



# Investigation of antioxidant activity and release kinetics of curcumin from tara gum/ polyvinyl alcohol active film



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## ABSTRACT

A curcumin incorporated film was developed from tara gum and polyvinyl alcohol. The mechanical properties, water-vapor permeability, total phenolic content and antioxidant properties of the resultant films were analyzed. The release of curcumin from films in 50% ethanol simulants was also studied. The average film thickness ranged from 0.078 mm to 0.091 mm after curcumin addition and the elongation at break of the film increased from 6.07% to 29.88% with a 1% content of curcumin. Although the water-vapor permeability decreased slightly, the antioxidant properties of the film were enhanced. The DPPH scavenging capacity was increased from 1.81% to 35.16% as the curcumin content increased to 5%. The release studies indicated that the release rate and curcumin diffusion coefficient increased with increasing curcumin content, and a higher temperature accelerated curcumin migration. The benefits of curcumin-incorporated films may yield new designs for the protection of fatty foods.

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## 1. Introduction

Biobased films are considered a promising alternative to synthetic films because of their biodegradable and safe features. The most frequently used materials for biobased films are polysaccharides, proteins and lipids. These materials could be used individually or in combination to produce films (Nesterenko, Alric, Silvestre, & Durrieu, 2013). The trend for active packaging arises because of consumers' increasing demand for fresh, safe and convenient food (Boateng, Burgos-Amador, Okeke, & Pawar, 2015). Active packaging often incorporates functional ingredients, such as nutritional supplements, antimicrobial, antioxidants and ultraviolet protection agents to prolong shelf life and to enhance the sensory properties of food and beverages (Koontz et al., 2010).

The incorporation of antioxidants in packaging film is often used because oxidation is a major problem that affects food quality. Antioxidants that are loaded on films could migrate from the packaging to the food, which prevents direct contact with food and reduces the amount of chemical additives used in food (Ciannamea, Stefani, & Ruseckaite, 2016). Because of consumer health concerns, current research has focused on natural active compounds rather than synthetic compounds. These natural compounds include

those in green-tea extract (Siripatrawan & Harte, 2010), quercetin (Souza et al., 2015), grapeseed extract (Moradi et al., 2012),  $\alpha$ -tocopherol (Melo, Arrivetti, Alencar, & Skibsted, 2016), pomegranate-rind extract (Qin et al., 2015), curcumin (Etxabide, Coma, Guerrero, Gardrat, & de la Caba, 2017) and essential oils (Moradi, Tajik, Rohani, & Mahmoudian, 2016). Among these natural active compounds, curcumin, which is a polyphenolic compound that is isolated from turmeric powder, has been used extensively in medicine because of its desirable antioxidant, antitumor and anti-inflammatory activities (Sonkaew, Sane, & Suppakul, 2012). Clinical trials have shown that curcumin is safe, even when consumed at a daily dosage of 12 g for 3 months (Goel, Kunnumakkara, & Aggarwal, 2008). Therefore, the incorporation of curcumin into biocompatible polymers to produce polymer composites has gained significant attention.

Sun et al. fabricated a poly(glycerol–sebacate–curcumin) polymer for possible use in healing brain gliomas (Sun et al., 2013); Baipai et al. prepared an antioxidant film made from curcumin, cellulose microcrystals and chitosan for wound healing (Bajpai, Chand, & Ahuja, 2015a). Bitencourt et al. prepared gelatin-based films that were activated with curcuma ethanol extract for food packaging (Bitencourt, Fávoro-Trindade, Sobral, & Carvalho, 2014). Suwantong et al. used electrospinning to obtain a cellulose-acetate fiber that contains curcumin as a herbal substance (Suwantong, Opanasopit, Ruktanonchai, & Supaphol, 2007). A major problem

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is that curcumin is insoluble in water, which limits its application, and therefore, many methods have been used to overcome this barrier to expand its application. Such methods involve using ionic liquid (Luo, Varaprasad, Reddy, Rajulu, & Zhang, 2012) or sodium phosphate buffer (Yang, Wu, Li, Zhou, & Wang, 2013) or dimethylsulfoxide (Bajpai, Chand, Ahuja, & Roy, 2015b) to dissolve curcumin or turn curcumin into curcumin nanoparticles or liposomal curcumin (Raghavendra, Jayaramudu, Varaprasad, Ramesh, & Raju, 2013). However, these methods are expensive and complex. A simple method is required to dissolve curcumin in alkaline solution and to introduce these onto films to expand its application. According to our previous study, the blend film of tara gum and polyvinyl alcohol possessed good mechanical and barrier properties, but no activity (Ma, Du, Yang, & Wang, 2017). Considering the desirable antioxidant properties of curcumin, we introduced it onto the blend film to obtain an active film.

The objectives of this study were to investigate the effects of curcumin on the properties of TG/PVA films, including the mechanical, barrier and antioxidant properties. The release of curcumin from the blend films into fatty food simulants was studied. Fatty food simulants (50% ethanol, v/v) was recommended by the US Food and Drug Administration. The effects of temperature on curcumin migration were also investigated and the migration process was described by Fickian diffusion.

## 2. Materials and experiments

### 2.1. Materials

Tara gum (TG) was purchased from Dymatic Fine Chemical Co., Ltd. (Guangzhou, China) with a molecular weight of around 1000 kDa. PVA (average Mw = 84,000–89,000 g/mol; degree of polymerization (DP) = 1700–1800; 88% alcoholysis) was obtained from Sinopec Shanghai Petrochemical Co., Ltd. (Shanghai, China). Curcumin (AR) and Folin-Ciocalteu reagent were purchased from Guangfu Fine Chemical Institute (Tianjin, China) and Hero Chemical Co. Ltd. (Shanghai, China), respectively. 2, 2-Diphenyl-1-picrylhydrazyl (DPPH) was acquired from Ruitaibio Co. Ltd. (Beijing, China).

### 2.2. Preparation of antioxidant films

TG, PVA and their blend solutions were prepared according to our previous study (Ma et al., 2017). TG was dissolved in distilled water for 3 h at 45 °C to obtain a 1 wt% solution, and a PVA solution (5 wt%) was prepared by dissolving PVA at 80 °C for 2 h under magnetic stirring. Curcumin was dissolved in NaOH solution (1 mol/L) at ambient temperature. PVA was added to the TG solution at a mass ratio of TG/PVA = 7/3. Different amounts of curcumin (0%, 1%, 1.5%, 2%, 2.5%, 3% and 5%) and 1.5 mL glycerol (30%) were added to the TG/PVA blend solutions. The mixed solution was stirred at 80 °C for 30 min. The final film-forming solution that contained curcumin was cast onto a plexiglass plate (26 cm × 26 cm × 4 cm) after bubble removal and dried at 50 °C in a vacuum-drying oven. The dried films were peeled off and conditioned at 43% relative humidity at ambient temperature.

### 2.3. Characterization of films

#### 2.3.1. Color and transparency measurement

Color parameters were measured by a portable colorimeter (Xrite2600d, MI, 101, USA). A white standard color plate for the instrument calibration was used as a background for color measurements of the films. The CIE color values recorded were: L = lightness (0 = black, 100 = white); a (−a = greenness, +a =

redness) and b (−b = blueness, +b = yellowness). The total color difference ( $\Delta E$ ) was calculated as follows:

$$\Delta E = \left[ (L - L^*)^2 + (a - a^*)^2 + (b - b^*)^2 \right]^{0.5} \quad (1)$$

where  $L^*$ ,  $a^*$  and  $b^*$  are color values of standard color plate.

#### 2.3.2. Mechanical properties

Tensile strength (TS) and elongation at break (EAB) of antioxidant films were determined by using an auto tensile tester (XLW-PC, PARAM, Jinan, China) equipment with a 500 N load cell at a strain rate of 300 mm/min. Five rectangular strips (15 mm × 80 mm) were prepared from each film to determine their mechanical properties. Average thickness of each film strip was measured by an ID-C112XBS micrometer (Mitutoyo Corp., Tokyo, Japan).

#### 2.3.3. Water vapor permeability (WVP)

The WVP of films was determined via the modified gravimetric method (Shojaee-Aliabadi et al., 2014). The films were initially cut into discs sealed in permeation cups containing anhydrous calcium chloride (0% RH) with a 1 cm air gap from the desiccant. These cups were conditioned inside desiccators maintained at 75% RH with a saturated solution of sodium chloride. Weight changes were measured over a seven-day period at 12 h intervals. The water vapor transmission rates (WVTR) can be calculated from the weight changes ( $\Delta m$ ) of exposed film (area  $A$ ) at a specified time interval ( $\Delta t$ ), as described by Eq. (2) (Bedane, Eić, Farmahini-Farahani, & Xiao, 2015):

$$WVTR = \Delta m / (\Delta t \cdot A) \quad (2)$$

and the water vapor permeability (WVP) can be calculated from the value of WVTR as follows:

$$WVP = WVTR \cdot X / \Delta p \quad (3)$$

where  $X$  is the average film thickness (mm), and  $\Delta p$  is the difference in water vapor pressure between the two sides of the film (Pa).

### 2.4. Total phenolic assay

Conditioned sample (0.2 g) was placed in conical flasks that contained 10 mL ethanol/water at 25 °C. The film solution (1 mL) was transferred to a 10-mL flask and diluted with water to a final volume of 10 mL. The total phenolic content of the film was determined by the Folin–Ciocalteu method (Ciannamea et al., 2016). Briefly, 1 mL of diluted film solution was mixed with 4 mL of Folin–Ciocalteu reagent (Hero Chemical Company, Shanghai, China). After 3 min, 5 mL of sodium carbonate solution [7.5% (w/v)] was added, and the mixture was maintained at room temperature for 2 h. Solution absorbance was measured at 765 nm using a spectrophotometer (UV-2600, Shimadzu, Kyoto, Japan). The total phenolic content (TPC) was expressed as mg gallic acid equivalents per gram of dried mass of the film sample:

$$TPC = C \times V \times n / M \quad (4)$$

where  $C$  is the concentration of gallic acid from the calibration curve (mg/L),  $V$  is the volume of diluted film extract used for measurement (L),  $n$  is the total dilution factor and  $M$  is the mass of the dried films (g).

### 2.5. Antioxidant activity

The antioxidant activities of the blend films were evaluated

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