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Gelatin-coated paper with antimicrobial and antioxidant effect for beef packaging

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ABSTRACT

In the present work, raw paper sheets were coated with biopolymeric solutions based on gelatin crosslinked with transglutaminase enzyme, containing glycerol as plasticizer, and citric acid as antimicrobial and antioxidant agent, for packaging beef. Thickness, grammage and bulk density analysis have shown the effectiveness in forming the coating film, which was confirmed by morphological analysis. The biopolymeric coating did not show improvement in mechanical properties, but the water vapor permeability was significantly reduced while maintaining the original optical properties of the paper sheets. The beef packaged in the active papers shown a lower microbial population at the end of the four days of storage, and had greater stability to lipid oxidation during the entire period. The active papers sheets held the desirable red color and significantly lower pH values compared with beef packaged in the uncoated control paper. The coated papers sheets still acted positively to prevent the loss of moisture from the beef to the environment.

1. Introduction

In recent years, the food distribution chain has shown great changes which has stimulated industries and researchers to concentrate their efforts in developing modern packaging systems. The greatest challenges faced lie in the prolongation of the durability of the packaged product while maintaining its desirable consumption characteristics, as well as its nutritional quality and microbiological safety. These packaging systems still need to meet the urgent demands of sustainability in order to minimize the environmental impacts caused by the sharp use of non-renewable source materials.

To replace the petroleum-based packaging with more sustainable alternatives, natural materials and their derivatives have drawn significant attention in recent years (Sirviö, Liimatainen, Niinimäki, & Hormi, 2013). Paper and cellulosic-based derivatives are green packaging materials due to their renewability, recycling and biodegradability. However, for application in foods and for safety reasons, the paper must be coated with fossil materials, besides the fact that these materials improve the mechanical and barrier properties (Zhang, Xiao, & Qian, 2014), which is required for use as packaging. Biopolymers originating from renewable sources have the potential to replace current synthetic paper coatings, as they offer favorable environmental benefits of recycling and reuse (Khwaldia, Basta, Aloui, & El-Saied, 2014).

Several biopolymers such as wheat gluten (Gastaldi, Chalier, Guillemin, & Gontard, 2007), alginate and soy protein (Arfa, Preziosi-Belloy, Chalier, & Gontard, 2007; Rhim, Lee, & Hong, 2006), modified starch (Arfa et al., 2007) and chitosan (Kjellgren, Gällstedt, Engström, & Järnström, 2006) have been used as raw material to prepare coating solutions for paper for food packaging purposes. Among the biomaterials used to prepare edible films and coatings, gelatin has been extensively used due to its wide range of functional characteristics such as excellent film-forming properties, high binding capability with water and emulsifying properties. In addition, its advantages include low cost, high availability, biocompatibility and biodegradability capabilities (Wang, Liu, Ye, Wang, & Li, 2015). Due to their hydrophilic character, films and coatings based on gelatin have high water vapor permeability. As a consequence, mechanical strength of gelatin films decreases when in contact with high moisture content surfaces, which restricts

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their application as packaging (Weng & Zheng, 2015). However, it has been further reported that the mechanical properties and permeability of the gelatin films can be improved by the crosslinking of the transglutaminase enzyme action, which is considered as a safe and effective cross-linking agent (Gómez-Guillén, Gimenez, Lopez-Caballero, & Montero, 2011). Transglutaminase has been applied for crosslinking of several proteins and their complexes, such as fish gelatin, soy protein isolated, whey protein isolate-gelatin and casein-gelatin, improving the functional properties of the materials (Wang, Liu, Ye, Wang, & Li, 2015). The development of composite materials, paper-biopolymer, can combine their different properties improving its performance. Besides, after use, these materials can be disposed off in an economically and ecologically acceptable way (Aloui, Khwaldia, Slama, & Hamdi, 2011; Johnston, Moraes, & Borrmann, 2005).

Meat is a highly perishable product due to its chemical composition susceptible to microorganisms attack (Lawrie & Ledward, 2006). The microbial growth accelerates changes in aroma, color and food texture, resulting in reduced shelf life and increasing the risk of foodborne diseases. Moreover, the oxidative processes lead to degradation of lipids, proteins, pigments, and constitutes major mechanisms of quality deterioration in meat and meat products (Liu, Dai, Zhu, & Li, 2010). To satisfy the demand of extending the shelf life of foods and reducing the deterioration, packaging techniques containing active nontoxic preservatives have gained prominence in recent years (Lorenzo, Batlle, & Gómez, 2014). This technology is based on the process of incorporation of certain components in packaging systems that release or absorb substances from/or to the packaged food or the surrounding environment, while preserving the quality of the food for a longer period than the conventional package (Barbosa-Pereira, Angulo, Lagarón, Paseiro-Losada, & Cruz, 2014; Realini & Marcos, 2014).

The use of packaging with incorporated antimicrobial agents is presented as a promising approach for controlling the growth of microorganisms present in the packaged food. The antimicrobial packaging has the function of increasing the lag phase and reducing the growth phase of microorganisms in order to prolong the shelf life and maintain the quality and food safety (Kerry, 2014). To preserve susceptible food from oxidation, the use of antioxidant active packaging has also been the subject of numerous studies (Hong, Lee, & Son, 2005). The antioxidants are incorporated in plastic films, paper or sachets where they will be released to protect food from oxidative degradation, inhibiting oxidation reactions by reacting with free radicals and peroxides and thus extending its shelf life (Brody, Strupinsky, & Kline, 2001; Lee, An, Lee, Park, & Lee, 2004; Vermeiren, Devlieghere, Beest, Kruijf, & Debevere, 1999). Recent studies have shown the associated antioxidant and antimicrobial potential of the citric acid in the preservation of meat products (González-Fandos, Herrera, & Maya, 2009; Vargas Júnior et al., 2015; Schwan, Dias, & Luiza, 2011).

In this context, the purpose of this study is to develop a paper sheet coated with biopolymeric film based on gelatin crosslinked with transglutaminase enzyme, containing citric acid as an active agent. The coated paper will provide antimicrobial and antioxidant effects for application as active packaging in fresh beef. This material can be used as paper packaging for rapid commercialization of chilled beef in supermarkets, for example, replacing the conventional plastic films as well as prolong the shelf-life of the beef. Therefore, it is expected that this active package will be able to meet the current demands of the consumer market and environmental requirements. Such requirements aim to prolong the food shelf life maintaining the desirable characteristics for consumption and microbiological safety, minimizing the use of raw materials from non-renewable sources such as plastic, reducing consequently the impact caused by excessive waste disposal of traditional packaging to the environment.

2. Materials and methods

2.1. Materials

Pork gelatin type A (220 bloom, mesh 30 and moisture content <13%) was kindly provided by The Gelnex Gelatin Specialists (Ita, SC, Brazil). Transglutaminase enzyme (TGase) ACTIVA® GS (obtained from *Streptomyces mobaraense*) was provided from Ajinomoto of Brazil Industry and Food Trade Ltda. (São Paulo, SP, Brazil). Analytical grade glycerol (99.5%) and anhydrous citric acid (99.5%) were purchased from Sigma-Aldrich. The sheet papers used in the experiments were obtained from a raw paper purchased in local market of trademark Royal Pack® on rolls of 30 cm × 7.5 m. The paper is composed of 100% virgin long fiber, with 0.035 mm thick, with non-stick surface.

2.2. Preparation of biopolymeric solutions

Biopolymeric solutions were prepared with 2% gelatin/distilled water (w/vol) using a magnetic stirrer at 25 °C, under which the gelatin was hydrated for 60 min. Gelatin solubilization was completed by increasing heating to approximately 60 °C for 30 min. Later, transglutaminase enzyme (0.1 g/10 g of gelatin), glycerol (2 g/10 g of gelatin) and citric acid were added in two different concentrations (0.5% and 1.0% relative to the solution mass), stirring constantly for 15 min. The concentration of citric acid used was based on previous study developed by Vargas Júnior et al. (2015). The enzyme was inactivated by heating the biopolymeric solutions to 85 °C for 15 min and then cooling to 60 °C for subsequent use as a coating for sheet papers. Vargas Júnior et al. (2015) also examined the thermal stability of citric acid, reporting a decomposition temperature of 231.54 °C, confirming that heating at 85 °C does not affect its stability.

2.3. Coating method

The biopolymeric solutions were applied to the raw paper sheets (rolls of 30 cm × 7.5 m) by spraying using a low pressure compressed air pistol, at an estimated rate of about 1 ml of biopolymeric solution/100 cm^2 of paper. After applying the solution, the paper sheets were dried at room temperature and controlled relative humidity (54 − 60%) for 24 h and subsequently conditioned in bobbins wrapped by plastic film. Table 1 shows the formulations used in the coatings of manufactured papers: control paper (CP), paper 1 (P1) paper 2 (P2) and paper 3 (P3).

2.4. Paper characterization

2.4.1. Surface morphology

Morphological analysis was obtained in a Jeol Scanning Electron Microscope (SEM), model JSM-6390LV, with a source of tungsten electrons and detector of secondary electrons. Samples of papers coated by each treatment were previously cut to an approximate size of 1 cm^2 and covered with a thin gold layer to be observed using 10 kV voltage.

2.4.2. Thickness, grammage and bulk density

The nominal thickness of paper (mm) was obtained by averaging five points measured in each sample (ends and middle) with the aid of an external digital micrometer Digimess® (Brazil) according to ISO 534 (International Organization for Standardization, 2011). For grammage analysis, five samples of each treatment were cut into 10 cm x 10 cm and were weighed using an analytical