THE PRODUCTION OF EDIBLE AND ANTIMICROBIAL ACTIVE PACKAGING MATERIAL WITH THYME OIL AND DETERMINATION OF PRINTABILITY PROPERTIES

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Abstract: *Increasing demands of consumers in food packaging enabled the development of active packaging practices. Antimicrobial and antifungal coatings or films are an active packaging application. In such packaging materials, natural or synthetic antimicrobial material is added to the film or coating to help the packaging protect food. The use of biofilms produced from biopolymers in food packaging is more suitable than polymers with many damages such as PET, PE. Film production of many biopolymers such as cellulose derivatives, chitosan, PVA, starch PLA can be realized. Thymus essential oils and extracts with antimicrobial and antioxidant properties are widely used in pharmaceutical, cosmetic, herbal tea, flavoring agents and perfume industry, also for flavorings and preservation of several food. In this study, it is aimed to produce antibacterial printable edible which is used in active packaging, using starch and thyme essential oil. For this purpose, starch edible films containing different rates (0.1, 2.5, 5, 10%) of thyme oil were produced by spin coating method. The transparency of prepared five different films were determined by UV spectroscopy. Antimicrobial properties of the obtained films were determined against S. aureus and E. coli. Edible films were printed with screen printing and printability parameters such as color, gloss, contact angle and surface tension were examined. Consequently, starch films loaded with thyme oil were successfully produced. It is concluded that edible films are colorless, transparent and have good printability. It was determined that the amount of thyme essential oil increased in the edible had a strong* *inhibitory feature against S. aureus and E. coli. With these features; produced films are considered to be suitable for active packaging applications.*

Keywords: active packaging, antibacterial, edible film, printability, thyme oil

1 INTRODUCTION

The long supply chain from the production area of food products to the consumer brings along some food safety problems. The deterioration of food by the effects of physical, chemical and biological factors threatens human health and safety. Food spoilage is appeared with bacterial growth, bad odor, loss of taste and color deterioration as a result of a number of factors, and the general quality and safety of the products are damaged and shelf life is shortened (Ahmed et al., 2017; Ozcan and Tutak, 2020). Bringing different features to packaging, which plays a critical role in the product supply chain, increases efficiency in transportation and protection tasks, while also providing access to fresh, delicious, safe, healthy and quality food products with a longer shelf life (McMillin, 2017).

Smart packaging is defined as a material that adds new functions compared to traditional packaging to improve the basic functions of the packaging, not only improving the basic functions, but also responding to the stimuli around the packaged product (Ozcan, 2020).

Edible films and coatings are also methods used in active packaging. Recently, biodegradable materials such as polysaccharides, lipids and proteins obtained from natural materials can add important functionality to packaging with their antimicrobial properties (Bifani et al., 2007; Vargas et al., 2008). Films and coatings that do not pose a problem in contact with food, edible, biodegradable, biocompatible, environmentally friendly, have the potential to reduce waste, provide barrier properties, and have protective, appearance and aesthetic properties will be very functional in food packaging (Shankar and Rhim, 2016; Kapetanakou et al., 2014). It is possible to find many studies showing that edible films and coatings obtained from some plant extracts or oils found in nature have antioxidant and antimicrobial activities and extend the shelf life of the food in the package (Gutiérrez, 2017; Sirocchi et al., 2017; Yang et al., 2016; u Nisa et al., 2015; Maryam Adilah and Nur Hanani, 2016). In recent years, researchers have been working on using plants such as thyme oil and basil, which are natural preservatives, as antimicrobial and antioxidant. Thyme, which is the general name of thymus, thymbra, origanum, coridothymus, satureja, genera from Lamiaceae family and known for its distinctive smell, is a plant suitable for active packaging applications with its high protection, deodorization, antimicrobial and antioxidant properties (Bakkali et al., 2008; Sánchez-González et al., 2011; Siripatrawan, 2016; Xie et al., 2008; Yuan et al.,2016).

2 MATERIAL AND METHODS

2. 1 Materials

Corn starch was provided by Sigma Aldrich (Darmstadt, Germany). Glycerol and Tween 80 were procured from MERCK (Darmstadt, Germany). Thyme oil was obtained from ARPAŞ ARİFOĞLU (Istanbul, Turkey). S. aureus was obtained from microbiology laboratory of Ankara University Dairy Technology Department, and E. coli from Ankara University Food Engineering Department. Bacteria stock cultures were transported to the laboratory in cold storage conditions.

2. 2 Methods

Starch-based biofilms were prepared according to the methods described by Ghasemlou et. al., 2013. In the preparation of the film, first of all, 10 g starch was added to 150 mL distilled water and gelatinized by stirring at 1000 rpm for approximately one hour at 90 °C. 2 g of glycerol, 2 mL of Tween and thyme oil in different proportions depending on the formulation were added to the mixture at the same temperature. The compositions of the biofilms are given in Table 1. It was dispersed at 20000 rpm for 8 minutes so that the mixture was completely homogeneous. The homogeneous mixture was formed into a film on the glass surface in the spin coating device. The resulting films were dried at room temperature for 2 days.

UV–Vis spectra of the biofilms were recorded in the range of 400–800 nm to evaluate the transparency of the films. The antimicrobial activity of biofilms was defined by inhibition zone method (disc diffusion method). Each bacteria culture was activated by inoculation in tryptic soy broth, at 37 °C for 24 hours. The inoculum (0.1 mL) was spread to the surface of Mueller-Hinton (MH) agar petri dishes by spread plate technique; then, 6-mm-diameter films cut from prepared films were placed onto petri dishes. Control samples were prepared under the same conditions. Disc film containing petri dishes and control samples were incubated at 37 °C for 24 hours. After incubation, petri dishes were checked for bacterial growth; inhibition zones around the disc films were evaluated qualitatively and quantitatively. Quantitative evaluation was performed according to the inhibition zone diameter. The zones around the disc films were evaluated as an indicator of inhibition of bacterial growth. The bio film that produced a large inhibition zone was considered to show a high antimicrobial activity. Contact angle and total surface energies of the biofilms were determined with Pocket Goniometer PGX+ in line with ASTM D5946.

Table 1: Formulations of the biofilms

Sample	*Thyme oil (%)*	*Starch (g)*	*Glycerol (g)*	*Tween (mL)*
F0	*0*	*10*	*2*	*2*
F1	*1*	*10*	*2*	*2*
F2	*2.5*	*10*	*2*	*2*
F3	*5*	*10*	*2*	*2*
F4	*10*	*10*	*2*	*2*

Solid printing was performed on obtained five different biofilms with a weaving density of 77 tpc, 75° scraping angle and 75° shore hardness by using an ARUS semi-automatic screen printing machine. The color characteristic of the prints was measured by X-Rite exact spectrophotometer spectral range 400 -700 nm, with D50 light, 2° observer angle, with polarize filter, 0/45-degree). and gloss values were measured by BYK Gardner gloss meter at 60° according to TAPPI T480 OM-15. Color differences (ΔE) were calculated according to the given below CIELab (2000) technique:

$$\Delta E_{00} = \sqrt{\left(\frac{\Delta L'}{k_L S_L}\right)^2 + \left(\frac{\Delta C'}{k_C S_C}\right)^2 + \left(\frac{\Delta H'}{k_H S_H}\right)^2 + R_T \frac{\Delta C'}{k_C S_C} \frac{\Delta H'}{k_H S_H}} \quad (1)$$

3 RESULTS

Biofilms containing different amounts of thyme oil have been prepared successfully. The transparency of the films obtained was examined by UV-Vis spectrophotometer. The UV-Vis spectrum of the films, which are very transparent even by eye, is given in Figure 1. As the films got thicker, the folding and transparency of the films decreased. For this reason, all biofilms were produced with a thickness of approximately 20 µm and their transparency was compared. When the results obtained were examined, it was determined that all biofilms had high transparency, but as the amount of thyme oil in them increased, there was a small decrease in transparency. This result is compatible with the literature (Wang et al., 2021).

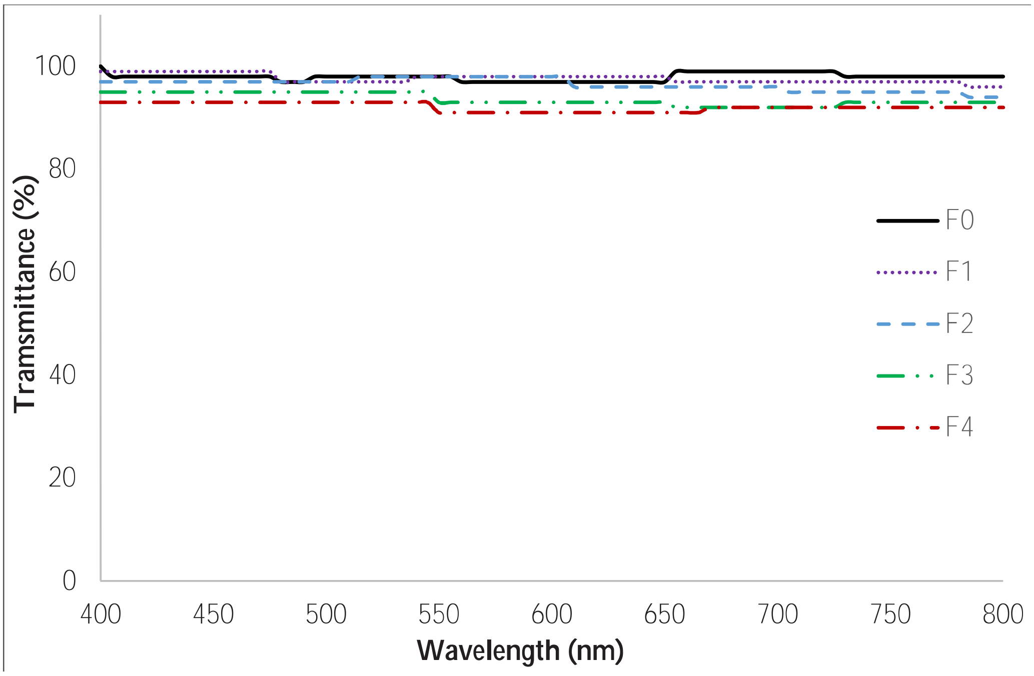

Figure 1: Transmittance of the antibacterail biofilms.

The antimicrobial activity of obtained biofilms was tested against both gram-positive (S. aureus) and gram-negative (E. coli) bacteria. Diameter of inhibition zone diameters of different ratio of thyme oil ingredient biofilms are shown in Table 2. It was seen that E. coli and S. aureus grown homogeneously in all regions of petri dishes in control samples. Inhibition zone against E. coli and S. aureus was not observed in films non-consisting thyme oil (F0). Because starch is natural carbohydrates, it is a food of microorganisms. On the other hand, thyme oil content biofilms showed inhibition zone against E. coli and S. aureus. This result showed that the thyme oil was antimicrobial against both gram-positive and gram-negative bacteria. When all the results were examined, it was observed that as the amount of thyme oil increased, the inhibition zone increased and the antimicrobial property increased. It was also concluded that thyme oil was slightly more effective on S. Aureus than E. Coli (Semeniuc et al., 2017).

Table 2: Antimicrobial activity of tyhme oil content biofilms against E. coli and S. aureus (inhibition zone diameter in centimeter)

Sample	***E. coli***	***S. aureus***
F0	-	-
F1	*1.2*	*1.8*
F2	*3.1*	*3.4*
F3	*3.9*	*4.4*
F4	*4.2*	*5.5*

The use of vegetable oils in the biofilms is a good way of create hydrophobic (Pelletier and Gandini, 2006). The most important parameter in determining the printing method and ink type to be used for printing is the surface energy and contact angle of the substrate. For this purpose, these properties of biofilms were examined and the results are given in Table 3.

Table 3: Contact Angle anda Surface Energies of tyhme oil content biofilms

Sample	Contact Angle (°)	Surface Energy (mJ/m²)
F0	72.3	38.9
F1	80.6	35.7
F2	82.4	34.2
F3	84.0	34.7
F4	85.9	33.3

When thyme oil was added to base formulation, the contact angle increased. This is due to the hydrophobic nature of the oil. It is seen that the contact angle increases when the amount of oil in the biofilm formulation is increased. This improves the offset and screen printing printability of the biofilms with less ink. Because the oleophilic groups in the offset and screen printing ink structure will interact more with the peppermint oil on the biofilm surface and a good ink surface relation will be provided and the printing process will be easier (Kandirmaz and Zelzele, 2020).

In addition, prints were made with the screen printing technique on the produced films and the color and gloss characters of the produced prints were measured and the results are given in Table 4. When Table 4 was examined, it was determined that the color shifted slightly towards yellow due to the oil's own color. When F0 and other prints containing thyme oil were compared, it was determined that the color difference value, ΔE, was below 3 and the difference between them could not be noticed with the human eye. When the gloss values were examined, it was determined that the added oil slightly reduced the gloss of the biofilm, but this reduction was within the standard deviation range. It was possible to print well on the obtained films with oil-based screen printing ink without any problems.

Table 4: Color and gloss properties of printed biofilms

Sample	L*	a*	b*	ΔE_{00}	Gloss
F0	47.5	74.3	-3.9	Ref.	15.6
F1	47.4	74.2	-2.6	0.43	15.5
F2	47.3	73.1	-2.2	1.46	15.5
F3	47.2	72.9	-1.8	1.15	15.4
F4	46.9	72.3	-1.1	1.35	15.2

4 CONCLUSIONS

As a result, thyme oil containing starch biofilms have been successfully produced. The obtained biofilms have transparent. Biofilms containing thyme oil have inhibitory properties against both gram positive and negative bacteria. The highest activity was observed against gram positive bacteria. When the printed colors of the films were examined, it was determined that the color difference was below 3, that is, there is no color difference that can be seen by the eye. As the amount of oil in the biofilm increased, the contact angle increased, the surface has become more sensitive to oil-based ink. The gloss values are not change with adding thyme oil. The oil based ink printability of the biofilm is increased by adding thyme oil. Screen prints were made onto biofilms without any problem.

5 REFERENCES

Ahmed, I., Lin, H., Zou, L., Brody, A., Li, Z., Qazi, I., Pavase, T. and Lv, L. (2017). A comprehensive review on the application of active packaging technologies to muscle foods. Food Control, 82, pp.163-178.

Bakkali, F., Averbeck, S., Averbeck, D. and Idaomar, M. (2008). Biological effects of essential oils–a review. Food and chemical toxicology, 46(2), pp.446-475.

Bifani, V., Ramírez, C., Ihl, M., Rubilar, M., García, A. and Zaritzky, N. (2007). Effects of murta (Ugni molinae Turcz) extract on gas and water vapor permeability of carboxymethylcellulose-based edible films. LWT - Food Science and Technology, 40(8), pp.1473-1481.

Ghasemlou, M., Aliheidari, N., Fahmi, R., Shojaee-Aliabadi, S., Keshavarz, B., Cran, M. J., Khaksar, R. (2013). Physical, mechanical and barrier properties of corn starch films incorporated with plant essential oils. Carbohydrate Polymers, 98(1), pp.1117–1126.

Gutiérrez, T. (2017). Surface and nutraceutical properties of edible films made from starchy sources with and without added blackberry pulp. Carbohydrate Polymers, 165, pp.169-179.

Kandirmaz, E. A., Zelzele, O. B. (2020). The production of ecofriendly biofilm with natural oil for food packaging. Proceedings - The Tenth International Symposium GRID 2020.

Kapetanakou, A. E., Manios, S. G., Skandamis, P. N. (2014). Application of edible films and coatings on food. Novel food preservation and microbial assessment techniques, 52, 237-273.

Maryam Adilah, Z. and Nur Hanani, Z. (2016). Active packaging of fish gelatin films with Morinda citrifolia oil. Food Bioscience, 16, pp.66-71.

McMillin, K. (2017). Advancements in meat packaging. Meat Science, 132, pp.153-162.

Ozcan, A. (2020). New approaches in smart packaging technologies. Proceedings - The Tenth International Symposium GRID 2020.

Ozcan, A. and Tutak, D. (2020). The effect of zeolite on inkjet coated paper surface properties and deinking. Nordic Pulp & Paper Research Journal, 35(3), pp.432-439.

Pelletier, H., Gandini, A. (2006). Preparation of acrylated and urethanated triacylglycerols. European Journal of Lipid Science and Technology, 108(5), pp.411-420.

Sánchez-González, L., Vargas, M., González-Martínez, C., Chiralt, A. and Chafer, M. (2011). Use of essential oils in bioactive edible coatings: a review. Food Engineering Reviews, 3(1), pp.1-16.

Semeniuc, C. A., Pop, C. R., Rotar, A. M. (2017). Antibacterial activity and interactions of plant essential oil combinations against Gram-positive and Gram-negative bacteria. Journal of Food and Drug Analysis, 25(2), pp.403-408.

Shankar, S., Rhim, J. W. (2016). Polymer nanocomposites for food packaging applications. In A. Dasari, & J. Njuguna (Eds.), Functional and physical properties of polymer nanocomposites (pp. 29e55). John Wiley and Sons, Ltd.

Siripatrawan, U. (2016). Active food packaging from chitosan incorporated with plant polyphenols. In Novel approaches of nanotechnology in food (pp. 465-507). Academic Press.

Sirocchi, V., Devlieghere, F., Peelman, N., Sagratini, G., Maggi, F., Vittori, S. and Ragaert, P. (2017). Effect of Rosmarinus officinalis L. essential oil combined with different packaging conditions to extend the shelf life of refrigerated beef meat. Food Chemistry, 221, pp.1069-1076.

u Nisa, I., Ashwar, B., Shah, A., Gani, A., Gani, A. and Masoodi, F. (2015). Development of potato starch based active packaging films loaded with antioxidants and its effect on shelf life of beef. Journal of Food Science and Technology, 52(11), pp.7245-7253.

Xie, Y., Zhao, X., Li, Y., Zhao, Q., Zhou, X. and Yuan, Q. (2008). CTAB-assisted synthesis of mesoporous F–N-codoped TiO2 powders with high visible-light-driven catalytic activity and adsorption capacity. Journal of Solid State Chemistry, 181(8), pp.1936-1942.

Vargas, M., Pastor, C., Chiralt, A., McClements, D. and González-Martínez, C. (2008). Recent Advances in Edible Coatings for Fresh and Minimally Processed Fruits. Critical Reviews in Food Science and Nutrition, 48(6), pp.496-511.

Wang, B., Sui, J., Yu, B., Yuan, C., Guo, L., Abd El-Aty, A. M., Cui, B. (2021). Physicochemical properties and antibacterial activity of corn starch-based films incorporated with Zanthoxylum bungeanum essential oil. Carbohydrate Polymers, 254, pp.117314.

Yang, H., Lee, J., Won, M. and Song, K. (2016). Antioxidant activities of distiller dried grains with solubles as protein films containing tea extracts and their application in the packaging of pork meat. Food Chemistry, 196, pp.174-179.

Yuan, G., Chen, X. and Li, D. (2016). Chitosan films and coatings containing essential oils: The antioxidant and antimicrobial activity, and application in food systems. Food Research International, 89, pp.117-128.