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Review

Hydrogel as an alternative structure for food packaging systems

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ABSTRACT

Hydrogels are three-dimensional, hydrophilic networks, comprising polymeric chains linked through physical or chemical bonds. In the area of food, hydrogels have great potential to be used in food packaging systems or as carriers of bioactive components. This paper reviews the nature of hydrogels, their 3D network conformation, their functional properties, and their potential applications in food packaging systems. Regarding their potential food packaging applications, hydrogels can present a conformation which allows their use as part of a packaging system to control the humidity generated by food products with high water content. Moreover, the incorporation of nanoparticles into hydrogels may grant them antimicrobial activity. Finally, although the current research in this field is still limited, the results obtained so far are promising for innovative and potential applications in the food field, which also include their integration into intelligent food packaging systems and their direct incorporation into food matrices as a flavor carrier system.

1. Introduction

Recently, bio-based hydrogels have been applied to food packaging. However, the use of superabsorbent polymers as bio-based hydrogels in the food industry remains limited. There are increasing prospects and interest to develop bio-based hydrogels and innovative technologies that reduce the use of fossil fuel related to conventional non-biodegradable petroleum-based materials (Rahman, Netravali, Tiimob, Apalangya, & Rangari, 2016; Rhim, Park, & Ha, 2013; Santana et al., 2017). In recent years, the industry has shifted toward the enhancement of the performance of polymers for food packaging applications, through the improvement of their biodegradability, their swelling properties, and their mechanical and thermal properties. Hydrogels are polymeric matrixes with the ability to absorb water or any water-compatible fluids up to 100% of their weight. Moreover, superabsorbent hydrogels are a kind of hydrogel which are constituted by polymers that can absorb over 100% and up to thousands of times their dry weight in water (Feng, Ma, Wu, Wang, & Lei, 2014).

The main role of these materials in food packaging is humidity control inside of a packaging. Their potential application in the food preservation field include to remove the moisture or exudates generated inside a package because of vegetables transpiration and water loss caused by physicochemical changes in packaged foods, or by the permeation of water vapor due to environmental conditions, especially, in the case of fresh foods (Ahmed, 2015; de Azeredo, 2013; Rooney, 1995). Thus, the use of hydrogel can decrease water activity (A_w), slowing down the growth of mold, yeast, and spoilage bacteria on foods such as ready-to-eat meals (e.g., freshly cut vegetables) and hygroscopic products (e.g., powder food), or to decrease the softening of dry crispy products (e.g., fried potatoes, biscuits) (de Azeredo, 2013).

To increase hydrogel applications, diverse materials and methodologies have been studied. Most studies focused on improving specific features of the polymers used to prepare hydrogels, as well as decreasing their environmental impact by finding biodegradable alternatives (Macieira et al., 2010; Rodríguez-Vázquez, Vega-Ruiz, Ramos-Zúñiga, Saldaña-Koppel, & Quiñones-Olvera, 2015; Thombare, Jha, Mishra, & Siddiqui, 2016; Zhao et al., 2009). In this regard, a considerable number of papers have been published between 1996 and

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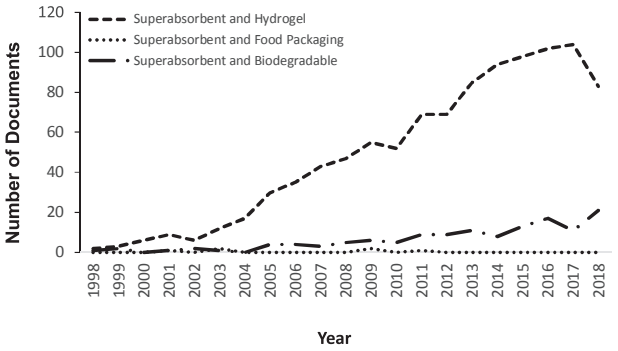

Fig. 1. Number of scientific documents (research and review articles) retrieved from the Web of Science™ Core Collection between 1998 and 2018, using the descriptors "superabsorbent", "hydrogel", "food packaging", and "degradable".

2016 (Fig. 1).

The use of hydrogel comprises an interesting strategy for food packaging systems, especially for the control of water content, which greatly limits food durability. In this regard, the development of hydrogels based on superabsorbent polymers allows the creation of alternative packaging systems (Gregorová, Saha, Kitano, & Sáha, 2015; Otoni, Espitia, Avena-Bustillos, & McHugh, 2016).

According to function, food packaging can be classified into three groups: traditional, active, and intelligent packaging (Fig. 2). Traditionally, the purpose of food packaging is containment, protection, convenience, and communication. Active packaging interacts directly with the contained food product or with the headspace to reduce or inhibit microbial growth, which leads to the extension of shelf-life and/or the preservation of the food's sensory characteristics.

On the other hand, intelligent packaging communicates quality and safety information to the consumer (Cierpiszewski, 2016; Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback, 2008). Despite the technological advances in active and intelligent packaging, its use is associated with health risks (Barska & Wyrwa, 2017; Cierpiszewski, 2016), which could be mitigated through the use of natural products.

Although studies on food shelf-life extension using hydrogels from natural polymers are limited, this approach is promising, since the features of these materials may add considerable value to packaged products due to their biocompatibility and low toxicity (Caló & Khutoryanskiy, 2015; Wang & Wang, 2009). In this regard, the integration of hydrogels into packaging systems constitutes an evolving and eco-friendly alternative technology.

Improving food quality, safety, and stability, while reducing the environmental impact is challenging, but in this direction, some promising alternatives have been well targeted. Currently, the development of active packaging systems from low-cost, renewable, natural compounds (polysaccharides, proteins) has gained interest and increasing consumer acceptance, which has generated new potential applications within the food industry. However, the most humidity-absorbent material currently incorporated in food packaging systems are based mainly on sachet technologies (from non-biodegradable sources) that can pose health risks and that have a negative impact in the environment (Guilherme et al., 2015; Ozdemir & Floros, 2004; Ozdemir, 1999).

This review presents the fundamentals of hydrogels conformation, their functional properties and potential application as part of food packaging systems. Finally, the future trends regarding the integration of hydrogels into active packaging systems for food preservation are presented.

2. Hydrogel nature, definition and classification

Hydrogels are three-dimensional networks of polymeric chains, randomly crosslinked by physical or chemical bonds (Ahmed, 2015; Chang, Duan, Cai, & Zhang, 2010; Kalia, 2016; Pourjavadi, Harzandi, & Hosseinzadeh, 2004). They can present a linear or branched structure and are characterized by the ability to absorb large amounts of water or biological fluids, thus, balance swelling and form insoluble structures due to their three-dimensional network (Hebeish, Hashem, El-Hady, & Sharaf, 2013).

Hydrogels are defined as colloidal gels, as they disperse in water and present viscoelastic and structural properties due to the behavior of the polymeric chain bonds and to the solvents incorporated into the system (Ahmed, Aggor, Awad, & El-Aref, 2013; Laftah, Hashim, & Ibrahim, 2011). The three-dimensional structure of hydrogels comprises two phases: a liquid phase, usually water or biological fluids, and a solid phase, based on a polymer or polymeric blends. In the swollen state and when exposed to certain pressures, its three-dimensional structure allows the absorption of target fluids without structural changes (Bao, Ma, & Li, 2011; Croisier & Jérôme, 2013). The water sorption capacity of hydrogel depends on several factors, such as the nature and density of the used polymer and crosslinks (Buwalda et al., 2014).

Some of the most important hydrogels performance features include mechanical resistance, swelling potential, and moisture retention capacity (Chen, Tang, Liu, & Tan, 2016; Guilherme et al., 2015; Gulrez, Al-Assaf, & Phillips, 2011; Ng, Yeong, & Naing, 2016). These properties are of interest to several industrial sectors and have contributed to the development of several common applications such as cosmetics, wastewater treatment, tissue engineering, drug release, biosensing, agriculture, and biomedicine (Alves & Mano, 2008; Caló & Khutoryanskiy, 2015; Chang & Zhang, 2011; Chen et al., 2016; Dash, Chiellini,

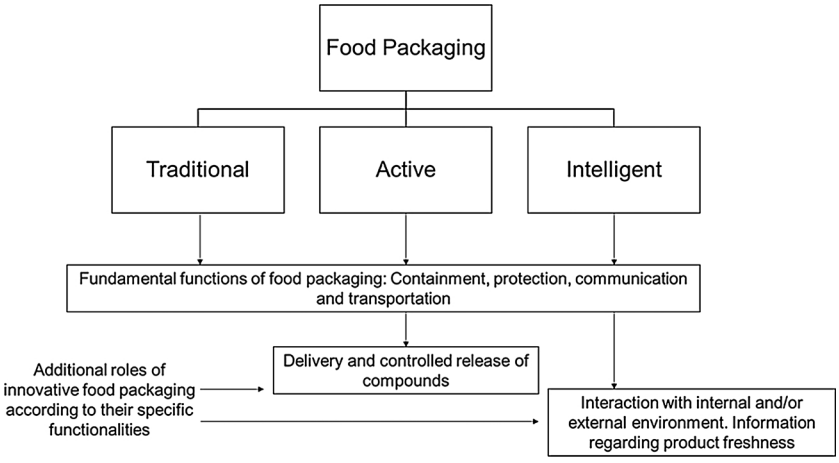

Fig. 2. Main functions of food packaging.

Table 1
Classification of hydrogels.

Main Feature	Classification	References
Nature of the polymeric matrix	• Natural • Semi-natural polymers • Synthetic • Hybrid	Bansal et al. (2011), Bao et al. (2011), Ma et al. (2015), Cuadri et al. (2016), Demitri et al. (2016), Feng et al. (2014), Guilherme et al. (2015), Huang et al. (2015), Chen et al. (2015), Thombare et al. (2016), Wang and Wang (2010), Wang et al. (2013), Zheng et al. (2007), Zohuriaan-Mehr et al. (2009), Salimi & Kurdtabar (2009)
Crosslink	• Chemical • Physical	Ranjha & Khan, 2013)
Biodegradability	• Biodegradable • Non-biodegradable	Byard et al. (2017), Feng et al., 2014, Lee et al. (2000), Pourjamal et al. (2016)
Polymeric composition	• Homopolymers • Copolymers • Multicomponents	
Physical appearance	• Sponge • Tablets • Film • Multiparticulated system (granules, microspheres, nanospheres)	Abaee et al. (2017), Byard et al. (2017), Feng et al. (2014), Lee et al. (2000), Pourjamal et al. (2016)
Electrical charge	• Neutral/nonionic nature • Ionic	Jensen and Lura (2006)
Stimulus responsiveness	• Sensitive to chemical factors, such as pH, antigens, and enzymes. • Sensitive to physical factors such as temperature, pressure, photo irradiation, and electric/magnetic field.	Argin et al. (2014), Berger et al. (2004), Bueno et al. (2013), Chang et al. (2010), Kalia (2016)

Ottenbrite, & Chiellini, 2011; Deligkaris, Tadele, Olthuis, & van den Berg, 2010; Feng et al., 2014; Hebeish et al., 2013; Hoare & Kohane, 2008; Nascimento Porto Neto et al., 2017; Nunes et al., 2016; Rodríguez-Vázquez et al., 2015; Souza et al., 2013).

Regarding the formation of hydrogel matrices, most hydrogels are produced from synthetic compounds, and their matrices are predominantly bound by chemical crosslinking. The most common compounds are petroleum derivatives, including polyacrylamide, poly (sodium acrylate), poly (acrylic acid), and polyvinylpyrrolidone (Kabiri, Omidian, Zohuriaan-Mehr, & Doroudiani, 2011; Ullah, Othman, Javed, Ahmad, & Akil, 2015), and efforts have been made to increase hydrogel production from bio-based and biodegradable polymers.

Different classification criteria might be used to distinguish different Hydrogels. They might be classified according to their sources in renewable and non-renewable, which is different from a classification that considers their nature (natural and synthetic). Other hydrogel classifications include: a) the nature of the polymeric matrix, b) crosslinking type, c) biodegradability, d) composition, e) physical appearance, f) electrical charge, and g) stimulus responsiveness (Table 1).

2.1. According to the nature of the polymeric matrix

Hydrogels can be prepared using natural macromolecules, synthetic polymers or a mix of both, in which case they are designated as hybrid hydrogels (Demitri, De Benedictis, Madaghiele, Corcione, & Maffezzoli, 2016). Most available hydrogels are produced from synthetic sources, due to their mechanical and chemical properties, which confer a wide application spectrum. On the other hand, hydrogels produced from natural polymers have higher biocompatibility and biodegradability and lower toxicity than synthetic ones. Consequently, the production of hybrid hydrogels has increased significantly, bringing together the desirable characteristics of both. The use of copolymers and multicomponent blends is a strategy to design hydrogels with controllable, resistant, and nontoxic properties (Huang, Li, & Zhou, 2015).

According to Zheng, Li, Zhang, and Wang, (2007), the main classes of natural polymers used to produce hydrogels are: i) proteins such as collagen, gelatin, soy, and fish proteins (Zohuriaan-Mehr, Pourjavadi, Salimi, & Kurdtabar, 2009); and ii) carbohydrates such as starch (Chen et al., 2015), cellulose and its derivatives (Bao et al., 2011; Ma, Li, & Bao, 2015), sodium alginate (Wang & Wang, 2010), guar gum (Thombare et al., 2016), xanthan gum (Feng et al., 2014), and chitosan and its derivatives (Bansal, Sharma, Sharma, Pal, & Malviya, 2011; Cuadri, Bengoechea, Romero, & Guerrero, 2016; Guilherme et al., 2015; Wang, Huang, Kang, & Wang, 2013). Moreover, an interesting raw material for producing hydrogels is the bacterial cellulose. Bacterial cellulose is a polymer synthetized by bacteria from the genus *Acetobacter, Rhizobium, Agrobacterium* and *Sarcina* (Gao et al., 2015). Its main characteristic is its physical structure as a non-woven fibrous multilayer which has the ability to absorb water in high amounts. Moreover, bacterial cellulose has a promising application due to its interesting mechanical properties, which include high tensile strength, high water retention ability, and biocompatibility (Chang & Zhang, 2011).

These naturals polymers may be blended, to achieve high chain flexibility for water transport, which is required for the application of hydrogels especially to the food preservation field (Guilherme et al., 2015). The blending capacity results from molecular modifications through the reactions of functional groups (amino and hydroxyl), which provide hydrogels with various possible structural and morphological profiles; thus, this feature enhances hydrogels potential absorption and liquid retention capacity (Deligkaris et al., 2010; Kalia & Sabaa, 2013; Omidian, Hashemi, Sammes, & Meldrum, 1999).

2.2. According to the type of crosslinking

Hydrogels can be formed by chemical crosslink, where the polymer chains interact with each other to form permanent junctions through the action of crosslinking agents or by physical crosslink, where physical interactions and chemical bonds are formed between the polymer chains, by weak junctions, such as Van der Walls forces and hydrogen interactions. This kind of linkage forms transient junctions and produces generally biocompatible materials and, hence, avoids the use of other compounds (Ranjha & Khan, 2013).

2.3. According to hydrogel biodegradability

Hydrogels are obtained from biodegradable polymers, usually originating from natural sources (proteins and carbohydrates). However, several synthetic polymers used in the food industry present biodegradability, such as poly(aldehyde guluronate) (Lee, Bouhadir, & Mooney, 2000), polyanhydrides, poly(N-Isopropyl Acrylamide) (Pourjamal, Fathi, Entezami, Hasanzadeh, & Shadjou, 2016), poly

(acrylic acid) (Feng et al., 2014), poly(lactic acid), poly(glycolic acid), acrylamide, 2- hydroxyethyl methacrylate, methoxyl poly(ethylene glycol) (MPEG), 2- hydroxypropyl methacrylate (HPMA) and acrylamide (Byard, Williams, McKenzie, Blanazs, & Armes, 2017).

2.4. According to the polymeric composition

Hydrogels can be based on homopolymer (the polymer network derives from a single species of monomer), copolymers (comprising two or more different monomer types, arranged along the polymer network chain), or multicomponent (formed by two independent, crosslinked, synthetic and/or natural polymer component, in a network form).

2.5. Physical appearance

Depending on their application, functionality, and polymerization process, hydrogels may present different structural conformations, such as nanoparticles, microparticles, coatings and films (Abaee, Mohammadian, & Jafari, 2017).

2.6. Electrical charge

Hydrogels can have a neutral/nonionic, ionic, and amphiphilic nature. In addition, their electrical charge influences their retention capacity of absorbed water (Jensen & Lura, 2006).

2.7. Stimulus responsiveness

Hydrogels based on a stimulus-response system are sensitive to environmental changes such as temperature, light, magnetic field, pH, and other factors that influence the normal characteristics of the system where the hydrogel is integrated (Argin, Kofinas, & Lo, 2014; Berger, Reist, Mayer, Felt, & Gurny, 2004; Bueno, Bentini, Catalani, & Petri, 2013). Such systems are of great interest in creating intelligent/smart food packaging systems (Chang et al., 2010; Kalia, 2016).

3. Hydrogel three-dimensional network conformation

Hydrogel networks are formed due to the polymeric associations resulting from covalent bonds (chemical crosslink), hydrogen bonds, and Van der Waals force (physical crosslink) (Croisier & Jérôme, 2013).

One of the main drawbacks of hydrogel three-dimensional conformation is its stability during the swelling state. In this regard, studies exploring the strategies to decrease the polymeric solubility in swelling equilibrium are needed. To this end, new formulations and methodologies to obtain miscible blends must be developed, since miscibility promotes specific interactions influencing hydrogel stability (by interfering with tension and solubility) (Margaritis & Kalfoglou, 1988; Vogelsanger et al., 2003). Some interactions can maintain system homogeneity, thus, favoring the use of these blends in the food industry (Liu et al., 2016; Sonego, Santagapita, Perullini, & Jobbágy, 2016).

The ideal formulation of hydrogels should present a high interaction between the polymeric chains in terms of thermodynamic miscibility profiles (da Silva Neiro, Dragunski, Rubira, & Muniz, 2000) and three-dimensional behavior, that is, the movement of polymer chains based on molecular weight and crystallinity (Marentette & Brown, 1998).

A description of each of the main methods for hydrogel three-dimensional structure conformation are presented as follows:

3.1. Hydrogel three-dimensional structure conformation through the chemical method

According to Jenkins, Kratochvíl, Stepto, and Suter, (1996), a crosslink is generally a covalent linkage between molecules. However, this term has also been used to describe weaker chemical bonds of crystal portions and even physical and entangled interactions. The main advantage of this method is that the chemical crosslinking increases the resistance of hydrogel to dissolution in water-based solutions, contributing to its physical integrity during the swelling equilibrium over time (Lien, Li, & Huang, 2008). In this case, covalent bonds are generated between the polymeric chains of the hydrogel. The main methods used to achieve chemical crosslinking are radical polymerization, the reaction of complementary groups, grafting, and enzymatic reactions (Hennink & Van Nostrum, 2012).

Chemical crosslinking-based bonding depends on the type and number of the functional groups and on the reactivity of the crosslinking agents, thus, generating a considerable number of linkages (Cai, Zuo, & Tang, 2005). Crosslinking agents are generally small, multifunctional molecules that react with polymers, favoring interaction with previously activated functional groups. Chemical crosslinking results in insoluble scaffolds and improved hydrogel mechanical resistance. They can be generated by photosensitive agents in a reaction catalyzed by enzymes and by polymer-polymer interaction with functional groups (Croisier & Jérôme, 2013; Hennink & Van Nostrum, 2012; Ranjha & Khan, 2013).

The main factors underlying the structural profile of chemical hydrogels are the crosslinking density and ratio of the agent/polymer used for hydrogel formation (Dash et al., 2011).

Chemical crosslinking is normally used to form permanent three-dimensional structures, conferring to the hydrogel thermal stability and mechanical resistance. However, such hydrogels are non-degradable and cannot be recycled easily (Chang & Zhang, 2011). Nonetheless, since they require a purification step, which is usually performed after hydrogel elaboration and aims to eliminate the residues of crosslinking agents or any other chemical compounds, hydrogels resulted from the chemical method are still not applied in the food industry due to possible toxicity issues (Berger et al., 2004; Kalia, 2016).

3.2. Physical method for the conformation of the hydrogel three-dimensional structure

In recent years, there has been growing interest in hydrogels developed through physical bonds, as they offer a wider spectrum of properties and applications than chemically crosslinked polymers do. Moreover, the physical interaction technique generates fewer toxic products since chemical reagents are not needed (Ranjha & Khan, 2013). Thus, depending on the polymers involved in the blend, it may be possible to elaborate eco-friendly products of great economic and environmental interest.

According to Berger et al. (2004), Croisier and Jérôme (2013) and Hennink and Van Nostrum (2012), the main characteristic of hydrogels obtained using physical methods (non-covalent) is the reversibility of their linkage.

Two conditions are crucial for the structure arrangement by physical methods: i) the interactions between polymeric chains must be strong enough to favor the existence of semi-permanent bonding sites during the formation of the three-dimensional network; and ii) the assembled three-dimensional network should allow the absorption of a considerable amount of water into the polymeric network (Fajardo et al., 2012).

Therefore, when the aim is to obtain a stiff hydrogel, the number of non-covalent interactions should be high, whereas limited non-covalent interactions are required to obtain a soft or weak hydrogel. Three main factors control this tunable characteristic of physical hydrogels: the pH of the system, the polymer concentration, and its temperature (Croisier & Jérôme, 2013).

Hydrogels obtained by physical methods are designated as hydrogels complexed with polyelectrolytes since they result from the ionic interactions between polymers; to obtain this type of hydrogels, the system must include two polyelectrolytes with opposite charges (Hoare & Kohane, 2008). Chitosan is one of the main polymeric matrixes used when applying this method for hydrogel conformation since the

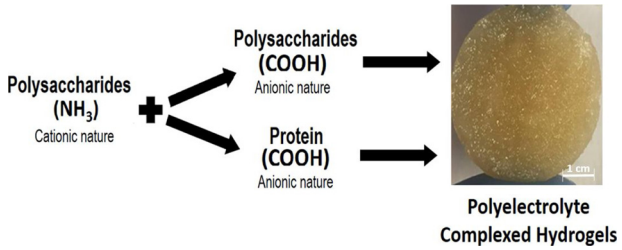

Fig. 3. Ionic natural polymers and the formation of natural polyelectrolytes with no crosslinking agent needed for ionic interaction.

structure of chitosan contains amino groups that ensure the cationic nature required to interact with the anionic group of other polyelectrolytes and form mixtures with a wide range of macromolecules, especially polysaccharides and proteins. In this context, chitosan allows obtaining numerous structural, morphological, and functional hydrogel profiles, associated with high absorption potential (Berger et al., 2004). In this regard, Fig. 3 presents one example of polysaccharide versatility toward the formation of a network between amino and carboxyl groups and the consequent formation of a hydrogel complexed with polyelectrolytes, this depends on the pH and the pKa of the interacting species.

In spite of the advantages mentioned above, the use of hydrogels in the food industry is limited by their low mechanical resistance and quick dissolution. Some previous publications have reported the potential application of hydrogels developed by the physical method intended in the food field. In this context, Farris et al. (2011) showed that gelatin–pectin composite films from polyion-complex hydrogels crosslinked (0.3 wt.% glutaraldehyde) swelled ten times less than uncrosslinked ones (215% versus 1950%, respectively) after 25 h. In the same study, hydrogel film-shape based on pectin and gelatin obtained by electrostatic interactions showed a swelling ratio of 300%, higher than crosslinked gelatin (215%), but lower than gelatin samples (1950%). After 25 h, gelatin-only hydrogel film-shape disintegrated. Thus, electrostatic self-assembly is a widely established route to generate supramolecular structures with enhanced properties and polyanionic interactions are an excellent alternative to obtaining a biodegradable hydrogel.

4. Hydrogels functional properties

One of the main functional properties of hydrogels is the high-water absorption, which is related directly to the swelling capacity. These properties are highly relevant in terms of the potential integration of polymeric hydrogels into food packaging systems and are generated by the presence of hydrophilic groups, such as hydroxyl (−OH), amine (NH_2), amide (−CONH-, $-CONH_2$) and sulfate (-SO_3 H).

Several factors, such as chemical structure, solvent quality, the degree of bonding between polymeric chains and specific stimuli caused by changes in the external environment, can influence hydrogel properties (Laftah et al., 2011). In this context, the water absorption capacity can be manipulated through the type of polymer, crosslinking degree, and specific conditions of the elaboration process such as temperature and pH (Kabiri et al., 2011).

According to Jensen (2011), superabsorbent hydrogels can absorb large amounts (> 100%) of specific substances from the environment (e.g., water) and retain them inside the polymer matrix for a certain period, even under mechanical pressure. When the polymeric chains' hydrophilic groups are submerged in water, they become ionized, hence, promoting electrostatic repulsion and leading to a higher degree of swelling than other polymers (> 1500% in some cases). The crosslinked network prevents water loss, thus, maintaining the swelling equilibrium (Bhattacharya et al., 2013). When the polyelectrolytic complex is in contact with aqueous solutions, the acting osmotic pressure becomes proportional to the ionic concentration of the medium (Friedrich, 2012). In the dry state, ions are strongly linked to the polymer network, leading to a higher osmotic pressure inside the hydrogel matrix. During water absorption, the ions distance themselves, thus, reducing osmotic pressure. Since the interactions between polymeric chains become weaker, the swelling process is limited by the mechanical strength of the polymeric matrix. Therefore, the mechanical behavior of hydrogels is driven by the polymer elasticity during swelling (Guilherme et al., 2015). The formation of hydrogen and covalent bonds increases with the increasing concentration of hydrophilic groups, leading to the consequent increase in the hydrophilicity of the polymeric matrix (Zheng et al., 2007). Moreover, in some cases, a temperature increase may improve the swelling degree, probably because of environmental entropy and the improved detachment of chains, considering environmental entropy as the disorder that can be measured in an specific system (Botaro, Santos, & Oliveira, 2009).

The degree of crosslinking exerts a direct influence on the thermal, mechanical, and swelling properties of hydrogels in different media (Singh & Pal, 2012), mostly due to the crosslinking effect on the solubility and spatial arrangement of the molecules (Kioussis & Kofinas, 2005). The swelling process is controlled by polymer-solvent interactions, which in turn are driven by the solubility and three-dimensional conformation. Therefore, the swelling potential is influenced by the molecular conformation of the polymeric matrix and by environmental conditions such as electrical charge, pH, temperature, and ionic force. In addition, the electrical charge influences the retention of absorbed water (Jensen & Lura, 2006).

Other factors to consider during hydrogel development and performance assessment are particle size, drying process, and porosity, which define the structural array of the three-dimensional network (Bao et al., 2011).

Particle size plays a relevant role in the swelling properties of hydrogels, especially in their kinetics; thus, the size of hydrogel particles should be adequate to the specific target application. With particles larger than 80 mesh, swelling occurs slowly and the liquid is retained in the system for extended periods. Those sizes may be adequate when using hydrogels in agriculture which requires swelling and slow release to generate adequate soil humidity (Ahmed, 2015). After deswelling, the formed empty spaces facilitate adequate gas interchanges. On the other hand, particles smaller than 80 mesh facilitate rapid fluid absorption with decreased retention due mostly to the increased superficial contact area (Omidian et al., 1999). Hydrogel particle size may be applied to personal care (hygiene) products, to facilitate rapid absorption of fluids such as blood or urine (Kabiri et al., 2011). Similar characteristics are required when applying hydrogels to food packaging systems, especially when the main purpose is the absorption of biological fluids (exudates) from fresh food products, such as meat, chicken, fruits, and vegetables (Bodbodak & Rafiee, 2016).

Other important properties to assess hydrogel performance include mechanical resistance, wettability, degradability, and thermal resistance (Table 2).

Regarding the mechanical properties of hydrogels, after hydration, hydrogels should be able to maintain their structural integrity over the product's shelf-life (Huber et al., 2017; Klein et al., 2016; Konwar, Gogoi, Majumdar, & Chowdhury, 2015). Previous studies have shown that the mechanical resistance of hydrogels can be increased by crosslinking, addition of nanocomposites (such as nanoclay), copolymerization, and the use of a pH-sensitive network (Barrett et al., 2013; Chen, Zhu, Zhao, Wang, & Zheng, 2013; Paulino et al., 2012).

Several previous studies on polymers have focused on degradability (Ahmed, 2015). The use of natural polymers is an interesting alternative for the production of biodegradable matrices intended to be used as food packaging systems while meeting the actual demand for sustainable packaging materials (Chen et al., 2016; Zohuriaan-Mehr et al., 2009). The improvement of polymers through functional group incorporation (Cuadri et al., 2016; Kalia, 2016; Spagnol et al., 2012), polyelectrolytes complexation, and monomeric reactions has been used

Table 2
Main functional properties of hydrogels.

Property	Main characteristics	Analytical methodology	Reference
Mechanical resistance	Determination of the structural integrity, as well as tensile strength in specific environment	Texture analyzer	Gupta, Tummalapalli, Deopura, and Alam (2014) Oun and Rhim (2017)
Swelling index (degree of swelling or swelling behavior)	Determination of the hydrogel fluid absorption capability and its swelling ability	Gravimetric method	Zhao et al. (2009), Huber et al. (2017), Fekete, Borsa, Takács, and Wojnárovits (2016), Fekete, Borsa, Takács, and Wojnárovits (2017), Ge et al. (2018), Oun and Rhim (2017)
Wettability	Determination of the level of interaction of the hydrogel surface with the fluid	Goniometer method	Konwar et al. (2015).
Thermal resistance	Determination of the structural thermal resistance at specific conditions of temperature	DSC, TGA	Klein et al. (2016), Konwar et al. (2015). Farris et al. (2011), Oun and Rhim, (2017)
Physical/morphological characterization	Visualization of hydrogel and hydrogel-based films structures	Scanning electron microscopy (SEM) Atomic force microscopy (AFM) Field emission scanning electron microscope	Farris et al. (2011), Fekete et al. (2016), Fekete et al. (2017), Gregorová et al. (2015), Oun and Rhim (2017)
Chemical characterization	Determination of the main chemical functional groups Confirmation functional groups, their molecular weights and chemical structures	ATR-FTIR spectrometer Gel permeation chromatography (GPC) and nuclear magnetic resonance (NMR)	Fekete et al. (2016), Fekete et al. (2017), Gregorová et al. (2015), Amonpattaratkit, Khunmanee, Kim, and Park, (2017)

as a strategy for obtaining degradable polymers (Paulino et al., 2009). Natural and biodegradable polymers, such as starch, guar gum, chitosan, xanthan gum, and sodium alginate, have been treated to obtain sustainable resistant hydrogels. The composite incorporation into natural polymer matrices has been a relevant strategy; however, studies in this matter are limited still (Kabiri et al., 2011).

5. Potential application of hydrogel in food packaging systems

Hydrogels can potentially be applied to food packaging systems. Recently, moisture-absorbent systems for food products based on "absorbent pads" with water-removal features have raised interest, especially due to their potential to reduce the risk of microbial contamination while preserving the sensorial characteristics of packaged food (Otoni et al., 2016). This active food packaging can have multiple functions, such as absorption of fluids emanated from food, modification of the packaging headspace, as well as antimicrobial activity.

Absorbent materials are applied mainly to food packaging systems in which a plastic tray or container receive the exudate released from food during storage. In this configuration, the hydrogel placed inside of the food packaging absorbs the released exudate while controlling the A_w of the food product (Otoni et al., 2016).

The use of absorbent materials in food packaging systems follows four essential requirements: i) following absorption, the absorbent material should retain the exudate in the three-dimensional structure; ii) absorbent materials should maintain good visual presentation and the sensory attributes of the packaged food, at a low cost; iii) the absorbent material should present certain performance properties that ensure the structural integrity of the food packaging system during storage; iv) the absorbent material should increase the shelf-life of stored food products and avoid microbial growth on food surface (Fernández et al., 2009; Roy, Saha, Kitano, & Saha, 2012).

Considering the requirements above, hydrogels constitute an attractive alternative for use in food packaging systems, in which this kind of absorbent materials can offer new opportunities for the efficient design of food packaging systems with desirable properties (such as shelf-life, biodegradability, and mechanical resistance). According to Roy et al. (2012), the integration of hydrogels into food packaging systems is an emerging and promising strategy, where hydrogels constitute innovative, environmentally friendly materials that can extend food products shelf-life.

Most hydrogels for food packaging are elaborated as a film-shaped structure. One of the methods for hydrogel film production involves the casting of the desired hydrogel, generally without the addition of chemical crosslinker agents (Kalia, 2016). During this process, the temperature is one of the main physical agents controlled. Usually, hydrogels developed through this method and using chitosan, PVP (polyvinylpyrrolidone), and CMC (carboxymethylcellulose) are transparent, biodegradable, flexible, hydrophilic, and highly permeable. One of the main applications of hydrogels in active packaging systems is to control the humidity generated by meat products, fresh fruits, vegetables, and other food products with high water content (Bodbodak & Rafiee, 2016).

Furthermore, Otoni et al. (2016) have described the main characteristics of active packaging systems based on the use of absorbent materials, such as absorbent pads. This system comprises two permeable layers (upper and lower layer), and an interlayer (inner layer) containing the absorbent material. The external layers are generally constituted of impermeable thermoplastic materials or cellulose to protect the absorbent material and prevent it from coming into direct contact with the packaged food. Moreover, the holes in the external layers help the exudate flow toward the inner layer (hydrogel or any other absorbent material). Fig. 4a shows a schematic representation of such packaging and Fig. 4b shows images from a goniometer, demonstrating the swelling process of a hydrogel based on xanthan-chitosan blend using distilled water. These figures show a simulation of exudate flow from packaged food to the inner layer (hydrogel), exudate absorption, and hydrogel swelling considering the architecture of the absorbent packaging system.

5.1. Application of hydrogels in vitro and in food matrices

The potential applications of the hydrogel in food packaging systems have been studied *in vitro* to determine their antimicrobial activity when incorporated with different antimicrobial compounds, such as nanoparticles (e.g., silver) or bioactive compounds, as well as in food matrices to determine the potential of hydrogel films potential for food preservation. Characterization studies have shown that despite the

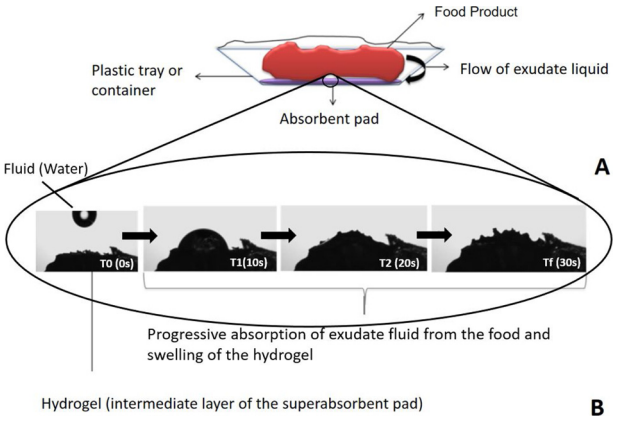

Fig. 4. Schematic representation of absorbent packaging architecture (a). Source: Otoni et al. (2016) with permission; and hydrogel swelling process (images from goniometer assay using Xanthan-Chitosan hydrogels and distilled water) (b).

potential applications of the hydrogel in the food industry and numerous other fields, their main drawbacks, such as limited mechanical and water resistance, still remain. One strategy to overcome these drawbacks consists of incorporating nanoparticles into the polymeric structure of the hydrogel.

Regarding *in vitro* studies, Wang and Rhim (2015) have developed agar/alginate/collagen ternary blend incorporated with silver nanoparticles (AgNPs) and grapefruit seed extract (GSE) for food packaging applications. The developed hydrogel film was tested against *Listeria monocytogenes* (Gram-positive) and *Escherichia coli* (Gram-negative), and the results showed that hydrogel films incorporated with AgNPs exerted a stronger biological activity against *E. coli* than *L. monocytogenes*. Moreover, GSE incorporation was associated with a better antimicrobial effect against *L. monocytogenes* than to *E. coli*. The combination of both antimicrobial agents resulted in a hydrogel film with a synergistic effect against the targeted microorganisms (Wang & Rhim, 2015). Similarly, Oun and Rhim (2017) developed hydrogel films based on carrageenan/KCl incorporated with ZnO and CuO nanoparticles. The results showed that the incorporation of metallic nanoparticles resulted in hydrogels with antimicrobial activity against *E. coli* and to a lesser extent to *L. monocytogenes*. Previous studies have shown that the antimicrobial activity of metallic nanoparticles is due mainly to the presence of positively charged ions, which react with the negatively charged bacterial membrane surface (Oun & Rhim, 2017). Moreover, Tyliszczak et al. (2017) have reported the development of a hydrogel based on chitosan and gelatin, incorporated with AgNPs. The antibacterial properties of the developed hydrogels were tested against *Staphylococcus aureus* and *Enterococcus faecalis*, indicating that the hydrogel incorporated with 3 mL presented activity against *S. aureus*, while *E. faecalis* was resistant to the tested treatments.

Wang and Rhim (2015) assessed the effect of developed hydrogel films based on agar/alginate/collagen and incorporated with AgNPs and GSE on the shelf-life of fresh potatoes. They showed that fresh potatoes present a high post-harvest respiration rate, which generates condensed water on the surface of the plastic containers—an effect known as fogging. However, the integration of the developed hydrogels into the food packaging system prevented the fogging and greening of the potatoes due to chlorophyll generation after light exposure (Wang & Rhim, 2015). Furthermore, hydrogel films based on antioxidant compounds (ferulic acid) have been developed to prevent lipid oxidation in butter (Benito-Peña et al., 2016).

5.2. Biodegradable packaging

The selection of adequate polymers ensures the production of biodegradable packaging materials with a suitable degradation rate. This depends on a number of factors, such as monomer type, binder nature, crystallinity, and initial molecular weight of the polymer, since the loss of polymer mass is initiated only when oligomeric fragments are small enough to be water soluble (Simionescu & Ivanov, 2016).

Polylactic acid synthesis was first reported by Kulkarni, Pani, Neuman, and Leonard, (1966). This was a milestone for unleashing the production of synthetic biodegradable polymers throughout the world (e.g., polyanhydrides and polyorthoesters). Since then, degradable polymers containing water-soluble elements have been described. Sawhney and Hubbell (1990) successfully increased hydrophilicity and degradation rate by synthesizing lactide, glycolide, and α-caprolactone copolymerized with polyethylene glycol (PEG).

Due to their biopolymeric nature, some hydrogels are biodegraded since they are constituted of bio-polysaccharides and are structured by means of glycosidic bonds, which can be degraded by enzymatic action (Croisier & Jérôme, 2013). Chitosan is one of the most commonly used biopolymers for developing hydrogel matrices. Important linkages, such as glucosamine–glucosamine, glucosamine–*N*-acetyl–glucosamine, and *N*-acetyl–glucosamine–*N*-acetyl–glucosamine, are targeted by enzymatic action. Lysozyme and bacterial enzymes are identified as being responsible for chitosan biodegradation. Moreover, higher plants present chitinases, enzymes that act on *N*-acetyl–glucosamine residues. The

Table 3
Potential uses of hydrogel in the food field.

Main goal	Main characteristic	Hydrogel composition	Reference
Food freshness indicator	Generated information regarding food freshness Based on metabolites production of in the food Detection changes in the pH, chemical degradation or microbial growth	Poly(N,N-dimethyl acrylamide-co-methacryloyl sulfadimethoxine) (poly(DMA-co-SDM)) hydrogels incorporated with methacryloyl sulfadimethoxine monomer (SDM) with a pH-responsive group	Baek et al. (2017)
Stability and retention of volatiles substances	Flavor encapsulation (nanoemulsions in hydrogels) Controlled release occurs by pH modification (e.g., in contact with saliva)	Flavored nanoemulsions incorporated in low methoxyl (LM) pectin and whey protein isolate (WPI) at pH 4.0 Orange oil, medium-chain triglyceride (MCT) oil, and WPI (stable nanoemulsions)	Kwan and Davidov-Pardo (2018)
Improvement of bioavailability of lipophilic compounds	Incorporation of lipophilic bioactive compounds (e.g. β-carotene) in food matrix, improving their bioavailability	Hydrogels based on polysaccharides (starch and xanthan gum) to incorporate β-carotene emulsion	Park et al. (2018)
Method for aflatoxin B_1 detection	Detection of aflatoxin B_1 in food sample when the hydrogel causes collapse of the network, and occurs the release of urease into the analyzed solution. Then, the released urease can catalyze the hydrolysis of urea and result in the rise of pH value. The change of pH value has a direct relationship to the concentration of aflatoxin B_1. Main advantages: Cheap, available, and well-established pH testing system with the robust performance of DNA-crosslinked hydrogel.	DNA hydrogel	Zhao et al. (2018)

Table 4
Commercial absorbent food packaging systems.
Source: Based on Janjarasskul and Suppakul, 2016; Lee, 2011; Day, 2003; Fernande, 2000; Vermeiren et al., 1999.

Food packaging system	Raw Material	Format	Commercial Product	Others Products
Desiccant: to maintain the relative humidity	Silica gels; Clays; Calcium oxide or Calcium sulfate.	Blister; Canisters; Disks; Foam tray; Polymeric films; Sachets; Tablets; Tape.	A combined saturated solution formulation of soluble substance - Humidipak® (Humidipak Inc., Minnesota).	Cryovacs® Food Packaging Systems (Cryovac Division of WR Grace and Co., Duncan); Pad-Loc®Super Absorbent Pads; MiniPax® (Multisorb Technologies, EUA); Desimax® (Multisorb Technologies, EUA); SuperDryFoil™ (Baltimore Innovations Ltd.); Acryl One (Maruto Sangyo Co., Tokyo, Japan); Activ-Blister® (CSP Technologies, Inc., Auburn, AL); Formpack® (Amcor Ltd., Australia).
Humidity control: to maintain dry surface	SAH combined particles (eg. polyacrylates and graft copolymer of starch) crosslinked networks of flexible chains and accept water diffused into the structure with swelling.	Film/membrane; Granular; Microperforated film; Sachet; Humectants between two layers of a plastic film.	They are usually embedded in the porous drip sheets or pads - ThermaFreeze® (ThermaRite Pty).	Xtend® (Stepack, Israel); Toppan Sheet® (Toppan Printing, Japan); SupaSorb® (ThermaRite Pty); Luquasorb® (BASF); Fresh-R-Pax (Maxwell Chase Technologies); Pebax® (Arkema, France); Thermarite® (Pty Ltd, Austrália); Toppan™ (Japan); Peaksor® (Peakfresh produts, Australian).
Purge pads: to remove the excess water on food surface.	Powder type of potassium polyacrylate.	Sheets; Pads	Humectant between moisture-permeable film layers (eg. viscous polypropylene glycol sandwiched between polyvinyl alcohol films - Pitchit®, Showa Denko, Japan).	Toppan Sheet® (Toppan Printing, Japan); Cryovacs® Food Packaging Systems (Cryovac Division of WR Grace and Co., Duncan); Luquasorb® (Stockhausen GmbH Krefeld and BASF Corporation, Germany); UltraZap® (Xtenda Pak pads, Paper Pak Industries, Canada).

extent of the chitosan biodegradation is determined by the degree of deacetylation, since a high level of deacetylation results in low biodegradation (Kean & Thanou, 2010).

6. Other potential applications in the food industry

Other potential applications in the food industry include the use of hydrogel as part of an intelligent packaging system or as a carrier system incorporated directly into food matrices (Table 3).

When used as part of an intelligent packaging system, their main purpose is to provide information regarding the freshness of the contained food products or as part of a simple detection method to determine the presence of contaminants, such as aflatoxin (Baek, Kim, Jeon, & Seo, 2017; Zhao et al., 2018). Other novel applications of hydrogels include their use as carriers of flavors or bioactive compounds, such as β-carotenes, that are incorporated usually in nanoemulsions (Kwan & Davidov-Pardo, 2018; Park, Mun, & Kim, 2018).

7. Future trends

Hydrogels can potentially be used in the food industry as part of food packaging systems, and most are produced from synthetic polymers, which ensure higher physical-chemical stability than natural ones. However, hydrogels based on natural compounds are renewable, have biological properties (biocompatibility and biodegradable), can be produced on large scale due to raw material abundance, and present interesting structural properties.

Nonetheless, before competing in the market with traditional materials, it is necessary to study the functional properties of hydrogels, such as swelling equilibrium and physical interactions, thus, avoiding the use of toxic reagents, especially in the crosslinking process (Athawale, Bailkeri, & Athawale, 2001; Kabiri, Zohuriaan-Mehr, Bouhendi, Jamshidi, & Ahmad-Khanbeigi, 2009).

Numerous food packaging systems for moisture control and food protection are available in the market (Table 4).

The current demand for protection against microorganism growth and the preservation of the sensorial characteristics of food products is met by using sachets and polymeric films desiccants such as SuperDryFoil™, Formpack®. Microperforated films (Xtend®, Pebax®) and have also been used on fresh products to control condensation. To avoid microorganism growth, minimally processed or fresh meat products, purge absorbers/pads, or polymeric packages, such as film, trays, or lids (e.g., Cryovacs®, UltraZap®) are commercialized.

Moreover, consumers look for natural materials, produced using renewable resources, and characterized by low toxicity and high biodegradability. In the development of competitive food packaging systems using hydrogels, two main challenges still remain, which are the search for new materials and methodologies to obtain stable and biodegradable hydrogels and the use of hydrogels as delivery systems for active compounds, such as antimicrobials, antioxidants, flavorings, and colorants.

Although many studies have focused on the application of biodegradable hydrogels in different fields, regarding food packaging systems, both challenges remain to be explored to an adequate extent.

On the other hand, over the years, acrylates and their derivate monomers have often been selected as raw materials to form hydrogels (Laftah & Hashim, 2014). However, the increasing demand for “eco-friendliness” induced a shift from artificial to natural polymers, especially polysaccharides (Mekonnen, Mussone, Khalil, & Bressler, 2013; Rinaudo, 2006). Copolymerization between inorganic/organic chains enhances the properties of composite materials and may even create new properties (e.g., the use of acrylate monomers and polysaccharides is an option for combining the properties of both materials) (Guilherme et al., 2015; Rodrigues et al., 2014). Another possibility consists of the use of nanomaterials to improve hydrogel performance. According to Spagnol et al. (2012), the addition of cellulose nanofibrils (obtained

from cotton fibers) to a polymer matrix based on chitosan-graft-poly acrylic acid significantly improved swelling capacity. Besides the established hydrogel potential for controlled solute release, due showed responsive behavior in relation to pH and salt solution. Such features lead these materials to be considered as smart materials and increase their technological applications. In fact, nanotechnology has a high potential in the food packaging industry, where it has been applied for several purposes, such as improved barriers, mechanical, thermal, and biodegradable properties and applications in active and intelligent food packaging (ex. nanosensors) (Avella et al., 2005). Nanomaterials such as cellulose nanocrystals, carbon nanotubes, cellulose nanowhiskers, chitin whiskers, montmorillonite, microfibrillated cellulose, and starch nanocrystals have already been applied in food packaging. Thus, nanocomposites are among the available alternatives for enhancing hydrogels (Cushen, Kerry, Morris, Cruz-Romero, & Cummins, 2012).

The applications of graphene-based composite hydrogels and modified graphene nano-materials properties have been exhaustively studied. This material is considered an active and smart material and has been used by several industrial sectors (Lu, Zhang, Guo, & Li, 2017).

Other future possibilities for this kind of systems is the synthesis of hydrogels at the nanoscale (nanogels or microgels). In this regard, the principle of controlled drug release can be adapted to food preservation in order to release active compounds with biological activity against foodborne pathogens or food-spoilage microorganisms (Landfester & Musyanovych, 2010).

Moreover, the potential use of hydrogels in biomedicine, as carriers of antimicrobial substances set a starting point of evidence for the high potential of hydrogels as potential carriers of active substances and their likely application when incorporated with antimicrobials in the field of food preservation. Finally, research studies in this field will increase the industry's prospects and interest in developing new products (e.g., active food packaging) based on degradable hydrogels.

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