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## Bring some colour to your package: Freshness indicators based on anthocyanin extracts

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## ABSTRACT

**Background:** Nowadays, consumers are more aware about what they eat and how it is packaged. Food quality is of paramount importance and consumers are eager to be constantly updated about the perishability and shelf-life of the food products they buy. This gave rise to intelligent packaging (IP) materials and devices that can communicate with all people throughout the entire food route. One of the main colour-based IP systems is based on the use of anthocyanin (ATH)-based natural dyes by exploiting their pH-dependent colour changes that can be easily readable by the naked eye.

**Scope and approach:** This review focuses on recent studies regarding ATH-based sensor packaging materials development for IP applications, offering valuable insights regarding the main factors that affect the efficacy of the developed IP, such as the material where the ATH is anchored, ATH type and concentration, the ingredients used in the formulation of the sensor and the storage conditions of the food product. Additionally, the last ATH-based IP developed are also summed and discussed in detail to identify the major bottlenecks that need to be overcome for accelerating the commercial application of ATH-based sensors in IP.

**Key findings and conclusions:** Although ATH-based indicators are able to correctly indicate food shelf-life in several products at a laboratory scale, their industrial production is still impaired by the production methods used for this type of sensors. Furthermore, ATHs are labile compounds that require stabilization in the packaging material to yield stable, standardized and durable IP materials.

## 1. Introduction

In the last decades, consumers became more and more aware about what they eat. Besides requesting minimally processed and highly nutritional foods, they also demand chemically and microbiologically safer foods with increased shelf-life. All these consumers' needs in conjunction with food globalization, have driven the food packaging industry for the development of new types of packaging materials that are not only able to contain the food and protect it from the environment, but are also able to increase product's shelf-life (active packaging) and communicate with the consumer in terms of the safety and freshness of the product (intelligent packaging, IP) (Borzi, Torrieri, Wrona, & Nerín, 2019; Poyatos-Racionero, Ros-Lis, Vivancos, & Martínez-Máñez, 2018; Wu, Wang, Hu, & Nerín, 2018).

In addition, food packaging innovations have been driven towards more environmental-friendly solutions by preferring natural substances

over synthetic ones, both in the packaging materials (Han, Ruiz-Garcia, Qian, & Yang, 2018) as well as in the compounds that are incorporated in the package to make it an active and/or intelligent package. For this purpose, a great amount of biopolymers with improved properties regarding barrier or mechanical features have been created from lipids, polysaccharides and/or proteins and applied to different types of food in last years (Gómez-Estaca, Gavara, Catalá, & Hernández-Muñoz, 2016; Mohamed, El-Sakhawy, & El-Sakhawy, 2020; Nerin, Becerril, Manso, & Silva, 2016). Regarding the use of natural compounds as active agents, there is a vast amount of research describing the use of compounds derived from plants for the development of antioxidant and/or antimicrobial food packaging materials (Gherardi, Becerril, Nerin, & Bosetti, 2016; Silva, Domingues, & Nerín, 2018; Wrona et al., 2019).

On the other hand, when focusing on intelligent food packaging materials, the preferred natural compounds include those that are capable of responding to changes in either the packaged food or its

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environment inside the package such as temperature, pH, microbial growth, volatile substances, etc., and which provide readily visible and comprehensive information to the user. The singularity of intelligent packaging is its ability to communicate with the food product, without interacting with it (Müller & Schmid, 2019). As long as the food is packaged, this communication is constant and uninterrupted throughout all the supply chain, from the producer to the retailer, and finally to consumers: as the package and the food product move together throughout the entire chain, the package is the ideal vehicle to communicate any alteration in the conditions of the packaged food product (Yam, Takhistov, & Miltz, 2005). One of the main food attributes that retailers and consumers are most worried about is freshness, as this is the main indicator that ensures both food quality and food safety.

## 2. Freshness indicators

Direct freshness indicators have emerged as a reliable tool to guarantee the safety and quality of the packaged food and to reduce food waste. Freshness indicators have been designed to monitor changes in quality or freshness of packaged food, such as microbial growth or the generation of undesirable substances, and transmit this information to the customer (Fang, Zhao, Warner, & Johnson, 2017; Kuswandi, 2017; Müller & Schmid, 2019; Poyatos-Racionero et al., 2018). Data provided can be used to determine the shelf-life of the product together with the “use-by-date” or serve to optimize storage and distribution conditions (Kuswandi, 2017; Vanderroost, Ragaert, Devlieghere, & De Meulenaer, 2014). The mode of action of a freshness indicator is based on the detection of qualitative or quantitative changes in the concentration of one or more substances inside the packaging that are related to food deterioration, and a subsequent transduction of these changes to perceptible signals that can be visually detected by consumers. In the case of colorimetric indicators (Fig. 1) for example, changes in the concentration of compounds such as glucose, volatile nitrogen compounds, biogenic amines, carbon dioxide or sulphide compounds are transduced into colour changes that can be noticed by consumers.

A freshness indicator should fulfill several requirements for a successful development and implementation in the food industry that includes: (i) a suitable performance that allows a sensitive and fast monitoring of changes in the selected substances and a good correlation between the detected changes and the quality of the food product; (ii) an easy implementation in the food industry driven by low prices and the need of simple technology for their inclusion in the packaging material and; (iii) a simple reading and interpretation by consumers, without the need to use any specific device. This last requirement is broadly fulfilled by colorimetric indicators, being this the main reason of the high progress done in this type of devices, some of them already launched into the market as Freshcode®, Freshtag® or Ripesense®.

A freshness indicator based on pH sensitive dyes is one of the strategies used to detect the deterioration of several types of foods such as fish, meat and dairy products (Alizadeh-Sani, Mohammadian, Rhim, & Jafari, 2020). When a certain food begins its deterioration process, a pH change occurs; this change can be monitored by a pH indicator that is embedded in a solid support or on the packaging material itself

(Balbinot-Alfaro et al., 2019). These changes in the pH indicator usually translate into a colour change, meaning that most pH-based freshness indicators are colorimetric sensors. This approach has been widely used for raw fish or meat products as they increase their pH during the decay process due to the growth of spoilage microorganisms that yields nitrogen compounds.

Although most freshness indicators based on pH changes used synthetic dyes, in recent years, research has been conducted to substitute these synthetic dyes by naturally occurring pH indicators, especially anthocyanin-rich extracts obtained from plants. Besides their natural origin, anthocyanins (ATH) present several advantages as they are easy to obtain, widely distributed in nature and cheap, making them a good alternative to synthetic dyes. Additionally, anthocyanins also circumvent the toxicity issues related with the use of synthetic dyes as anthocyanin toxicity has not been shown in human intervention studies and according to the last re-evaluation of the Scientific Panel on Food Additives and Nutrient Sources Added to Food from the European Food Safety Authority, anthocyanins (mostly grape-skin and black currant extracts) were considered unlikely to be of safety concern (EFSA, 2013; Wallace & Giusti, 2015).

The development of pH-sensitive smart packaging based on anthocyanins and other natural food colourants have been recently reviewed (Alizadeh-Sani et al., 2020; Bhargava, Sharanagat, Mor, & Kumar, 2020; Kalpana, Priyadarshini, Maria Leena, Moses, & Anandharamkrishnan, 2019; Roy & Rhim, 2020). Although most of these reviews summarize the main characteristics of anthocyanin extracts and their incorporation in biopolymers for the development of novel smart packaging, they are not focused on the mechanistics behind the development of anthocyanin-based smart food packaging materials such as the interactions between the anthocyanin extract and the polymeric matrix, the interaction of the ATH-based indicator with the food product and the issues concerning ATH migration and indicator stability, and colour assessment and its relation to food spoilage.

## 3. Anthocyanins

Anthocyanins are the most important pigments of vascular plants that provide many of the blue, red, orange, pink and violet colours observed in flowers, berries, leaves, stems and roots of many plants (Castañeda-Ovando, Pacheco-Hernández, Páez-Hernández, Rodríguez, & Galán-Vidal, 2009). Chemically, these water soluble pigments belong to the widespread class of phenolic compounds collectively named flavonoids whose structure is based on the flavylum cation. The structure of anthocyanins consists on a core called anthocyanidin, which is composed of two aromatic rings linked to an oxygenated heterocycle, that can be bonded to different sugar moieties (Smeriglio, Barreca, Bellocco, & Trombetta, 2016) (Fig. S1). The flavylum ion is the chromophore moiety of the anthocyanin and is responsible for the high instability and susceptibility to degradation of these coloured substances (Smeriglio et al., 2016).

To date, 23 different anthocyaninidins have been described with variations in the number and degree of methylation of hydroxyl groups in one of the rings. In vascular plants, the six most common anthocyanins are pelargonidin, cyanidin, delphinidin, malvidin, peonidin and

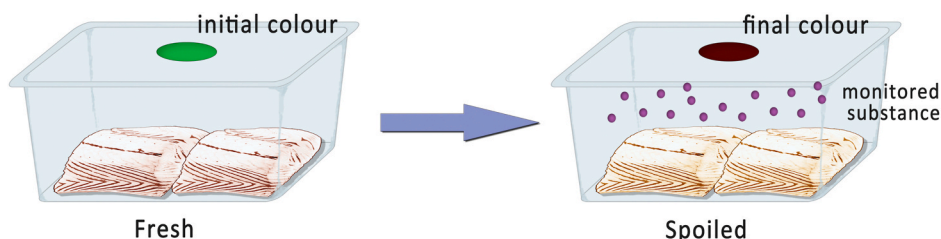


Fig. 1. Schematic operating mode of a colorimetric freshness indicator.

petunidin (Clifford, 2000) (Fig. S1), being cyanidins (which contain two hydroxyl groups) the most widely distributed species. Regarding anthocyanins, there are reports of more than 500 different types (Castañeda-Ovando et al., 2009). The main differences between them are the number of hydroxylated groups, the nature and the number of bonded sugars and the number and position of the aliphatic or aromatic carboxylates bonded to the sugar moieties (Castañeda-Ovando et al., 2009). These differences imply consequent variations in stability, and hues (Rodríguez-Amaya, 2019). The relative abundance of anthocyanins varies greatly according to the plant species, time of harvest or growing conditions, with light being one of the most important environmental factors that affects ATH biosynthesis (Gu, Wang, Hu, & Hao, 2019; Tsuda, 2012).

The attractive colours, high water solubility and biological properties of ATH together with their natural origin and low toxicity have attracted a great interest by researchers, industry and consumers towards their application in the food industry and in food products. Nowadays, ATH are widely used as food colorants in some types of processed foods and beverages replacing older pigment substances of synthetic origin. Furthermore, ATH can be used as antioxidants in food products by adding them directly into the food or by their incorporation into the packaging material (Ahmad, Gokulakrishnan, Giriprasad, & Yattoo, 2015; Lorenzo et al., 2018; Wang, Yong, et al., 2019). In fact, the use of ATH as food additives is reinforced by their proven benefits to human health that include anti-inflammatory, antioxidant, antiobesity, antiangiogenesis, anticancer, antidiabetes, antimicrobial, neuroprotective and immunomodulatory properties (Li, Wu, Fu, & Reddivari, 2019).

As mentioned above, anthocyanins possess low stability due to the presence of the flavylium ion and its peculiar electron distribution (Smeriglio et al., 2016). This instability, besides the undesirable interactions with food products, limits the use of ATH in the food industry. The ATH chemical structure is critical to its stability, as it has been proved that the increase of methoxyl groups enhances stability, whereas the increase of hydroxyl groups yields the opposite effect (Tarone, Cazarin, & Marostica Junior, 2020). Additionally, the anthocyanin glycosyl acylation has demonstrated to strengthen the *in vitro* and *in vivo* chemical stability of ATH (Zhao et al., 2017). Hence, a correct selection of the plant raw material is essential to obtain extracts with high content of chemically stable anthocyanins. Nowadays, black carrot, red cabbage or purple sweet potato are used extensively as sources of anthocyanins because of their high contents of stable acetylated anthocyanins. Anthocyanin stability is improved in nature by interaction with other anthocyanins, metallic ions, phenolic compounds and other non-phenolic substances like pectin polysaccharides as happen (Castañeda-Ovando et al., 2009; Cavalcanti, Santos, & Meireles, 2011; Cortez, Luna-Vital, Margulis, & Gonzalez de Mejia, 2017). These associations not only enhance the stability but can also lead to changes or increments in colour intensity. This phenomenon, which is called copigmentation is the main responsible for the final colours that plants show in nature. Interactions might also be used in the food industry to increase the stability and improve the colour characteristics of the extracts (Chung, Rojanasasithara, Mutilangi, & McClements, 2015; Cortez et al., 2017; Koh, Xu, & Wicker, 2020; Xu et al., 2019).

The use of proper ATH extraction methods is also essential to avoid high ATH degradation rates and to obtain large yields. The classical procedures are based in the extraction by a solvent, generally of polar type such as methanol, ethanol, acetone or water, acidified with a vast array of both organic and inorganic acids. However, efforts have been made to use other more advantageous modern techniques to reduce the amount of solvent used, improve the kinetics of the extraction, diminish the need for purification and concentration steps or increase the extraction yield (Silva, Costa, Calhau, Morais, & Pintado, 2017). Among them, ultrasound-assisted, microwave-assisted and pressurized liquid extractions have demonstrated great potential for ATH extraction. Notwithstanding, these techniques have limited effectiveness and

should be applied at low temperatures to avoid ATH degradation (Silva et al., 2017).

Due to their ionic structure, ATHs in solution can be found as different chemical species depending on the pH (Fig. 2). This singularity has also important consequences in ATH stability and colour. Regarding stability, it is widely accepted that the predominant species at low pH display much higher stability than the more common species at higher pH (Cavalcanti et al., 2011; Khoo, Azlan, Tang, & Lim, 2017). Concerning colour, the different hues observed for the different species make them behave as natural occurring pH indicators. It is known that ATH usually displays red colours at low pH, whereas at higher pH, the colour spectra changes to purple and blue colours (Cavalcanti et al., 2011). As mentioned before, this particular feature of ATHs has been the one most exploited in the development of freshness indicator based on pH indicators.

#### 4. Freshness indicators based on anthocyanins extracts

In the last three years, the pH-dependent colour change properties of anthocyanins have attracted attention for their use as colour indicators in food packaging applications. The development of fully functional anthocyanin-based freshness indicators is a very challenging process, where ATH extract and the polymer used as substrate play a decisive role. In order for it to work correctly, the ATH extract should be compatible with the polymer and able to respond to the freshness alterations suffered by the food product during its storage and distribution. Additionally, as these indicators are based on colour changes, colour alterations must be unequivocal and easily readable by the consumers/retailers and should always be related to a quality/shelf-life changes in the packaged food product. Other properties of ATH-based indicator as water resistance or stability are also crucial for a successful performance.

##### 4.1. Anthocyanin extracts

The selection of the plant used as source of ATH is essential for the development of an ATH-based indicator, since it will determine the type of predominant ATH. As explained in the previous section, the type of ATH restrains the stability of the extract and produce hue variations. As can be seen in Fig. 3, the extracts obtained from different plants exhibit different hue variations at different pH which could affect the use of the ATH-based indicator.

The most commonly used plants for this purpose are red cabbage and purple sweet potato, accounting for more than 35% of all studies (Table S1), due to their high contents of cyanidins and peonidins in its monoacylated and diacylated forms. These ATHs possess improved thermal- and photostability properties and exhibit a wider pH-dependent colour spectrum when compared to the non-acylated anthocyanins (Li, Wu, et al., 2019; Strauch et al., 2019). Traditional sources of ATH such as blackberries and grapes are also being used as well as other more uncommon sources such as black soybean seed coat extracts (Wang, Xia et al., 2019), purple and black eggplant peels (Yong, Wang, Zhang, et al., 2019) or jambolan fruits (Merz et al., 2020). Another very interesting approach is the use of residues from juice processing as the primary source of ATH, such as the ones obtained from mulberry, blueberry, blackberry and chokeberry (Andretta, Luchese, Tessaro, & Spada, 2019; Halász & Csóka, 2018; Kurek et al., 2018; Luchese, Abdalla, Spada, & Tessaro, 2018; Luchese, Sperotto, Spada, & Tessaro, 2017; Zeng et al., 2019). Additionally, it was found that the use of blackberry processing residue (pomace) could reduce the degradation of ATH and improve the yield and colour quality (Kurek et al., 2018).

The concentration of ATH extract in the ATH-based indicator is an essential factor that should be adjusted for a correct functioning of the indicator. It has been demonstrated that excessively low concentrations of ATH produce less distinguishable colour changes (Ge et al., 2019; Kurek et al., 2018; Merz et al., 2020; Prietto et al., 2018; Zhang, Huang,

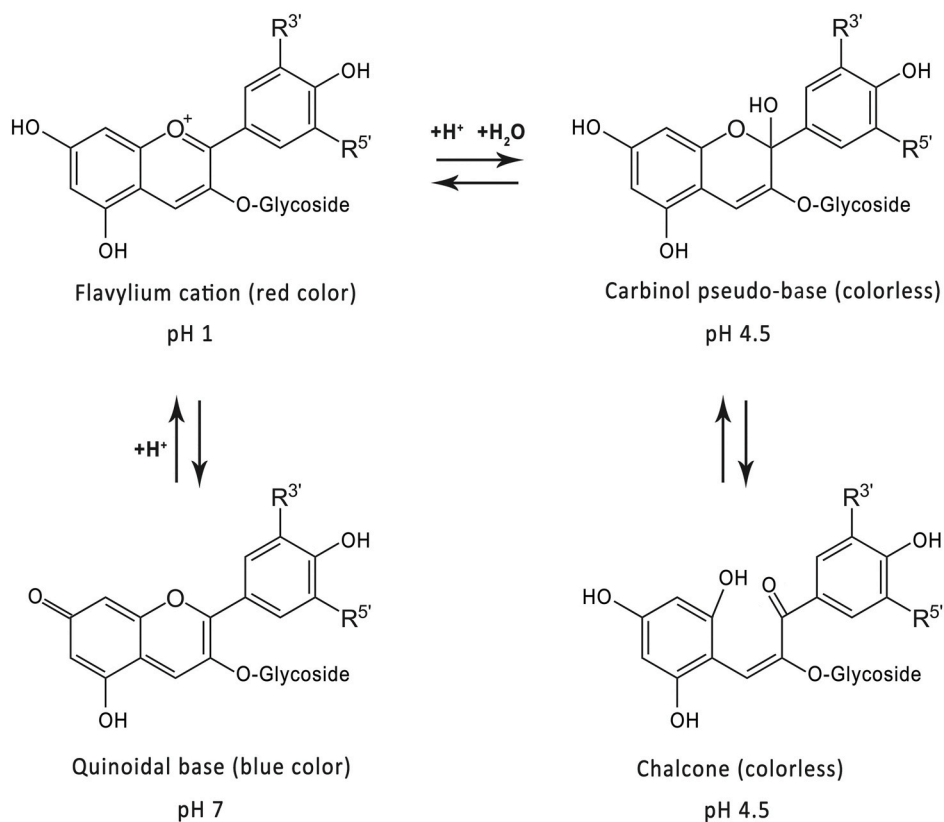


Fig. 2. Anthocyanin chemical species depending on the pH.



Fig. 3. Colour variations of (a) roselle and (b) purple sweet potato extract at different pH. Adapted from Jiang et al. (2020) and Zhai et al. (2017).

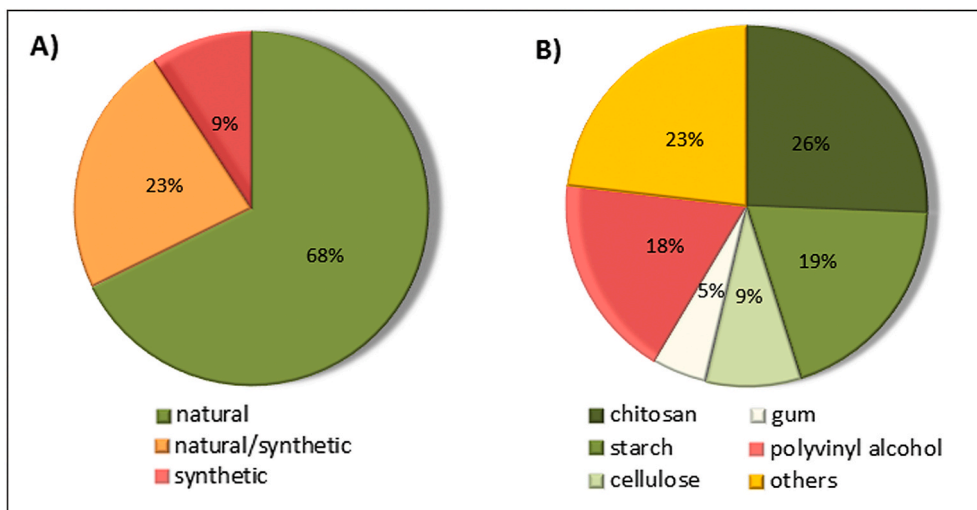


Fig. 4. A) Percentage distribution of polymer use in ATH-based indicators according to their natural or synthetic origin. B) Main polymer types used in ATH-based indicators.

Yan, Hu, & Ren, 2020) whereas too high concentrations have lower pronounced variations, which can be attributed to the interference caused by the colour of anthocyanin itself (Pourjavaher, Almasi, Meshkini, Pirsai, & Parandi, 2017; Qin et al., 2019; Zhang et al., 2020). Because the concentration of ATH used for the development of the intelligent packaging materials is generally given as entire plant extract not as ATH content, it is difficult to compare between different works and establish concentrations ranges that could be more effective. Besides, concentrations of ATH extracts are generally expressed in terms of film forming solution or coating. It would be more convenient to give the data of ATH content per film surface unit for a better comparison between the different final indicator materials.

#### 4.2. Polymer substrate

Another critical component of a freshness indicator based on pH dyes is the substrate used to anchor the dye. On the same mind of the new trends for food packaging, most of the ATH-based indicators have been made from polymers of natural origin (Fig. 4a). The use of starch and chitosan stands out, although cellulose, gelatin or several gums, have also been employed for this purpose (Fig. 4b). Regarding polymers of synthetic origin, polyvinyl alcohol (PVA) alone or in combination was predominantly used in a percentage similar to starch (Fig. 5b). These water-based polymers have been the preferred ones, mostly due to the fact that the water solubility of anthocyanins facilitates their incorporation in these polymers.

It should be noted that the polymer matrix must comply with some basic requirements to be used in the development of ATH-based indicators. In particular, it should be stable at low pH to assure anthocyanin stability, almost colourless to avoid masking ATH colour, moisture sensitive to allow the correct mode of action of the ATH-based indicator, and possess adequate mechanical properties. Generally, to obtain materials with these properties, two or more different polymers are combined and different additives are added. Some works have combined polymers of natural origin and others have mixed those from natural origin with other water soluble synthetic polymers, especially PVA (Fig. 4 and Table S2). Novel substances such as bentonite nanoclays (Koosha & Hamed, 2019), montmorillonite (Kang et al., 2018), chitosan nanoparticles or chitin nanocrystals (Ge et al., 2019; Ma, Liang, Cao, & Wang, 2018; Wu, Tsai, et al., 2020, 2019) (Table S2) have been added to improve mechanical, structural and barrier properties. It should be noted that many works use glycerol as plasticizer and linking agents such as boric acid, glutaraldehyde, polyphosphate or trimetaphosphate (Jiang et al., 2020; Kang et al., 2018; Mustafa et al., 2020; Pereira, de Arruda, & Stefani, 2015; Vo, Dang, & Chen, 2019; Zhang, Lu, & Chen, 2014).

The final properties of the material and the mode of action of the ATH-based indicator not only depend on the innate characteristics of the material or the ATH extract, but also from the foreseeable interactions between both of them. These interactions play a fundamental role in the final structural, mechanical properties and water sensitivity of the material, determine the ATH retention ability and may cause changes in the pH-dependent change of colour of ATH and in ATH retention in the material.

With respect to the alterations in the mechanical and structural properties of the polymers resulting from ATH addition, different results have been described depending on the combinations of polymers, ATH extracts and concentrations used. In some cases, no changes have been observed in polymer parameters such as tensile strength, elongation at break or water vapor permeability (Andretta et al., 2019; Chen & Gu, 2013; Uranga, Etxabide, Guerrero, & de la Caba, 2018; Yong, Wang, Zhang, et al., 2019). On the other hand, an improvement or decline in the properties of the material have been observed after the addition of the ATH extract (Andretta et al., 2019; Chi et al., 2020; Jiang et al., 2020; Kang et al., 2020; Koosha & Hamed, 2019; Liu et al., 2019; Luchese et al., 2018; Pourjavaher et al., 2017; Qin et al., 2019; Rawdkuen, Faseha, Benjakul, & Kaewprachu, 2020; Vo et al., 2019; Wei, Cheng, Ho, Tsai, & Mi, 2017; Wu et al., 2019, 2020; Yoshida, Maciel, Mendonça, & Franco, 2014; Zeng et al., 2019; Zhai et al., 2017; Zhang et al., 2019; Zhang et al., 2020). Additionally, some authors have described that these polymer alterations depend on the concentration of ATH included (Ge et al., 2019; Ma et al., 2018; Ma & Wang, 2016; Yong, Wang, Bai, et al., 2019; Yong, Wang, Zhang, et al., 2019; Zhang et al., 2020). However, although these changes in the polymer occur, they do not seem to play a critical factor in the development of ATH-based indicators, since indicator materials tend to be included as a label being a minor component of the package (Pereira et al., 2015). The effect of ATH-polymer interactions in the water sensitivity of the material, particularly in its swelling (water uptake) ability and its resistance in the presence of water, is an important issue considering that indicators based on pH changes are generally applied to foods with high water content. Higher swelling could interfere with the indicator colour response efficiency leading to a faster ATH release (Ebrahimi Tirtashi et al., 2019; Pourjavaher et al., 2017); while low resistance to water would limit the applicability of ATH-based indicators to foods with high water content such as meat or fish.

The addition of ATH into several polymer materials has revealed different effects in the water uptake and solubility of the material. Increasing water absorption and solubility rates after ATH addition have been explained by a less compact structure with weaker bonds between polymer components and by the high hydrophilicity of ATH (Andretta et al., 2019; Maftoonazad & Ramaswamy, 2019; Pourjavaher et al., 2017; Rawdkuen et al., 2020; Zhang et al., 2020). On the other hand, the reduction of water intake and solubility due to ATH addition has been attributed to ATH acetylation, and intermolecular interactions between the polymer components and ATH that decrease the interaction between the polymer and water (Ebrahimi Tirtashi et al., 2019; Halász & Csóka, 2018; Jiang et al., 2020; Moazami Goodarzi et al., 2020; Qin et al., 2019; Wang, Yong, et al., 2019; Wei et al., 2017). These variable results indicated that it is essential to evaluate the sensitivity to water of the intelligent material containing the ATH-based indicator to achieve a stable and efficient indicator in humid conditions.

As mentioned above, ATH affinity to water along with the swelling ability of some polymeric materials can lead to ATH migration from the anchoring substrate to the surface of the intelligent film due to the presence of water, thus affecting the correct performance of the indicator (Guo et al., 2020; Liang, Sun, Cao, Li, & Wang, 2019). In fact, some migration phenomena can be observed in photographs of ATH-based

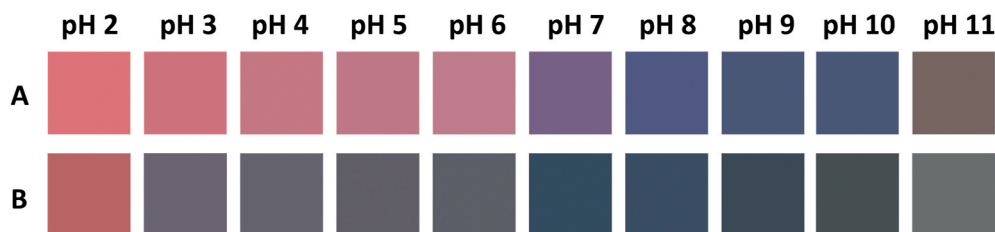


Fig. 5. Example of the colour variation of carrot ATHs (a) before and (b) after inclusion in cellulose/chitosan polymer. Adapted from Ebrahimi Tirtashi et al. (2019). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

indicators that have been submerged in buffers with different pH (Chi et al., 2020; Choi, Lee, Lacroix, & Han, 2017; Pereira et al., 2015; da Silva, Mastrantonio, Costa, & de Moraes, 2019; Vo et al., 2019; Wei et al., 2017). Although this poses a serious problem when developing this type of ATH-based intelligent packaging materials, only a few research works carried out released studies to evaluate this major bottleneck and propose solutions. For instance, Guo et al. (2020) observed migration of purple sweet potato ATH from fiber mats of zein and pullulan. This migration was initially high, due to the purple sweet potato located on or near the fiber surface, followed by a gradual increase in the release ascribed to the entrapped dye in the fiber. Migration was also detected in  $\kappa$ -carrageenan films containing *Lycium ruthenicum* Murr extract (Liu et al., 2019) and also in chitosan films containing black chokeberry (Halász & Csóka, 2018). Other study also mentions the reduction in chroma values which was attributed to the migration of red cabbage extracts from PVA films after storage at 5 °C for 30 days (Maftoonazad & Ramaswamy, 2019). To increase chemical interactions between ATH and polymeric matrix and, consequently, diminish ATH migration, low percentages of anionic polymers or other molecules have been added to film forming components. Using this approach, sodium carboxymethyl cellulose or pectin polymers have been added to films based on gum or chitosan, respectively, to achieve low release rates (Liang et al., 2019; Maciel, Yoshida, & Franco, 2015). Regarding anionic molecules, negative charged oxidized nanocrystals and soy protein isolates have been added to konjac glucomannan films or gellan gum-based films, respectively, to obtain a reduction of ATH migration (Wu, Tsai, et al., 2020; Wu, Tsai, et al., 2020). Finally, chemical interactions between polymer matrices and anthocyanins can also result in some variation in the hue of ATH at different pHs and affect the colour variation efficiency of the ATH-based indicator (Pourjavaher et al., 2017). For instance, electrostatic interactions and hydrogen bonds observed between hydroxyl, amino and amide groups of chitosan, gelatin, pullulan, polyvinyl alcohol or cellulose (Ge et al., 2019; Guo et al., 2020; Liang et al., 2019; Merz et al., 2020; Pourjavaher et al., 2017; Silva-Pereira, Teixeira, Pereira-Júnior, & Stefani, 2015; Wang et al., 2019a; Wu et al., 2019) could affect the electronic transitions responsible for the UV/Vis absorption bands and cause variations in the coloration of the ATH extracts. Fig. 5 shows an example of this phenomenon in black carrot extract included in a cellulose/chitosan polymer.

The most widely used method for the laboratory production of ATH-based intelligent films is casting. This method consists of pouring the polymeric film-forming solution containing ATH on small plates and subsequent drying. Despite of their simplicity, the casting methodology is not a common method used in the packaging industry since it is difficult to scale-up to produce large films and requires long drying times (De Moraes, Scheibe, Sereno, & Laurindo, 2013). Another method that has been used in some works is the dip coating method (Ebrahimi Tirtashi et al., 2019; Moazami Goodarzi et al., 2020; Kang et al., 2018; Kuswandi, Asih, Pratoko, Kristiningrum, & Moradi, 2020; Mohammadlinejad, Almasi, & Moradi, 2020; Moradi, Tajik, Almasi, Forough, & Ezati, 2019; Pourjavaher et al., 2017). This procedure is based on the coating of a polymeric material with a layer of ATH by immersing it into an ATH solution that can contain other polymeric substances to facilitate ATH adherence to the film. The polymeric materials used are generally very porous and absorbent such as paper or bacterial cellulose, to ensure the absorption of enough ATH concentration for the correct performance of the intelligent packaging. For example, Kang et al. (2018) used a less porous material (polyethylene vinyl alcohol and montmorillonite) and had to employ three or five layers of ATH colorimetric coating to obtain good film coloration. Electrospinning have also been used to prepare fibrous films of zein, polycaprolactone-polyethylene oxide, PVA or pullulan-zein (Guo et al., 2020; Maftoonazad & Ramaswamy, 2019; Prietto et al., 2018; da Silva et al., 2019) containing ATH. Fibrous mats have the advantage of possessing high porosity, small pore size and high surface-to-volume ratio that makes them very suitable for the loading of high amounts of active substances (Becerril, Nerín, & Silva, 2020).

#### 4.3. Application of ATH-based indicators to food

Many of the developed ATH-based indicators have been tested for monitoring the quality and freshness of selected types of packaged foods (Table 1). The food products tested display an increase or decrease on their pH during their shelf-life, with this pH change being related to ATH changes in colour. Besides, as mentioned before, they possess high water content, which is necessary to create a humid environment for the pH sensing of the ATH to occur. In particular, ATH-based indicators have been applied mainly to pork meat, fish, shrimp and milk (Table 1). Additionally, ATH-based sensors application to chicken meat and rutab fruit has also been described (Table 1). In the case of meat, fish and shrimp, ATH monitors pH increase resulting mainly from microbial growth and the subsequent production of alkaline compounds, mainly volatile nitrogen containing substances. The high volatility of these alkaline compounds allows the placement of the ATH-based pH indicator in the headspace of the food package, which means without direct contact between the food product and the ATH-based indicator. In the case of milk, ATH detects the reduction of pH that produces the growth of microorganisms, mainly acid lactic bacteria.

The application of ATH-based indicators on food packaging has demonstrated that these new intelligent materials change their colour along with the deterioration of the food product. However, their operation varies greatly between works, which can be considered reasonable taking into account the use of different ATH extracts, polymers and ATH concentrations. Some materials showed very distinguishable colour changes while others have colour variations more difficult to perceive by the naked eye. Some examples of ATH-based indicators applied to food products with colour changes that can be easily perceived by the consumers are shown in Table 2. It is important to point out that most research studies not only describe the relation between colour change and microbial spoilage but have also linked it to the expected shelf-life of the food product, which is the ultimate goal for a freshness indicator. In the case of pork, shrimp and fish, the authors have established that the end of the shelf-life of these products is attained when the concentration of volatile basic nitrogen substances (total volatile basic nitrogen value, TVBN) is higher than the limit established by current legislation and when the number of microorganism is higher than 7 log CFU/g (Jiang et al., 2020; Kang et al., 2018; Qin et al., 2019; Zeng et al., 2019). In the case of milk, the end of shelf-life has been established according to the total acid content (TA) value. Milk with TA values higher than 20°D is not accepted by qualified judges while milk with values higher than 30°D is not accepted by common consumers (Ebrahimi Tirtashi et al., 2019; Kuswandi et al., 2020; Moazami Goodarzi et al., 2020).

As mentioned, ATH-based indicators applied to meat and products can be used without direct contact between the indicator and the food product. Taking advantage of this feature, the most used approach is the placement of ATH-based indicators in the inner side of the package without any contact with the food (Table 1) to only detect changes of pH caused by volatile substances released by the food during its storage and subsequent deterioration. Only some works have tested these intelligent materials directly on the surface of meat or fish products (Luchese et al., 2018; Vo et al., 2019; Wu, Tsai, et al., 2020) (Table 1); probably because it involves more disadvantages. In direct contact, the hydrophilic polymer indicators are exposed to higher water concentrations that could produce a high water uptake. Besides, the ATH extract could migrate to the food product easily as described by Luchese et al., 2018 after direct contact between chicken meat and ATH-based indicator containing blueberry residue. Along with that, food compounds could interfere with the correct functioning of the indicator or yield higher ATH degradation. In the headspace, the indicator is exposed to a more limited number of substances that can interact with ATH. Although indirect contact is the preferred mode of application of ATH-based sensors, there are some situations where direct contact is needed. For instance, in the use of ATH based indicators in milk, direct contact is required (Table 1), although the indicator is not continuously submerged in the milk but is only

**Table 1**  
ATH-based indicators applied to food products.

Tested food	Source of ATH	Polymeric material	ATH extract concentration	Type of contact food-indicator	Storage temperature	Reference
Milk	Red cabbage	PVA/chitosan film cross-linked with tripolyphosphate	25% total volume of the mixture of hydrogels	Immersion	25 °C	Pereira et al. (2015)
		PVA/starch film cross-linked with boric acid	No data	Immersion	No data	Mustafa et al. (2020)
	Grape skins	Bacterial cellulose coated with ATH	Extract solution	Immersion	25 °C	Kuswandi et al. (2020)
		Tara gum/cellulose nanocrystals film	10 g/100 g (tara gum basis)	Immersion	Room temperature	Ma and Wang (2016)
	Black carrot	Paper coated with a solution of chitosan/tetraethyl orthosilicate/ZnO nanoparticles containing ATH extract	10.5 mg total ATH content/100 mL chitosan solution	Immersion	20 °C	Ebrahimi Tirtashi et al. (2019)
		Paper coated with a solution of starch containing ATH extract	10 mg total ATH content/100 mL filmogenic solution	Immersion	20 °C	Moazami Goodarzi et al. (2020)
		Chitosan film	1, 2 and 3% (w/w) (chitosan basis)	Immersion	40 °C	Yong, Wang, Zhang, et al. (2019)
Pork	Purple and black eggplant peel	κ-carrageenan film	1.5, 2.5, 3.5 and 4.5% (w/w) (κ-carrageenan basis)	Direct contact	25 °C	Liu et al. (2019)
	<i>Lycium ruthenicum</i>	κ-carrageenan film	1.5, 2.5, 3.5 and 4.5% (w/w) (κ-carrageenan basis)	Direct contact	25 °C	Liu et al. (2019)
	Murr fruit	Zein/pullulan fiber mat	20% (w/w) (pullulan fibers basis)	Headspace	25 °C	Guo et al. (2020)
	Black carrot	Agar/potato starch film	No data	Direct contact	25 °C	Choi et al. (2017)
shrimp	Purple sweet potato	Chitosan/PVA cross-linked with tripolyphosphate	25 mL/75 mL filmogenic solution	Direct contact	room temperature	(Vo et al., 2019)
	Red cabbage	Starch/PVA films	2.5% (w/w) total mixed hydrogels	Headspace	25 °C	(Zhang et al., 2019)
	Roselle calyx	Cassava starch film	1, 2 and 4% (w/w) (starch basis)	Headspace	25 °C	Qin et al. (2019)
	<i>Lycium ruthenicum</i>	κ-carrageenan/hydroxypropylcellulose film	6% (w/w) (κ-carrageenan and hydroxypropylcellulose basis)	Headspace	25 °C	Chi et al. (2020)
	Murr	Cassava starch film	0.5% (w/w) filmogenic suspension	Headspace	4 °C	Golasz, Silva, and Botelho Da Silva (2013)
	Grape skin	κ-carrageenan/hydroxypropylcellulose film	2, 4, 8 and 16% (w/w) κ-carrageenan and hydroxypropyl ethylcellulose basis)	Headspace	25 °C	Sun et al. (2019)
	<i>Prunus maackii</i> fruit	Chitosan cross-linked with glutaraldehyde coated with ATH extract	Extract solution	Headspace	25 °C	(Zhang et al., 2014)
	<i>Bauhinia blakeana</i> Dunn flower	Polyethylene vinyl alcohol/montmorillonite film coated with a solution of polyethylene vinyl alcohol containing the ATH extract	100 mL ATH solution mixed with 100 mL polyethylene vinyl alcohol solution	Headspace	25 °C	Kang et al. (2018)
	Mulberry	Gelatine/Oxidized chitin nanocrystals film	20, 40, 80, and 100 mg/100 mL filmogenic solution	Headspace	25 °C	Ge et al. (2019)
	Black rice	Chitosan/oxidized chitin nanocrystals	1%, 3% and 5% (w/w, chitin and chitosan basis)	Direct contact	25 °C	(Wu, Tsai, et al., 2020)
fish	<i>Echium amoenum</i> flowers	Bacterial cellulose coated with ATH extract solution	1:1 dilution of extract solution	Headspace	4 °C	Mohammadalinejad et al. (2020)
	Jambolan fruit	Chitosan/PVA film	0.3 g of ATH/100 g macromolecules	Headspace	−20, 4 and 20 °C	Merz et al. (2020)
	Red cabbage and purple sweet potato	Corn starch/PVA film	0.25, 0.5 and 1% (w/v filmogenic solution)	Headspace	25 °C	(Zhang et al., 2020)
	Rose petals	PVA/okra mucilage polysaccharide film	25 mL/100 mL filmogenic solution	Headspace	25 °C	Kang et al. (2020)
	<i>Lycium ruthenicum</i>	κ-carrageenan film	1.5, 2.5, 3.5 and 4.5% (w/w, κ-carrageenan basis)	Direct contact	25 °C	Liu et al. (2019)
	Murr fruit	Chitosan/oxidized chitin nanocrystals	1%, 3% and 5% (w/w, chitin and chitosan basis)	Direct contact	25 °C	(Wu et al., 2019)
	Black rice	Starch/PVA film	4% (v/v) filmogenic solution	Headspace	4 °C	(Chen, Zhang, Bhandari, & Yang, 2020)
	Purple sweet potatoes/curcumin	Carboxymethylcellulose/starch film cross-linked with trimetaphosphate	0.1, 0.3, 0.5, 0.7, and 0.9 g/100 g filmogenic solution	Headspace	25 °C	Jiang et al. (2020)
	Purple sweet potato	Bacterial cellulose nanofibers coated with ATH extract solution	Extract solution of 6 mg/mL of total anthocyanin content	Headspace	4 °C	Moradi et al. (2019)
	black carrot	Gelatine/oxidized chitin nanocrystals film	20, 40, 80, and 100 mg/100 mL filmogenic solution	Headspace	25 °C	Ge et al. (2019)
Roselle calyx	Starch/PVA film		Headspace	4 °C	Zhai et al. (2017)	

(continued on next page)

**Table 1** (continued)

Tested food	Source of ATH	Polymeric material	ATH extract concentration	Type of contact food-indicator	Storage temperature	Reference
			30, 60 and 120 mg of ATH/100 g of starch			
	Mulberry	Gelatin/PVA film	30, mg/100 mL filmogenic solution	Headspace	25 °C	Zeng et al. (2019)
		PVA/chitosan nanoparticles film	10, 20, 30 or 40% (w/w, PVA basis)	Headspace	Room temperature	Ma et al. (2018)
	Red cabbage	Chitosan/corn starch film	5 mL/100 mL of hydrogel mixture	Headspace	Refrigeration (4–7 °C) and room temperature (25 °C)	Pereira et al. (2015)
	<i>Vitis amurensis</i> husk	Tara gum/cellulose nanocrystals film	10% (w/w, tara gum basis)	Headspace	Room temperature	Ma, Ren, Gu, and Wang (2017)
Chicken meat	Blueberry residue	Cassava starch film	4 g/100 mL of filmogenic solution	Direct contact and headspace	6 °C	Luchese et al. (2018)
	Butterfly pea flower	Sago film	15% w/v of filmogenic solution	Headspace	23 °C	Ahmad
Rutab fruit	Red cabbage	PVA nanofiber mat	30% (g/g PVA)	Direct contact	25 °C	Maftoonazad and Ramaswamy (2019)

**Table 2**  
Color response of some ATH-based indicators applied to food products.

ATH-based indicator	Tested food	Colour response	Reference
Black carrot-starch film	Milk		Moazami Goodarzi et al. (2020)
Zein/pullulan fibers	Pork		Guo et al. (2020)
Mulberry coated PVA/Montmorillonite film	Shrimp		Kang et al. (2018)
Rose-PVA/okra mucilage polysaccharide	Shrimp		Kang et al. (2020)
Purple sweet potato-carboxymethylcellulose/starch	Fish		Jiang et al. (2020)
Black carrot coated bacterial cellulose nanofibers	Fish		Moradi et al. (2019)

(s) spoiled.

immersed some seconds at different times of storage. This method reduces the contact times and therefore the possible associated adverse effects.

The evaluation of the practical applicability of ATH-based indicators in food spoilage has been carried out in many works at much higher temperatures (20–25 °C) than those used commonly to store these fresh food products (Table 1). As the spoilage process in fresh products and the volatility of the chemical compounds largely depends on the storage temperature, tests at refrigeration temperatures should be performed to assure the correct performance of the pH indicator. In order to fulfill this goal, several works have been carried out at refrigerated temperatures using food products such as pork (Golasz et al., 2013), shrimp (Merz et al., 2020; Mohammadalinejad et al., 2020), fish (Chen et al., 2020;

Moradi et al., 2019; Pereira et al., 2015; Zhai et al., 2017) and rutab fruit (Maftoonazad & Ramaswamy, 2019) (Table 1) with good results.

The colour stability of a colorimetric indicator is considered as an essential parameter to give appropriate visual feedback to the consumer (Mohammadalinejad et al., 2020). Therefore, the study of the colour evolution during storage of ATH-based indicators is compulsory for a correct applicability, even more when considering the low chemical stability of ATH. Even though only a few works addressed this topic, the results indicate that materials containing ATH change their colour during storage being the variations more noticeable at higher temperatures. Changes were detected in films containing purple sweet potato after 180 days (Chen et al., 2020), jambolan fruit after 15 days (Merz et al., 2020) and roselle after 16 days (Zhang et al., 2019). Zhai et al.



(2017) also observed colour changes after 14 days in films stored at 4 °C containing roselle but to a lesser extent. Mohammadinejad et al. (2020) detected colour variations in the two first days of storage in an indicator containing *E. amoenun*, with subsequent colour maintenance in the following 4 days. In sum, More research should be conducted to study the stability of ATH-based indicators and to apply strategies for its improvement as encapsulation or copigmentation.

## 5. Conclusions

ATH-based indicators have demonstrated a great potential for monitoring freshness of several widely consumed fresh products such as seafood, meat or milk. ATH can track pH changes associated with food decay by a highly noticeable colour change that can be easily perceived by consumers. Compared to other pH-based freshness indicators, ATH have the advantage of being of natural origin, cheap and easy to incorporate in eco-friendly water-based polymers such as chitosan, cellulose or starch.

However, there are still some issues that should be overcome for a successful implementation of ATH-based indicators. Namely, the stability of ATH-based indicators should be assured during storage and distribution times, ATH losses due to migration should be limited and, the indicator water resistance should be improved. An increment of ATH stability could be achieved by copigmentation with other substances or by the encapsulation of ATH. Migration could be reduced by the incorporation of negatively charged substances in the polymer matrix that interact with ATH by covalent or non-covalent bonds. Encapsulation might be also used to control or reduce ATH release from the polymer. Besides, food trials should be extended to ensure the correct performance of ATH-based indicators considering that meat and fish products can vary their pH due to different factors such as breed, feeding, slaughter conditions or further processing. At the industrial level, more research is needed to scale-up the production of ATH-based indicators and to develop technically and economically feasible methods to include them in the common packaging used in food. Regarding this topic, it could be interesting to evaluate the behaviour of these indicators under real storage conditions such as modified atmosphere, refrigerated storage, changes in temperature during storage and distribution, among others.

## Author contributions

RB is responsible for the conceptualization and the writing of the original draft FS and CN are responsible for the review and editing of the manuscript.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.tifs.2021.02.042>.

## References

- Ahmad, S. R., Gokulakrishnan, P., Giriprasad, R., & Yattoo, M. A. (2015). Fruit-based natural antioxidants in meat and meat products: A review. *Critical Reviews in Food Science and Nutrition*, 55(11), 1503–1513. <https://doi.org/10.1080/10408398.2012.701674>
- Alizadeh-Sani, M., Mohammadian, E., Rhim, J. W., & Jafari, S. M. (2020). pH-sensitive (halochromic) smart packaging films based on natural food colorants for the

- monitoring of food quality and safety. *Trends in Food Science & Technology*, 105, 93–144. <https://doi.org/10.1016/j.tifs.2020.08.014>
- Andretta, R., Luchese, C. L., Tessaro, I. C., & Spada, J. C. (2019). Development and characterization of pH-indicator films based on cassava starch and blueberry residue by thermocompression. *Food Hydrocolloids*, 93, 317–324. <https://doi.org/10.1016/j.foodhyd.2019.02.019>
- Balbinot-Alfaro, E., Craveiro, D. V., Lima, K. O., Costa, H. L. G., Lopes, D. R., & Prentice, C. (2019). Intelligent packaging with pH indicator potential. *Food Engineering Reviews*, 11(4), 235–244. <https://doi.org/10.1007/s12393-019-09198-9>
- Becerril, R., Nerín, C., & Silva, F. (2020). Encapsulation systems for antimicrobial food packaging components: An update. *Molecules*, 25(5), 1134–1174. <https://doi.org/10.3390/molecules25051134>
- Bhargava, N., Sharanagat, V. S., Mor, R. S., & Kumar, K. (2020). Active and intelligent biodegradable packaging films using food and food waste-derived bioactive compounds: A review. *Trends in Food Science & Technology*, 105, 385–401. <https://doi.org/10.1016/j.tifs.2020.09.015>
- Borzi, F., Torrieri, E., Wrona, M., & Nerín, C. (2019). Polyamide modified with green tea extract for fresh minced meat active packaging applications. *Food Chemistry*, 300, 125242. <https://doi.org/10.1016/j.foodchem.2019.125242>
- Castañeda-Ovando, A., Pacheco-Hernández, M. de L., Páez-Hernández, M. E., Rodríguez, J. A., & Galán-Vidal, C. A. (2009). Chemical studies of anthocyanins: A review. *Food Chemistry*, 113(4), 859–871. <https://doi.org/10.1016/j.foodchem.2008.09.001>
- Cavalcanti, R. N., Santos, D. T., & Meireles, M. A. A. (2011). Non-thermal stabilization mechanisms of anthocyanins in model and food systems—An overview. *Food Research International*, 44(2), 499–509. <https://doi.org/10.1016/j.foodres.2010.12.007>
- Chen, X., & Gu, Z. (2013). Absorption-type optical pH sensitive film based on immobilized purple cabbage pigment. *Sensors and Actuators B: Chemical*, 178, 207–211. <https://doi.org/10.1016/j.snb.2012.12.094>
- Chen, H. zhi, Zhang, M., Bhandari, B., & Yang, C. hui (2020). Novel pH-sensitive films containing curcumin and anthocyanins to monitor fish freshness. *Food Hydrocolloids*, 100, 105438. <https://doi.org/10.1016/j.foodhyd.2019.105438>
- Chi, W., Cao, L., Sun, G., Meng, F., Zhang, C., Li, J., et al. (2020). Developing a highly pH-sensitive κ-carrageenan-based intelligent film incorporating grape skin powder via a cleaner process. *Journal of Cleaner Production*, 244, 118862. <https://doi.org/10.1016/j.jclepro.2019.118862>
- Choi, I., Lee, J. Y., Lacroix, M., & Han, J. (2017). Intelligent pH indicator film composed of agar/potato starch and anthocyanin extracts from purple sweet potato. *Food Chemistry*, 218, 122–128. <https://doi.org/10.1016/j.foodchem.2016.09.050>
- Chung, C., Rojanasasithara, T., Mutilangi, W., & McClements, D. J. (2015). Enhanced stability of anthocyanin-based color in model beverage systems through whey protein isolate complexation. *Food Research International*, 76, 761–768. <https://doi.org/10.1016/j.foodres.2015.07.003>
- Clifford, M. N. (2000). Anthocyanins – nature, occurrence and dietary burden. *Journal of the Science of Food and Agriculture*, 80(7), 1063–1072. [https://doi.org/10.1002/\(SICI\)1097-0010\(20000515\)80:7<1063::AID-JSFA605>3.0.CO;2-Q](https://doi.org/10.1002/(SICI)1097-0010(20000515)80:7<1063::AID-JSFA605>3.0.CO;2-Q)
- Cortez, R., Luna-Vital, D. A., Margulis, D., & Gonzalez de Mejia, E. (2017). Natural pigments: Stabilization methods of anthocyanins for food applications. *Comprehensive Reviews in Food Science and Food Safety*, 16(1), 180–198. <https://doi.org/10.1111/1541-4337.12244>
- De Moraes, J. O., Scheibe, A. S., Sereno, A., & Laurindo, J. B. (2013). Scale-up of the production of cassava starch based films using tape-casting. *Journal of Food Engineering*, 119(4), 800–808. <https://doi.org/10.1016/j.jfoodeng.2013.07.009>
- Ebrahimi Tirtashi, F., Moradi, M., Tajik, H., Forough, M., Ezati, P., & Kuswandi, B. (2019). Cellulose/chitosan pH-responsive indicator incorporated with carrot anthocyanins for intelligent food packaging. *International Journal of Biological Macromolecules*, 136, 920–926. <https://doi.org/10.1016/j.ijbiomac.2019.06.148>
- EFSA. (2013). Scientific Opinion on the re-evaluation of anthocyanins (E 163) as a food additive. *EFSA Journal*, 11(4), 3145–3196. <https://doi.org/10.2903/j.efsa.2013.3145>
- Fang, Z., Zhao, Y., Warner, R. D., & Johnson, S. K. (2017). Active and intelligent packaging in meat industry. *Trends in Food Science & Technology*, 61, 60–71. <https://doi.org/10.1016/j.tifs.2017.01.002>
- Ge, Y., Li, Y., Bai, Y., Yuan, C., Wu, C., & Hu, Y. (2019). Intelligent gelatin/oxidized chitin nanocrystals nanocomposite films containing black rice bran anthocyanins for fish freshness monitorings. *International Journal of Biological Macromolecules*, 155, 1296–1306. <https://doi.org/10.1016/j.ijbiomac.2019.11.101>
- Gherardi, R., Becerril, R., Nerin, C., & Bosetti, O. (2016). Development of a multilayer antimicrobial packaging material for tomato puree using an innovative technology. *Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology*, 72. <https://doi.org/10.1016/j.lwt.2016.04.063>
- Golasz, L. B., Silva, J. D. a, & Botelho Da Silva, S. (2013). Film with anthocyanins as an indicator of chilled pork deterioration. *Ciencia y Tecnología Alimentaria*, 33(Supl. 1), 155–162.
- Gómez-Estaca, J., Gavara, R., Catalá, R., & Hernández-Muñoz, P. (2016). The potential of proteins for producing food packaging materials: A review. *Packaging Technology and Science*, 29(4–5), 203–224. <https://doi.org/10.1002/pts.2198>
- Di Gu, K., Wang, C. K., Hu, D. G., & Hao, Y. J. (2019). How do anthocyanins paint our horticultural products? *Scientia Horticulturae*, 249, 257–262. <https://doi.org/10.1016/j.scienta.2019.01.034>
- Guo, M., Wang, H., Wang, Q., Chen, M., Li, L., Li, X., et al. (2020). Intelligent double-layer fiber mats with high colorimetric response sensitivity for food freshness monitoring and preservation. *Food Hydrocolloids*, 101, 105468. <https://doi.org/10.1016/j.foodhyd.2019.105468>

- Halász, K., & Csóka, L. (2018). Black chokeberry (*Aronia melanocarpa*) pomace extract immobilized in chitosan for colorimetric pH indicator film application. *Food Packaging and Shelf Life*, 16, 185–193. <https://doi.org/10.1016/j.foodpack.2018.03.002>
- Han, J. W., Ruiz-García, L., Qian, J. P., & Yang, X. T. (2018). Food packaging: A comprehensive review and future trends. *Comprehensive Reviews in Food Science and Food Safety*, 17(4), 860–877. <https://doi.org/10.1111/1541-4337.12343>
- Jiang, G., Hou, X., Zeng, X., Zhang, C., Wu, H., Shen, G., ... Zhang, Z. (2020). Preparation and characterization of indicator films from carboxymethyl-cellulose/starch and purple sweet potato (*Ipomoea batatas* (L.) lam) anthocyanins for monitoring fish freshness. *International Journal of Biological Macromolecules*, 143, 359–372. <https://doi.org/10.1016/j.ijbiomac.2019.12.024>
- Kalpna, S., Priyadarshini, S. R., Maria Leena, M., Moses, J. A., & Anandharamkrishnan, C. (2019). Intelligent packaging: Trends and applications in food systems. *Trends in Food Science & Technology*, 93, 145–157. <https://doi.org/10.1016/j.tifs.2019.09.008>
- Kang, S., Wang, H., Guo, M., Zhang, L., Chen, M., Jiang, S., ... Jiang, S. (2018). Ethylene-vinyl alcohol copolymer-montmorillonite multilayer barrier film coated with mulberry anthocyanin for freshness monitoring. *Journal of Agricultural and Food Chemistry*, 66(50), 13268–13276. <https://doi.org/10.1021/acs.jafc.8b05189>
- Kang, S., Wang, H., Xia, L., Chen, M., Li, L., Cheng, J., ... Jiang, S. (2020). Colorimetric film based on polyvinyl alcohol/okra mucilage polysaccharide incorporated with rose anthocyanins for shrimp freshness monitoring. *Carbohydrate Polymers*, 229, 115402. <https://doi.org/10.1016/j.carbpol.2019.115402>
- Kho, H. E., Azlan, A., Tang, S. T., & Lim, S. M. (2017). Anthocyanidins and anthocyanins: Colored pigments as food, pharmaceutical ingredients, and the potential health benefits. *Food & Nutrition Research*, 61, 1361779. <https://doi.org/10.1080/16546628.2017.1361779>
- Koh, J., Xu, Z., & Wicker, L. (2020). Binding kinetics of blueberry pectin-anthocyanins and stabilization by non-covalent interactions. *Food Hydrocolloids*, 99, 105354. <https://doi.org/10.1016/j.foodhyd.2019.105354>
- Koosha, M., & Hamedii, S. (2019). Intelligent Chitosan/PVA nanocomposite films containing black carrot anthocyanin and bentonite nanoclays with improved mechanical, thermal and antibacterial properties. *Progress in Organic Coatings*, 127, 338–347. <https://doi.org/10.1016/j.porgcoat.2018.11.028>
- Kurek, M., Garofulić, I. E., Bakić, M. T., Šćetar, M., Uzelac, V. D., & Galić, K. (2018). Development and evaluation of a novel antioxidant and pH indicator film based on chitosan and food waste sources of antioxidants. *Food Hydrocolloids*, 84, 238–246. <https://doi.org/10.1016/j.foodhyd.2018.05.050>
- Kuswandi, B. (2017). Freshness sensors for food packaging. *Reference module in food science*. <https://doi.org/10.1016/b978-0-08-100596-5.21876-3>
- Kuswandi, B., Asih, N. P. N., Pratoko, D. K., Kristiningrum, N., & Moradi, M. (2020). Edible pH sensor based on immobilized red cabbage anthocyanins into bacterial cellulose membrane for intelligent food packaging. *Packaging Technology and Science*, 1–12. <https://doi.org/10.1002/pts.2507>
- Liang, T., Sun, G., Cao, L., Li, J., & Wang, L. (2019). A pH and NH<sub>3</sub> sensing intelligent film based on Artemisia sphaerocephala Krasch. gum and red cabbage anthocyanins anchored by carboxymethyl cellulose sodium added as a host complex. *Food Hydrocolloids*, 87, 858–868. <https://doi.org/10.1016/j.foodhyd.2018.08.028>
- Liu, J., Wang, H., Guo, M., Li, L., Chen, M., Jiang, S., ... Jiang, S. (2019). Extract from *Lycium ruthenicum* Murr. Incorporating κ-carrageenan colorimetric film with a wide pH-sensing range for food freshness monitoring. *Food Hydrocolloids*, 94, 1–10. <https://doi.org/10.1016/j.foodhyd.2019.03.008>
- Li, S., Wu, B., Fu, W., & Reddivari, L. (2019). The anti-inflammatory effects of dietary anthocyanins against ulcerative colitis. *International Journal of Molecular Sciences*, 20. <https://doi.org/10.3390/ijms20102588>
- Li, A., Xiao, R., He, S., An, X., He, Y., Wang, C., ... He, J. (2019). Research advances of purple sweet potato anthocyanins: Extraction, identification, stability, bioactivity, application, and biotransformation. *Molecules*, 24(21), 3816. <https://doi.org/10.3390/molecules24213816>
- Lorenzo, J. M., Pateiro, M., Domínguez, R., Barba, F. J., Putnik, P., Kovačević, D. B., ... Franco, D. (2018). Berries extracts as natural antioxidants in meat products: A review. *Food Research International*, 106, 1095–1104. <https://doi.org/10.1016/j.foodres.2017.12.005>
- Luchese, C. L., Abdalla, V. F., Spada, J. C., & Tessaro, I. C. (2018). Evaluation of blueberry residue incorporated cassava starch film as pH indicator in different simulants and foodstuffs. *Food Hydrocolloids*, 82, 209–218. <https://doi.org/10.1016/j.foodhyd.2018.04.010>
- Luchese, C. L., Sperotto, N., Spada, J. C., & Tessaro, I. C. (2017). Effect of blueberry agro-industrial waste addition to corn starch-based films for the production of a pH-indicator film. *International Journal of Biological Macromolecules*, 104, 11–18. <https://doi.org/10.1016/j.ijbiomac.2017.05.149>
- Maciel, V. B. V., Yoshida, C. M. P., & Franco, T. T. (2015). Chitosan/pectin polyelectrolyte complex as a pH indicator. *Carbohydrate Polymers*, 132, 537–545. <https://doi.org/10.1016/j.carbpol.2015.06.047>
- Maftoonazad, N., & Ramasamy, H. (2019). Design and testing of an electrospun nanofiber mat as a pH biosensor and monitor the pH associated quality in fresh date fruit (Rutab). *Polymer Testing*, 75, 76–84. <https://doi.org/10.1016/j.polymertesting.2019.01.011>
- Ma, Q., Liang, T., Cao, L., & Wang, L. (2018). Intelligent poly (vinyl alcohol)-chitosan nanoparticles-mulberry extracts films capable of monitoring pH variations. *International Journal of Biological Macromolecules*, 108, 576–584. <https://doi.org/10.1016/j.ijbiomac.2017.12.049>
- Ma, Q., Ren, Y., Gu, Z., & Wang, L. (2017). Developing an intelligent film containing *Vitis amurensis* husk extracts: The effects of pH value of the film-forming solution. *Journal of Cleaner Production*, 166, 851–859. <https://doi.org/10.1016/j.jclepro.2017.08.099>
- Ma, Q., & Wang, L. (2016). Preparation of a visual pH-sensing film based on tara gum incorporating cellulose and extracts from grape skins. *Sensors and Actuators B: Chemical*, 235, 401–407. <https://doi.org/10.1016/j.snb.2016.05.107>
- Merz, B., Capello, C., Leandro, G. C., Moritz, D. E., Monteiro, A. R., & Valencia, G. A. (2020). A novel colorimetric indicator film based on chitosan, polyvinyl alcohol and anthocyanins from jambolan (*Syzygium cumini*) fruit for monitoring shrimp freshness. *International Journal of Biological Macromolecules*, 153, 625–632. <https://doi.org/10.1016/j.ijbiomac.2020.03.048>
- Moazami Goodarzi, M., Moradi, M., Tajik, H., Forough, M., Ezati, P., & Kuswandi, B. (2020). Development of an easy-to-use colorimetric pH label with starch and carrot anthocyanins for milk shelf life assessment. *International Journal of Biological Macromolecules*, 153, 240–247. <https://doi.org/10.1016/j.ijbiomac.2020.03.014>
- Mohamed, S. A. A., El-Sakhawy, M., & El-Sakhawy, M. A. M. (2020). Polysaccharides, protein and lipid -based natural edible films in food packaging: A review. *Carbohydrate Polymers*, 238, 116178. <https://doi.org/10.1016/j.carbpol.2020.116178>
- Mohammadalinejad, S., Almasi, H., & Moradi, M. (2020). Immobilization of Echium amoenum anthocyanins into bacterial cellulose film: A novel colorimetric pH indicator for freshness/spoilage monitoring of shrimp. *Food Control*, 113. <https://doi.org/10.1016/j.foodcont.2020.107169>
- Moradi, M., Tajik, H., Almasi, H., Forough, M., & Ezati, P. (2019). A novel pH-sensing indicator based on bacterial cellulose nanofibers and black carrot anthocyanins for monitoring fish freshness. *Carbohydrate Polymers*, 222, 115030. <https://doi.org/10.1016/j.carbpol.2019.115030>
- Müller, P., & Schmid, M. (2019). Intelligent packaging in the food sector: A brief overview.  *Foods*, 8(1), 16. <https://doi.org/10.3390/foods8010016>
- Mustafa, P., Niazi, M. B. K., Jahan, Z., Samin, G., Hussain, A., Ahmed, T., et al. (2020). PVA/starch/propolis/anthocyanins rosemary extract composite films as active and intelligent food packaging materials. *Journal of Food Safety*, 40(1), Article e12725. <https://doi.org/10.1111/jfs.12725>
- Nerin, C., Becerril, R., Manso, S., & Silva, F. (2016). Ethyl lauroyl arginate (LAE): Antimicrobial activity and applications in food systems. *Antimicrobial food packaging*. <https://doi.org/10.1016/B978-0-12-800723-5.00023-1>
- Pereira, V. A., de Arruda, I. N. Q., & Stefani, R. (2015). Active chitosan/PVA films with anthocyanins from Brassica oleracea (Red cabbage) as Time-Temperature Indicators for application in intelligent food packaging. *Food Hydrocolloids*, 43, 180–188. <https://doi.org/10.1016/j.foodhyd.2014.05.014>
- Pourjavaher, S., Almasi, H., Meshkini, S., Pirsä, S., & Parandi, E. (2017). Development of a colorimetric pH indicator based on bacterial cellulose nanofibers and red cabbage (*Brassica oleracea*) extract. *Carbohydrate Polymers*, 156, 193–201. <https://doi.org/10.1016/j.carbpol.2016.09.027>
- Poyatos-Racionero, E., Ros-Lis, J. V., Vivancos, J. L., & Martínez-Mañez, R. (2018). Recent advances on intelligent packaging as tools to reduce food waste. *Journal of Cleaner Production*, 172, 3398–3409. <https://doi.org/10.1016/j.jclepro.2017.11.075>
- Prietto, L., Pinto, V. Z., El Halal, S. L. M., de Moraes, M. G., Costa, J. A. V., Lim, L.-T., ... Zavareze, E. da R. (2018). Ultrafine fibers of zein and anthocyanins as natural pH indicator. *Journal of the Science of Food and Agriculture*, 98(7), 2735–2741. <https://doi.org/10.1002/jsfa.8769>
- Qin, Y., Liu, Y., Yong, H., Liu, J., Zhang, X., & Liu, J. (2019). Preparation and characterization of active and intelligent packaging films based on cassava starch and anthocyanins from *Lycium ruthenicum* Murr. *International Journal of Biological Macromolecules*, 134, 80–90. <https://doi.org/10.1016/j.ijbiomac.2019.05.029>
- Rawdkuen, S., Faseha, A., Benjakul, S., & Kaewprachu, P. (2020). Application of anthocyanin as a color indicator in gelatin films. *Food Bioscience*, 36, 100603. <https://doi.org/10.1016/j.fbio.2020.100603>
- Rodriguez-Amaya, D. B. (2019). Update on natural food pigments - a mini-review on carotenoids, anthocyanins, and betalains. *Food Research International*, 124, 200–205. <https://doi.org/10.1016/j.foodres.2018.05.028>
- Roy, S., & Rhim, J.-W. (2020). Anthocyanin food colorant and its application in pH-responsive color change indicator films. *Critical Reviews in Food Science and Nutrition*, 1–29. <https://doi.org/10.1080/10408398.2020.1776211>
- Silva-Pereira, M. C., Teixeira, J. A., Pereira-Júnior, V. A., & Stefani, R. (2015). Chitosan/corn starch blend films with extract from *Brassica oleracea* (red cabbage) as a visual indicator of fish deterioration. *Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology*, 61(1), 258–262. <https://doi.org/10.1016/j.lwt.2014.11.041>
- Silva, S., Costa, E. M., Calhau, C., Morais, R. M., & Pintado, M. E. (2017). Anthocyanin extraction from plant tissues: A review. *Critical Reviews in Food Science and Nutrition*, 57(14), 3072–3083. <https://doi.org/10.1080/10408398.2015.1087963>
- Silva, F., Domingues, F. C., & Nerín, C. (2018). Control microbial growth on fresh chicken meat using pinosylvin inclusion complexes based packaging absorbent pads. *Lebensmittel-Wissenschaft und -Technologie- Food Science and Technology*, 89, 148–154. <https://doi.org/10.1016/j.lwt.2017.10.043>
- Silva, C. K. da, Mastrantonio, D. J. da S., Costa, J. A. V., & de Moraes, M. G. (2019). Innovative pH sensors developed from ultrafine fibers containing açai (*Euterpe oleracea*) extract. *Food Chemistry*, 294, 397–404. <https://doi.org/10.1016/j.foodchem.2019.05.059>
- Smeriglio, A., Barreca, D., Bellocchio, E., & Trombetta, D. (2016). Chemistry, pharmacology and health benefits of anthocyanins. *Phytotherapy Research*, 30, 1265–1286. <https://doi.org/10.1002/ptr.5642>
- Strauch, R. C., Mengist, M. F., Pan, K., Yousef, G. G., Iorizzo, M., Brown, A. F., et al. (2019). Variation in anthocyanin profiles of 27 genotypes of red cabbage over two growing seasons. *Food Chemistry*, 301, 125289. <https://doi.org/10.1016/j.foodchem.2019.125289>
- Sun, G., Chi, W., Zhang, C., Xu, S., Li, J., & Wang, L. (2019). Developing a green film with pH-sensitivity and antioxidant activity based on κ-carrageenan and hydroxypropyl

- methylcellulose incorporating Prunus maackii juice. *Food Hydrocolloids*, 94, 345–353. <https://doi.org/10.1016/j.foodhyd.2019.03.039>
- Tarone, A. G., Cazarin, C. B. B., & Marostica Junior, M. R. (2020). Anthocyanins: New techniques and challenges in microencapsulation. *Food Research International*, 133, 109092. <https://doi.org/10.1016/j.foodres.2020.109092>
- Tsuda, T. (2012). Dietary anthocyanin-rich plants: Biochemical basis and recent progress in health benefits studies. *Molecular Nutrition & Food Research*, 56, 159–170. <https://doi.org/10.1002/mnfr.201100526>
- Uranga, J., Etxabide, A., Guerrero, P., & de la Caba, K. (2018). Development of active fish gelatin films with anthocyanins by compression molding. *Food Hydrocolloids*, 84, 313–320. <https://doi.org/10.1016/j.foodhyd.2018.06.018>
- Vanderroost, M., Ragaert, P., Devlieghere, F., & De Meulenaer, B. (2014). Intelligent food packaging: The next generation. *Trends in Food Science & Technology*, 39(1), 47–62. <https://doi.org/10.1016/j.tifs.2014.06.009>
- Vo, Dang, & Chen. (2019). Synthesis of intelligent pH indicative films from chitosan/poly(vinyl alcohol)/anthocyanin extracted from red cabbage. *Polymers*, 11(7), 1088. <https://doi.org/10.3390/polym11071088>
- Wallace, T. C., & Giusti, M. M. (2015). Anthocyanins. *Advances in Nutrition*, 6(5), 619–622. <https://doi.org/10.3945/an.115.009233>
- Wang, S., Xia, P., Wang, S., Liang, J., Sun, Y., Yue, P., et al. (2019a). Packaging films formulated with gelatin and anthocyanins nanocomplexes: Physical properties, antioxidant activity and its application for olive oil protection. *Food Hydrocolloids*, 96, 617–624. <https://doi.org/10.1016/j.foodhyd.2019.06.004>
- Wang, X., Yong, H., Gao, L., Li, L., Jin, M., & Liu, J. (2019b). Preparation and characterization of antioxidant and pH-sensitive films based on chitosan and black soybean seed coat extract. *Food Hydrocolloids*, 89, 56–66. <https://doi.org/10.1016/j.foodhyd.2018.10.019>
- Wei, Y. C., Cheng, C. H., Ho, Y. C., Tsai, M. L., & Mi, F. L. (2017). Active gellan gum/purple sweet potato composite films capable of monitoring pH variations. *Food Hydrocolloids*, 69, 491–502. <https://doi.org/10.1016/j.foodhyd.2017.03.010>
- Wrona, M., Blasco, S., Becerril, R., Nerin, C., Sales, E., & Asensio, E. (2019). Antioxidant and antimicrobial markers by UPLC®–ESI-Q-TOF-MSE of a new multilayer active packaging based on Arctostaphylos uva-ursi. *Talanta*, 196. <https://doi.org/10.1016/j.talanta.2018.12.057>
- Wu, C., Li, Y., Sun, J., Lu, Y., Tong, C., Wang, L., et al. (2020). Novel konjac glucomannan films with oxidized chitin nanocrystals immobilized red cabbage anthocyanins for intelligent food packaging. *Food Hydrocolloids*, 98, 105245. <https://doi.org/10.1016/j.foodhyd.2019.105245>
- Wu, C., Sun, J., Zheng, P., Kang, X., Chen, M., Li, Y., ... Pang, J. (2019). Preparation of an intelligent film based on chitosan/oxidized chitin nanocrystals incorporating black rice bran anthocyanins for seafood spoilage monitoring. *Carbohydrate Polymers*, 222, 115006. <https://doi.org/10.1016/j.carbpol.2019.115006>
- Wu, L.-T., Tsai, I.-L., Ho, Y.-C., Hang, Y.-H., Lin, C., Tsai, M.-L., ... Pang, J. (2020). Active and intelligent gellan gum-based packaging films for controlling anthocyanins release and monitoring food freshness. *Carbohydrate Polymers*, 254, 117410. <https://doi.org/10.1016/j.carbpol.2020.117410>
- Wu, Y.-M., Wang, Z.-W., Hu, C.-Y., & Nerin, C. (2018). Influence of factors on release of antimicrobials from antimicrobial packaging materials. *Critical Reviews in Food Science and Nutrition*, 58(7), 1108–1121. <https://doi.org/10.1080/10408398.2016.1241215>
- Xu, X. J., Fang, S., Li, Y. H., Zhang, F., Shao, Z. P., Zeng, Y. T., ... Meng, Y. C. (2019). Effects of low acyl and high acyl gellan gum on the thermal stability of purple sweet potato anthocyanins in the presence of ascorbic acid. *Food Hydrocolloids*, 86, 116–123. <https://doi.org/10.1016/j.foodhyd.2018.03.007>
- Yam, K. L., Takhistov, P. T., & Miltz, J. (2005). Intelligent packaging: Concepts and applications. *Journal of Food Science*, 70(1), R1–R10. <https://doi.org/10.1111/j.1365-2621.2005.tb09052.x>
- Yong, H., Wang, X., Bai, R., Miao, Z., Zhang, X., & Liu, J. (2019). Development of antioxidant and intelligent pH-sensing packaging films by incorporating purple-fleshed sweet potato extract into chitosan matrix. *Food Hydrocolloids*, 90, 216–224. <https://doi.org/10.1016/j.foodhyd.2018.12.015>
- Yong, H., Wang, X., Zhang, X., Liu, Y., Qin, Y., & Liu, J. (2019). Effects of anthocyanin-rich purple and black eggplant extracts on the physical, antioxidant and pH-sensitive properties of chitosan film. *Food Hydrocolloids*, 94, 93–104. <https://doi.org/10.1016/j.foodhyd.2019.03.012>
- Yoshida, C. M. P., Maciel, V. B. V., Mendonça, M. E. D., & Franco, T. T. (2014). Chitosan biobased and intelligent films: Monitoring pH variations. *Lebensmittel-Wissenschaft und -Technologie: Food Science and Technology*, 55(1), 83–89. <https://doi.org/10.1016/j.lwt.2013.09.015>
- Zeng, P., Chen, X., Qin, Y. R., Zhang, Y. H., Wang, X. P., Wang, J. Y., ... Zhang, Y. S. (2019). Preparation and characterization of a novel colorimetric indicator film based on gelatin/polyvinyl alcohol incorporating mulberry anthocyanin extracts for monitoring fish freshness. *Food Research International*, 126, 108604. <https://doi.org/10.1016/j.foodres.2019.108604>
- Zhai, X., Shi, J., Zou, X., Wang, S., Jiang, C., Zhang, J., ... Holmes, M. (2017). Novel colorimetric films based on starch/polyvinyl alcohol incorporated with roselle anthocyanins for fish freshness monitoring. *Food Hydrocolloids*, 69, 308–317. <https://doi.org/10.1016/j.foodhyd.2017.02.014>
- Zhang, K., Huang, T. S., Yan, H., Hu, X., & Ren, T. (2020). Novel pH-sensitive films based on starch/polyvinyl alcohol and food anthocyanins as a visual indicator of shrimp deterioration. *International Journal of Biological Macromolecules*, 145, 768–776. <https://doi.org/10.1016/j.ijbiomac.2019.12.159>
- Zhang, X., Lu, S., & Chen, X. (2014). A visual pH sensing film using natural dyes from Bauhinia blakeana Dunn. *Sensors and Actuators B: Chemical*, 198, 268–273. <https://doi.org/10.1016/j.snb.2014.02.094>
- Zhang, J., Zou, X., Zhai, X., Huang, X. W., Jiang, C., & Holmes, M. (2019). Preparation of an intelligent pH film based on biodegradable polymers and roselle anthocyanins for monitoring pork freshness. *Food Chemistry*, 272, 306–312. <https://doi.org/10.1016/j.foodchem.2018.08.041>
- Zhao, C. L., Yu, Y. Q., Chen, Z. J., Wen, G. S., Wei, F. G., Zheng, Q., ... Xiao, X. L. (2017). Stability-increasing effects of anthocyanin glycosyl acylation. *Food Chemistry*, 214, 119–128. <https://doi.org/10.1016/j.foodchem.2016.07.073>