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Development of active and biodegradable film of ternary-based for food application

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ABSTRACT

The effectiveness of plastic packaging in protecting food is quite appreciable, but its non-biodegradable characteristic raises concerns about environmental impacts. This has drawn attention to the development of alternative materials for food packaging from bio-based polymers. Chitosan, a polysaccharide with biodegradable, biocompatible, and non-toxic properties, is widely used in the formulation of food films. The objective of this work was to create a biodegradable and sustainable chitosan-based film whose active and intelligent action is obtained from red cabbage anthocyanins and the addition of propolis. The edible film's thickness and total polyphenol content were 61.0 ±0.1μm and 20.08 ±0.5 mgAG g^{-1}, respectively. The content of phenolic compounds and the biodegradation showed significant results (p <0.05), besides the good thermal stability to 200 °C and transparency. The proposed formulation developed an edible, biodegradable, and active (antioxidant) film with interesting heat-sealing resistance, moisture barrier and gas transfer, which contributes to increasing food shelf life.

Keywords: anthocyanin, biodegradable, biopolymer, chitosan, packaging, propolis

INTRODUCTION

Bio-sourced packaging materials are an interesting alternative to conventional polymers **[1]**, **[2]**, **[3]**. Bio-based polyesters are economically competing with conventional ones due to their biodegradability, availability, compostable nature, barrier properties and good mechanical **[4]**, **[5]**. Chitosan is a polysaccharide obtained by partial deacetylation of chitin found in insect exoskeletons, fungi, and crustacean shells **[6]**, and is the second most abundant natural polysaccharide after cellulose **[7]**. The excellent properties, biodegradability, biocompatibility and non-toxicity of chitosan allow its potential application in the food industry as packaging or coatings that improve the preservation of these products, drawing on its antimicrobial property against a wide range of bacteria, moulds and yeasts **[8]**, **[9]**, **[10]**. Chitosan can interact with the bacterial cell membrane and destabilize it, which makes chitosan films powerful vehicles for antimicrobial compounds that have difficulty reaching their cellular target due to low solubility or permeability **[11]**. Among other advantages is observed high film-forming ability **[12]**, antioxidant activity **[11]**, **[13]**, gas barrier properties **[13]**, **[14]**, and moisture barrier **[15]**, which make it a promising compound for different food products. Propolis is a complex mixture of resinous, gummy and balsamic substances of various consistency, texture and colour. It comprises 50% resin and vegetable balsam, 30% wax, 10% essential and aromatic oils, 5% pollen, and 5% other substances, including organic residues. It presents several antioxidant substances (caffeic acid, feluric acid and caffeic phenyl ester acid) besides flavonoids in its ethanolic extracts **[16]**. Anthocyanins, members of the phenolic family, are responsible for different nuclei in flowers, vegetables and fruits **[17]**. These have been used as active compounds in film formulations for food coatings due to their strong antioxidant power and antimicrobial potential **[18]**, **[19]**. In addition, anthocyanins can change their chemical structures and colours at different pH values, which is suitable for use in pH-sensing smart food packaging **[20]**, **[21]**. Different anthocyanins have been applied in the development of smart food packaging, such as anthocyanins extracted from red cabbage **[22]**, **[23]**, **[24]**. Thus, it is observed that both compounds used in this study (chitosan and anthocyanins), present important properties in forming material for active and intelligent packaging, with the main objective of increasing the shelf life of foods **[21]**.

Scientific hypothesis

Food film, based on chitosan, propolis and red cabbage, with active and biodegradable properties, can increase the shelf life of food and replace non-biodegradable plastic materials.

MATERIAL AND METHODOLOGY

Samples

The edible film is made from biopolymer materials such as chitosan, propolis and red cabbage.

Chemicals

All reagents used in the execution of this research were of analytical grade and obtained from the Food Preservation & Innovation Laboratory on Campus 2 of the University of Blumenau, Brazil. The acid acetic p.a. was bought from Dinamica Quimica Contemporânea Ltda, and the chitosan (low molecular weight) with a deacetylation degree ≥75% was purchased from Sigma-Aldrich.

Animals, Plants and Biological Materials

It was used extract from red cabbage (*Brassica oleracea*), and propolis produced in the Vale do Itajaí region, South Brazil.

Instruments

A sphere spectrophotometer (SP60, Lovibond) was used for colour testing. Scanning electron microscopy (SEM) was performed with a VEGA 3 SEM scanning electron microscope, Tescan. A HERMLE universal centrifuge, Z300K and UV-1800 spectrophotometer, SHIMADZU were used for total polyphenol content analyses. Thermogravimetric (TGA) analysis was performed in a simultaneous DTA-TG and DTG-60 analyzer, SHIMADZU. An analytical balance (Ohaus Corp. Pine Brook, N. J. USA) was used and for thickness measurements a digital micrometre (0.1 µm resolution) (Mi-tutoyo Co., Kawasaki-Shi, Japan).

Laboratory Methods

The project was carried out in the Biochemical Engineering, Chemical Analysis, and Food Preservation & Innovation laboratories at the University of Blumenau, in southern Brazil.

Film colour was determined **[21]** with a CIE-Lab colour scale applied at 3 different points on each sample to measure L* (clarity), a* (reddish-green) and b* (yellowish-green). Illuminant D65 and an observation angle of 10° were applied for the measurements. Scanning electron microscopy (SEM) of the surface and the morphology of the cross sections of the films were obtained according to **[21]**. In this procedure, the samples were coated with a thin gold layer prior to observation (Q150R ES, Quorum), and an accelerating voltage of 10 kV was applied. Images of the films' top surface and cross section were taken at magnifications of 5000x and 350x.

Total polyphenol content was determined by the Folin Ciocalteau method, with results expressed as mg gallic acid, 100 g^{-1} fresh mass (FM). Briefly, samples (10 g) were macerated and mixed with 20 mL of 96% ethanol. The solution was centrifuged at 4 °C and 120 rpm for 10 min. The supernatant was collected and submitted to the Folin Ciocalteu reagent and sodium carbonate 20% (w/v). The absorbance reading of the samples was performed at a wavelength of 756nm and gallic acid was used as a standard solution.

The biodegradability test was adapted **[25]**, using film samples (C) cut into squares (11cm^2), which were kept inserted 1cm below the soil for 14 days. The soil material used was collected from the University of Blumenau (Latitude: -26.9333 Longitude: -49.0500).

Thermogravimetric analysis (TGA) of the samples was performed according to **[21]**, and the heating rate was set to 10 °C min^{-1} in the range of 30 °C to 600 °C.

Description of the Experiment

Sample preparation: The anthocyanins were extracted from a solution containing 100 g of red cabbage, previously crushed, and 200 mL of distilled water with 1% glacial acetic acid. The solution was stored under refrigeration (4 °C) for 24 hours and then vacuum filtered with Whatman strip filter paper nº 1.

The films were produced following the casting methodology. For this, 4g of chitosan were dissolved in 3 different solutions: (1) 150 mL of a 1% acetic acid solution (control, C), (2) 150 mL of a 1% acetic acid and 1.5 mL propolis extract solution (CP) and (3) 150 mL of red cabbage extract and 1.5 mL propolis extract solution (CPC).

These solutions were kept under stirring (100 rpm) for 5 hours and then, after adding 1.2 mL of glycerol, remained under stirring for another 1 hour. The filmogenic solutions developed were poured into 30 cm x 10 cm refractories and kept at 20 °C, without light, for 7 days. After this period, the films were removed from the refractories. Thickness measurements were taken at 6 different locations of each film sample using a digital micrometre (resolution of 0.1 µm) (Mitutoyo Co., Kawasaki-Shi, Japan).

Number of samples analyzed: The number of samples analyzed was 3.

Number of repeated analyses: Three repeated analyzed were performed for each treatment factor. The total sample analyzed was 9 samples.

Number of experiment replication: The number of experiment replication was 3.

Design of the experiment: The three film samples, developed and analyzed, consisted of the following formulations: chitosan-based (C), which represents the control sample, chitosan and propolis (CP), and chitosan, propolis and red cabbage (CPC).

Statistical Analysis

Data were analyzed using Statistica software (version 7.0, StatSoft Inc., Tulsa, USA). All measurements were performed in triplicate and reported as the mean ± standard deviation (SD). Significance among mean values was determined at 5% level ($p \leq 0.05$) of significance by one-way analysis of variance (ANOVA) with following post-hoc Tukey's test.

RESULTS AND DISCUSSION

Analysis of colour parameters

Different types of anthocyanins are observed in nature, of which six are the most widespread (cyanidin, delphinidin, pelargonidin, peonidin, petunidin, and malvidin). Being polar in nature, anthocyanins are soluble in polar solvents such as methanol, ethanol and water. This is the reason why it was extracted with solutinic acetic acid. These solvents are being acidified to stabilize the anthocyanins in the flavylium cation **[17]**. Table 1 shows the results obtained for the colour parameters. Chitosan is a light and soft polymer, so the C sample had a lighter and translucent colour. On the other hand, the CP sample, because it contains propolis, has a more orange tone **[26]**, and the CPC film showed a more intense colouration due to the presence of anthocyanins from red cabbage **[27]**, whose initially red tone, over time, due to the acid medium, becomes orange **[21]**.

Table 1 Colour parameters values for chitosan (C), chitosan and propolis (CP), and chitosan, propolis and red cabbage (CPC) films according to the CIE-Lab scale.

Samples	L*	a*	b*	CIE-lab
C	93.5 ±2.1^{a}	-3.1 ±0.9^{b}	9.6 ±0.8^{b}	
CP	84.7 ±2.6^{a}	-3.5 ±0.6^{b}	40.5 ±1.4^{a}	
CPC	55.7 ±1.5^{b}	25.5 ±2.7^{a}	46.9 ±1.5^{a}	

Note: Different lowercase letters denote significant difference among the films samples ($p \leq 0.05$). L* (clarity), a* (reddish-green) and b* (yellowish-green).

Figure 1 shows the films developed (chitosan = C, CP = chitosan + propolis) and CPC = chitosan + propolis + red cabbage), demonstrating the different colouration and opacity.

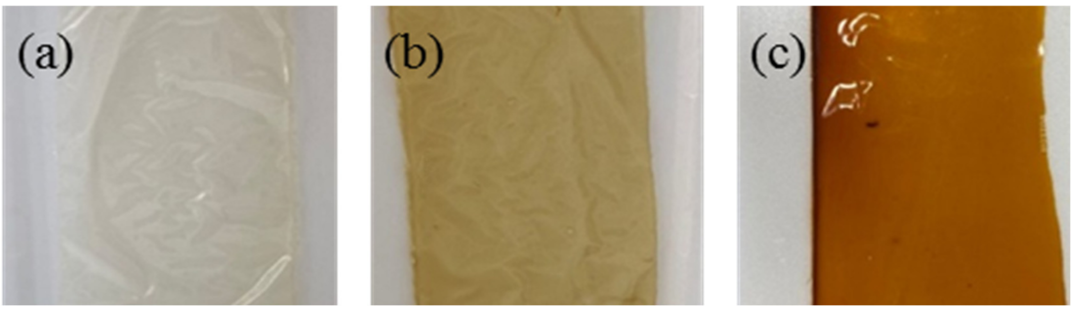

Figure 1 Films developed, (a) chitosan (C), (b) chitosan and propolis (CP), and (c) chitosan, propolis and red cabbage (CPC).

Studies have shown the variation in the colouration of materials composed of anthocyanins **[23]** when submitted to different pH environments, highlighting the potential of these materials for application in intelligent packaging **[27]**. Translucent films are desirable for food application, as it makes it possible to better visualization of the internal contents of the package. More opaque packaging, in turn, reduces the incidence of light, which is interesting for specific products by protecting them against deterioration caused by photooxidative reactions **[28]**.

Studies demonstrate the antimicrobial action of films developed and applied to food, using propolis **[29]**, antioxidant action **[30]** and intelligent function **[31]** when using extracts composed of anthocyanins, resulting in a smart material **[32]**.

Scanning electron microscope (SEM)

Morphological images of the surface and cross-section of the produced biodegradable films are shown in Figure 2.

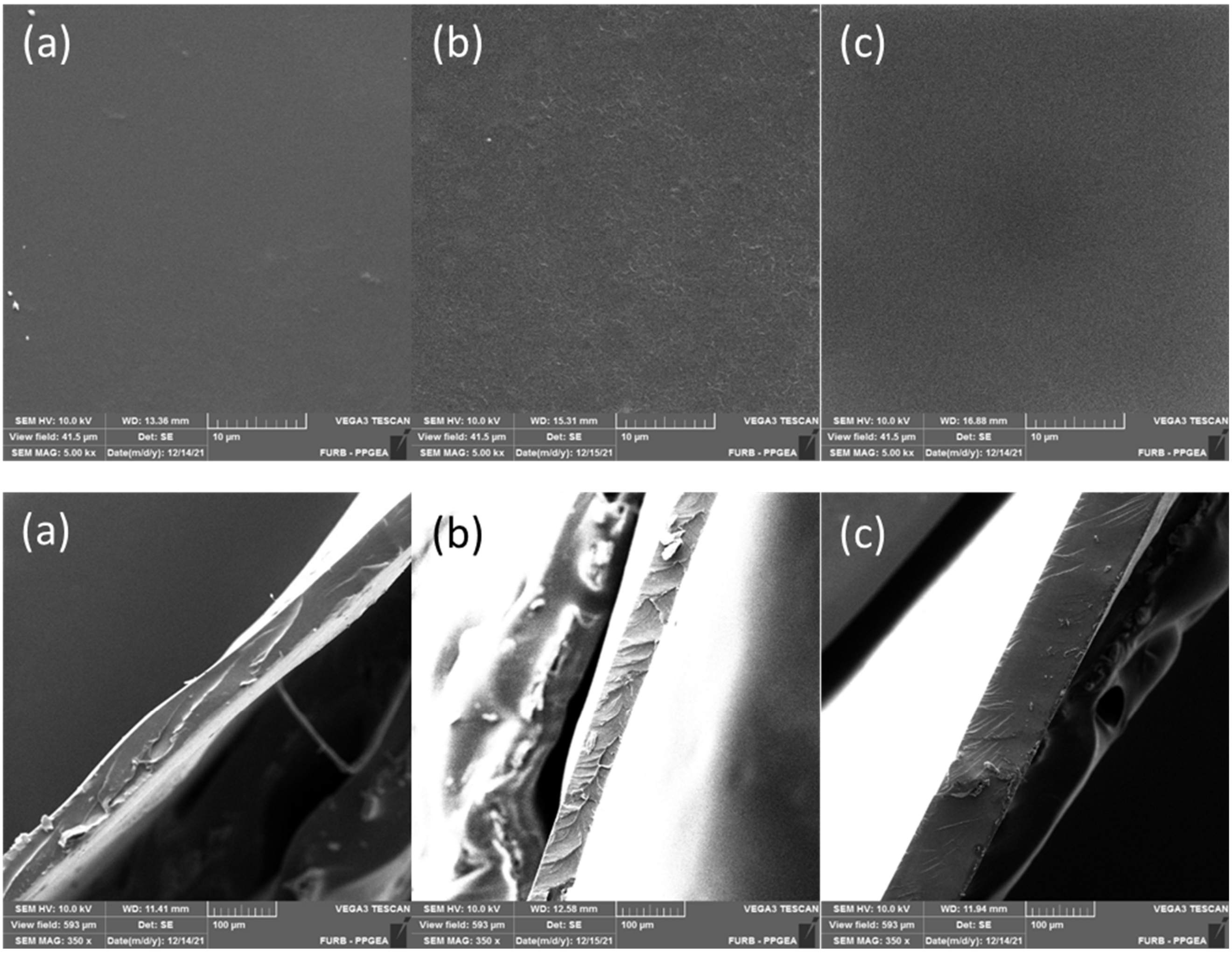

Figure 2 SEM of the surface area (upper image) and cross-section (bottom image) of chitosan film (a), chitosan and propolis (b) and chitosan, propolis and red cabbage (c).

Figure 2 shows a compact and homogeneous structure in all films, indicating good compatibility between all components of the formulations. The addition of propolis makes the structure less uniform, suggesting that there is interaction between the components of chitosan and propolis. However, purple cabbage extract results in a continuous and uniform surface **[33]**, identical to the addition of propolis **[34]**.

The film samples had a thickness of 59.0 to 62.0 ±0.1 µm, similar to that verified by **[33]**. The physical characteristics of the films are very important for marketing and food quality, including when associated with other conservation methods **[35]**, **[36]**. The biopolymer-based films' thickness, colour, and opacity properties are parameters of considerable influence in food packaging and coating applications since they can positively or negatively affect both the quality of the applied food and consumer decisions **[26]**.

Total polyphenol content

The total polyphenolic content quantified in films C, CP and CPC are presented in Table 2. As cited in other studies, propolis **[37]** and red cabbage **[38]** have active agents, such as flavonoids and phenolic compounds.

Table 2 Total polyphenol content in chitosan (C), chitosan and propolis (CP), and chitosan, propolis and red cabbage (CPC) films.

Samples	Total polyphenol content mgAG g^{-1}
C	00.0 ±0.0^{c}
CP	12.4 ±0.5^{b}
CPC	20.1 ±0.5^{a}

Note: Different lowercase letters denote significant difference among the films samples ($p \leq 0.05$). All values are expressed as the mean ±SD (standard deviation).

The results demonstrate that propolis and red cabbage extract present interesting amounts of phenolic compounds **[39]**, responsible for the antioxidant activity of the films **[40]**. The antioxidant property of the films contributes to the prevention of food oxidation and, therefore, extends the shelf life of food products **[41]**. Studies show increased antioxidant activity with the addition of propolis **[42]**. The total phenolic content and DPPH in chitosan films increased significantly (p <0.05) when propolis was added, demonstrating the increase proportional to the concentration of propolis **[43]**. Chitosan has also been reported to have intrinsic antimicrobial activities **[44]** against Gram-negative and Gram-positive bacteria **[45]**.

Biodegradability test

It was observed that after 2 weeks, the C film inside the soil degraded. Figure 3 shows the film's images on day 0 (a) and after 14 days submerged in soil (b). The film has a high potential for degradability **[46]**, which can be explained by its natural composition (biopolymers and chitosan), which facilitates its degradation in a moist, nutrient-rich environment such as soil **[47]**. The films developed with chitosan in this study are non-toxic, are biodegradable and mechanically stable. Thus, they could be used as food packaging material capable of protecting the food content and the environment **[48]**. The presence of propolis in the films did not alter the degradation properties of the soil. Thus, the chitosan-propolis composite films can be considered rapidly degrading materials when discarded in nature **[26]**.

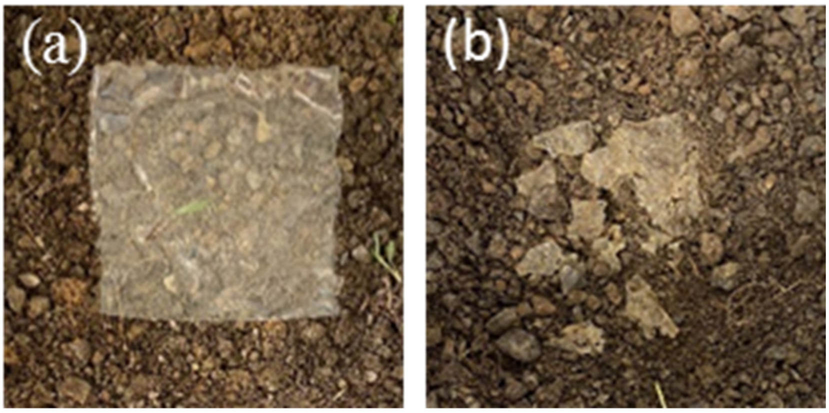

Figure 3 Biodegradability test of C-film (chitosan), (a) day 0 and (b) after 14 days submerged in soil.

Thermogravimetric analysis (TGA)

The thermal property of film reflects its ability to resist decomposition at high temperatures. TGA is frequently used to investigate the thermal stability of film **[49]**. The thermal degradation results of the C, CP and CPC films are shown in Figure 4.

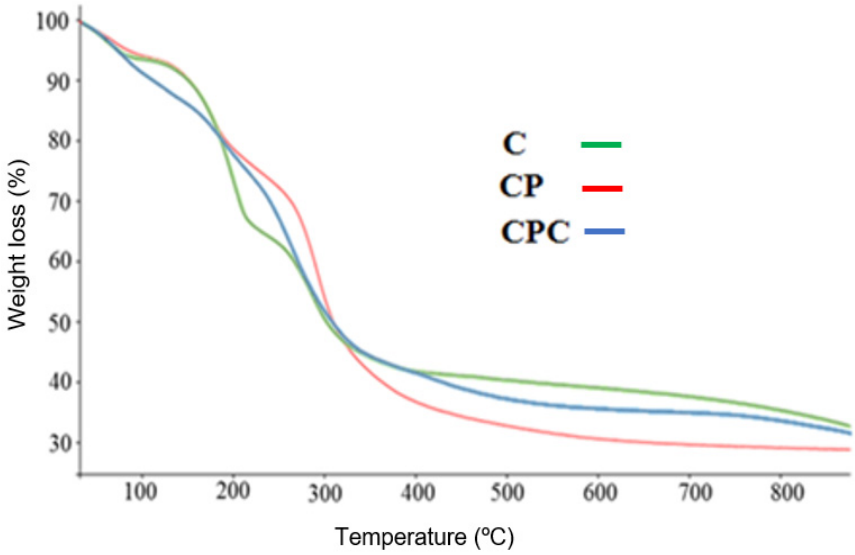

Figure 4 Thermogravimetric analysis of chitosan (C), chitosan and propolis (CP), and chitosan, propolis and red cabbage (CPC) films.

All films showed mass loss due to thermal degradation up to 400 °C. The C film showed a gradual mass reduction of 32%, the CP film of 37% and the CPC of 38% until reaching the temperature of 400 °C **[50]**. This is due to the loss of moisture from the film and from the glycerol. The three films obtained good thermal stability up to 200 °C, which can be attributed to the more compact structure formed by the components, making it difficult for the samples to lose moisture **[51]**. The results were similar to those **[26]** that used chitosan and bio-waste enriched with propolis extract, which showed the first mass loss between 30 and 100 °C, due to the evaporation of water and ethanol in their structures, and the second mass loss (46.0% and 66.9%), between 100 and 700 °C, due to the decomposition of their structure. Studies proved that increased natural extracts such as banana peels extract **[52]**, and purple-fleshed sweet potato extract **[53]**, can reduce chitosan film's weight loss. Films with heat-sealing properties show resistance to thermal degradation and potential application for food packaging **[54]**.

CONCLUSION

The colouration of the films showed a significant difference ($p < 0.05$), demonstrating the influence of propolis and red cabbage extract (anthocyanins) on the colouration and transparency of the material. It is observed that the interaction of the formulation components contributed to obtaining a film with uniform texture and homogeneous colouration, becoming stable at temperatures close to 200 °C, which also indicates the possibility of being heat-sealed. The three compounds used in the development of the film have excellent properties (active, intelligent, edible and biodegradable) that contribute to increasing the shelf life of foods. In addition, the film produced showed good degradability in soil (within 14 days), demonstrating potential as a substitute for petroleum-based plastics.

REFERENCES

1. Finardi, S., Hoffmann, T. G., Angioletti, B. L., Mueller, E., Lazzaris, R. S., Bertoli, S. L., Hlebová, M., Khayrullin, M., Nikolaeva, N., Shariati, M. A., & Krebs de Souza, C. (2022). Development and application of antioxidant coating on fragaria spp. stored under isothermal conditions. In Journal of microbiology, biotechnology and food sciences (Vol. 11, Issue 4, p. e5432). Slovak University of Agriculture in Nitra. https://doi.org/10.55251/jmbfs.5432
2. Pergentino Dos Santos, S., Angioletti, B. L., Hoffmann, T. G., Gonçalves, M. J., Bertoli, S. L., Hlebová, M., Khayrullin, M., Gribkova, V., Shariati, M. A., & Krebs De Souza, C. (2022). Whey based biopolymeric coating as an alternative to improve quality of fresh fruits (malpighia emarginata d.c.) from southern brazil. In Journal of microbiology, biotechnology and food sciences (Vol. 11, Issue 5, p. e5433). Slovak University of Agriculture in Nitra. https://doi.org/10.55251/jmbfs.5433
3. Angioletti, B. L., Pergentino, S., Hoffmann, T. G., Goncalves, M. J., Carvalho, L. F., Bertoli, S. L., Souza, C. K. (2020). Aloe vera gel as natural additive to improve oxidative stability in refrigerated beef burger stored in aerobic and vacuum packaging. In: 2020 Virtual AIChE Annual Meeting, San Francisco.

4. Pergentino, S., Angioletti, B. L., Hoffmann, T. G., Goncalves, M. J., Carvalho, L. F., Bertoli, S. L., Souza, C. K. (2020). Influence of whey protein edible film and refrigeration temperature on quality of acerola in natura during postharvest storage. In: 2020 Virtual AIChE Annual Meeting, San Francisco.
5. Sid, S., Mor, R. S., Kishore, A., & Sharanagat, V. S. (2021). Bio-sourced polymers as alternatives to conventional food packaging materials: A review. In Trends in Food Science & Technology (Vol. 115, pp. 87–104). Elsevier BV. https://doi.org/10.1016/j.tifs.2021.06.026
6. Sarkar, S., Das, D., Dutta, P., Kalita, J., Wann, S. B., & Manna, P. (2020). Chitosan: A promising therapeutic agent and effective drug delivery system in managing diabetes mellitus. In Carbohydrate Polymers (Vol. 247, p. 116594). Elsevier BV. https://doi.org/10.1016/j.carbpol.2020.116594
7. Baek, J., Wahid-Pedro, F., Kim, K., Kim, K., & Tam, K. C. (2019). Phosphorylated-CNC/modified-chitosan nanocomplexes for the stabilization of Pickering emulsions. In Carbohydrate Polymers (Vol. 206, pp. 520–527). Elsevier BV. https://doi.org/10.1016/j.carbpol.2018.11.006
8. Hoffmann, T. G., Amaral D. P., Angioletti, B., Bertoli, S. L., Peres, L. V., Reiter, M. G. R., & de Souza, C. K. (2019). Potentials nanocomposites in food packaging. In Chemical Engineering Transactions (Vol. 75, pp. 253–258). https://doi.org/10.3303/CET1975043
9. Souza, V. G. L., Pires, J. R. A., Rodrigues, C., Coelhoso, I. M., & Fernando, A. L. (2020). Chitosan composites in packaging industry-current trends and future challenges. In Polymers (Vol. 12, Issue 2, p. 417). MDPI AG. https://doi.org/10.3390/polym12020417
10. Alirezalu, K., Hesari, J., Nemati, Z., Munekata, P. E. S., Barba, F. J., & Lorenzo, J. M. (2019). Combined effect of natural antioxidants and antimicrobial compounds during refrigerated storage of nitrite-free frankfurter-type sausage. In Food Research International (Vol. 120, pp. 839–850). Elsevier BV. https://doi.org/10.1016/j.foodres.2018.11.048
11. Sahariah, P., & Másson, M. (2017). Antimicrobial Chitosan and Chitosan Derivatives: A Review of the Structure–Activity Relationship. In Biomacromolecules (Vol. 18, Issue 11, pp. 3846–3868). American Chemical Society (ACS). https://doi.org/10.1021/acs.biomac.7b01058
12. Shankar, S., Wang, L.-F., & Rhim, J.-W. (2017). Preparation and properties of carbohydrate-based composite films incorporated with CuO nanoparticles. In Carbohydrate Polymers (Vol. 169, pp. 264–271). Elsevier BV. https://doi.org/10.1016/j.carbpol.2017.04.025
13. Karagöz, Ş., & Demirdöven, A. (2019). Effect of chitosan coatings with and without Stevia rebaudiana and modified atmosphere packaging on quality of cold stored fresh-cut apples. In LWT (Vol. 108, pp. 332–337). Elsevier BV. https://doi.org/10.1016/j.lwt.2019.03.040
14. Lin, L., Gu, Y., & Cui, H. (2018). Novel electrospun gelatin-glycerin-ε-Poly-lysine nanofibers for controlling Listeria monocytogenes on beef. In Food Packaging and Shelf Life (Vol. 18, pp. 21–30). Elsevier BV. https://doi.org/10.1016/j.fpsl.2018.08.004.
15. Xiong, Y., Chen, M., Warner, R. D., & Fang, Z. (2020). Incorporating nisin and grape seed extract in chitosan-gelatine edible coating and its effect on cold storage of fresh pork. In Food Control (Vol. 110, p. 107018). Elsevier BV. https://doi.org/10.1016/j.foodcont.2019.107018
16. Alves Pinto, L. D. M., Taironi Do Prado, N. R., & De Carvalho, L. B. (2011). Propriedades, usos e aplicações da própolis. In Revista Eletrônica de Farmácia (Vol. 8, Issue 3). Universidade Federal de Goias. https://doi.org/10.5216/ref.v8i3.15805
17. Yousuf, B., Gul, K., Wani, A. A., & Singh, P. (2015). Health Benefits of Anthocyanins and Their Encapsulation for Potential Use in Food Systems: A Review. In Critical Reviews in Food Science and Nutrition (Vol. 56, Issue 13, pp. 2223–2230). Informa UK Limited. https://doi.org/10.1080/10408398.2013.805316
18. Kurek, M., Garofulić, I. E., Bakić, M. T., Ščetar, M., Uzelac, V. D., & Galić, K. (2018). Development and evaluation of a novel antioxidant and pH indicator film based on chitosan and food waste sources of antioxidants. In Food Hydrocolloids (Vol. 84, pp. 238–246). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2018.05.050
19. Wang, X., Yong, H., Gao, L., Li, L., Jin, M., & Liu, J. (2019). Preparation and characterization of antioxidant and pH-sensitive films based on chitosan and black soybean seed coat extract. In Food Hydrocolloids (Vol. 89, pp. 56–66). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2018.10.019
20. Zhang, J., Zou, X., Zhai, X., Huang, X., Jiang, C., & Holmes, M. (2019). Preparation of an intelligent pH film based on biodegradable polymers and roselle anthocyanins for monitoring pork freshness. In Food Chemistry (Vol. 272, pp. 306–312). Elsevier BV. https://doi.org/10.1016/j.foodchem.2018.08.041
21. Hoffmann, T. G., Angioletti, B. L., Bertoli, S. L., & de Souza, C. K. (2021). Intelligent pH-sensing film based on jaboticaba peels extract incorporated on a biopolymeric matrix. In Journal of Food Science and

Technology (Vol. 59, Issue 3, pp. 1001–1010). Springer Science and Business Media LLC. https://doi.org/10.1007/s13197-021-05104-6

22. Liang, T., Sun, G., Cao, L., Li, J., & Wang, L. (2019). A pH and NH3 sensing intelligent film based on Artemisia sphaerocephala Krasch. gum and red cabbage anthocyanins anchored by carboxymethyl cellulose sodium added as a host complex. In Food Hydrocolloids (Vol. 87, pp. 858–868). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2018.08.028

23. Pereira, V. A., Jr., de Arruda, I. N. Q., & Stefani, R. (2015). Active chitosan/PVA films with anthocyanins from Brassica oleraceae (Red Cabbage) as Time–Temperature Indicators for application in intelligent food packaging. In Food Hydrocolloids (Vol. 43, pp. 180–188). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2014.05.014

24. Pourjavaher, S., Almasi, H., Meshkini, S., Pirsa, S., & Parandi, E. (2017). Development of a colourimetric pH indicator based on bacterial cellulose nanofibers and red cabbage (Brassica oleraceae) extract. In Carbohydrate Polymers (Vol. 156, pp. 193–201). Elsevier BV. https://doi.org/10.1016/j.carbpol.2016.09.027

25. Medina Jaramillo, C., Gutiérrez, T. J., Goyanes, S., Bernal, C., & Famá, L. (2016). Biodegradability and plasticizing effect of yerba mate extract on cassava starch edible films. In Carbohydrate Polymers (Vol. 151, pp. 150–159). Elsevier BV. https://doi.org/10.1016/j.carbpol.2016.05.025

26. De Carli, C., Aylanc, V., Mouffok, K. M., Santamaria-Echart, A., Barreiro, F., Tomás, A., Pereira, C., Rodrigues, P., Vilas-Boas, M., & Falcão, S. I. (2022). Production of chitosan-based biodegradable active films using bio-waste enriched with polyphenol propolis extract envisaging food packaging applications. In International Journal of Biological Macromolecules (Vol. 213, pp. 486–497). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2022.05.155

27. Chen, M., Yan, T., Huang, J., Zhou, Y., & Hu, Y. (2021). Fabrication of halochromic smart films by immobilizing red cabbage anthocyanins into chitosan/oxidized-chitin nanocrystals composites for real-time hairtail and shrimp freshness monitoring. In International Journal of Biological Macromolecules (Vol. 179, pp. 90–100). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2021.02.170

28. Souza, V., Fernando, A., Pires, J., Rodrigues, P., Lopes, A., Fernandes, F. (2017). Physical properties of chitosan films incorporated with natural antioxidants. Industrial Crops And Products, (Vol. 107, pp. 565-572). Elsevier BV. http://dx.doi.org/10.1016/j.indcrop.2017.04.056

29. Shahabi, N., Soleimani, S., & Ghorbani, M. (2023). Investigating functional properties of halloysite nanotubes and propolis used in reinforced composite film based on soy protein/basil seed gum for food packaging application. In International Journal of Biological Macromolecules (Vol. 231, p. 123350). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2023.123350

30. Cheng, M., Cui, Y., Yan, X., Zhang, R., Wang, J., & Wang, X. (2022). Effect of dual-modified cassava starches on intelligent packaging films containing red cabbage extracts. In Food Hydrocolloids (Vol. 124, p. 107225). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2021.107225

31. Freitas, P. A. V., de Oliveira, T. V., Silva, R. R. A., Fialho e Moraes, A. R., Pires, A. C. dos S., Soares, R. R. A., Junior, N. S., & Soares, N. F. F. (2020). Effect of pH on the intelligent film-forming solutions produced with red cabbage extract and hydroxypropylmethylcellulose. In Food Packaging and Shelf Life (Vol. 26, p. 100604). Elsevier BV. https://doi.org/10.1016/j.fpsl.2020.100604

32. Zhao, L., Liu, Y., Zhao, L., & Wang, Y. (2022). Anthocyanin-based pH-sensitive smart packaging films for monitoring food freshness. In Journal of Agriculture and Food Research (Vol. 9, p. 100340). Elsevier BV. https://doi.org/10.1016/j.jafr.2022.100340

33. Hashim, S. B. H., Tahir, H. E., Lui, L., Zhang, J., Zhai, X., Mahdi, A. A., Ibrahim, N. A., Mahunu, G. K., Hassan, M. M., Xiaobo, Z., & Jiyong, S. (2023). Smart films of carbohydrate-based/sunflower wax/purple Chinese cabbage anthocyanins: A biomarker of chicken freshness. In Food Chemistry (Vol. 399, p. 133824). Elsevier BV. https://doi.org/10.1016/j.foodchem.2022.133824

34. Marangoni Júnior, L., Jamróz, E., Gonçalves, S. de Á., da Silva, R. G., Alves, R. M. V., & Vieira, R. P. (2022). Preparation and characterization of sodium alginate films with propolis extract and nano-SiO_2. In Food Hydrocolloids for Health (Vol. 2, p. 100094). Elsevier BV. https://doi.org/10.1016/j.fhfh.2022.100094

35. Hoffmann, T. G., Ronzoni A. F., da Silva D. L., Bertoli S. L., & de Souza C. K. (2021). Cooling Kinetics and Mass Transfer in Postharvest Preservation of Fresh Fruits and Vegetables Under Refrigerated Conditions. Chemical Engineering Transactions, 87, 115–120. https://doi.org/10.3303/CET2187020

36. Hoffmann, T.G., Meinert, C., Ormelez, F., Campani, C., Bertoli, S.L., Ender, L., De Souza, C.K. (2023). Fresh food shelf-life improvement by humidity regulation in domestic refrigeration. Procedia Computer Science (Vol.217, pp. 826-834). Elsevier BV. https://doi.org/10.1016/j.procs.2022.12.279

37. Bertotto, C., Bilck, A. P., Yamashita, F., Anjos, O., Bakar Siddique, M. A., Harrison, S. M., Brunton, N. P., & Carpes, S. T. (2022). Development of a biodegradable plastic film extruded with the addition of a Brazilian propolis by-product. In LWT (Vol. 157, p. 113124). Elsevier BV. https://doi.org/10.1016/j.lwt.2022.113124
38. Ren, G., He, Y., Lv, J., Zhu, Y., Xue, Z., Zhan, Y., Sun, Y., Luo, X., Li, T., Song, Y., Niu, F., Huang, M., Fang, S., Fu, L., & Xie, H. (2023). Highly biologically active and pH-sensitive collagen hydrolysate-chitosan film loaded with red cabbage extracts realizing dynamic visualization and preservation of shrimp freshness. In International Journal of Biological Macromolecules (Vol. 233, p. 123414). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2023.123414
39. Fernández-Marín, R., Labidi, J., Andrés, M. Á., & Fernandes, S. C. M. (2020). Using α-chitin nanocrystals to improve the final properties of poly (vinyl alcohol) films with Origanum vulgare essential oil. In Polymer Degradation and Stability (Vol. 179, p. 109227). Elsevier BV. https://doi.org/10.1016/j.polymdegradstab.2020.109227
40. Kanatt, S. R., Rao, M. S., Chawla, S. P., & Sharma, A. (2012). Active chitosan–polyvinyl alcohol films with natural extracts. In Food Hydrocolloids (Vol. 29, Issue 2, pp. 290–297). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2012.03.005
41. Ghosh, T., Priyadarshi, R., Krebs de Souza, C., Angioletti, B. L., & Rhim, J.-W. (2022). Advances in pullulan utilization for sustainable applications in food packaging and preservation: A mini-review. In Trends in Food Science & Technology (Vol. 125, pp. 43–53). Elsevier BV. https://doi.org/10.1016/j.tifs.2022.05.001
42. Correa-Pacheco, Z. N., Bautista-Baños, S., Ramos-García, M. de L., Martínez-González, M. del C., & Hernández-Romano, J. (2019). Physicochemical characterization and antimicrobial activity of edible propolis-chitosan nanoparticle films. In Progress in Organic Coatings (Vol. 137, p. 105326). Elsevier BV. https://doi.org/10.1016/j.porgcoat.2019.105326
43. Siripatrawan, U., & Vitchayakitti, W. (2016). Improving functional properties of chitosan films as active food packaging by incorporating with propolis. In Food Hydrocolloids (Vol. 61, pp. 695–702). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2016.06.001
44. Elsabee, M. Z., & Abdou, E. S. (2013). Chitosan based edible films and coatings: A review. In Materials Science and Engineering: C (Vol. 33, Issue 4, pp. 1819–1841). Elsevier BV. https://doi.org/10.1016/j.msec.2013.01.010
45. Hernández-Muñoz, P., Almenar, E., Valle, V. D., Velez, D., & Gavara, R. (2008). Effect of chitosan coating combined with postharvest calcium treatment on strawberry (Fragaria×ananassa) quality during refrigerated storage. In Food Chemistry (Vol. 110, Issue 2, pp. 428–435). Elsevier BV. https://doi.org/10.1016/j.foodchem.2008.02.020
46. Gan, P. G., Sam, S. T., Abdullah, M. F., Omar, M. F., & Tan, W. K. (2021). Water resistance and biodegradation properties of conventionally-heated and microwave-cured cross-linked cellulose nanocrystal/chitosan composite films. In Polymer Degradation and Stability (Vol. 188, p. 109563). Elsevier BV. https://doi.org/10.1016/j.polymdegradstab.2021.109563
47. Tu, H., Li, X., Xie, K., Zhang, J., Liu, Y., shao, X., Lin, X., Zhang, R., & Duan, B. (2023). High strength and biodegradable dielectric film with synergistic alignment of chitosan nanofibrous networks and BNNSs. In Carbohydrate Polymers (Vol. 299, p. 120234). Elsevier BV. https://doi.org/10.1016/j.carbpol.2022.120234
48. Akyuz, L., Kaya, M., Ilk, S., Cakmak, Y. S., Salaberria, A. M., Labidi, J., Yılmaz, B. A., & Sargin, I. (2018). Effect of different animal fat and plant oil additives on physicochemical, mechanical, antimicrobial and antioxidant properties of chitosan films. In International Journal of Biological Macromolecules (Vol. 111, pp. 475–484). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2018.01.045
49. Ruggero, F., Carretti, E., Gori, R., Lotti, T., & Lubello, C. (2020). Monitoring of degradation of starch-based biopolymer film under different composting conditions, using TGA, FTIR and SEM analysis. In Chemosphere (Vol. 246, p. 125770). Elsevier BV. https://doi.org/10.1016/j.chemosphere.2019.125770
50. Ardjoum, N., Chibani, N., Shankar, S., Fadhel, Y. B., Djidjelli, H., & Lacroix, M. (2021). Development of antimicrobial films based on poly(lactic acid) incorporated with Thymus vulgaris essential oil and ethanolic extract of Mediterranean propolis. In International Journal of Biological Macromolecules (Vol. 185, pp. 535–542). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2021.06.194
51. Thirunathan, P., Arnz, P., Husny, J., Gianfrancesco, A., & Perdana, J. (2018). Thermogravimetric analysis for rapid assessment of moisture diffusivity in polydisperse powder and thin film matrices. In Food Chemistry (Vol. 242, pp. 519–526). Elsevier BV. https://doi.org/10.1016/j.foodchem.2017.09.089
52. Zhang, W., Li, X., & Jiang, W. (2020). Development of antioxidant chitosan film with banana peels extract and its application as coating in maintaining the storage quality of apple. In International Journal of

Biological Macromolecules (Vol. 154, pp. 1205–1214). Elsevier BV. https://doi.org/10.1016/j.ijbiomac.2019.10.275

53. Yong, H., Wang, X., Bai, R., Miao, Z., Zhang, X., & Liu, J. (2019). Development of antioxidant and intelligent pH-sensing packaging films by incorporating purple-fleshed sweet potato extract into chitosan matrix. In Food Hydrocolloids (Vol. 90, pp. 216–224). Elsevier BV. https://doi.org/10.1016/j.foodhyd.2018.12.015

54. Lim, W. S., Ock, S. Y., Park, G. D., Lee, I. W., Lee, M. H., & Park, H. J. (2020). Heat-sealing property of cassava starch film plasticized with glycerol and sorbitol. In Food Packaging and Shelf Life (Vol. 26, p. 100556). Elsevier BV. https://doi.org/10.1016/j.fpsl.2020.100556

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