Vol. 18, No. 3

ISSN 2064-7964

## THE INFLUENCE OF THE TYPE AND CONCENTRATION OF PLASTICIZER ON THE PROPERTIES OF BIOPOLYMER FILMS BASED ON WILD FLAX (CAMELINA SATIVA)

<sup>1</sup>Šuput Danijela, <sup>1</sup>Lončar Biljana, <sup>1</sup>Popović Senka, <sup>1</sup>Hromiš Nevena, <sup>2</sup>Rakita Slađana, <sup>2</sup>Spasevski Nedeljka

<sup>1</sup>University of Novi Sad, Faculty of Technology Novi Sad, Bulevar cara Lazara 1, 21000 Novi Sad, Serbia, <sup>2</sup>University of Novi Sad, Institute of Food Technology, Bulevar cara Lazara 1, 21000 Novi Sad, Serbia, e-mail: suput.danijela@gmail.com

Received: 13 <sup>th</sup> June	Accepted: 19th September

#### ABSTRACT

The development of biodegradable packaging materials using naturally occurring, renewable biopolymers has gained attraction due to consumer demand for high-quality products and concerns about environmental waste problems. However, the inferior mechanical properties and low water resistance of packaging materials based on natural polymers pose a significant obstacle to their wider use. One of the ways to improve the properties of biopolymer-based packaging materials is the addition of plasticizers during their synthesis. In this work, the influence of the type and concentration of plasticizer on the properties of new biopolymer films based on wild flax (*Camelina sativa*) was investigated. *Camelina sativa* oil cake (CSoC) remains after edible oil cold pressing as a by-product. One of the possibilities for its valorization is the synthesis of biopolymer materials. During the synthesis, different plasticizers - glycerol and polyethylene glycol 400 - were added in different concentrations - 20%-60%. The obtained CSoC-based biopolymer films were analyzed for the following properties: Moisture content, solubility, thickness, tensile strength, elongation at break, and water vapor permeability. The results obtained showed significant differences when different plasticizers were applied at different concentrations. The biopolymer film with optimal properties was obtained by adding glycerol at a concentration of 40%.

Keywords: biopolymer films, Camelina sativa, plasticizers, properties

#### **1. INTRODUCTION**

Recent research has concentrated on creating eco-friendly, edible, and biodegradable materials due to growing concerns about the quality and shelf life of food products as well as awareness of environmental problems associated with plastic materials disposal [1]. Renewable resources including polysaccharides (starch, alginates, pectin, carrageenans, chitosan), proteins (casein, whey, collagen, gelatin, corn, soy, wheat), and lipids (fat, wax, oil) are commonly used to create biopolymer materials [2]. Reducing the environmental damage caused by non-biodegradable plastic waste is largely dependent on new biodegradable films manufactured from biopolymers [3]. Furthermore, there is significant interest in the field of bio-based polymers derived from different agricultural and food commodities/waste in creating materials with filmforming and packaging attributes that could be used for food safety and shelf-life prolongation [4]. Therefore, the cakes remaining after the cold pressing of the edible oil can be valorized as an ideal substrate for obtaining biopolymer films. Their advantage is a rich nutritional profile in terms of bioactive compounds, but also a composite composition. One of the top contenders to be employed in the future bio-economy is Camelina sativa, an annual oilseed crop in the Brassicaceae family [5]. Camelina sativa has been gaining attraction because of many advantages over traditional oilseed crops, including significant agrotechnical and industrial benefits [6] and its extremely rich nutritional profile: oil (30%-49%), proteins (24%-31%), tocopherols, phytosterols, phenolic compounds,  $\omega$ -3 and  $\omega$ -6 acids, and other substances [7-9]. The US Food and Drug Administration has granted camelina seed oil the Generally Recognized as Safe (GRAS) designation [10]. After oil is extracted, large amounts of by-products (such as cakes) are left over. About 10% fat, 45% crude protein, 13% fiber, 5% minerals, and a few other ingredients make up camelina cake [11].

Vol. 18, No. 3

ISSN 2064-7964

Applications for composite films are numerous, and one of them is food packing since composite films can unite two different kinds of biopolymers, each with unique properties, so that the resulting biopolymerderived films may protect a wider range of food products [12]. Films made from biodegradable biopolymers exhibit certain disadvantages in terms of their mechanical, barrier, and physicochemical properties compared to their plastic counterparts. To enhance these, lipids, plasticizers, nanoparticles, and antibacterial substances can be added. One of the directions of biodegradable materials for food packaging design looks to optimize film properties by studying the effect of plasticizers. Low molecular weight chemicals are typically utilized as plasticizers since they must be miscible in the biopolymer [13]. To modify the functional qualities of films by enhancing their extensibility, dispensability, flexibility, elasticity, stiffness, and mechanical properties, plasticizers are added to biopolymer materials [14]. Since polyols, such as glycerol, sorbitol, mannitol, and xylitol, are especially effective at plasticizing, numerous researchers have concentrated on their application. Additionally, monosaccharides like glucose, mannose, fructose, and sucrose could also act as plasticizers. Films have also been produced using fatty acids as plasticizers [15]. The IUPAC defines a plasticizer as a material added to a material to improve its workability, flexibility, or distensibility [16]. Reducing the relative number of polymer-polymer connections causes the polymer to become more malleable by lowering the rigidity of the three-dimensional hard structure and permitting the polymer to bend without rupturing. Thus, plasticizers enhance polymers' flexibility, processability, durability, and, in certain situations, cost [17]. Plasticizers enable processing on various types of machinery (extrusion, calendering, injection molding), and reduce extrusion pressure and mixing time. They also improve fracture and impact resistance while decreasing physical attributes like hardness and elastic modulus. The flexibility of the polymer chain also affects viscosity, density, and dielectric constant [18].

Edible films have been synthesized using a range of widely used polyol-plasticizers; a tiny amount of these plasticizers can be simply injected between polymer chains [2]. Because of its three hydroxyl groups, glycerol is a simple polyol that is soluble in water, polar, non-volatile, and tolerant of high temperatures and is recognized by the Food and Drug Administration as a food additive, for the plasticization of biodegradable films [18]. It is a little hydrophilic molecule that can be placed between polymer chains increasing intermolecular space while decreasing intermolecular hydrogen bonding. The distance between the matrix chains grows and the number of direct interactions decreases as glycerol is inserted into the matrix network. By increasing the concentration of plasticizers in a film-forming solution, less stiff, less rigid, and more flexible films are synthesized [15]. Polyethylene Glycol (PEG) is a biocompatible, hydrophilic, and antifouling plasticizer. Compatibility, mechanical qualities (tensile and elongation strength), and thermal stability are all enhanced by the inclusion of PEG in film-forming structures. Furthermore, because PEG is hydrophilic, it can also degrade bioplastics quickly when added. PEG 400 has the advantage of creating biopolymers that are firm and elastic when compared to other plasticizers, like glycerol, and it can raise the strain value [19].

This work aims to answer the question of which of the two plastificators - glycerol or PEG 400 - is more suitable for optimizing the mechanical, barrier, and physico-chemical properties of biopolymer films based on *Camelina sativa* oilseed cake.

## 2. MATERIALS AND METHODS

#### 2.1. Materials

*Camelina sativa* seeds were kindly supplied by the Institute of Field and Vegetable Crops (Novi Sad, Serbia). The remained cake after the seed's cold pressing was used as a film-making substrate. All other reagents used in this study were of analytical grade.

Vol. 18, No. 3

ISSN 2064-7964

### 2024

### 2.2. Biopolymer film synthesis

Laboratory casting method was applied when synthesizing biopolymer films based on *Camelina sativa* oilseed cake (CSoC): Filmogenic suspension was prepared by dispersing CSoC (5 %, w/v) in distilled water with the addition of plasticizers glycerol and PEG 400 in various concentrations (20% -60%) accounted on the weight of CSoC. In this way, 10 samples were obtained labeled as 20%G, 30%G, 40%G, 50%G, 60%G, 20%PEG, 30%PEG, 40%PEG, 50%PEG, and 60%PEG. Further, all of the sample's pH was adjusted to pH 12 by adding NaOH solution. pH was determined using a pH meter (Metrohm AG, Switzerland). The obtained suspension was incubated in a water bath at 100 °C for 20 min. Finally, the obtained film-forming suspension was passed through a nylon filter to remove the undissolved particles and poured onto Petri plates, covered with Teflon. Biopolymer films based on CSoC were obtained after 5-day drying at room conditions  $(23\pm2 °C; 50\pm5 % RH)$  and processed for further analysis.

#### 2.3. Biopolymer film characterization

Mechanical properties – tensile strength and elongation at break

Instron Universal Testing Instrument 4301 (Instron Engineering Corp., Canton, MA, USA) was used for mechanical properties (tensile strength (TS) and elongation at break (EB)) determination according to standard method EN ISO 527-3:2018 [20]. The initial distance between the instrument clamps was set to 50 mm and the test speed was set to 50 mm/min. The result was expressed as the mean value of three independent measurements  $\pm$  SD, for each sample.

Barrier properties - water vapor permeability

The water vapor permeability (WVP) of the biopolymer film samples was determined according to the standard gravimetric (dish) method ISO 2528:2017 [21] as absorbed moisture (the difference in the mass of the silica gel before and after treatment) divided by the surface area of the film sample. The results were expressed as grams per square meter per hour (g/m<sup>2</sup>h). The result was expressed as the mean value of three independent measurements  $\pm$  SD, for each sample.

## Physico-chemical properties

#### Moisture content

Film samples were cut into squares  $(1\text{cm}^2)$ , weighed at analytical balance  $(m_1)$ , and dried in an air-circulating oven at  $(105\pm2)^{\circ}$ C (Instrumentaria, Zagreb, Croatia) to assess constant weight  $(m_2)$ . The moisture content (MC) was determined as the percentage of weight reduction after film drying, expressed on the total weight of the film:

(1)

MC (%) =  $\frac{m_1 - m_2}{m_1} \cdot 100$ 

The result was expressed as the mean value of three independent measurements  $\pm$  SD, for each sample.

Film solubility

Dried film samples  $(m_1)$ , after moisture content determination, were immersed in 30 ml of deionized water and left for 24h at room conditions. After 24 h, the water was decanted and the films were dried again in an air-circulating oven (Instrumentaria, Zagreb, Croatia) at  $(105 \pm 2)^{\circ}$ C to a constant mass  $(m_2)$ . The total solubility of the films (%) was calculated as:

(2)

$$S(\%) = \frac{m_1 - m_2}{m_1} \cdot 100$$

The result was expressed as the mean value of three independent measurements  $\pm$  SD, for each sample.

ISSN 2064-7964

#### 2.4. Statistical analysis -standard scores

Standard scores were assessed by ranking 10 samples by comparing their raw data (TS, EB, WVP, MC, and S) to extreme values. The ranking depended on sample data and the highest and lowest values observed for each output value. The lowest values of WVP, MC, and S, and the highest values of TS, and EB were set as optimal output variable values.

#### 3. RESULTS AND DISCUSSION

#### Mechanical properties - tensile strength and elongation at break

The results of the mechanical properties of biopolymer films based on *Camelina sativa* oilseed cake, depending on the type and concentration of added plasticizers, are shown in Tab. 1. The obtained TS values were in the range of 2.61-3.54 MPa with glycerol addition and in the range 0.83-4.56 MPa with PEG 400 addition. The obtained value ranges are following the nature of the raw material from which the films were obtained – oilseed cakes. Literature findings point to 1.37 MPa for biopolymer films based on pumpkin oilseed cake [22]; 4.37 MPa for sunflower oilseed cake [23]; and 1.7 MPa for cardoon seed oilseed cake [24]. Obtained low values direct application of these materials in the form of packaging coatings rather than film. Plasticizers have an impact on the biopolymer film's tensile strength, or its capacity to bear stresses without breaking. The TS of biopolymer films decreased as plasticizer concentration increased. This could be explained by the fact that the plasticizer weakens the solid dispersion system, which lowers the stability of the internal hydrogen bonds in the intermolecular bonds. As a result, the produced material has poor mechanical properties, which lowers the tank strength of the bioplastics produced [19]. Reduced TS values were more pronounced in samples with added PEG.

The EB values in this paper ranged from 3.74% to 10.25% with glycerol addition and in the range of 0.62-1.35 % with PEG 400 addition. The obtained EB values were in correlation with literature values for EB values of films based on other oilseed cakes: about 5% for films based on pumpkin oilseed cake [25]; about 10-20% for films based on rapeseed oilseed cake [26]; and 20-30% for films based on sunflower oilseed cake [23]. The addition of plasticizers increased EB values which is favourable and more pronounced in samples with added glycerol. Plasticizer addition disrupts hydrogen bonds (intermolecular and intramolecular) positioning itself between the polymer molecules thereby interfering with the interaction of the polymers and increasing the flexibility of the film [15, 19]. The hygroscopic nature of glycerol contributed more plasticization effect compared to any other plasticizer, thus increasing the mobility of polymer chains and leading to increased stretchability and flexibility of films [27]. It can be concluded that by providing flexibility and stretchability, lowering intermolecular pressures, and boosting polymer chain mobility, plasticizers help to reduce brittleness in films and make them easier to handle and store [28]. Film mechanical qualities are all directly impacted by the use of plasticizers; the higher the concentration, the greater the elongation at break and the lower the tensile strength [13].

Plastificator	Tensile	Elongation at	Water vapor	Moisture content	Solubility
	strength (MPa)	Break (%)	permeability (g/m <sup>2</sup> h)	(%)	(%)
20%G	3.54	3.74	5.13	20.38	34.72
30%G	3.78	5.42	5.44	27.96	38.59
40%G	3.84	6.46	6.17	19.77	39.68
50%G	3.07	7.24	9.17	30.55	51.41

 Table 1. Characterization of the obtained CSoC-based film properties regarding the addition of glycerol and PEG 400 in various concentrations

DOI: https://doi.org/10.14232/analecta.2024.3.69-76

Vol. 18, No. 3

ISSN 2064-7964

2024

Plastificator	Tensile	Elongation at	Water vapor	Moisture content	Solubility
	strength (MPa)	Break (%)	permeability (g/m <sup>2</sup> h)	(%)	(%)
60%G	2.61	10.25	13.05	22.85	54.32
20%PEG	4.56	0.62	4.71	18.40	43.64
30%PEG	3.22	0.94	5.10	10.34	46.15
40%PEG	1.62	1.00	7.19	19.79	52.39
50%PEG	1.26	1.25	8.72	10.28	54.49
60%PEG	0.83	1.35	9.75	13.89	59.16

#### Barrier properties - water vapor permeability

One of the permeability characteristics is water vapor permeability (WVP). Lower WVP values indicated better water vapor barrier properties of the films. The obtained values for films with glycerol addition were in the range (5.13-13.05 g/m<sup>2</sup>h), and for films with PEG 400 addition were in the range (4.71-9.75 g/m<sup>2</sup>h) (Tab. 1) and aligned with literature findings for sunflower oil cake-based films (8.62-11.63 g/m<sup>2</sup>h) [23], pumpkin oil cake-based materials (14.7 g/m<sup>2</sup>h) [29] and argan seed protein concentrate (7.5 g/m<sup>2</sup>h) [30]. The increase in the concentration of both added plasticizers was directly correlated with the observed increases in WVP which could be explained by networks biopolymers create that are not dense, which makes them more permeable after the addition of plasticizers. These increase free volume and lower the vitreous transition temperature of the system by weakening the intermolecular interactions between neighboring macromolecular chains [13]. Further, water vapor permeability is in correlation with water vapor adsorption. Films with no added plasticizer had the lowest monolayer value while films containing gradually higher plasticizer concentration had gradually higher monolayer value, which is a measure of the number of sorption sites in a film and indicates the maximum amount of water that can be absorbed in a single layer of dry film [31]. Adding plasticizers provides more active sites via exposure of hydroxyl groups, where water molecules can be absorbed.

## **Physico-chemical properties**

According to the obtained results (Tab 1.), moisture content (MC) exhibited higher values for biopolymer films based on *Camelina sativa* with the addition of glycerol as the plasticizer (in the range 19.77-27.96%) compared to biopolymer films based on *Camelina sativa* with the addition of PEG 400 (in the range 10.28-19.79%). The obtained results were in agreement with MC found in the literature for other films based on oilseed cakes: sunflower oilseed cake-based films (13.76-20.47 %) [23], pumpkin oilseed cake-based films (19.78 %) [22] and of cardoon protein-based films (17 %) [24].

Water solubility values were in similar ranges (34.72-54.32%) and (43.64-59.16%) for biopolymer films based on *Camelina sativa* with the addition of glycerol and PEG 400 as the plasticizers, respectively. The addition of both plasticizers caused solubility to increase which was a negative consequence since higher water solubility signifies lower water resistance. Anyway, the obtained values are relatively low, which was confirmed by the fact that at the end of exposure of film samples to water for 24 hours, as required by the test method, the entirety of the film was preserved, and there was no dissolution or dissolution of parts of the film.

Vol. 18, No. 3

ISSN 2064-7964

#### Standard score analysis

The standard score (SS) was obtained by summing the normalized scores for each variable (TS, EB, WVP, MC, S) to determine of ideal plastificator and the optimal concentration applied. The total Z-score values, which mathematically summarize all segment Z-scores, display the best combination of all tested responses. SS function approach to a value of 1 suggests a higher chance that the processing inputs that were examined were optimal. Process optimization relies on standard score results.



Figure 1. Z-Score Analysis of the obtained CSoC-based film properties regarding the addition of glycerol and PEG 400 in various concentrations

According to the results of the standard score analysis (Fig. 1), the highest SS value was obtained for the sample with the addition of 40% glycerol. The obtained SS value reached 0.713. This samples output variable values were: TS = 3.84 MPa; EB = 6.46 %; WVP = 6.17 g/m<sup>2</sup>h; MC = 19.77 %; and S = 39.68 %.

### 4. CONCLUSIONS

In each step of the synthesis of the film-forming solution, it is possible to vary the process parameters, as well as the type and amount of added auxiliary components. The ultimate goal was the valorization of agroindustrial waste - *Camelina sativa* oil cake, the residue leftover after cold pressing of the oil to obtain biopolymer films, where in this step the properties of the films were optimized depending on the type and concentration of the added plasticizer. For this purpose, films were synthesized with 20-60% added glycerol and PEG 400. The obtained results showed that the addition of both plasticizers affected the mechanical properties – tensile strength decrease and elongation at break increase. Water vapor permeability was negatively affected by plasticizer addition since the WVP values increased by plasticizer addition and caused higher film permeation. This result was in agreement with solubility results in which the addition of both plasticizers caused solubility to increase which was a negative consequence since higher water solubility signifies lower water resistance. Standard score analysis indicated sample with the addition of 40% glycerol was optimal in film-making synthesis to be applied as packaging material for further research.

Vol. 18, No. 3

ISSN 2064-7964

### ACKNOWLEDGEMENT

This research was funded by the Provincial Secretariat for High Education and Scientific Research Activity, Autonomous Province of Vojvodina, Republic of Serbia, grant number 142-451-3059/2023-01/02, and by Ministry of Science, Technological Development and Innovation of the Republic of Serbia, grant number 451-03-66/2024-03/200134.

## REFERENCES

[1] S. Galus, E.A. Arik Kibar, M. Gniewosz, K. Krasniewska, Novel materials in the preparation of edible films and coatings—A review, Coatings, 10, (2020), pp. 674. <u>https://doi.org/10.3390/coatings10070674</u>
[2] D. Praseptiangga, A. Rahmawati, G.J. Manuhara, L.U. Khasanah, R. Utami, IOP Conference Series: Earth and Environmental Sciences, 828, (2021), pp. 012034. <u>https://doi.org/10.1088/1755-1315/828/1/012034</u>

[3] P.Y. Soo, N.M. Sarbon, Preparation and characterization of edible chicken skin gelatin film incorporated with rice flour, Food Packaging and Shelf Life, (2018), pp.1–8. <u>https://doi.org/10.1016/j.fpsl.2017.12.009</u>

[4] R. Saranya, P. Shaiju, E. Kevin, P. Ramesh Babu, Bio-based and biodegradable polymers—State-of-theart, challenges and emerging trends, Current Opinion in Green and Sustainable Chemistry, 21, (2020), pp. 75–81. <u>https://doi.org/10.1016/j.cogsc.2019.12.005</u>

[5] C. Lopez, H. Sotin, H. Rabesona, B. Novales, J.-M. Le Quéré, M. Froissard, J.-D. Faure, S. Guyot, M. Anton, Oil Bodies from Chia (Salvia hispanica L.) and Camelina (Camelina sativa L.) Seeds for Innovative Food Applications: Microstructure, Composition and Physical Stability, Foods, 12, (2023), pp. 211. https://doi.org/10.3390/foods12010211

[6] Y.A. Mohammed, C. Chen, R.K. Afshar, Nutrient requirements of Camelina for biodiesel feedstock in Central Montana, Agronomy Journal, 109, (2017), pp. 309–316. https://doi.org/10.2134/agronj2016.03.0163

[7] Z. Hossain, E.N. Johnson, L. Wang, R.E. Blackshaw, Y. Gan, Comparative analysis of oil and protein content and seed yield of five Brassicaceae oilseeds on the Canadian prairie, Industrial Crops and Products, 136, (2019), pp.77–86. <u>https://doi.org/10.1016/j.indcrop.2019.05.001</u>

[8] M. Berti, R. Gesch, C. Eynck, J. Anderson, S. Cermak, S. Camelina uses, genetics, genomics, production, and management, Industrial Crops and Products, 94, (2016), pp. 690-710. https://doi.org/10.1016/j.indcrop.2016.09.034

[9] D. Kurasiak-Popowska, K. Stuper-Szablewska, The phytochemical quality of Camelina sativa seed and oil, Acta Agriculturae Scandinavica, Section B — Soil & Plant Science, 70 (1), (2020), pp. 39-47. https://doi.org/10.1080/09064710.2019.1665706

[10] A. Ubeyitogullari, O.N. Ciftci, Fabrication of bioaerogels from camelina seed mucilage for food applications, Food Hydrocolloids, 102, (2020), pp. 105597. <u>https://doi.org/10.1016/j.foodhyd.2019.105597</u>
[11] M. Sydor, D. Kurasiak-Popowska, K. Stuper-Szablewska, T. Rogozinski, Camelina sativa. Status quo and future perspectives, Industrial Crops & Products, 187, (2022), pp. 115531. https://doi.org/10.1016/j.indcrop.2022.115531

[12] V. Dharini, S. Periyar Selvam, J. Jayaramudu, R. Sadiku Emmanuel, Functional properties of clay nanofillers used in the biopolymer-based composite films for active food packaging applications, Applied Clay Science, 226, (2022), 106555. <u>https://doi.org/10.1016/j.clay.2022.106555</u>

[13] J. Castro-Rosas, A.M. Cruz-Galvez, C.A. Gomez-Aldapa, R.N. Falfan-Cortes, F.A. Guzman-Ortiz, M.L. Rodriguez-Marin, Biopolymer films and the effects of added lipids, nanoparticles and antimicrobials on their mechanical and barrier properties: a review, International Journal of Food Science and Technology, 51 (9) (2016), pp. 1967-1978. <u>https://doi.org/10.1111/ijfs.13183</u>

[14] Z.N. Hanani, J.A. O'Mahony, Y.H. Roos, P.M. Oliveira, J.P. Kerry, Extrusion of gelatin-based composite films: Effects of processing temperature and pH of film forming solution on mechanical and

DOI: https://doi.org/10.14232/analecta.2024.3.69-76

Vol. 18, No. 3

ISSN 2064-7964

barrier properties of manufactured films, Food Packaging and Shelf Life, 2 (2), (2014), pp.91–101. https://doi.org/10.1016/j.fpsl.2014.09.001

[15] N. Suderman, M.I.N. Isa, N.M. Sarbon, The effect of plasticizers on the functional properties of biodegradable gelatin-based film: A review, Food Bioscience 24, (2018), 111–119. https://doi.org/10.1016/j.fbio.2018.06.006

[16] R. Wolf, B.L. Kaul, Plastics, Additives, Ullmann's Encyclopedia of Industrial Chemistry, 2012, pp. 619–671. <u>https://doi.org/10.1002/14356007.a20</u>

[17] S.S. Muobom, A.-M.S. Umar, Y. Soongseok, A.-P. Brolin, A Review on Plasticizers and Eco-Friendly Bioplasticizers: Biomass Sources and Market, International Journal of Engineering Research & Technology (IJERT), 9 (05), (2020), pp.1138-1144. <u>https://doi.org/10.17577/IJERTV9IS050788</u>

[18] M. Bocque, C. Voirin, V. Lapinte, S. Caillol, J.-J. Robin, Petro-Based and Bio-Based Plasticizers: Chemical Structures to Plasticizing Properties. Journal of Polymer Science, Part A: Polymer chemistry, 54, (2016), pp. 11–33. <u>https://doi.org/10.1002/pola.27917</u>

[19] D.S. Maulana, A.S. Mubarak, D.Y. Pujiastuti, The concentration of polyethylene glycol (PEG) 400 on cellulose-based carrageenan waste on biodegradability and mechanical properties bioplastic, IOP Conf. Series: Earth and Environmental Science, 679, (2021), pp. 012008. <u>https://doi.org.10.1088/1755-1315/679/1/012008</u>

[20] ISO 527-3:2018. Plastics - Determination of tensile properties. Part 3: Test conditions for films and sheets. International Organization for Standardization. Geneva, Switzerland.

[21] ISO 2528:2017. Sheet materials. Determination of water vapor transmission rate (WVTR). Gravimetric (dish) method. International Organization for Standardization. Geneva, Switzerland.

[22] S. Bulut, Research of obtaining, characterization and optimization of properties of active, biodegradable, packaging material based on pumpkin oil cake, Doctoral Thesis, University of Novi Sad, Faculty of Technology Novi Sad, Serbia (2021).

[23] D. Šuput, V. Lazić, S. Popović, N. Hromiš, S. Bulut, L. Pezo, J. Banićević, Effect of process parameters on biopolymer films based on sunflower oil cake, Journal on processing and energy in agriculture, 22 (3) (2018), pp. 125-128.

[24] S.F. Mirpoor, D. Zannini, G. Santagata, C.V.L. Giosafatto, Cardoon seed oil cake proteins as substrate for microbial transglutaminase: Their application as matrix for bio-based packaging to extend the shelf-life of peanuts, Food Hydrocolloids, 147 (2014), pp. 109339. <u>https://doi.org/10.1016/j.foodhyd.2023.109339</u>

[25] S. Popović, The study of production and characterization of biodegradable, composite films based on plant proteins. Doctoral Thesis, University of Novi Sad, Faculty of Technology Novi Sad, Serbia (2013).

[26] S.-A. Jang, G.-O. Lim, K.B. Song, Preparation and Mechanical Properties of Edible Rapeseed Protein Films, Journal of Food Science, 76 (2011), pp. C218-C223. <u>https://doi.org/10.1111/j.1750-3841.2010.02026.x</u>

[27] R. Sothornvit, Edible films and coatings for packaging applications, Polymers for Packaging Applications, Apple Academic Press Inc, NJ, 2014, pp. 173-195.

[28] L.M. Perez, G.N. Piccirilli, N.J. Delorenzi, R.A. Verdini, Effect of different combinations of glycerol and/or trehalose on physical and structural properties of whey protein concentrate-based edible films, Food Hydrocolloids, 56 (2016), pp. 352–359. https://doi.org/ <u>https://doi.org/10.1016/j.foodhyd.2015.12.037</u>

[29] S. Bulut, S. Popović, N. Hromiš, D. Šuput, D. Adamović, V. Lazić, Incorporation of essential oils into pumpkin oil cake-based materials in order to improve their properties and reduce water sensitivity, Hemijska Industrija, 74 (5) (2020), pp. 313-325. <u>https://doi.org/10.2298/HEMIND200622026B</u>

[30] S.F. Mirpoor, C.V.L. Giosafatto, L. Mariniello, A. D'Agostino, M. D'Agostino, M. Cammarota, C. Schiraldi, R. Porta, Argan (Argania spinosa L.) Seed Oil Cake as a Potential Source of Protein-Based Film Matrix for Pharmaco-Cosmetic Applications, International Journal of Molecular Science, 23 (15) (2022), pp. 8478. <u>https://doi.org/10.3390/ijms23158478</u>

[31] S. Mali, S. Grossmann, F. Ysamashita, M.V.E. Grossmann, Water sorption and mechanical properties cassava starch films and their relation to plasticizing effect, Carbohydrate Polymers, 50 (2005), pp. 283–289. https://doi.org/10.1016/j.carbpol.2005.01.003