

Preparation and investigation of curcumin-containing PLA composite films for food packaging application

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Abstract. Polylactic acid (PLA) is a biocompatible and biodegradable natural polymer known as a promising biopolymer that can be used for food packaging due to its good physical and physicochemical properties. This biopolymer possesses excellent thermal processability, good oxygen barrier capacity, and complete biodegradability, and it is one of the materials used as a food packaging material. PLA has a significant potential for optimizing physical properties through material modifications, including in combination with natural additives as different polyphenols. Polyphenols are a class of naturally occurring compounds found in plants, which exhibit a wide range of beneficial properties, including antioxidant, anti-inflammatory, and antimicrobial activities. In the present paper PLA-based composite films with incorporated polyphenol curcumin with different concentrations were investigated. PLA composite films were prepared using a solution casting method. The films obtained were charged in a corona discharge. The electret properties of the charged samples were studied. The possible surface potential decay mechanisms responsible for the electret's behavior were discussed. It was established that the surface potential decay depends on the corona polarity and the type of films.

Key words: polylactic acid, curcumin, corona discharge, electret properties.

Introduction

The development of the polymer industry in the last century has led to the production of many different materials, including a vast variety of food packaging applications. With the creation of new food processing technologies, the demand for plastic packaging has increased exponentially. However, this increase in demand has also resulted in significant ecological problems, caused by the disposal of a vast variety of polymer products. The search for a solution to the environmental impact of petrol-based plastics has led the scientific community to develop a variety of biodegradable alternatives (Mangaraj et al., 2019). These types of polymers are usually produced by the utilization of biomass, created by different microorganisms or synthesized from different natural monomers, which makes them not only indepen-

dent from the oil industry, but also allows them to further decrease their environmental impact by utilizing different natural byproducts from other biobased industries (Bhargava et al., 2020).

The food packaging industry is one of the biggest users of petrol-based plastics, with millions of tons being produced annually (Siracusa et al., 2008). This high demand also results in the creation of vast amounts of waste, as most packaging products are disposed of after use and cannot be properly recycled. With the demand for food packaging rising each consecutive year, the need for novel, cheap, and widely utilized biodegradable polymer products has become more and more evident. However, any large-scale application of biodegradable food packaging is limited by several factors, such as the high cost of production, as well as different limitations in their physical,

thermal, barrier, and mechanical properties. These limitations can be mitigated through different modifications of the base biodegradable polymer product, such as the addition of different smart and active materials (Motelica et al., 2020). These additions can improve the desired properties of the biopolymers, thus increasing their applicability. Several scientific reviews (Jiang et al., 2023; Omerović et al., 2021; Nilsen-Nygaard et al., 2021) demonstrate the potential of different active packaging materials, with Ordoñez et al. (2022) demonstrating the use of different phenolic materials for the creation of active packaging materials.

The polyphenol curcumin has shown great potential amongst the different bioactive compounds (de Oliveira Filho et al., 2021), with its ability to show undesirable transformations in the packaged product through a change in color (Kumar et al., 2018). Additionally, the inclusion of curcumin has been shown to improve the antioxidant properties of the base material (Chen et al., 2018), thus further increasing their applicability in the food packaging industry.

The presented paper studies the impact of the inclusion of different concentrations of curcumin on the electret properties of composite polylactic acid films for food packaging applications.

Materials and methods

Materials

Biopolymer PLA and polyphenol curcumin were purchased from Sigma-Aldrich and were used as delivered. All other chemicals were of analytical grade.

Film creation

The biodegradable and biocompatible polymer polylactic acid was chosen for the creation of the films. A solution of the polymer in chloroform was prepared at a concentration of 2% w/v. The polyphenol curcumin was selected as the active component of the film. During the solution preparation, the surfactant Tween 20, also known as Polysorbate 20, was added at a concentration of 1.5% w/v.

The following methodology was utilized for the creation of the samples: preparation of a set volume of polymer solution in chloroform with a concentration of 2% w/v, with added Tween 20 with a concentration of 1.5% w/v. This solution was used for the creation of control films, which

would be used as a baseline for comparison with films containing the active component. Films containing the active component were created by the addition of curcumin at concentrations of 10, 15, and 20%. These solutions were mixed on a magnetic stirrer for at least 12 hours until the complete dissolution of the polymer, followed by a homogenization in a centrifuge at 12 000 rpm for 10 minutes, carried out right before the casting of the solutions in glass petri dishes with a set size. The cast films were dried at a temperature of 35°C and set humidity until the complete evaporation of the solvent. The obtained composite films were kept in a dry desiccator (RH = 20%) until further use.

Corona charging

The obtained composite films were charged in a corona discharge at room temperature. The charging in a corona discharge was carried out by means of a conventional corona triode system, consisting of a corona electrode (needle), a grounded plate electrode, and a metal grid (controlling electrode) placed between them. The samples were placed on the grounded plate electrode and were charged for 1 minute. The voltage supplied to the corona electrode was set to 5 kV, and the voltage of the grid was set to 1 kV for 1 minute. Both voltages were of positive polarity.

Scanning electron microscopy (SEM)

The morphology of the created biodegradable composite films was determined with the use of scanning electron microscopy (SEM) (Prisma E SEM, Thermo Scientific, Waltham, MA, USA). Two milligrams of each of the tested samples were attached to an aluminum holder and subsequently coated with carbon and gold, with the use of a turbomolecular pumped sputter coater system Quorum Q150T Plus (Quorum Technologies, West Sussex, UK). The resulting images were captured with the use of a back-scattered electron detector (Prisma E SEM, Thermo Scientific, Waltham, MA, USA) at an accelerating voltage of 15 kV at different magnifications.

Conductivity measurement

The surface and volume resistance of the composite samples of PLA were measured with a Keithley electrometer at different voltages. The conductivity of the films was calculated from the collected results.

Results and Discussion

Electret properties

The impact of storage time on the decrease of the surface potential for polymer composite PLA films with 0, 10, 15, and 20wt%. The % curcumin content was studied. Time dependences of the normalized surface potential of PLA and PLA with 20% curcumin were investigated during a period of 6 hours. The surface potential of the created electrets was measured every 5 minutes

during the first half hour, as the decrease is rapid during this time period. Six samples of each type of composite film were used for the determination of the surface potential by averaging the values of the normalized surface potential at each time interval. The statistical error of the averaged values (which was determined to be lower than 5%) was also calculated. Time dependences of the normalized surface potential for positively charged samples are presented in Fig. 1.

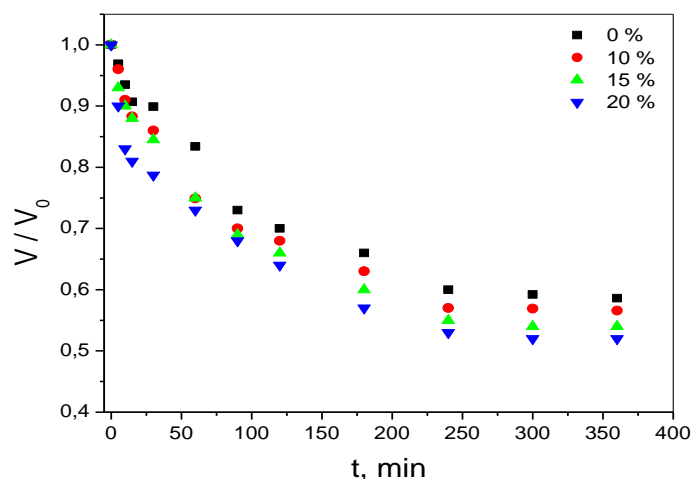


Fig. 1. Time dependences of the normalized surface potential for PLA composite films charged in a positive corona.

From the results shown in Fig. 1 can be determined that the values of the normalized surface potential were exponentially decreasing for the first 75 minutes, after which the decay rate decreased and became practically stable within the sixth hour. The collected results show the existence of a variety of surface states, localized on the sample surface, which can hold entrapped charges. The exponential decrease observed in the initial time period can be explained by the release of weakly trapped charges from shallow energy states. After this initial rapid release period, the surface potential stabilized to a set steady state value, resulting from charges entrapped within the deepest traps. Such exponential decay followed by slow linear reduction of charge of electrets was observed by researchers, such as Sessler (2001) and Viraneva et al. (2025).

It was also observed that the values of the normalized surface potential were the highest for pure PLA electrets and decreased when the polyphenol curcumin was included. The lowest values of the normalized surface potential were observed

for the PLA films with 20% curcumin. This is probably due to the structure of the films and their conductivity. The conductivity of all investigated composite films is presented in Fig. 2.

The results presented in Fig. 2 and Fig. 3 show that:

- The increase of the curcumin concentration leads to a corresponding increase in the conductivity regardless of the applied voltage.
- The lowest value of both the surface and volume conductivity was observed for PLA films without added curcumin.
- The increase in voltage results in a decrease in the conductivity in all studied PLA samples, regardless of curcumin concentration.
- The values of the surface conductivity were significantly higher than those of the volume conductivity, regardless of the applied voltage and curcumin concentration.

These results suggest that the decrease in the surface potential with time is caused entirely by surface processes.

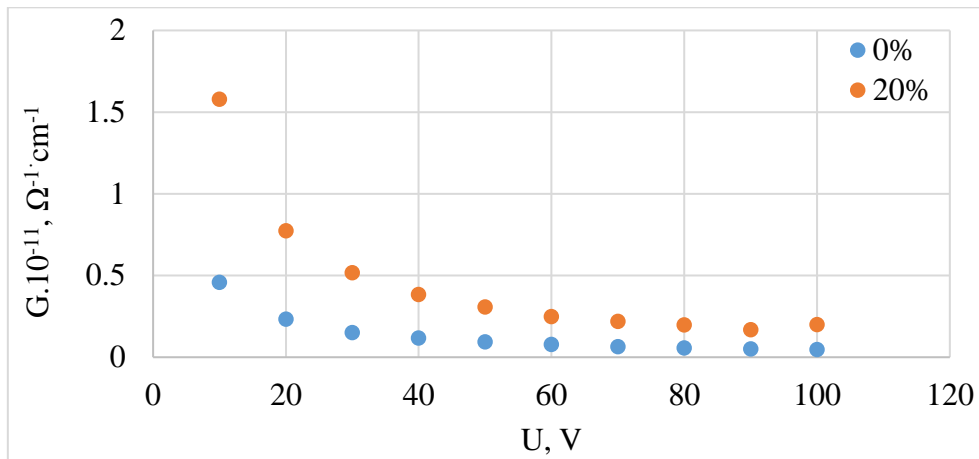


Fig. 2. Dependency of the volume conductivity on the voltage for composite PLA films.

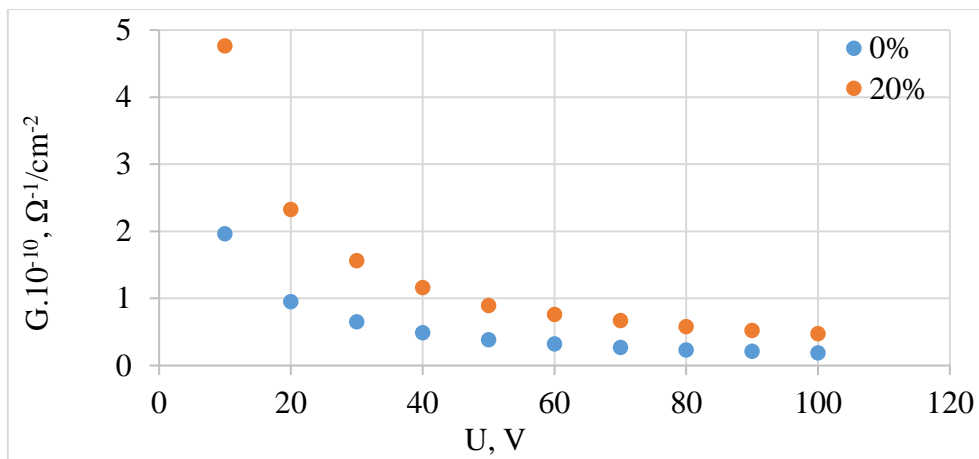


Fig. 3. Dependency of the surface conductivity on the voltage for composite PLA films.

Scanning electron microscopy (SEM)

SEM is an intuitive technique for observing the morphology of samples, which was performed

to provide some information about the physical structures of pure films of PLA and PLA films containing curcumin with a concentration of 20%.

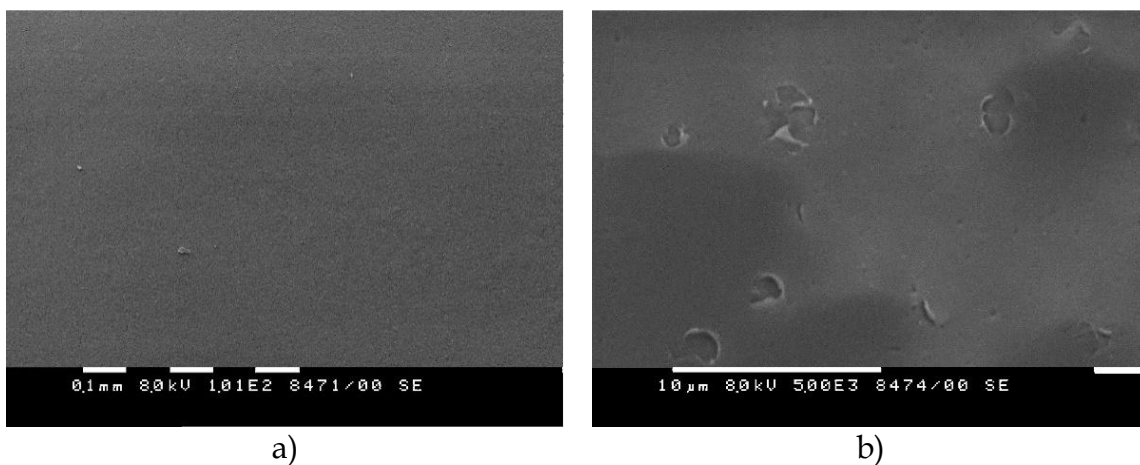


Fig. 4. SEM images of a) PLA, b) PLA with 20 % curcumin.

The images obtained for the PLA films show the smooth surface structure of the pure PLA films (Fig. 4a). The SEM images of films containing curcumin (Fig. 4b) show that the inclusion of the polyphenol in the structure of the polymer films increased their surface roughness. The incorporation of curcumin in the structure led to a significant increase in the surface roughness, which was possibly due to the higher content of hydroxyl groups in the used polyphenol, leading to the easier interaction with PLA matrix. The increase in roughness in PLA films containing curcumin has also been observed in other papers (Reddy et al., 2019; Roy et al., 2020), whose results show that the increase of curcumin concentration impacts not only the surface but also other properties of the films. These results suggest a maximum concentration of curcumin, which can be included within PLA structures without the deterioration of their desired properties.

Conclusions

Overall, the inclusion of curcumin in films of PLA was shown to have a significant impact on their surface and conductivity properties. The results also demonstrate that the relatively high amount of incorporated polyphenol (20 %) does not significantly impact the desired electret properties of the PLA film, thus allowing for the creation of biodegradable films containing larger amounts of active ingredients.

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