Food Bioscience 24 (2018) 111–119

Contents lists available at ScienceDirect

Food Bioscience

journal homepage: www.elsevier.com/locate/fbio

The effect of plasticizers on the functional properties of biodegradable gelatin-based film: A review

N. Suderman[a], M.I.N. Isa[b], N.M. Sarbon[a,*]

[a] School of Food Science and Technology, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia
[b] School of Fundamental Science, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia

ARTICLE INFO

Keywords:
Biodegradable films
Gelatin film
Plasticizers
Plasticization effect

ABSTRACT

Food packaging derived from a petroleum base represents a serious environmental problem. Finding alternative sustainable solutions is a must. Therefore, the current study has focused on the production of biodegradable food packaging from renewable materials, primarily gelatin. The effect of the biomaterials used on functional properties of the films produced needs thorough investigation. Gelatin represents interesting biomaterials for developing biodegradable food packaging, mainly due to their good film forming properties and abundantly in nature. However, the incorporation of gelatin in biodegradable films for food packaging may give some drawback on certain properties of the film such as tensile strength and water vapour permeability. Thus, addition of plasticizers into the film materials improves the functional properties of films by increasing their extensibility, dispensability, flexibility, elasticity, and rigidity. This study aims to review the current findings on how plasticizers impact the functional properties of biodegradable gelatin-based films. Plasticizers incorporation in the films may affect the continuity of the polymer matrix, leading to physical changes, where the films become more flexible and stretchable. Generally, the plasticization effect of plasticizers strengthens the film structure, in which the tensile strength and elongation of the films are improved and water barrier properties are reduced.

1. Introduction

Packages are used as a marketing tools to communicate with consumers, protect products from the deteriorating effects of the external environment, contain products of various sizes and shapes, and provide the consumer with ease of use and time-saving convenience (Otles & Yalcin, 2008). Food packaging provides a physical barrier between the outside environment and food products, thereby guaranteeing the hygiene and prolong the shelf life of perishable items, especially those prone to microbiological and oxidative deterioration (Gómez-Guillén et al., 2009). The most common materials used for food packaging are plastic, paper, glass, aluminium, fibreboard and steel. Petroleum-based plastics are commonly used as they offer various advantages over other packaging materials in terms of low weight, stability and sturdiness. However, they lead to serious environmental issues because they generate extensive volumes of non-biodegradable waste (Siracusa, Rocculi, Romani, & Rosa, 2008).

New biodegradable films made from biopolymers play a crucial role in lowering the environmental impact of non-biodegradable plastic waste (Soo and Sarbon, 2018). The main biopolymers employed in the development of biodegradable films are protein and polysaccharides. Polysaccharides studied thus far include chitosan (Aider, 2010; Martins, Cerqueira, & Vicente, 2012), carboxymethyl cellulose (Nazmi, Isa, & Sarbon, 2017), and starch (Soo & Sarbon, 2018; Tongdeesoontorn, Mauer, Wongruong, Sriburi, & Rachtanapun, 2011). Commonly studied proteins for developing biodegradable films include soy protein (Guerrero & De la Caba, 2010), milk protein, such as casein and whey protein (Kokoszka, Debeaufort, Lenart, & Voilley, 2010), and gelatin (Hanani & Ross, & Kerry, 2014a; Hanani, O'Mahony, Roos, Oliveira, & Kerry, 2014b; Nor, Nazmi, & Sarbon, 2017).

Gelatin has been studied extensively for its ability to form film, good functional properties, and usefulness as an outer barrier to protect food from drying, exposure to oxygen and light (Bakry, Isa, & Sarbon, 2017). Films composed of gelatin possess good mechanical properties but have been found to be moisture sensitive and exhibit poor barrier properties against water vapour (Gómez-Guillén et al., 2009). This causes negative feedback when applied to high moisture food products because films may dissolve, swell or disintegrate upon contact with water. Therefore, current trends in designing biodegradable materials for food packaging look for optimize film properties by studying the effect of plasticizers.

Plasticizers are molecules of low volatility which are added to biopolymer materials to allow the modification of the functional properties

* Corresponding author.
E-mail address: norizah@umt.edu.my (N.M. Sarbon).

https://doi.org/10.1016/j.fbio.2018.06.006
Received 17 September 2017; Received in revised form 6 June 2018; Accepted 12 June 2018

of films by increasing their extensibility, dispensability, flexibility, elasticity, rigidity and mechanical properties (Hanani et al., 2014a, 2014b). Polyols have been reported to be particularly efficient in plasticizing hydrophilic polymers (Ghasemlou, Khodaiyan, & Oromiehie, 2011; Tihminlioglu, Atik, & Özen, 2010). For this reason, many recent researchers have focused on the usage of polyols such as glycerol (Li et al., 2011; Muscat, Adhikari, Adhikari, & Chaudhary, 2012), sorbitol (Bakry et al., 2017; Mikus et al., 2014), mannitol (Liew, Tan, & Peh, 2014; Mikus et al., 2014) and xylitol (Tong, Xiao, & Lim, 2013). There are also plasticizers from monosaccharides such as glucose, mannose, fructose, and sucrose (Piermaria et al., 2011; Qiao, Tang, & Sun, 2011). Films have also been produced using fatty acids as plasticizers (Jiménez, Fabra, Talens, & Chiralt, 2012; Limpisophon, Tanaka, & Osako, 2010).

However, plasticizers may also serve as mechanical antiplasticizers at low concentrations (2.5%), resulting stiffer film blends (Chang, Karim, & Seow, 2006). Mechanical antiplasticization has significant effects on physical properties and textural characteristics of edible film (Cheng et al., 2002). Most water compatible diluents (polyols, monosaccharides, disaccharides, oligosaccharides) has been reported to have plasticizing rather than antiplasticizing effects on both physical and mechanical properties of biopolymer-based film (Chang et al., 2006). Therefore, water compatibility diluents are mostly used in film development. However, different types of polymer may respond differently to the low concentration of plasticizers. The absence of antiplasticizing effects may be due to the limited range of diluent concentrations examined, since those plasticizers are usually applied at optimum levels to enhance film flexibility and workability.

Studies have shown that the addition of plasticizers in various percentages have improved the mechanical properties of gelatin films (Nor et al., 2017). For instance, the different concentration of glycerol used in film formulation has different effects on the tensile strength values of gelatin films such as glycerol concentration at 10% (108.28 ± 6.38 MPa) (Fakhoury et al., 2012); 20% (1.75 MPa); 25% (1.67 ± 0.12) (Al-Hassan & Norziah, 2012) and 30% (2.91 ± 0.43 MPa) (Soo & Sarbon, 2018). The blend of different plasticizers in film formulation could also improve the functional properties of the gelatin film that was produced (Ghasemlou et al., 2011). For example, the used of the glycerol with sorbitol (Al-Hassan & Norziah, 2012) (1.28–25.03 MPa) and glycerol with Polyethylene glycol (PEG) (33–80 MPa) (Cao, Yang, & Fu, 2009) showed the improvement on tensile strength with concentration increased. As a small hydrophilic molecule, glycerol can be inserted between protein chains, hence acting as a plasticizer. The distance between the protein chains increases and direct interactions are reduced as glycerol become interspaced in a protein network (Guo et al., 2012).

In addition, a study conducted by Rezaei and Motamedzadegan (2015) revealed that the addition of sorbitol to bovine gelatin films with incorporation of clay nanoparticles reduced the Young's modulus (YM) value of the films. There were no significant differences observed when sorbitol concentration increased from 10% up to 25%. However, when sorbitol concentration was raised from 25% to 30%, young's modulus of bovine gelatin films has improved. As for the sago starch/ fish gelatin film with incorporation of 25% sorbitol, showed that the YM value were ranged between 0.91 and 1.71 Pa with varied ratio of blended sago starch and fish gelatin film. The results however, showed higher value when compared to addition of 25% of glycerol into sago starch/fish gelatin film with YM value between 0.12 and 0.20 Pa for different ration of blended sago starch and fish gelatin film (Al-Hassan & Norziah, 2012). To conclude, the study by Rezaei and Motamedzadegan (2015) was in corresponding with findings by Al-Hassan and Norziah (2012) which stated that glycerol has better plasticizing effect than sorbitol with respect to mechanical properties in bovine and fish gelatin film. The results was supported with study by Mali, Sakanaka, Yamashita, and Grossmann (2005) which also mentioned that the lower stress and Young's Modulus values were obtained in films plasticized with glycerol, indicating that glycerol exerted a more effective plasticization.

Falguera, Quintero, Jiménez, Muñoz, and Ibarz (2011) found that food scientists worldwide had developed great interest in recent years in the effects of various types of plasticizers for improving biodegradable film properties. Previous studies on the microstructure, interactions, crystalline structure of gelatin films had been conducted to improve film properties, which include water vapour permeability (Nunez-Flores et al., 2013; Nor et al., 2017), gas permeability (Ninan, Joseph, & Abubacker, 2010), tensile strength (Nor et al., 2017), Fourier transform infrared spectroscopy (Nunez-Flores et al., 2013), X-ray diffraction (Tongdeesoontorn et al., 2011) and light transmission (Ahmad, Benjakul, Prodpran, & Agustini, 2012). Due to growing interest in the study of gelatin biodegradable films, this paper aims to review recent research into the functional properties of gelatin biodegradable film and how they are affected by plasticizers.

2. Biodegradable film packaging

The primary biopolymers used to develop biodegradable films include proteins, lipids, and polysaccharides. Biomaterials or biopolymers are polymers that are bio-based and biodegradable in nature. Polymers were usually produced from renewable feedstock, biomass in general, and had considerable ecological benefits, like decreased carbon dioxide emissions (Imre & Pukánszky, 2013). Biomaterials derived from food ingredients, such as proteins, lipids and polysaccharides, were edible and had a high potential to replace petrochemical-based plastics as edible film packaging (Hanani et al., 2014a, 2014b).

The materials used play an important role in determining the quality of film produced. Biomaterials that could be used for film making included polysaccharides, proteins, lipids and polyesters, or combinations of these materials (Ghanbarzadeh & Almasi, 2011). Biomaterials could stand alone (i.e. single polymers) as films or could be blended with other polymers to improve the characteristics of the film. Single biodegradable films that have been examined include chitosan (Leceta, Guerrero, Ibarburu, Dueñas, & De la Caba, 2013), gelatin (Nor et al., 2017; Nur Hazirah, Isa, & Sarbon, 2016), and starch (Müller, Laurindo, & Yamashita, 2009). The most commonly blended biomaterials include gelatin/ chitosan (Abruzzo et al., 2012), starch/ gelatin (Soo & Sarbon, 2018), gelatin/ carboxymethyl cellulose (Nazmi et al., 2017), pectin/gelatin (Farris et al., 2011), and SPI/ gelatin (Guerrero, Hanani, Kerry, & De la Caba, 2011).

Among all biomaterials, protein is the material commonly studied because of its good functional properties as a food packaging material. Proteins confers a broad range of functional properties, especially a high intermolecular binding potential, due to its unique structure (based on 20 different monomers) (Vieira, Da Silva, Santos, & Beppu, 2011). Protein-based biodegradable films have high potential in forming numerous linkages and could form bonds at different positions (Kokoszka et al., 2010; Vieira et al., 2011). Protein-based films exhibit poor water resistance and lower mechanical strength than that of synthetic films. However, proteins provide films with greater mechanical and barrier properties as compared to polysaccharides (Guilbert, Cuq and Gontard, 1997). Protein-based films possessed better barrier properties for carbon dioxide and oxygen, as well as improved mechanical properties, when compared with polysaccharide films.

Various types of proteins have been used for the development of edible/biodegradable films, such as soya protein (Guerrero et al., 2011), corn zein (Cho, Lee, & Rhee, 2010), sodium caseinate (Fabra, Talens, & Chiralt, 2010), pea proteins (Kowalczyk & Baraniak, 2011), sunflower protein (Salgado, Ortiz, Petruccelli, & Mauri, 2010) and gelatins (Nor et al., 2017; Nur Hazirah et al., 2016). Among protein films, gelatin-based films likely to have a huge potential for commercial application as green packaging films due to their unique characteristics (Mikkonen et al., 2012). Compared to other materials that had high melting temperatures, gelatin was thermo-reversible and had ability to