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Green tea-boron nitride incorporated pumpkin pectin-alginate food packaging film preparation and characterization

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Petroleum-based packaging poses a major environmental threat to life on Earth. This type of packaging does not degrade in nature for many years and causes environmental pollution as it is difficult/costly to recycle. Packaging is used in food storage to preserve food quality, ensure food safety and extend shelf life. With the increasing need for food in the world, the demand for packaging materials is also increasing. It is therefore difficult to strike a balance between the use and recycling of packaging. Efforts should be made to design food packaging according to the type of food, to produce it in an environmentally friendly way and to be applicable in food packaging systems. Within the scope of this study, food packaging films were produced to utilize agricultural residues left by farmers after harvest and integrate them into the economy. In this study, pectin was obtained from pumpkins and mixed with alginate to make packaging films. Green tea extract (GTE) was incorporated as an enhance water permeability and strength, while boron nitride nanoparticles (nB) were selected to improve the physical attributes of the films. The use of green tea decreased the moisture content and swelling of the films while enhancing their mechanical strength. The incorporation of nB into the films enhanced their opacity and mechanical strength, while simultaneously decreasing their moisture content, swelling percentage, and water vapor permeability rate (WVPR). Future research suggests that these films may serve as food packaging materials.

Keywords: Pumpkin pectin films; boron nitride; green tea extract; food package

INTRODUCTION

Packaging is very important for the food industry. The fact that it protects the quality of the food, complies with the safety standard and extends the shelf life of the food makes it indispensable. As the demand for food increases, so does the dependence on packaging. In order to protect nature, some innovative approaches and changes (smart-active packaging) should be taken in the packaging sector [1].

Due to their biodegradability and abundant availability in various natural resources, biopolymers have attracted significant interest in the packaging industries, making them effective alternatives. Biopolymers derived from natural sources are useful for food packaging applications due to the environmentally friendly and biodegradable properties of natural materials with antimicrobial activity such as polysaccharides, proteins and lipids [2]. However, most materials alone have low thermal, chemical and mechanical resistance. Therefore, packaging properties need to be improved by blending different materials. Especially in recent years, with the developments in the field of zero waste and sustainable environment, the production of biopolymers from wastes, which replace polymeric materials, has come to the forefront both economically and environmentally [3].

Pumpkin belongs to the family *Cucurbitaceaceae* which consists of 800 species and 130 genera. It is widely cultivated in Asia and the Americas: China, India, Russia, Ukraine and the USA. In the world, it is recorded as one of the most produced agricultural products with 27.4 million tons of production. This constitutes approximately 61% of total world production [4, 5]. Recently, there has been an increase in the production of pumpkin in our country. When used on an industrial scale, it can be frozen or canned. When pumpkin is processed in the industry, the edible part of the fruit usually needs to be separated from the skin (2.6-16%), seeds (3.1-4.4%) and by-products (25%) [6-8]. During the pumpkin processing, the peels rich in pectin content are discarded. Hydrochloric acid, sulfuric acid or nitric acid are generally preferred when extracting pectin from waste [9-11].

Boron nitride is a stable material with physicochemical properties similar to those of graphene in hexagonal shape. It is a well-known nanomaterial with antibacterial properties due to its strong mechanical strength and high surface area. It was reported that the use of boron nitride led to

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an increase in the antibacterial activity of the film and that it was not harmful to human cells due to its low cytotoxicity [12]. Behera *et al.* [13], prepared chitosan films with boron nitride nanobiocomposite and it was observed that the nB additive decreased the water solubility and moisture absorption of the film. Green tea is a significant material for packaging owing to its antioxidant capabilities, optical characteristics, antibacterial effects, and pH sensitivity. GTE augments the interaction between macromolecular polymers and increases the physical and chemical properties of the matrix [15]. The research indicates that it can enhance barrier performance in active packaging films and significantly decrease the water vapor transmission rate [16].

EXPERIMENTAL

Pumpkin pectin extraction

This method used by Güzel and Akpınar [17] in their study in 2019 was modified. Pumpkin peels were cut and dried in an oven at 40 ºC for two days. They were ground with an electronic grinder and sieved. 100 mL of citric acid solution with a pH of about 1 was prepared to extract pectin. 10 g of pumpkin powder was weighed and added to the citric acid solution. It was stirred at 80 ºC for 60 minutes and filtered. 100 mL of 96% ethanol was added to the remaining filtrate and left in the refrigerator for 12 h. The pectin was centrifuged at 3000 rpm for 15 min; the supernatant was discarded and the pectin remaining at the bottom was washed for neutralization. The washing was performed twice with 70% acidic ethanol (0.5% HCl) and 96% ethanol. Then it was centrifuged again, and the precipitated pectin was dried in an oven at 50 ºC. The pectin obtained was stored in a refrigerator.

Film preparation

Alginate (A), pumpkin pectin (PP), green tea extract (GTE) and nano boron nitride (nB) added films were produced by solution casting method. Packaging solutions containing 20% of alginate and 20% of pectin by mass, not exceeding 2% solid biopolymer content in total, were prepared. Some modifications were applied on the methods with reference to the studies of da Silva *et al.* [18] and Bierhlaz *et al.* [19]. A-PP solution was stirred at 50 ºC for 1 h to dissolve; nB was added at different ratios 1%, 0.5%, 0.1% by mass and GTE at 1 mL, 2 mL, 3 mL by volume was also added to the prepared film (Table 1). 1% $CaCl_2$ was added as a crosslinking agent into the solution. In the last step, glycerol was used as plasticizing agent. The films were dried in an oven set at 40 ºC for 1 day. 30 min. The removed films were allowed to dry at room temperature. The films produced were assigned as A, A-PP, APP-G1, APP-G2, APP-G3, APP-B1, APP-B2, APP-B3, accordingly (Table 1).

Table 1. Film composition

Code	A (wt.%)	PP (wt.%)	GTE (vol%)	nB (wt.%)
APP	1.6	0.4		
APPB1	1.6	0.4		0.1
APPB2	1.6	0.4		0.5
APPB3	1.6	0.4		1
APPG1	1.6	0.4	1	
APPG2	1.6	0.4	2	
APPG3	1.6	0.4	3	

Film characterization

The thickness of the films was measured with a digital micrometer (Dasqua 4310). The thickness was measured randomly from at least three points and the values were averaged. The film sample was cut in a rectangular shape (10 mm × 40 mm). It was placed in a UV-vis spectrophotometer (Shimadzu UV-1280) cell. At a wavelength of 600 nm the light transmittance, i.e. opacity, of the films was measured. Opacity values were calculated according to Abs600 absorbance per unit thickness (Abs600/Thickness). The moisture content of the films trapped in the standard medium was found as a percentage. It is a gravimetric method. The films were left at 105 ºC for 24 h to reach constant weight. Based on the difference between initial and final weight, the moisture content was found as a percentage. A swelling test was performed to determine the water resistance of the films and how much water they trap. To get rid of excess moisture, the samples were dried in an oven at 65 ºC for 12 h. Film specimens were cut into 10 × 40 mm pieces. The tensile strength of the films was determined according to ASTM Standard D882 using a Universal Mechanical Testing Machine (ANKARIN). The room temperature and relative humidity was 55% during the analysis. At least three replicates of each film sample were tested.

Materıals

Alginate was from Sigma Aldrich; citric acid, ethanol of analytical grade were purchased from Merck Chemicals, Türkiye. Boron nitride nanopowder (purity 99.7%, 790 nm) was from Nanografi, Türkiye. Fresh pumpkins were purchased from a local market in Çanakkale.

RESULTS AND DISCUSSION

There are tests necessary for determining the use of food packaging films. Morphology, mechanical strength and color properties also have effects on their use as packaging. Fig. 1 shows the SEM micrographs and mechanical strength of the prepared films. SEM analysis of the green tea-filled film is seen in Fig. 1a. Fig. 1b shows the analysis of the nB-doped film. As can be seen from the SEM analysis, the particle distribution on the film surface is homogeneous. However, there is clustering in some regions at nB loading. The film surface is non-porous and smooth. This is important for the barrier properties of the packaging.

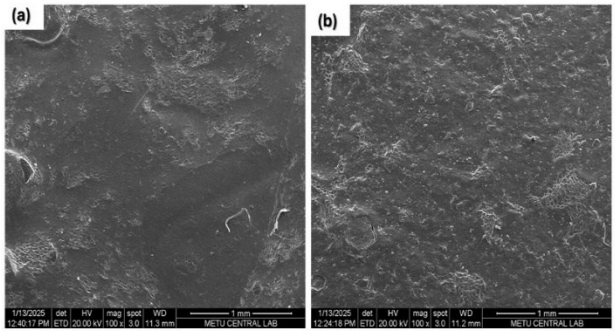

Fig. 1. SEM analysis of APP-G1 (a), APP-B1 (b) films

As seen in Fig. 2a, the opacity of APP-B films was between 37.579-76.536 Abs600/mm while APP-G was between 6.774-14.250 Abs600/mm. It was noticed that as the ratio of nB and GTE added to the films increased, the films became opaquer, and the light transmittance decreased (Fig. 2b). Visually, the appearance of the films generally reflects good properties. Therefore, consumers typically preferred transparent packaging. Films with high opacity protect food from light, air, and heat, thereby extending shelf life [20].

Table 2 shows the color properties of the films. For the APP-G films, compared to the control film, the color of the films changed towards green as the green tea extract content increased. In one study, the color analysis of starch-pectin based films with 0.25% and 0.5% green tea extract (w/v) (L*=87.82 and ΔE=12.11; L*=86.91 and ΔE= 16.16), respectively, showed similar L* and ΔE values [21]. For the APP-B film, compared to the control films, the color of the films became whiter when the B ratio added to the films increased. L* values between 87.87-92.68 indicate that the films are light colored. If ΔE > 3, the color differences of the films are easily distinguishable with the naked eye. In Table 2, all of the ΔE values of the APP-B films are above 10 and the color changes are clear. a* values are generally > 0, indicating a slight shift to red tones. b* values are also positive, indicating a shift to yellow tones (Table 2).

Table 2. Color analysis of films

Code	L*	a*	b*	ΔE
APP (Control/ Blank)	92,21	0,23	-2,22	0,43
APPB1	87,87	0,35	7,01	11,72
APPB2	89,19	-0,03	7,91	12,41
APPB3	88,31	0,76	14,23	18,78
APPG1	91,60	-0,25	2,39	13,76
APPG2	92,68	0,16	-1,92	17,95
APPG3	90,43	0,19	-2,18	17,19

The film thicknesses measured were 0.094 mm for A-PP, 0.0243-0.0350 mm for APP-B, and 0.061-0.125 mm for APP-G. The incorporation of nB significantly lowered the films' thickness, whereas the inclusion of GTE significantly enhanced the films' thickness. The tensile strength of the A-PP control film was determined to be 17.247 MPa. The tensile strength of APP-B films ranged from 16.008 to 45.152 MPa, while that of APP-G films ranged from 4.488 to 33.262 MPa (Fig. 2). Addition of nB and GTE to the films increased the strength of the films. The addition of GTE did not greatly affect the durability of the films, similar situations were observed in some studies [22]. APP-B films alone increased the mechanical strength by approximately 300% compared to A-PP films. The combination of pectin and alginate was able to improve the mechanical profiles of the films due to the compatibility between the components [23]. Moreover, it is thought that double cross-linking of the films may have enhanced mechanical strength.

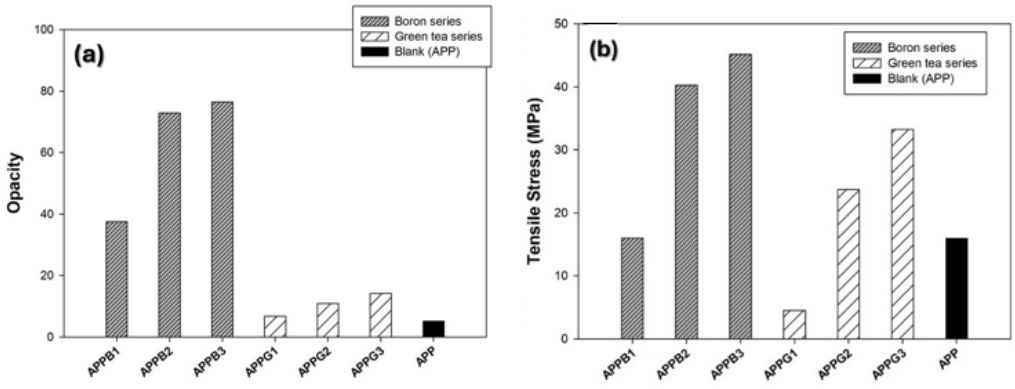

Fig. 2. Opacity (a) and appearances, (b) tensile stress APP, APP-B and APP-G films

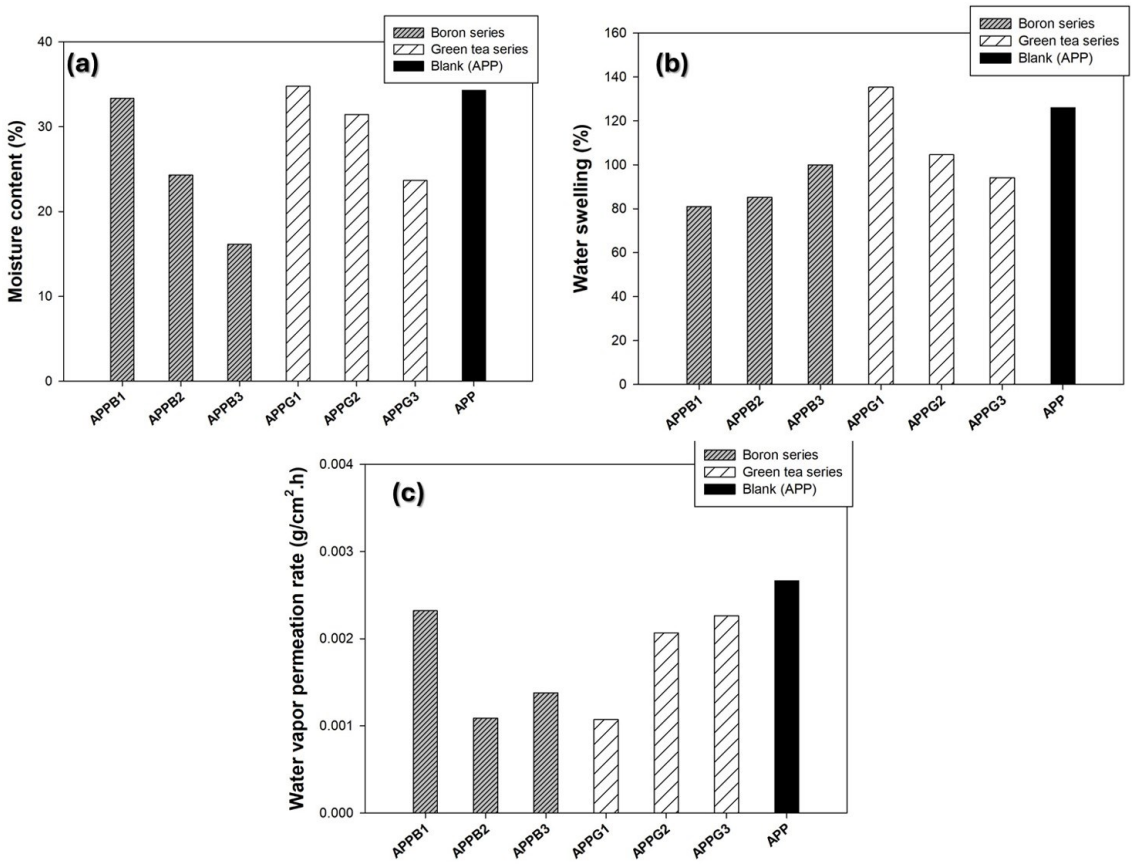

Fig. 3. Moisture content (a), water swelling (b) and water vapor permeation rate (c) of films

The barrier properties of the films are given in Fig. 3. The moisture content of APP-B films was between 16.129-33.333%, while that of APP-G films was between 23.684-34.783%. As the amount of nB increased, the moisture content significantly decreased. A similar situation was observed with the addition of green tea extract (Fig. 3a). In a previous study, Wen *et al*. [24] developed composite films by adding 2, 5 and 10% GTE to polyvinyl alcohol-based films. The moisture percentages of the films ranged between 28.7-21.7%, and the moisture percentage decreased as the addition of GTE increased. This is due to the crosslinking of polysaccharides with polyphenols [25]. Tong *et al*. [26] found 13.47% moisture content in the film produced from orange peel pectin/sodium alginate.

Swelling has the potential to directly affect the water resistance of packaging films under humid atmosphere conditions [25]. The swelling percentages ranged from 126.087% for the control group A-PP film, 85.185% to 126.667% for APP-B films, and 94.118% to 135.294% for APP-G films. The interactions that occur according to the addition of GTE and nB to A and PP containing films determine the behavior of the matrix towards water. Compared to the control group, the addition of nB decreased the swelling percentage, while the addition of GTE increased the swelling percentage of the films (Fig. 3b). The presence of phenolic compounds may explain the affinity of GTE for water molecules and the reason for the increase in swelling [22].

Since the films are intended to be used as food packaging, the water vapor permeation rate (WVPR) varies according to the moisture content and shelf life of the product to be packaged. The packaging films developed in this study are intended to be used in meat products. Therefore, films need a moderate barrier. The WVPR results show the differences in the barrier performance of the film content. The addition of nB reduced the WVPR (Fig. 3c). However, as the concentration of nB in films increases, the WVTR values slightly increase because of the hydrogen bonding effect. The best barrier property was also found in APP-G1 with the addition of GTE. This reduction may be due to the formation of hydrogen bonds between the A, PP, B and GTE matrix and oxygen, which may reduce the diffusion of water molecules in the film [27, 28].

Green tea extract which is rich in phenolic compounds, is a component with various biological activities such as antioxidant and pH sensitivity. It is seen as a potential natural additive to enhance pH sensitivity. Tea polyphenols undergo structural transformation in response to pH changes [24]. If the pH of the solution is < 7, the color of the films is

yellowish-green. If the pH of the solution was > 7, as the pH values increased, the films turned dark brown. The pH-sensitive color changes of APP-G3 film observed at pH 4, 8 and 12 are presented in Table 3.

Table 3. Color analysis at different pH ranges

pH	L*	a*	b*	ΔE	Color
4	77.55	-3.62	22.45	4.22	
8	82.97	-2.51	14.17	8.45	
12	55.01	6.41	16.85	2919	

In Table 3, L* values decreased as the pH values increased and accordingly, the films exhibited a darker color. The negative a* values indicate that the films are in a color transition towards green and the positive b* values indicate that the films are in a color transition towards yellow. According to the results obtained, APP-G3 films proved to be pH sensitive.

CONCLUSION

In this study, the potential of films composed of alginate and pectin for use as food packaging was determined by tests. In the scope of the study, it was observed that especially nB and GTE additives increased the opacity and mechanical strength of the film and decreased moisture content. In future studies, the changes in the packages in contact with real food will be examined and their shelf life will be determined. The fact that the materials selected within the scope of the study are biodegradable and all additives are food-safe, makes the potential of the produced biobased packaging important in terms of both environmental and sustainable economy.

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