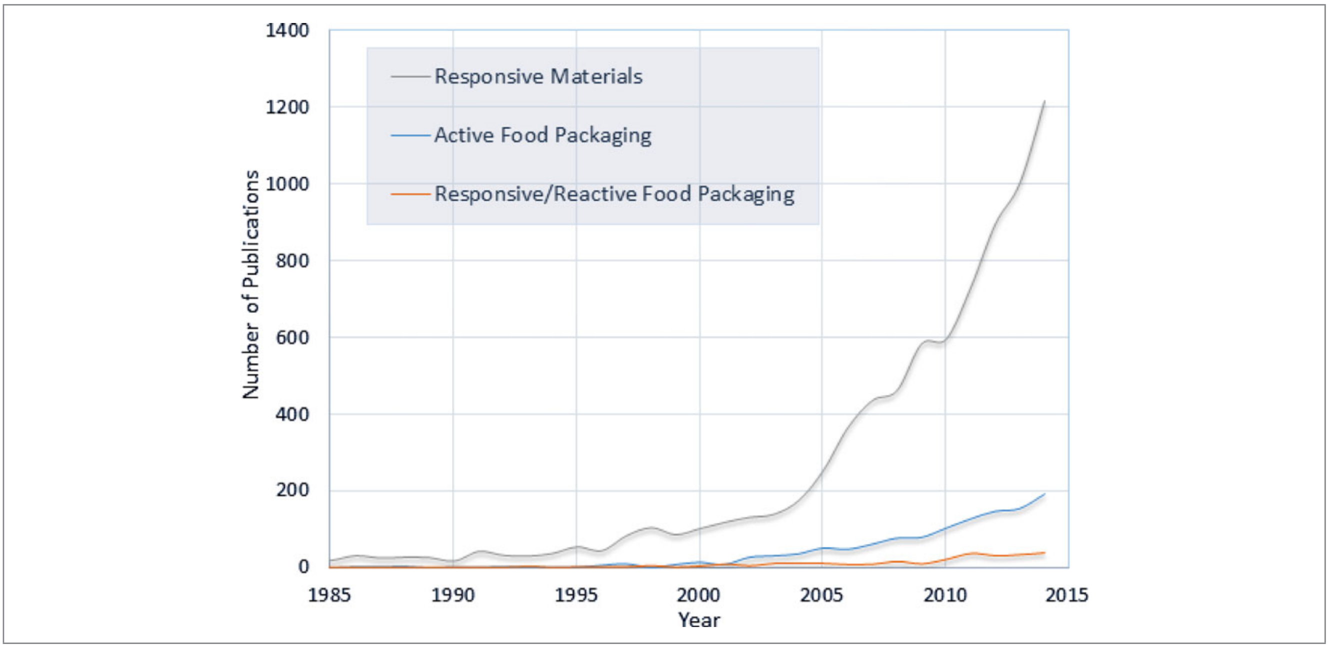

Figure 2–Publication trends in food packaging, responsive (reactive) packaging, and responsive materials from 1985 to 2014. Data collected from Scopus using the keywords "responsive materials," "active food packaging," and "reactive food packaging."

considered responsive (specifically chemoresponsive) due to the material choice. The difference between this type of system and a typical active system based on diffusion and migration of a soluble compound into the food product is subtle, but the distinction is important due to the engineered goal of the packaging. A diffusion-based active packaging system begins operating as soon as the food product comes into contact with the package and does not respond to any change in food quality or food safety. For example, a milk carton with a silver particle embedded film would be active packaging because the antimicrobial activity of the particles only depends on the diffusion of silver ions into the food product. An example of a responsive packaging system using water as a trigger would be sealed produce packages. A produce package with a water-soluble film could release antimicrobials into the food only after the build-up of moisture inside the package. Responsive packaging is discussed in great detail later in this review.

Active packaging can be broken down into 2 categories, chemoactive and bioactive, depending on the active agent. Chemoactive packages are designed to have an effect on the chemical composition of the food product or package headspace atmosphere and include modified atmosphere packaging, ethylene scavengers, and moisture control systems (Kruijf and others 2002; Kerry and others 2006). The term bioactive has been widely used and refers to materials or compounds that directly interact with biological molecules like bacteria and can influence biological processes (Cao and Hench 1996; Baker 2012). An example of bioactive food packaging is the incorporation of antimicrobial compounds into packaging films (Coma 2008).

The most common active packaging used by the food industry is modified atmosphere packaging (MAP) (Restuccia and others 2010). For MAP, air within a package is replaced with a gas mixture often composed of CO_2, O_2, and/or N_2 (Sivertsvik and others 2002). CO_2 is the most commonly used gas in MAP due to its antimicrobial properties as it effectively alters cell membranes, inhibits enzyme activity, and blocks microbial nutrient transfers (Sivertsvik and others 2002; Thompson 2010). These types of packaging systems are commonly used with meat products to improve shelf life (Kerry and others 2006). Researchers have worked to introduce many new materials into active packaging systems like nanoparticles, antimicrobials, and reactive films. A brief overview of these materials and their uses in active packaging is provided below.

Nanocomposites

Nanocomposites are polymer matrices reinforced with nanomaterials such as nanoparticles (Sozer and Kokini 2009). By definition these materials have at least one dimension on the nanometer scale (10^{-9}m) and are composed of a variety of materials including metal ions (such as silver, copper, and gold) and metal oxides (TiO_2, MgO) (Rhim and others 2013). Incorporation of nanoparticles provides many benefits including material reinforcement, antimicrobial properties, and vapor-sensing (Potyrailo and Naik 2013; Rhim and others 2013). Silver nanoparticles are common in antimicrobial films as they are effective against a wide variety of organisms (De Azeredo 2009; Becaro and others 2015). These particles operate through a variety of mechanisms including disruption of cell membranes, damage to bacterial DNA, and release of bioactive Ag^+ ions (De Azeredo 2009; Dallas and others 2011). Recently, these materials have been incorporated into petrochemical plastics, clays, and other materials (Jorda-Beneyto and others 2013; Keshavarzian and others 2014; Kim and Cha 2014; Bodaghi and others 2015).

Antimicrobial films

Antimicrobial compounds have also been incorporated into films for use in active packaging. These films are considered active because they rely on diffusion through the packaging medium as opposed to a triggered release of antimicrobials via responsive materials. Early research in this area utilized films incorporated with antibacterial/antifungal compounds like sodium benzoate and benomyl (Kruijf and others 2002). More recently, edible and inedible films have been explored that utilize natural antimicrobial ingredients like clove, pepper, cinnamon, coffee, and others (Seydim and Sarikus 2006; Kechichian and others 2010; Jo and others 2015; Manso and others 2015). Chitosan, another biologically derived material, has also been thoroughly researched due to its natural antimicrobial activities and its nontoxicity (Aider 2010; Lei and others 2014; Van Den Broek and others 2015). Direct implementation of antimicrobial enzymes like lysozyme and other biocatalysts in active packaging has also been explored (Fernández and others 2008). Very recently, bacteriophages have been embedded into acetate cellulose films for use against *Salmonella* Typhimurium (Gouvêa and others 2015). Cinnamon oil-embedded polymer films have also been recently produced at the pilot-scale to repel larger pests and insect larvae (Jo and others 2015).

Gas scavengers

Gas-scavenging packaging seeks to remove gases that cause degradation of food products and create inhospitable environments for microbial growth (Vermeiren and others 2003). For example, oxygen penetration into food is a major problem as it facilitates growth of aerobic bacteria, causes browning of meat products, rancidity of fats, and other undesired effects (Busolo and Lagaron 2012). Oxygen scavenging materials like photocatalytic TiO_2 nanoparticles, iron, and many other oxygen-reactive materials have been studied for use in packaging films (Xiao-e and others 2004; Miltz and Perry 2005; Busolo and Lagaron 2012; Di Maio and others 2015). Gas scavengers for ethylene, a produce ripening agent, have also been researched in order to increase the shelf-life of fruits and vegetables (Baker and others 1978; Terry and others 2007). Recently, packaging systems have been explored that actively release antioxidants to control oxygen levels within the package (Gómez-Estaca and others 2014).

Responsive Food Packaging Technologies

Design requirements

Responsive food packaging is based on the integration of a sensor or sensing interface on the packaging film for real-time and continuous quality monitoring. Such integration is implemented following 3 important design features: prevention of cross-contamination, selection of the target analytes, and choice of the transduction system.

Cross-contamination. Unlike the design of biosensors for blood, urine, or water analysis, food packaging sensors require the protection of the analyzed samples (food products) from any contamination that can be caused by the sensing process or sensor components. This can be achieved through the use of nontoxic, food safe sensing compounds, or by the incorporation of a porous separation membrane between the food and the sensing interface (Figure 3).

Food analytes. The primary challenge facing responsive food packaging systems is engineering responsive materials to respond to specific chemical or biological targets present in food. Within the food industry, rapid and accurate detection of these risks is imperative and many techniques are utilized at all levels in the food supply chain (Cho and others 2013). A great deal of research has been conducted to monitor and control these threats, and assessing food quality begins with detection of specific target compounds or analytes unique to spoilage pathogens or common contaminants (Velusamy and others 2010; Cho and others 2013). The choice of the target analyte for detection requires a prior understanding of the microbial agents involved, their occurrence in different products and conditions, and the reaction products released during the spoilage process. For example, food poisoning is, in part, caused by the ingestion of toxins or byproducts released during microbial food spoilage. The most common byproducts are biogenic amines such as tyramine, trimethylamine, 2-phenylethylamine, histamine, putrescine, cadaverine, spermine, spermidine, tryptamine, and agmatine (Ruiz-Capillas and Jiménez-Colmenero 2005; Bulushi and others 2009; Linares and others 2011). These amines result from the decarboxylation of amino acids in food (such as meat, fish, and milk) by bacteria (including pseudomonas and lactic acid bacteria). Hence, amines are very useful targets for responsive packaging due to their small size and chemical reactivity. Analyte size is an important factor to consider because only small molecules such as the microbial reaction products (biogenic molecules) will pass through the nanoporous barrier membrane discussed in the previous section. A short overview of other common food analytes can be found in Table 1. By incorporating triggers for these analytes directly into food packaging systems, food quality can be directly monitored by consumers or treated directly by reactive packaging.

Transduction system. Once analytes are identified and appropriately targeted, a transduction system must be put in place. A transduction system is a device that converts a signal or energy from one form to another (mechanical, electrical, chemical, magnetic, optical or thermal). In the case of biosensors, the transducer translates biological processes into measurable signals (Velusamy and others 2010). Within the food industry, transduction systems for sensing purposes come in many forms including electrochemical, optical, and colorimetric (Terry and others 2005; Serna-Cock and Perenguez-Verdugo 2011). Incorporating transducers into food packaging systems has been a daunting task due to the requirements for safety, performance durability, and the challenges related to the readout mode. Two main detection methods are emerging as potential tools for responsive food packaging: (i) colorimetric systems with naked-eye assessment. These systems utilize dyes, polymers, nanoparticles, or another medium to illicit a color change in response to a stimulus; (ii) spectroscopic systems that take advantage of changes in the optical properties of certain materials to produce a signal. These signals can then be analyzed with a variety of hand-held equipment such as Raman, infrared, or ultraviolet spectrometers. These signal-producing systems can be incorporated into entire packaging films whereby the entire package emits a color change or signal. Or, these systems can be placed strategically in the package in the form of sensor "chips" or "windows" which can then be visualized or scanned. The idea of a sensor "window" is depicted in Figure 3.

Advances in stimuli–responsive materials have recently revolutionized the sensing technologies these innovations are poised to influence food packaging systems (Lau and Wang 2013; Blum and others 2014). Triggerable packaging systems can identify and mitigate foodborne threats as they occur. Research and development of the specific mechanisms for identification and mitigation are discussed later in this review. Given the importance and potential of stimuli-responsive materials in food packaging not only for detection (transduction) but also for triggering alteration of the food product, those will be addressed in an independent section below.

Stimuli–responsive materials

Responsive materials can take many forms including self-assembled nanoparticles, hydrogels, supramolecular materials, surface-grafted polymers, and layered films (Hu and Liu 2010; Zelzer and others 2013). These materials are capable of exhibiting changes in physical or chemical properties in response to external stimuli including pH, temperature, ions, light,

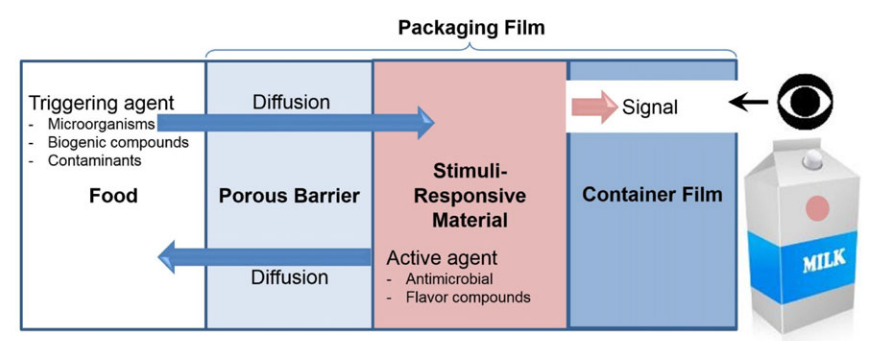

Figure 3–Schematic of responsive food packaging design. Upon diffusion of target analytes, the material triggers a signal visible or readable by the consumer, or releases corrective agents back into the food product.

Table 1–Overview of common target analytes used for sensing in food safety and quality monitoring

Analyte type	Microbial agent/molecule	Food product	Reference
Bacteria, viruses, parasites	*Escherichia coli*	Meat and dairy products	Silbert and others (2006), Pyun and others (1998), Johnson and others (2009)
	Salmonella groups B, D, and E	Range of foods	Bokken and others (2003), Zhao and others (2005), Crump and others (2011)
	Bacillus cereus	Milk	Ray and Bhunia (2007), Pickering and others (2012)
	Psychrobacter spp., Pseudomonas spp.	Refrigerated milk and raw meat	Davies and others (1998)
	Listeria monocytogenes	Processed meat and dairy products	Farber and Peterkin (1991), Heo and others (2014)
	Clostridium perfringens	Raw meat	Pickering and others (2012)
	Toxoplasma gondii	Swine	Gubbels and others (2003), Gebreyes and others (2008)
	Vibrio spp.	Seafood	Vora and others (2005), Talkington and others (2011)
	Staphylococcus aureus	Meat products	Lowy (1998), Le Loir and others (2003)
	Campylobacter spp.	Poultry and raw milk	Nelson and others (2007), Ge and others (2003)
	Shigella spp.	Range of foods	Sivapalasingam and others (2006)
	Norovirus	Range of foods	Glass and others (2009), Tuan and others (2010)
Chemical or biological molecules	Oxygen (redox compounds)	Milk	Cavallo and others (2014)
	Gaseous amines	Fish	Kuswandi and others (2012)
	Lactate oxidase	Wine and Yogurt	Serra and others (1999)
	Acetyl cholinesterase	Range of foods	Gulla and others (2002)
	Lead	Range of foods	Clarkson (1971)
	Mercury	Fish	Ye and Yin (2008), Pentreath (1976), Ely (1971)
Yeast, mold, mycototoxins	*Aspergillus versicolor, Penicillium chrysogenum*	Yogurt, cheese	Yong and Cousin (1995)
	Geotrichum candidum, Candida lipolyticum	Milk	Pitt and others (2009)
	Mucor circinoides, Rhodotorula mucilaginosa	Yogurt	Pitt and others (2009)
	Cladosporium cladosporioides, Debaryomyc es hansenii	Cheese	Deak (2007)

mechanical stressors, and biological molecules (Bajpai and others 2011). Triggered changes in steric conformation, conductance, charge, physical structure (shrinking/swelling), hydrophobicity, or chemical functionality, in turn, provide measurable responses that can be used for detection or for release of altering compounds (Hu and Liu 2010; Zelzer and others 2013). In this section we will discuss responsive materials research and the response mechanisms of these materials, along with their potential use in responsive food packaging.

Hydrogels. Hydrogels are composed of highly absorbent hydrophilic polymers that contain 90% to 99% water. They are composed of a variety of macromolecular building blocks including cross-linked polymers, thin films, and colloidal assemblies (Ulijn and others 2007). They are unique in that they experience macroscopic shrinking, swelling, or degradation from a variety of triggers including pH, temperature, and light (Hendrickson and Lyon 2009). These macroscopic changes occur as the intermolecular bonding sites within the gels are stressed or as the polymers units change conformation (Ulijn and others 2007). Upon shrinking/swelling, hydrogels can be designed to release embedded compounds, generate a detectable signal like fluorescence, or change specific optical properties such as diffraction and absorbance (Wang and others 2004; Liang and others 2009). Recently, hydrogel research has focused on making these materials sensitive to biological molecules for use in tissue engineering, biological sensing, and drug delivery systems (Ulijn and others 2007; Hendrickson and Lyon 2009). Bio-based hydrogels derived from xanthan gum, chitosan, carboxymethylcellulose, and other materials have been explored for use as sustainable, biodegradable packaging films (Silvestre and Cimmino 2013; Biscarat and others 2015; Gregorova and others 2015). The use of functionalized hydrogels in responsive packaging systems is discussed later in this review.

Surfaces. Stimuli–responsive surfaces are composed of responsive materials embedded on substrates like silicon oxide or gold (Mendes 2008). These surfaces have gained research interest due to the fact that they can be widely applied in biosensors, micro- and nanofluidic devices, electrochemical sensors, and biomedical devices (Nath and Chilkoti 2002; Mendes 2008). These surfaces often exist as self-assembled monolayers (SAMs) or polymer films and can be tuned to respond to biological molecules (Bossi and others 2001). Specifically, surfaces operate by directly immobilizing biological molecules, incorporating biological recognition elements, or incorporating electrochemically active materials (Mendes and others 2007; Mu and others 2007). Currently these systems are primarily used for sensing purposes, but functionalized polymeric films are popular in antimicrobial active packaging (Cunliffe and others 1999; Quintavalla and Vicini 2002).

Particles. Particle-based responsive systems are based upon interactions of particles in solution with a particular target or other agent. When a stimulus is presented, particles will aggregate or change their size thus changing their optical properties or resulting in a color change visible to the naked eye (Saha and others 2012; Stoffelen and others 2014). Unlike nanocomposites that utilize nanoparticles as antimicrobials and structural support, particle-based responsive systems use highly functionalized interactions that can be used in high-specificity sensors, self-assembled materials, catalysis, and other applications (Daniel and Astruc 2004; Nie and others 2010; Saha and others 2012). Particle-based responsive systems have not been incorporated into responsive food packaging systems yet, most likely due to the success of active nanocomposites and health concerns related to metal nanoparticles (Elsaesser and Howard 2012). Since responsive systems require contact between the food product and the responsive material, contamination and potential toxicity are the major hindrances for using inorganic nanoparticles in food packaging technologies today.

Supramolecules. Supramolecular materials are characterized by self-assembled, non-covalent, spatially organized molecular units (Lehn 2002; Zelzer and others 2013). Supramolecular chemistry has been a very popular research topic in the past 2 decades and supramolecular materials have been developed in the form of gels, vesicles, micelles, nanoparticles, and polymers (Blanazs and others 2009). They can be engineered to assemble, dissemble, change conformation, or affect chemical functionality in response to biological stimuli (Blanazs and others 2009). Like other responsive materials, these changes produce measurable signals (colorimetric response, change in optical properties, and so on) or trigger the release of compounds (Apostolovic and others 2010). Supramolecular building blocks have been widely investigated and are discussed above in this review. Other types of supramolecules, particularly polymers, are gaining popularity and can potentially be applied to recycling systems and finely tuned materials (Aida and others 2012). One example of signal-producing supramolecular polymer is polydiacetylene. Polydiacetylenes are π-conjugated polymers that exhibit a visible color change in response to external stimuli including pH, solvents, temperature, and biological molecules (Silbert and others 2006; Pires and others 2011). This color change is facilitated by the change in conformation of the conjugated backbone of the polymer caused by steric interactions between head groups on the chain itself. These materials are currently used in regenerative medicine, electronics, and biological sensing (Hirst and others 2008). These polymers have been utilized for sensing purposes and have been explored for use in smart packaging systems (Hill and others 2013).

Current Applications in Responsive Food Packaging

Another challenge to responsive food packaging is the incorporation of responsive materials into the packaging system. Responsive systems must be made food-safe, accurate, and must be produced cheaply with minimum impact on the packaging fabrication chain. In this section we review current responsive packaging research.

Bioresponsive packaging systems

The use of bioresponsive technologies in food packaging provides a unique "biology to material" communication (Ulijn and others 2007). Additionally, bioresponsive materials can be applied to both responsive packaging systems as biologically triggered sensors and chemical release systems. In this section we present an overview of bioresponsive technologies ideal for food packaging. Enzymes have emerged as a popular biomolecule for sensing; drug delivery, and other applications, and many enzyme-responsive materials (ERMs) have been designed in past decade. Materials can be considered enzyme responsive if they change their functionality as a result of the direct action of an enzyme (Zelzer and others 2013). Such changes include cleavage of a protective group, structural rearrangement, and/or triggered self-assembly (Zelzer and others 2013). For example, enzymatic activity of alkaline phosphatase was effectively monitored via biodegradable polymers by Tanaka and others (2010). Upon digestion with alkaline phosphatase the synthesized polymer released fluorescein which could then be easily seen with the naked eye under ultraviolet light. The authors did not discuss the efficacy of this polymer for food packaging applications. Also, Schneider and others (2011; 2012) have designed multiple hydrogels that can be broken down via cellulase enzymes from *Aspergillus* molds and bacteria in order to release a red signaling dye. As discussed by the authors, these hydrogels have a potential to be applied to food packaging systems, but no tests were conducted using food or food packaging. A DNA-embedded responsive hydrogel was recently developed by Shin and others (2014) for the detection of viruses. Upon introduction of the virus, the DNA strands within the gel melted allowing for macroscopic changes in the gel. This type of system can potentially be applied to foodborne viruses like norovirus. Another bacterial sensor using color-changing polydiacetylenes incorporated into a reactive peptide membrane has been created by Pires and others (2011). This sensing approach was accomplished by inoculating the polydiacetylenes with bacterial supernatants. Color change was caused by the breakdown of the peptide membrane by bacterial toxins. These sensors were also successfully incorporated into cellulose paper substrates for food packaging purposes (Pires and others 2011).

Chemoresponsive packaging systems

We define chemoresponsive packaging systems as systems triggered by nonfood chemicals. These chemicals include contaminants and non-biological byproducts of microbial or fungal activity like acids or gases.

Contaminants. A number of chemical toxins or contaminants can be introduced into food during production, processing, transportation, or storage. A common example of a chemical contaminant is the food packaging disinfectant hydrogen peroxide (H_2O_2 vapor). Sanchez and Trogler (2008) developed a boronate-based polymer for the detection of H_2O_2 through a fluorescent response caused by peroxide driven oxidative cleavage of the boronate functional groups. However, direct incorporation of this sensor into packaging needs first to address issues related to polymer stability and potential safety concerns.

Biogenic chemicals. Other systems have been designed that interact with molecules present as a direct result of biological activity. While not considered discretely bioresponsive, these materials have great potential for food packaging applications, specifically the detection or mitigation of biological activity in food. Hydrogels have been tuned with fluorophores in response to specific biogenic ions like Cu^{2+}, K^+, and $SO_4{}^{2-}$ thus enabling their detection (Onoda and others 2007; Yin and others 2009). For biogenic acids and bases, pH-triggered hydrogels have been designed for the release of antimicrobial compounds by Fuciños and others (2014a,b). Upon addition of acid, the gels collapse and release embedded pimaricin. The results suggest that these gels could have great potential for responsive packaging applications.

Biogenic gases. The most popular use of responsive materials in food packaging is the monitoring of gaseous compounds present in the headspace of the package. Depending on the type of food present in the package gases like O_2, CO_2, and ethylene can be very good indicators of spoilage as they are produced or consumed by bacteria. Using this platform for sensing eliminates concerns over proper sampling, cross-contamination, and transducer choice. The major drawback of these sensors is specificity (cannot identify particular spoilage agents) and sensitivity. A colorimetric polypropylene sensor has been developed by Cavallo and others (2014) for the detection of reducing substances produced by spoilage bacteria in milk. In this sensor, the dye bromophenol blue was made colorless as oxygen was removed by bacteria. In another work by Kuswandi and others (2013), a blue to green color change of bromophenol blue (BPB) was used as an in-package colorimetric indicator of volatile organic compounds like acetic acid present during guava spoilage. Other research groups have developed in-package sensors for fish spoilage by measuring the release of volatile organic compounds by spoilage bacteria (Pacquit and others 2007; Huang and others 2011; Kuswandi and others 2012). Specifically, fish spoilage odors consist of C6–C9 alcohols and volatile amines like trimethylamine, dimethylamine, and ammonia. Sensing was accomplished by incorporating dyes for the specific volatile compounds or tracking headspace pH and relating spoilage to total viable basic nitrogen (TVB-N) (Pacquit and others 2007; Huang and others 2011).

Detection of carbon dioxide in modified atmosphere and conventional packaging has also gained considerable attention (Puligundla and others 2012; Meng and others 2014). For example, pH responsive materials (bromothymol blue and methyl red) were incorporated into dessert packages to measure spoilage via headspace CO_2 monitoring (Nopwinyuwong and others 2010). Headspace CO_2 was also measured by a combination of a pH indicator and phosphorescent reporter dye to boost sensitivity (Borchert and others 2013). Chitosan was also used as a CO_2 sensor by utilizing its aqueous solubility in different CO_2-rich atmospheres (Jung and others 2012). When combined with an indicator, the transparency of the chitosan solution, as a result of higher chitosan solubility, was related to CO_2 concentration. Other research has been conducted using oxygen indicators in food packaging (Mills 2005). For example, Lawrie and others (2013) have developed a printable oxygen indicator that utilizes oxidation of a photocatalyzed dye. When exposed to UV light the blue dye becomes bleached and color recovery is then related to oxygen content. A similar system was developed by Vu and Won (2013) but was incorporated with an aliginate polymer to prevent dye leaching from colorimetric films. Another indicator based on electrochromic polyviologen films has also been developed (Roberts and others 2011). Upon addition of an electronic pulse the films change from pale to highly colored surfaces. This change in color is in turn affected by reduction of O_2. These films have a distinct advantage over simple dye-based sensors due to the fact that they are more sensitive and are less susceptible to leaching into the food product.

Thermoresponsive and mechanoresponsive systems

Thermoresponsive systems respond to changes in temperature often with a color change visible to the naked eye. A particular type of thermoresponsive materials used in food packaging are the time-temperature indicators (TTIs) that indicate both a change in temperature and the length of time the product was exposed to the targeted temperature (Wanihsuksombat and others 2010). TTIs have been developed to expressly monitor temperature of packaged food products and are the most commonly produced responsive packaging technology. These sensors can be based on enzymes, nanoparticle reactions, or reactive materials and are designed to indicate if a food product has been introduced to temperature conditions favorable for fungal/microbial growth (Wanihsuksombat and others 2010; Kim and others 2012; Pereira Jr and others 2015). Additionally, many advances have been made recently on other types of thermoresponsive materials. Unlike TTIs, these systems are designed to change color or release compounds once a target temperature is reached instead of tracking both temperature and the time spent at that temperature. Although the majority of these materials are not designed for food packaging purposes (Roy and others 2013; Yaseen and Lu 2013), recent reports show promising perspectives of temperature triggered materials in food packaging (Fuciños and others 2014a).

Another sensing system that is of interest to the responsive food packaging industry includes mechanoresponsive systems. Food packages are commonly dropped, mishandled or otherwise compromised during transportation. Mechanoresponive packaging could provide a sensing response to ensure that these packages are removed before being distributed to consumers. Mechanoresponsive materials respond to applied mechanical force and have been utilized to activate chemical reactions, assemble/dissemble small molecules, or mechanically break down chemical bonds (Weder 2011). For example, mechanoresponsive crystals have been developed that change the color of their luminescence when forces like shearing or elongation are applied (Sagara and Kato 2009). Other research has focused on mechanically-induced chemical changes in materials (Caruso and others 2009). Mechanoresponsive systems have yet to practically be applied to food packaging, but polydiacetylene based impact sensors have already been explored by Hill and others (2013).

Corrective responsive food packaging

Corrective responsive packaging systems utilize changes in stimuli-responsive materials (shrinking, swelling, changes in chemical functionality, dissolution, self-assembly, and more) to trigger the release of compounds. As evidenced by the above sections, informative responsive systems have dominated research and development with respect to the food packaging field likely due to regulations, costs, and other challenges regarding the introduction of curative agents into food products. More information regarding these challenges can be found below. Another major challenge to corrective responsive packaging is the popularity of controlled release active packaging systems (Buonocore and others 2004). Controlled release active packaging utilize polymer blends or other composite materials to manage diffusion of antimicrobials into packaging. This approach is preferred for release systems in food packaging and research in this area has grown over the past decade.

While corrective responsive systems are not widely explored in food packaging, development of these technologies is, however, very popular in the medical field. One specific technology that has potential in food packaging is the use of biodegradable polymeric particles for drug delivery. Briefly, drugs are enclosed in hollow nanosized capsules based on biodegradable materials. These capsules serve to protect the drugs from being broken down in the body allowing the encapsulated drugs to be released slowly or released in a specific location in the body. Nanoencapsulation systems have been explored for the delivery of antimicrobials, flavor compounds, and vitamins to food products (Donsì and others 2011; Fathi and others 2012). These capsules have not been incorporated into food packaging systems, but nanocapsules have been formulated with popular packaging materials like poly(lactic) acid (PLA) and chitosan (Siracusa and others 2008; Dutta and others 2009; Kumari and others 2010). However, these capsules like the capsules for drug delivery are formulated to facilitate controlled (active) rather than triggered (responsive) release of compounds. This technology becomes interesting for responsive packaging when nanocapsules are formulated from stimuli-responsive materials. Recently, nanocapsules have been formulated that respond to light, pH changes, temperature, and enzyme interactions (Fleige and others 2012). Once triggered these capsules rupture and release their drug payloads. These stimuli-responsive nanocapsules can have great potential in responsive packaging as release mechanisms for flavoring agents, antimicrobials and other compounds triggered by events that occur inside or outside of the food package. We expect advances in this technology in the food packaging sector as specific triggering mechanisms are established and advances are made in the synthesis of these materials.

Perspectives and Challenges

Current challenges in responsive food packaging

Efficiency. The first criterion in the development of responsive food packaging is the performance of the sensing mechanism, including sensitivity, limit of detection, and operational range. Beside the analytical parameters, false positive reactions are another important factor than needs to be minimized to avoid unnecessary food recalls. One of the major fields of research in sensing in general and responsive packaging in particular is the development of signal amplification systems to allow the detection of microorganisms and contaminants at an early stage of spoilage or contamination, which would then allow curative or preventive responses. The sensor must also be reliable in order to gain consumer trust in the product. The purpose of in-packaging sensors is to communicate food safety information directly to consumers. Finally, sensors that are broadly applicable to multiple threats will be more cost-effective and industrially competitive than systems that only detect one analyte.

Safety and regulations. Unlike the development of sensors and responsive surfaces for medical and environmental applications, responsive food packages require strict safety considerations (Muncke 2014; Williams 2014; Feichtinger and others 2015; Parisi and others 2015). The sensing surface needs to be in contact with the food to detect the targeted changes, without inducing changes to the food or releasing contaminants into it. This important requirement disqualifies the use of many traditional and effective sensing materials such as metal nanoparticles or carbon nanomaterials. Polymers and organic biocompatible materials will be increasingly used in research for the development of new packaging technologies as they circumvent safety issues and provide reasonably efficient sensing mediums.

Manufacturing. The translation of responsive packages at the industrial scale is barred by 2 major requirements: (i) the sensors or responsive labels need to be stable under the stress caused by the package manufacturing process including temperature, pressure, mechanical strain, and processing chemicals; (ii) the label needs to be integrated in a time- and cost-effective fashion by causing minimal changes to the existing production chain. Sensing labels that can be printed or directly attached to packaging substrates without major modification will be most successful in this regard.

Impact on the environment. The trend towards sustainability has greatly impacted the food industry (Baldwin 2011). Food packaging is no exception and today the industry is pressured by laws and regulations to adopt sustainable practices and utilize renewable materials. Also, concerns over plastic pollution in the oceans and public health issues with plasticizers like bisphenol A (BPA) have also had a major effects on the disposable plastics market (Staples and others 1998; do Sul and Costa 2014). As a result of these pressures, we expect to see convergence towards paper-based packaging and packaging based on renewable biomaterials. Thus, research on responsive packaging will be directed towards integration of sensors on paper and other renewable biomaterials (Yam and Lee 2012; Peelman and others 2013). Additionally, bio-based plastics have been explored for use as bio-nanocomposite materials for food packaging (Rhim and others 2013; Reddy and others 2013; Ghanbarzadeh and others 2014; Ahmadzadeh and others 2015). Bio-based active packaging materials like chitosan and plant-based essential oils have also drawn considerable attention from researchers due to their availability and ability to be further modified with natural, bioactive compounds (Khan and others 2014; Rodríguez and others 2014; L Mateescu and others 2015). Research in biodegradable food packaging materials has also increased over the past decade in hopes of reducing the waste generated in the packaging stream (Yates and Barlow 2013; Yu and others 2014; Martino and others 2015). Another important consideration is the impact of sensor media on material recycling, composting, and disposal.

Perspectives and future trends

Advanced materials and systems. Research in responsive materials has increased exponentially over the past 2 decades and the materials are likely to be incorporated into food packaging in the near future. In addition to the materials discussed earlier in this review there are many others that can be applied to responsive food packaging systems, specifically printed electronics and self-immolative materials. Printed electronics utilize common printing methods (screen printing, Inkjet, and so on) to deposit electrical devices on substrates. Common examples within the food industry include radiofrequency identification (RFID) tags and electronic noses. RFID tags have emerged as a great alternative to common barcodes due to their ability to incorporate a large range of information into a scanned code (Want 2006). Unlike traditional barcodes, information can be added or removed from an RFID tag even after it is printed/ installed. RFID devices exist in 2 forms, battery-powered (active) or those with no battery (passive) that are activated by the reader. Passive tags have major potential in food packaging systems because they provide unique, modifiable information on individual packaged units. Large-scale incorporation of RFID tags into food packaging systems has been limited, but significant developments have been made in recent years (Costa and others 2013; Vanderroost and others 2014). We classify RFID tags as informative packaging, but these tags have potential in responsive packaging as well. Very recently, a few research groups have combined RFID tags with sensors in food packaging. By combining RFID systems with polymers that react to food analytes like biogenic amines, a detectable signal can be acquired based on the change in electronic potential of the RFID (Fiddes and others 2014). Another sensor was prepared using responsive RFID tags to detect other vapors like water, ethanol, ammonia, and toluene (Fiddes and Yan 2013). By combining RFID systems with responsive materials, labels could not only provide up-to-date information on the contents of the food package, but also the quality of the food within the package.

Other research has focused on electrochemically active systems for food package monitoring. These systems, referred to as electronic noses, are small semiconductor systems that detect biological molecules through changes in sensor mass, conductance, or optical properties (Sozer and Kokini 2009). They have potential in informative packaging and have primarily been used to detect food odors and biogenic gases associated with spoilage in packaged meat products (Panigrahi and others 2006; Rajamäki and others 2006). Very recently, these sensors have been combined with colorimetric materials for visual detection of freshness. (Huang and others 2014) have developed a colorimetric sensor array for monitoring biogenic amines produced by spoilage of pork. Very recently, similar sensors have been developed for other pork products and boiled marinated turkeys by and (Salinas and others 2014aa,b). An electrochemical sensor for *E. coli* has been developed on a flexible sheet by (Basu and others 2014). These types of sensors have great potential for use in modified atmosphere packaging, and with further development these sensors can be used within a package.

Another type of advanced material with potential application in food packaging is self-immolative materials. Very recently, self-immolative polymers have emerged as a new form of tunable polymers that can be actively triggered by a variety of stimuli (Phillips and DiLauro 2014). These polymers consist of a series of monomers with a specific end cap. When this end cap is cleaved the polymer undergoes head-to-tail depolymerization. Research in this field has yielded many varieties of polymers with unique, responsive end caps. Bioresponsive self-immolative polymers have been recently developed as well. β-glucuronidase activity was successfully monitored by (Grinda and others 2012). Additionally, an active release of the drug Paclitaxel was accomplished from an enzyme-triggered self-immolative polymer designed by (Erez and others 2009). These materials could be utilized as high-sensitivity sensors or could be used as a trigger mechanism for the release of compounds in responsive packaging. These materials are not currently utilized in food packaging due to complicated synthesis operations and high cost.

Market. The global market for smart, active, and intelligent packaging was projected to grow rapidly between 2010 and 2015 from 15.7 million to 23.5 billion (marketsandmarkets.com 2011). This growth was stimulated by increased demand for modified atmosphere food packaging and time temperature indicators for both food and medical products. Major corporations active in this area include Multisorb Technologies (U.S.A.), Sealed Air Corporation (U.S.A.), M & G USA Corporation, and Amcor Limited (Australia) among others. These companies specialize in active packaging including oxygen absorbers, MAP, and TTIs.

A recent study conducted by IDTechEx estimated only the growth of smart (ergonomic) packaging without the inclusion of active packaging. The smart packaging market was estimated to grow from 75 million to 1.5 billion from 2013 to 2023 (Das and Chansin 2013). When active packaging was excluded, TTIs dominated the smart packaging market. With an increase in smart packaging growth, comes a significant increase in the number of companies dedicated to smart packaging, specifically responsive packaging. New Zealand-based *Ripesense Limited* has developed a color-changing sensor (also called Ripsense) that responds to volatile organic compounds released from pears. Other sensor-based responsive food packaging labels have been marketed by Insignia Technologies (Scotland). Freshpoint (Switzerland) produces TTIs (marketed as CoolVu^{TM} and OnVu^{TM}) and oxygen sensors marketed as O_2Sense^{TM}. These sensors include CO_2 monitors and temperature-sensitive labels. TTIs have also been developed by larger companies, such as 3M marketing TTIs directly under the name MonitorMark^{TM}.

Conclusions

Current research in bioresponsive and stimuli-responsive materials is expected to translate into reactive food packaging over the next few years. Commercialization of these materials in food packaging will be possible as new technologies become more reliable and cost-effective. Currently, a very limited number of companies produce responsive packaging systems as compared to active packaging companies. However, with the recent advances discussed in this review, and the increasing demand from consumer and regulatory agencies, we expect the responsive packaging market to grow steadily over the next decade. Furthermore, responsive packaging will have a tremendous impact on many aspects of the food industry by reducing spoilage, food waste, food recalls, and foodborne illness outbreaks. The dairy industry, in particular, will benefit from new functional packaging. Beyond the food industry, the growth of responsive packaging is expected to have an impact on other market segments as well, namely the paper, plastics and the bioplastics industries.

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The authors are not aware of any affiliations, memberships, funding, financial holdings or any other conflicts of interest that might be perceived as affecting the objectivity of this review.

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