FOOD PACKAGING AND SHELF LIFE 1 (2014) 170–178

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: http://www.elsevier.com/locate/fpsl

Diffusion of carotenoids from mono and bilayer polyethylene active packaging into soybean oil

Citlali Colín-Chávez, Herlinda Soto-Valdez *, Elizabeth Peralta

Centro de Investigación en Alimentación y Desarrollo, A.C. CTAOV, Carret. a la Victoria km. 0.6, Apdo. Postal 1735, Hermosillo, Sonora 83304, Mexico

ARTICLE INFO

Article history:
Received 9 January 2014
Accepted 14 January 2014
Available online 14 February 2014

Keywords:
Carotenoids
Antioxidants
Active packaging
Polyethylene
Diffusion
Activation energy

ABSTRACT

The present work aims to study the kinetics of the diffusion of carotenoids from a monolayer low-density polyethylene film (MM) and a coextruded high-density polyethylene/low-density polyethylene film (BM) to soybean oil. Also, the effect of temperature on the diffusion rate was evaluated. The diffusion coefficients for total carotenoids were 8.8–28.66 and (3.99–21.30) $\times 10^{-11}$ cm^2/s for the MM films at 30 and 40 °C. Meanwhile, the diffusion coefficients were <1.56, 1.56 $\pm$ 0.43, 6.75 $\pm$ 0.45, and (17.70 $\pm$ 6.66) $\times 10^{-11}$ cm^2/s at 10, 25, 30, and 40 °C, respectively, for the BM films. The addition of a high-density polyethylene layer as a coextruded film delayed the release of carotenoids and the time to reach the equilibrium. The effect of temperature on the diffusion coefficients followed an Arrhenius-type model with activation energy of 118.97 kJ/mol for the BM films. This release can be more effective for fatty food stored/commercialized in regions with a temperate to hot climate.

1. Introduction

Antioxidant active packaging may be able to prevent or delay the oxidation of food components susceptible to this type of reaction. This approach requires the deliberate incorporation of antioxidants into packaging materials and the further migration of antioxidants to the food. Therefore, the food shelf life is extended and safety is improved while quality is maintained (Colín-Chávez, Soto-Valdez, Peralta, Lizardi-Mendoza, & Balandrán-Quintana, 2013a; Robertson, 2012; Suppakul, Miltz, Sonneveld, & Bigger, 2003). There are different interactions between food and packaging. Migration is one of them and consists of transferring components from the packaging material to the food. It is a molecular diffusion process explained by the Fick' second law, and there are two basic parameters derived from this process: the diffusion coefficient (*D*), which expresses how fast the antioxidant is diffused and the partition coefficient ($K_{p,s}$), which expresses how much migrant is diffused when diffusion reaches the equilibrium between the packaging and food (Colín-Chávez et al., 2013a; Castle, 2007).

Polyethylene is the most widely used plastic in food packaging and has the simplest structure based on –CH_2– units (Robertson, 2012). Polyethylene can be branched or linear, like low-density polyethylene (LDPE) and high-density polyethylene (HDPE), respectively. Both materials have different crystallinities. LDPE has a structure with many branched chains that interrupt the regular arrangement of the atoms, generating a low degree of crystallinity (50–70%) and introducing amorphous zones. HDPE has a linear and ordered structure with few short-chain branches, favoring a parallel chain configuration, a higher degree of crystallinity (90%) and fewer amorphous zones (Colín-Chávez et al., 2013a). Thus, multilayer structures are made from different

* Corresponding author. Tel.: +52 662 289 2400x222; fax: +52 662 280 0057.
E-mail address: hsoto@ciad.mx (H. Soto-Valdez).

2214-2894/$ – see front matter
http://dx.doi.org/10.1016/j.fpsl.2014.01.008

polyethylene types to optimize the properties of food packaging (Butler & Morris, 2009).

Vegetable oils are composed principally of triglycerides of fatty acids with some degree of unsaturation. They can be used for cooking, as ingredient of foods and directly as food. The unsaturated fatty acids found in oils are susceptible to oxidation that causes quality deterioration. Thus, oils must be adequately protected and one alternative is by the use of antioxidant active packaging (Piergiovanni & Limbo, 2010). Many reports on antioxidant active packaging have focused on the migration of synthetic antioxidants, such as butylated hydroxytoluene (BHT) and butylated hydroxyanisole (BHA) (Huang & Weng, 1998; Soto-Cantú et al., 2008; Tawfik & Huyghebaert, 1999; Torres-Arreola, Soto-Valdez, Peralta, Cárdenas-López, & Ezquerra-Brauer, 2007; Wessling, Nielsen, & Giacin, 2001). Nowadays, natural antioxidants are replacing synthetic antioxidants in the form of extracts like packaging combined with flavonoid-cocoa extract (Calatayud et al., 2013), astaxanthin-marigold extract (Colín-Chávez, Soto-Valdez, Peralta, Lizardi-Mendoza, & Balandrán-Quintana, 2013b), rosemary extract (Camo, Beltrán, & Roncalés, 2008), oregano extract (Licciardello, Muratore, Mercea, Tosa, & Nerin, 2012), citronella essential oil (Licciardello et al., 2012), and barley husks (Pereira de Abreu, Paseiro Losada, Maroto, & Cruz, 2010).

Carotenoids are a family of pigmented (red, yellow and orange) lipophilic compounds that have antioxidant activity and are widely distributed in green plants, flowers, fruits, and vegetables. They are divided into two classes, xanthophylls (contain oxygen) and carotens (hydrocarbons). There are several natural sources of carotenoids like Azteca marigold (*Tagetes erecta*) flowers, which main carotenoids are the xanthophylls lutein and zeaxanthin. Transformation of these compounds has been performed to obtain commercial extracts rich in astaxanthin which is also a xanthophyll (Rodriguez, 1999; Rodriguez et al., 2001; Schloemer et al., 2002). The antioxidant activity of carotenoids consists in its ability to quench singlet oxygen, quench photosensitizers and scavenge free radicals. These properties can be useful for food preservation in the food industry. The family of carotenoids is composed by more than 700 members, but no more than 10 have been applied commercially in the food and feed industry as pigments (Christaki, Bonos, Giannenas, & Florou-Paneri, 2013). In the food packaging field there are few publications about the use of carotenoids as polymer stabilizers or active compounds. For example, two papers reported the incorporation of the carotens β-carotene and lycopene in poly(lactic acid) and polypropylene, respectively, as polymer stabilizers (Cerruti, Malinconico, Rychly, Matisova-Rychla, & Carfagna, 2009; López-Rubio & Lagaron, 2010). Other two papers reported our previous work which describes the addition of an Azteca marigold extract in mono and bilayer polyethylene films as a source of the xanthophyll astaxanthin and its diffusion into 95% ethanol (Colín-Chávez et al., 2013b). The first report showed a positive effect of the films on the soybean oil oxidative stability. This effect is the result of the migration of all the carotenoids, among them astaxanthin, present in the films. Diffusion of astaxanthin was reported in our second paper (Colín-Chávez et al., 2013a) and the present work reports the diffusion of total carotenoids from the mono and bilayer polyethylene films into soybean oil. Chromatographic methods are used for the quantification of migrants in most of the migration studies. Spectrophotometry has also been reported for diffusion of catechin and quercetin from ethylene-vinyl alcohol copolymer films into 95% ethanol (Chen, Lee, Zhu, & Yam, 2012) and α-tocopherol and quercetin from ethylene vinyl alcohol, ethylene vinyl acetate, LDPE, and polypropylene films into 95% ethanol (López-de-Dicastillo, Alonso, Catalá, Gavara, & Hernández-Muñoz, 2010). This method was implemented in the present work in order to quantify the diffusion of all carotenoids diffusing from the active packaging into soybean oil.

The present work aims to study the kinetics of diffusion of total carotenoids from monolayer and coextruded films, including polyethylenes with different densities, to soybean oil at different temperatures and to evaluate the effect of temperature on the diffusion.

2. Materials and methods

2.1. Materials

The marigold extract was provided by Industrias Vepinsa S.A. de C.V. (Los Mochis, Sin, Mexico). The astaxanthin (>97%) HPLC standard was obtained from Alexis® Biochemicals (New York, NY, USA). BHT (99%) was purchased from TCI America (Portland, OR, USA). Deodorized soybean oil was obtained from Industrializadora Oleofinos S.A. de C.V. (Zapopan, Jal, Mexico). All solvents to quantify total carotenoids were HPLC grade (J.T. Baker, Toluca, Edomex, Mexico).

2.2. Film fabrication

Two films were fabricated and details were reported in previous works (Colín-Chávez et al., 2013a, 2013b). A monolayer film of LDPE added with 2.90% of marigold extract (MM film) and a bilayer film of HDPE/LDPE added with 3.59% of marigold extract in the LDPE layer (BM films). Two more films with the same components but without addition of marigold extract were fabricated and used as controls. Thickness was determined with a micrometer model DTT, E.J. Cady & Co. (Illinois, USA). In the case of the bilayer films, a microscope (Olympus BX51, Center Valley, PA, USA) was used to measure the thickness of each layer.

2.3. Quantification of total carotenoids in the marigold extract and films

A spectrophotometric method for total carotenoids quantification was established using astaxanthin as standard. This is the main carotenoid in the marigold extract used in the current research (Colín-Chávez et al., 2013b). Standard solutions of astaxanthin in acetone at concentrations between 0.50 and 6.0 μg/mL were used to produce a calibration curve. Total carotenoids were quantified by measuring absorbance at 474 nm with a spectrophotometer Cary 50 Bio (Varian, Victoria, Australia). Acetone was used as a blank and a linear plot ($R^2 = 0.99$) was obtained. The limit of quantification (LOQ) of the method was established when a standard solution of astaxanthin gave an absorbance <0.0047 which corresponded to 0.033 μg/mL of carotenoids.