turnitin **Turnitin Originality Report**

64 Cold Plasma-Based Fabrication and Characterization by Shella Santoso

From Paper (Paper)

Processed on 26-Apr-2022 09:07 WIB
ID: 1820408143
Word Count: 11051

Similarity Index	Similarity by Source	
18%	Internet Sources:	8%
	Publications:	18%
	Student Papers:	2%

sources:

1 2% match (publications)
Xin-Jie Loke, Chao-Kai Chang, Chih-Yao Hou, Kuan-Chen Cheng, Chang-Wei Hsieh. "Plasma-treated polyethylene coated with polysaccharide and protein containing cinnamaldehyde for active packaging films and applications on tilapia (Orechromis niloticus) fillet preservation", Food Control, 2021

2 1% match (publications)
Bingren Tian, Jianhua Cheng, Tianping Zhang, Yumei Liu, Dejun Chen. "Multifunctional chitosan-based film loaded with hops β-acids: Preparation, characterization, controlled release and antibacterial mechanism", Food Hydrocolloids, 2022

3 1% match (Internet from 17-Apr-2022)
https://www.mdpi.com/2073-8994/13/9/1629/htm

4 1% match (publications)
Somenath Das, Vipin Kumar Singh, Abhishek Kumar Dwivedy, Anand Kumar Chaudhari et al. "Fabrication, characterization and practical efficacy of Myristica fragrans essential oil nanoemulsion delivery system against postharvest biodeterioration", Ecotoxicology and Environmental Safety, 2020

5 1% match (publications)
Hadi Almasi, Saeedeh Azizi, Sajed Amjadi. "Development and characterization of pectin films activated by nanoemulsion and Pickering emulsion stabilized marjoram (Origanum majorana L.) essential oil", Food Hydrocolloids, 2020

6 1% match (publications)
Liming Zhang, Zhanli Liu, Xiangyou Wang, Shuang Dong, Yang Sun, Zitong Zhao. "The properties of chitosan/zein blend film and effect of film on quality of mushroom (Agaricus bisporus)", Postharvest Biology and Technology, 2019

7 1% match (publications)
Victor J. Romero, Alberto Sanchez-Lite, Gerard Liraut. "Development of a Multi-Criteria Design Optimization Methodology for Automotive Plastics Parts", Polymers, 2021

8 1% match (publications)
Ilyes Dammak, Rodrigo Vinicius Lourenço, Paulo José do Amaral Sobral. "Active gelatin films incorporated with Pickering emulsions encapsulating hesperidin: Preparation and physicochemical characterization", Journal of Food Engineering, 2019

9 < 1% match (Internet from 26-Oct-2021)
https://www.mdpi.com/2079-6412/9/9/583/htm

10 < 1% match (Internet from 01-Mar-2020)
https://www.mdpi.com/1420-3049/24/3/466/html

11 < 1% match (publications)
Bingren Tian, Jie Wang, Qiang Liu, Yumei Liu, Dejun Chen. "Formation chitosan-based hydrogel film containing silicon for hops β-acids release as potential food packaging material", International Journal of Biological Macromolecules, 2021

12 < 1% match (publications)
Yue Yang, Qiyang Jiao, Lujin Wang, Yu Zhang, Bin Jiang, Dongmei Li, Zhibiao Feng, Chunhong Liu. "Preparation and evaluation of a novel high internal phase Pickering emulsion based on whey protein isolate nanofibrils derived by hydrothermal method", Food Hydrocolloids, 2022

13 < 1% match (publications)
Li-Wah Wong, Chih-Yao Hou, Chun-Chi Hsieh, Chao-Kai Chang, Yi-Shan Wu, Chang-Wei Hsieh. "Preparation of antimicrobial active packaging film by capacitively coupled plasma

treatment", LWT, 2020

14 < 1% match (Internet from 17-Feb-2020)
https://www.e3s-conferences.org/articles/e3sconf/pdf/2020/07/e3sconf_ismfr20_03014.pdf

15 < 1% match ()
Ginting, Binawati, Maira, Ratna, ., Mustanir, Helwati, Hira, Desiyana, Lydia Septa, Mujahid, Rohmat. "ISOLATION OF ESSENSIAL OIL OF NUTMEG (Myristica fragrans Houtt) and ANTIOXIDANT ACTIVITY TEST WITH DPPH", 'LPPM Unsyiah', 2018

16 < 1% match (publications)
Hajlaoui, Hafedh, Hedi Mighri, Mahjoub Aouni, Néji Gharsallah, and Adel Kadri. "Chemical composition and in vitro evaluation of antioxidant, antimicrobial, cytotoxicity and anti-acetylcholinesterase properties of Tunisian Origanum majorana L. essential oil", Microbial Pathogenesis, 2016.

17 < 1% match (publications)
Ilyes Dammak, Rosemary Aparecida de Carvalho, Carmen Sílvia Fávaro Trindade, Rodrigo Vinicius Lourenço et al. "Properties of active gelatin films incorporated with rutin-loaded nanoemulsions", International Journal of Biological Macromolecules, 2017

18 < 1% match (publications)
Yi Shen, Zhi-Jing Ni, Kiran Thakur, Jian-Guo Zhang, Fei Hu, Zhao-Jun Wei. "Preparation and characterization of clove essential oil loaded nanoemulsion and pickering emulsion activated pullulan-gelatin based edible film", International Journal of Biological Macromolecules, 2021

19 < 1% match (publications)
Isabela dos Santos Paglione, Marcella Vitoria Galindo, João Augusto Salviano de Medeiros, Fabio Yamashita et al. "Comparative study of the properties of soy protein concentrate films containing free and encapsulated oregano essential oil", Food Packaging and Shelf Life, 2019

20 < 1% match (publications)
"Releasing Systems in Active Food Packaging", Springer Science and Business Media LLC, 2022

21 < 1% match ()
"Antioxidant, cell-protective, and anti-melanogenic activities of leaf extracts from wild bitter melon (Linn. var. Ser) cultivars", Springer, 2014

22 < 1% match (publications)
Sajed Amjadi, Hadi Almasi, Ali Ghadertaj, Laleh Mehryar. " Whey protein isolate-based films incorporated with nanoemulsions of orange peel ( ) essential oil: Preparation and characterization ", Journal of Food Processing and Preservation, 2021

23 < 1% match (publications)
Sylwia Członka, Anna Strąkowska, Agnė Kairytė, Arūnas Kremensas. "Nutmeg filler as a natural compound for the production of polyurethane composite foams with antibacterial and anti-aging properties", Polymer Testing, 2020

24 < 1% match (Internet from 14-Oct-2021)
https://riunet.upv.es/bitstream/handle/10251/163234/Thesis%20-%20Elisa%20F.%20Ribeiro_versi%c3%b3n%203.pdf?isAllowed=y&sequence=1

25 < 1% match (publications)
weiqing lan, Yanan ZHAO, Jiali LIU, Jing XIE. "Effects of chitosan-grafted-phenolic acids coating on quality and microbiota composition of vacuum packaged sea bass (Lateolabrax japonicus) fillets during chilled storage", Journal of Food Protection, 2022

26 < 1% match (publications)
Franziska Grzegorzewski, Sascha Rohn, Lothar W. Kroh, Martin Geyer, Oliver Schlüter. "Surface morphology and chemical composition of lamb's lettuce (Valerianella locusta) after exposure to a low-pressure oxygen plasma", Food Chemistry, 2010

27 < 1% match (publications)
Bai-You Cheng, Guey-Shin Shyu, Shi-Ching Wu, Hsiao-Hsien Lin, Chia-Hsuan Hsu, Ben LePage, Wei-Ta Fang. "Fragmented Riverine Habitats in Taiwan Have Spatio-Temporal Consequences, Re-Distributing Caprimulgus affinis into Urban Areas Leading to a Human–Wildlife Conflict", Sustainability, 2019

< 1% match (publications)

28 Yanyu Hu, Shen Yang, Yucang Zhang, Linfan Shi, Zhongyang Ren, Gengxin Hao, Wuyin Weng. "Effects of microfluidization cycles on physicochemical properties of soy protein isolate-soy oil emulsion films", Food Hydrocolloids, 2022

29 < 1% match ()
Pankaj, Shashi. "Cold Plasma Treatment of Biodegradable films and smart packaging", Dublin Institute of Technology, 2015

30 < 1% match (Internet from 28-Jan-2022)
https://www.nature.com/articles/s41598-021-98074-w?code=16b3fbb6-f71e-4000-ad93-10e00bd0ec67&error=cookies_not_supported

31 < 1% match (publications)
Bahareh Rahmani, Hedayat Hosseini, Mohammadreza Khani, Mehdi Farhoodi et al. "Development and characterisation of chitosan or alginate-coated low density polyethylene films containing Satureja hortensis extract", International Journal of Biological Macromolecules, 2017

32 < 1% match (publications)
Sedigheh Farzaneh, Mohammadali Shirinbayan. "Processing and Quality Control of Masks: A Review", Polymers, 2022

33 < 1% match (publications)
Yingying Han, Miao Yu, Lijuan Wang. "Physical and antimicrobial properties of sodium alginate/carboxymethyl cellulose films incorporated with cinnamon essential oil", Food Packaging and Shelf Life, 2018

34 < 1% match (Internet from 20-Jul-2016)
https://issuu.com/erdenetsetsegerka/docs/food-science-and-technology-intern_20110225_061429

35 < 1% match (student papers from 26-Aug-2020)
Submitted to National Chung Hsing University on 2020-08-26

36 < 1% match (publications)
Sepideh Amirabadi, Jafar Mohammadzadeh Milani, Farshad Sohbatzadeh. "Application of dielectric barrier discharge plasma to hydrophobically modification of gum arabic with enhanced surface properties", Food Hydrocolloids, 2020

37 < 1% match (Internet from 01-Apr-2022)
https://mdpi-res.com/d_attachment/horticulturae/horticulturae-08-00019/article_deploy/horticulturae-08-00019.pdf

38 < 1% match (publications)
"Bio-based Packaging", Wiley, 2021

39 < 1% match (publications)
Bu-Seog Ju, Bub-Gyu Jeon, Sung-Wan Kim, Ho-Young Son. "Validation of Seismic Performance of Stainless Press-to-Connect Piping System under Cyclic Loadings", Applied Sciences, 2022

40 < 1% match (publications)
Sajad Shokri, Karim Parastouei, Maryam Taghdir, Sepideh Abbaszadeh. "Application an edible active coating based on chitosan- Ferulago angulata essential oil nanoemulsion to shelf life extension of Rainbow trout fillets stored at 4 °C", International Journal of Biological Macromolecules, 2020

41 < 1% match (Internet from 17-Feb-2022)
http://dspace.nuft.edu.Ua/bitstream/123456789/37001/1/UKRAINIAN%20FOOD%20JOURNAL%202021%20V.10%20Is.4.pdf

42 < 1% match (Internet from 26-Jun-2018)
https://upcommons.upc.edu/bitstream/handle/2117/105811/TMGGI1de1.pdf;jsessio

43 < 1% match (Internet from 03-Mar-2022)
https://www.dovepress.com/mussel-inspired-gold-nanoparticle-and-plgal-lysine-g-graphene-oxide-co-peer-reviewed-fulltext-article-IJN;

44 < 1% match (Internet from 24-Nov-2021)

https://www.researchgate.net/publication/350493807_Preparation_and_characterization_of_clove_essential_oil_loaded_nanoemulsion_and_gelatin_based_edible_film

45 < 1% match (publications)
Antonio Soldo, Victor Aguilar, Marta Miletić. "Macroscopic Stress-Strain Response and Strain-Localization Behavior of Biopolymer-Treated Soil", Polymers, 2022

46 < 1% match (publications)
Bo Young Chung, Ji Young Um, Jin Cheol Kim, Seok Young Kang, Min Je Jung, Hye One Kim, Chun Wook Park. "Anaphylaxis: Five Years' Experience in the Emergency Rooms of Five University Hospitals in Korea", Medicina, 2020

47 < 1% match (publications)
Manuel Viuda-Martos, Abd El-Nasser G. S. El Gendy, Esther Sendra, Juana Fernández-López et al. "Chemical Composition and Antioxidant and Anti-Listeria Activities of Essential Oils Obtained from Some Egyptian Plants", Journal of Agricultural and Food Chemistry, 2010

48 < 1% match (publications)
Seyedeh-Maryam Hasheminya, Jalal Dehghannya. "Development and characterization of novel edible films based on Cordia dichotoma gum incorporated with Salvia mirzayanii essential oil nanoemulsion", Carbohydrate Polymers, 2021

49 < 1% match (Internet from 26-Nov-2018)
http://amsdottorato.unibo.it/7633/1/Tappi_Silvia_tesi.pdf

50 < 1% match (Internet from 28-Mar-2016)
http://cdn.aphca.org/dmdocuments/PAP_13_SAADC2013%20Proceedings_130901.pdf

51 < 1% match (publications)
"Pectin: Technological and Physiological Properties", Springer Science and Business Media LLC, 2020

52 < 1% match (publications)
Sabina Galus. "Functional properties of soy protein isolate edible films as affected by rapeseed oil concentration", Food Hydrocolloids, 2018

paper text:

polymers Article Cold Plasma-Based Fabrication and Characterization of Active Films Containing Different Types of Myristica fragrans Essential Oil Emulsion Bara Yudhistira 1,2 , Andi Syahrullah Sulaimana 3 , Fuangfah Punthi 1, Chao-Kai Chang 1 , Chun-Ta Lung 1, Shella Permatasari Santoso 4,5, Mohsen Gavahian 6 and Chang-Wei Hsieh 1,7,* 1 Department of Food Science and Biotechnology, National Chung Hsing University, Taichung City 40227, Taiwan; barayudhistira@staff.uns.ac.id (B.Y.); fuangfahp3@gmail.com (F.P.); kai70219@nchu.edu.tw (C.-K.C.); as920227@gmail.com (C.-T.L.) 2 Department of Food Science and Technology, Sebelas Maret University, Surakarta City 57126, Indonesia 3 Department of Agro-Industrial Technology, Universitas Gadjah Mada, Yogyakarta 55281, Indonesia; andisyahrullahs@mail.ugm.ac.id 4 Department of Chemical Engineering, Widya Mandala Surabaya Catholic University, Surabaya 60114, Indonesia; shella@ukwms.ac.id 5 Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 10607, Taiwan 6 Department of Food Science, National Pingtung University of Science and Technology, Pingtung 91201, Taiwan; mg@mail.npust.edu.tw 7 Department of Medical Research, China Medical University Hospital, Taichung City 40402, Taiwan ?????????? * Correspondence: welson@nchu.edu.tw; Tel.: +886-4-22840385 (ext. 5010) ??????? Citation: Yudhistira, B.; Sulaimana, Abstract: Myristica fragrans essential oil (MFEO) is a potential active compound for application as A.S.; Punthi, F.; Chang, C.-K.; Lung, an active packaging material. A new approach was developed using a cold plasma treatment to C.-T.; Santoso, S.P.; Gavahian, M.; incorporate MFEO to improve the optical, physical, and bacterial inhibition properties of the film. Hsieh, C.-W. Cold Plasma-Based The MFEO was added as coarse emulsion (CE), nanoemulsion (NE), and Pickering emulsion (PE) at Fabrication and Characterization of different concentrations. The PE significantly affected ($p < 0.05$) the optical, physical, and chemical Active Films Containing Different properties compared with CE and NE films. The addition of MFEO to low-density polyethylene Types of Myristica fragrans Essential (LDPE) film significantly reduced

38**water vapor permeability (WVP) and oxygen permeability (OP**) Oil Emulsion. **Polymers**

2022, 14, 1618. and showed marked activity against E. coli and S. aureus ($p < 0.05$). The release rate of PE films after https://doi.org/10.3390/ 30 h was 70% lower than that of CE and NE films. Thus, it can be concluded that the fabrication of polym14081618 active packaging containing MFEO is a potential food

packaging material. Academic Editor: Ana Beltrán Sanahuja Keywords: active film; cold plasm; emulsion; essential oil; Myristica fragrans

39**Received: 22 March 2022 Accepted: 11 April 2022 Published**: 16 **April 2022**

1. Introduction **Publisher's Note: MDPI stays neutral**

20**Active film is a** new form **of food packaging**

technology that includes functional with regard to jurisdictional claims in published maps and institutional affil- additions to the packaging, such as antioxidants or antibacterial functions [1]; these are iations. distinguished by the slow

20**release of** biologically **active** molecules **into the food**

matrix over a long storage time [2].

20**Essential oils (EOs) are** defined as **volatile compounds**

, with preservation effects and several health benefits that can be isolated from plant materials through various techniques [3]; these are used in active packaging and have the status of in traditional medicine as an antibiotic, antioxidant, and antithrombotic agent [5]. active compounds in M. fragrans essential oil (MFEO) are sabinene, α-pinene, β-pinene, distributed limonene [6], myristicin, and safrole and it contains high phenolic content [7]. Moreover, conditions of the Creative Commons it has bacteriostatic properties [8]. Previous research by Balakrishnan et al. [9] showed eugenol, isoelemicin, isoeugenol, methoxy eugenol, myristic acid, and myristicin from MFEO can be used for the fabrication of silver nanoparticles against MDR (multidrug- 4.0/). resistant) Salmonella enterica. MFEO has bacteriostatic properties as evidenced by very low Polymers 2022, 14, 1618. https://doi.org/10.3390/polym14081618 https://www.mdpi.com/journal/polymers concentrations that can inhibit bacteria and yeasts including Arizona, Salmonella, Morganella, Entrobacter, Escherichia coli, Klebsia pesudomon, Pseudomonas aeruginosa, Staphylococcus aureus, Candida albicans, Cryptoccus neoformans, Aspergillus flavous, Tericophyton verruco, and Epider- mophyton floccodum. The highest inhibition effectiveness was on Escherichia coli. Based on a previous study, storage of Bovine loin (Longissimus dorsi) coated with an active film with polyvinyl alcohol, gelatin, and MFEO was shown to suppress the increase in total volatile nitrogen base and peroxide value and maintain color parameters, which in turn can extend the shelf life of meat [10]. It can be used as a potential candidate for inclusion in packaging. According to Liu et al. [11], EOs have volatile compounds that readily evaporate during film formation and storage. The major challenges in incorporating EO into films are the development of poor miscibility and transparency, phase separation in the film production process, and the

5**sensitivity of bioactive compounds** to **environmental factors**

[4]. Nanoencapsulation can cover the odor and taste of EOs and can mitigate the effect on food sensory properties and provide an effective distribution of the EO release properties [12].

5**Biopolymer-stabilized emulsions**, referred to **as Pickering emulsions** (PEs), **are interesting**; the advantages **of**

PEs include the increased stability to coalescence, increased load capacity, enhanced protection of the encapsulated component, and decreased release rate [2]. In a previous study, it was proven that the addition of PEs to a chitosan film increased the oxygen barrier property [13]. PEs have many advantages and are potential materials for use in packaging development to improve the function of packaging, particularly the protective effects and the release of active compounds. EOs have drawbacks, including high volatility and sensitivity to oxidation, light, and thermal decomposition [14]. For this reason, efforts are needed to improve the application of EOs; one method uses different wall materials, including biopolymers. For the core material of PEs, protein complexation with polysaccharides can be used. Protein matrix filler agents can use carbohydrates that can protect active compounds [15]. In a previous study, the encapsulation of marjoram EO using inulin (a polysaccharide) with whey protein isolate in pectin film showed

51**good mechanical and water barrier properties due to their highly dense and less permeable structure**

[16]. Cold plasma is an emerging technology that has been used for several purposes, including microbial inactivation and removal of hazardous chemicals [17]. It is an efficient, cost-effective, and ecologically acceptable means of substituting pollutants and dangerous coating techniques [18]. However, the application of this technology to develop essential oi-based packaging is a newly developed topic. The studies on active packaging prepared by cold plasma treatment use free EO or coarse emulsion (CE). Those studies are related to EOs in film, such as oregano EO [11], lemongrass EO [19], and marjoram EOs [16]. In addition, the use of active components from other natural sources has been investigated, including the use of cellulose nanofibers and filmogenic soy protein, which can improve the mechanical properties of the film [20] and

28**the physicochemical properties of soy protein isolate-oil emulsion films**

are affected by oil droplets and the heating temperature of soy protein isolate [21], edible coating of whey protein isolate nanofibers and carvacrol showed antibacterial activity that can maintain the quality of salted duck egg yolk [22]. Furthermore, other active components that were investigated are exopolysaccharides from Lactobacillus plantarum, which show inhibitory properties of α-glucosidase and α-amylase and have antioxidant activity [23]. According to the above, the application of the PE system to packaging prepared by cold plasma treatment is still rare. The addition of EO to the film is expected to increase the biological activity which necessitates research regarding the effect of adding external materials to improve film properties [24]. In this sense, the aim of this work was to use cold plasma to develop an active packaging containing MFEO and to characterize the properties of CEs, NEs, and PEs of MFEO-loaded LDPE films and to evaluate the effect of MFEO addition on the optical properties, physical properties, microbial inhibition characteristics, and release properties of LDPE films. 2. Materials and Methods 2.1. Materials Myristica fragrans essential oil (MFEO) with a 95% purity was purchased from Pulau Pinang, Malaysia. The

1**Staphylococcus aureus (S. aureus) strains 328 (BCRC 15211**) and **Escherichia coli** (E. coli **O1:K1:H7) strains NCTC 9001 (BCRC 10675**

)

1**were purchased from Bioresource Collection and Research Centre (BCRC) of the Food Industry Research and Development Institute in Hsinchu, Taiwan**

. Chemical reagents were obtained from Sigma- Aldrich (St. Louis, MO, USA). 2.2. Preparation of Emulsion CEs, NEs, and PEs were used in this study with MFEO concentrations of 1%, 3%, and 6%. The CEs and NEs were prepared using the

5**method of Noori et al**. [25]. The CE **was**

prepared by the gradual addition of MFEO (

5**1% wt) and Tween 80 (30% of** MFEO) into **distilled water with** stirring **at**

314.16 rad/s. Nanoemulsification used a sonicator (Sonopuls HD 4200, Bandelin, Berlin, Germany) to produce NEs operating at 20 kHz and 200 W. The method of Hosseinnia et al. [15] was used to prepare WPI/inulin-stabilized PE. A ratio of 1:1 for

5**WPI (2% wt) and inulin (2% wt) in distilled water was** used as **the**

biopolymer suspension. Then, 1 g of MFEO was added slowly to the biopolymer dispersion with stirring at 523.60 rad/s

5**to obtain** a **pre-emulsion with** a **core-coating ratio** of **1:4**

. Ultrasonication was conducted

34**for 20 min at room temperature** to **the** prepare PE, which **was then**

dried using a freeze dryer (FD50-6S-S, Kingmech, New Taipei, Taiwan) at 0.007 atm and −40 ◦C for 48 h. The PE was stored in an airtight container that was dark in color and stored in a refrigerator. Dynamic light scattering was used to assess the diameter and polydispersity index (PdI) of droplets (Zetasizer Nano ZS-90, Malvern Instruments, Worcestershire, UK) [19]. 2.3. Preparation of LDPE-Treated Film LDPE-treated film was prepared using the method of Wong et al. [26]. The films were first cut into 7 × 9 cm and cleaned using 75% alcohol. A

1**vacuum plasma reactor** uses a **cold radiofrequency plasma (13.56 MHz**

) (Model 1000W, Junsun Tech Co., New Taipei, Taiwan) and a pressure of 0.0643 Torr. The film was treated

26**for 60 s at a power of** 30 **W**

. After the nanocarriers were mixed using a magnetic stirrer (15 min) [16], the CE, NE, and PE

1**solutions (1 mL) were then evenly coated on plasma-treated LDPE films**

. The solution was spread evenly over the entire surface of the LDPE film using a glass tu stick triangle dish and dried for approximately 24 h. The control was an LDPE film without plasma treatment and without coating of MFEO [27]. 2.4. Optical Properties The method of Mendes et al. [19]

22**was used to determine the** optical characteristics (**L, a**, b, **and** yellowness index (YI)) **of the film**

using a color difference meter (ZE6000, Nippon Denshoku Co., Tokyo, Japan). The following formulas were used: ΔE = ΔL2 + Δa2 + Δb2 (1) √ YI = (142.86 × b)/L (2) Film opacity was determined using a UV-visible spectrophotometer (CT- 8600, Chrom Tech, Taipei, Taiwan) at an absorbance of 600 nm [28]. The water contact angle (WCA)

34**was measured in accordance with the method of** Grzegorzewski **et al**. [29] **and**

was determined using a water contact angle instrument (Si-plasma CAM-120, Creating Nano Tech., Tainan, Taiwan).

26**Drops of** liquid **distilled water were dispersed on the adaxial surface of each** film **using a** microliter **pipette**

. WCA analysis was performed in five different measurement positions for each film. 2.5. Physical and Mechanical Properties Film thickness was measured to the closest 0.001 mm using a hand-held digital micrometer (Mitutoyo 293-185-30 Quantumike, Digimatic, Elgoibar, Spain). The texture profile analyzer (TA-XT2i, Stable Micro Systems Ltd., Surrey, UK) detected the film's tensile strength (TS).

33**A load cell of 500 N** and **a gauge length of** 100 **mm**

were used in line with the ASTM D882-91 standard test procedure [30]. Shearing strength measurements

49**were performed** on **a texture** profile analyzer (**TA**-XT2i, **Stable Micro Systems** Ltd., **Surrey, UK) with a load cell** of 40 N **and a**

gauge length of 80 mm. Tensile and shear strength were determined using three replications of the sample and analysis. Tensile strength =

1**F/(W × D**) (3) **where F is the force** required to break **the** film (**N), W** denotes **the** film **width (mm), and D** denotes **the** film **thickness (mm**). The **film**

17**water vapor permeability (WVP) was determined** using **the ASTM E96** standard. The **film** was **placed in** an **aluminum** cell **containing silica gel and placed in a desiccator containing distilled water (100% RH**, 30 ◦C). The **aluminum cells were weighed** from day **0**

to day 10 to ensure steady-state permeation [31]. The following formula was used: WVP = ∆g/∆

8**t (x/A × ∆P**) (4) **where** ∆g/∆t **is the rate of weight change (g/h), x is the film thickness (mm), A is the permeation area (0.0032 m2), and** ∆P **is the partial pressure difference** of **water vapor saturation across the film (4244.9 Pa at 30 ◦C**

). The deoxidizer absorption method was used to measure the film oxygen permeability (OP). The bottle was filled with a deoxidizing agent (3 g) and the

6**film was** placed **on the bottle and** closed. **The bottles were** weighed before being **put** in **a desiccator at**

a temperature of 23 ◦C with 75% RH for 48 h [32]. The following formula was used: OP = (∆

6**m × d)/(A × t × P**) (5) **where** ∆m **is the weight variation** in **the test bottle** (kg), **d is the thickness of** the **film** (m), **A is the effective area of** the **film** (m2), **t is the time interval** (s), and **P is the partial pressure difference of oxygen on both sides of the**

film (Pa).

13**An attenuated total reflection Fourier transform infrared** spectroscopy (**Thermo Nicolet 6700 ATR-FTIR**

, Thermo Fisher Scientific, Taichung, Taiwan) was

50**used to determine the chemical composition of the**

film surface [30]. A

1**scanning electron microscopy (SEM) (JEOL JSM-7800F Prime Schottky**

) was performed for the evaluation of film morphology [26]. 2.

10**6. Antioxidant** Properties **The DPPH (2,2-diphenyl-1-picrylhydrazyl) method was used to measure the** antioxi- dant **activity**

of the films [33] with slight modifications [26]. A total of 35 mg of film was dissolved in 3 mL of distilled water, and then 1 mM of DPPH methanol solution was added and reacted for 30 min in the dark. The following formula was used: DPPH scavenging (%) = (absDPPH − absextract)/absDPPH) (6) The total phenolic content (TPC) was measured by weighing 100 mg of film and soaking it in 10 mL of distilled water prior to incubation for 3 h [34]. Then, 2 mL of the supernatant was analyzed using the Folin–Ciocalteu method. The solution was homog- enized and determined using a UV-visible spectrophotometer (CT- 8600, Chrom Tech, Taipei, Taiwan) at 725 nm. The TPC was expressed in mg gallic acid equivalent per 100 g sample [35]. 2.7. Antibacterial Assay

43**Staphylococcus aureus (S. aureus) and Escherichia coli (E. coli) were used to** examine **the** inhibitory effect **of** the film on **bacterial** growth. The cells **were**

cultured by

inoculating 100 µL from 10 mL of nutrient broth pre-cultures and incubated

1for 16–18 h. Each bacterial culture was adjusted to a cell concentration of 105 CFU/mL using Mueller–Hinton broth. Each film was dipped into 10 mL of cell suspension in the

microbe and shaken at 150 rpm. Finally, 100 µL of the suspension

13was placed on plate count agar and stored at 37 ◦C for 24 h

[36]. 2.8. Release Properties The MFEO release was measured from a film (2 × 2 cm2) inserted into a vial containing 95% ethanol (10 mL) and the vials were placed

19in the dark at room temperature for 34 h. Then, 1 mL of solution was

examined using a UV-visible spectrophotometer (CT-8600, Chrom Tech, Taipei, Taiwan) at 329 nm (with slightly modification) [1,16].

18The absorbance of the sample was compared with the total absorbance of the

film until a constant absorbance was reached, indicating that all active compounds were released to the simulant. The following formula was used: Release rate (%) = abstotal − abssample /abssample (7) ( )

11The Higuchi and Korsmeyer–Peppas equations were used to fit cumulative release

data over time to determine the EO's release kinetics. The following formula was used: Higuchi : M∞ = K1t Mt 1/2 (8) Korsmeyer − Peppas : MM∞t = K2tn (9) where Mt/

2M∞ is the percentage of EOs released at time t; k1 and k2 are constant characteristics of the bioactive-polymer system; n is the diffusion index, which denotes the parameters relating to the mechanism

of release. 2.9. Statistical Analysis The mean ± standard deviation is shown for all data. The statistical analysis was calculated using SPSS (version 20) software

13with one-way ANOVA and Duncan's multiple- range test (significance p < 0.05

). All parameters used three replicates for sample and

1analysis. 3. Results and Discussions 3.1. Emulsion Properties The

MFEO concentrations were maintained at constant levels (1%, 3%, and 6%); based on Shokri et al. [37], 3% of Ferulago angulata EO had good bacterial inhibition but poor antioxidant activity. Therefore, in this study, an attempt was made to increase the concentration of EO. As shown in Table 1, the CE droplet size was larger and significantly different from NE and PE ($p < 0.05$). It can be seen that the CE treatment has the same subset as the NE and PE treatments. The droplet size for all samples is in the range of 170.93–1731.23 nm, where NE and PE have droplet sizes below 250 nm. Emulsion stability in a film matrix is indicated by the small droplet size. Furthermore, it can reduce the rate of droplet aggregation, flocculation, and coalescence [11]. In addition, NE has the advantage of more stability and more transparency [16] based on the zeta potential parameters (Table 1) for all samples are in the range of 8.47–40.80 mV. The difference in the type of emulsion and the concentration of MFEO had a significant effect on the zeta potential ($p < 0.05$). In the CE treatment, there was a significant difference in the level of each different concentration ($p < 0.05$), while in the NE and PE treatments, the relative zeta potentials were not so different. In a previous study, increasing the concentration of Grammosciadium ptrocarpum Bioss. EO (GEO) decreased the zeta

potential [1] and the NE has a lower zeta potential than the CE [25]. Zeta potential estimates the interaction and surface charge characteristics at the molecular level, where an emulsion with a potential zeta value of ≤−30 mV or ≥+30 mV provides emulsion stability [38]. In a previous study, ultrasonic treatment of 300 W for 20 min on microgel particles can provide the highest potential. This is because ultrasonic treatment causes a redistribution of charged chains on the particle surface so that the interfacial and internal structures change [39]. PE can be stabilized by whey protein isolate nanofibrils where oil droplets can be dissolved and wrapped by whey protein isolate nanofibrils to

> 12**prevent oil droplets from coalescing. This** is because **fibrillation**

increases the zeta potential and flexibility of whey protein isolate nanofibrils, allowing

> 12**them to self-assemble at the oil-water interface to form a layer with higher electrostatic repulsion and the stretched structure can have more intermolecular hydrogen** bonds **and Van** der Waals **force**, enhancing **the** interface layer's **rigidity**

and preventing oil droplet coalescence, stabilizing the PE [40]. In this study, it was shown that the stable emulsion was only in CE 1% and CE 3% samples. This is possible because the power and time of ultrasonication in the production of each emulsion use the same power and time, so it is necessary to optimize the power and time of ultrasonication to obtain an emulsion with a zeta potential that leads to a stable emulsion. Table 1. The droplet

> 22**size, zeta potential, and polydispersity index of** MFEO emulsion **prepared** with different concentration **and**

emulsion type. Emulsion Droplet Size (nm) Zeta Potential (mV) Polydispersity Index CE 1% 1533.15 ± 0.31 a 40.80 ± 0.25 a 0.66 ± 0.37 a CE 3% 1615.70 ± 0.13 a 38.38 ± 0.29 b 0.81 ± 0.32 a CE 6% 1731.23 ± 0.24 a 26.54 ± 0.58 c 0.93 ± 0.11 a NE 1% 170.93 ± 0.11 b 8.47 ± 0.26 d 0.22 ± 0.01 c NE 3% 197.46 ± 0.52 b 12.10 ± 0.01 c,d 0.38 ± 0.02 b NE 6% 155.60 ± 0.10 b 11.13 ± 0.03 c,d 0.25 ± 0.02 c PE 1% 244.76 ± 0.86 b 21.30 ± 0.26 c 0.39 ± 0.01 b PE 3% 236.98 ± 0.07 b 18.34 ± 0.10 c,d 0.29 ± 0.04 c PE 6% 233.56 ± 0.11 b 19.43 ± 0.08 c,d 0.32 ± 0.04 b,c a–d The values

> 25**in the table** are **the** average ± **standard** error of **n = 3** samples and **the**

> 14**different lowercase letter in each column indicate significant differences (p < 0.05**

). CE: coarse emulsion; NE: nanoemulsion; PE: Pickering emulsion. As shown in Table 1, the PdI parameters in the CE treatment resulted in a significant difference compared with

> 18**NE and PE (p < 0.05**). In **the** CE treatment, **the**

PdI did not differ significantly for different MFEO concentrations, while the PE treatment had the lowest PdI, especially at 3% and 6% MFEO concentrations. In a previous study, the PdI of ginger EO emulsion at NE (0.222) was lower than at CE (0.584). Ultrasonication produces a lower PdI in NE and a uniform particle size [25]. Emulsions that have a PdI lower than 0.3 indicate a stable emulsion and have uniformity of emulsion droplets [16]. 3.2. Optical Properties

> 9**As shown in Table 2, the** L **value of** the treated film **was significantly lower than that of** the control film (**p < 0.05**

). In the color parameters, especially L and a, it can be seen that the film with PE 6% and NE 6% treatment had the highest

> 19**value and was significantly different from the** film with **other treatments**

. In addition, the increase in MFEO concentration significantly increased the red–green (a) and blue–yellow (b) values ($p < 0.05$) (Table 2). An increase in EO causes the film to become more yellow. The ΔE in all the treated films was less than 2, indicating only a slight difference in film appearance [19]. PE films of marjoram essential oil (MEO) had higher ΔE than NE films [16], this demonstrated that emulsion type and concentration affect the color of films. The YI

> 44**increased as the** MFEO concentration **increased** and **was higher than the**

control film ($p < 0.05$) (Table 2). This shows something similar to the color parameter at the b value. In this study, in PE films the increase in yellowness was due to the natural bright yellow color of the whey protein isolate. This is because the type and concentration of EOs, as well as the presence of additives, directly affect the color of the emulsified film [16]. Table 2. Optical characteristics of LDPE film-treated cold plasma-stabilized MFEO. Emulsion L a b Δ E YI Opacity WCA Control CE 1% CE 3% CE 6% NE 1% NE 3% NE 6% PE 1% PE 3% PE 6% 93.01 ± 0.10 a 27.79 ± 0.16 b,c 24.76 ± 0.55 c 25.20 ± 0.35 c 25.20 ± 0.13 c 31.21 ± 0.65 b 30.95 ± 0.21 b 27.05 ± 0.55 b,c 26.72 ± 0.49 b,c 31.24 ± 0.02 b 0.10 ± 0.16 a −6.76 ± 0.16 d - −10.38 ± 0.16 e 0.45 ± 0.16 f 4.88 ± 0.01 e −11.80 ± 0.19 e 2.35 ± 0.18 c 1.70 ± 0.16 12.10 ± 0.18 d 1.05 ± 0.04 e 6.22 ± 0.01 d −10.09 ± 0.09 c 2.88 ± 0.88 c 0.08 ± 0.88 16.40 ± 0.49 c 1.07 ± 0.05 e 7.63 ± 0.01 b −9.47 ± 0.15 b 3.15 ± 0.61 c 0.35 ± 0.31 17.89 ± 0.35 b,c 1.06 ± 0.15 e 7.73 ± 0.01 b −10.78 ± 0.18 d 4.22 ± 0.29 b 0.31 ± 0.04 23.92 ± 0.08 b 1.10 ± 0.05 d 4.94 ± 0.05 d,e −11.01 ± 0.50 d 6.58 ± 0.40 a 0.49 ± 0.71 30.10 ± 0.66 a 1.15 ± 0.05 c 5.72 ± 0.10 d −12.18 ± 0.07 e 6.77 ± 0.10 a 0.36 ± 0.21 31.24 ± 0.21 a 1.15 ± 0.06 c 8.83 ± 0.09 a −13.70 ± 0.55 g 5.91 ± 0.14 a 0.50 ± 0.22 21.55 ± 0.60 b 1.46 ± 0.47 b 4.66 ± 0.05 e −12.86 ± 0.33 f 4.19 ± 0.69 b 0.40 ± 0.43 22.57 ± 0.55 b 1.44 ± 0.10 b 6.62 ± 0.01 c −11.68 ± 0.12 e 4.71 ± 0.54 b 0.40 ± 0.03 31.42 ± 0.04 a 2.34 ± 0.05 a 8.28 ± 0.07 a a–g The values

> 25**in the table** are **the** average ± **standard** error of **n = 3** samples and **the**

> 14**different lowercase letter in each column indicate significant differences (p < 0.05**

). CE: coarse emulsion; NE: nanoemulsion; PE: Pickering emulsion. In this study, the opacity of all film samples was significantly higher than the control (Table 2). The opacity parameter for films with 6% PE treatment has the highest value and is significantly different from other treatments.

> 41**An increase in the** concentration **of** MFEO **caused an increase** in **the**

opacity. A high opacity indicates that the films have low transparency. The PE films had the largest opacity because there was a matrix formed from inulin and WPI, and the existence of

> 52**an oil phase in the protein matrix** increases **light dispersion and the light-scattering** impact **of** the **oil**

[41]. From Table 2, it also can be seen that an increase in the MFEO concentration causes an increase in the WCA. As in the opacity parameter, the film with PE 6% treatment had the highest WCA

> 19**value and was significantly different from other treatments**

. The incorporation of hydrophobic compounds and plasma treatment increases the surface hydrophobicity [26,27]. However, according to Liu et al. [11], WCA is not only related to surface hydrophobicity but also influenced by porosity and roughness on the film surface. In addition,

> 36**plasma active species** may lower **the surface hydrophobicity by increasing** the number of **polar anchors on the surface**

[42]. Based on a previous study by Wong et al. [43], LDPE films increased in tensile strength parameters due to an

> 36**increase in plasma treatment time**, which was caused **by**

the formation of interfacial roughness and polar groups. The process of coating the

20**active ingredient is** easier **on the surface of the film**

with high roughness. This study also compared the surface morphology of the control LDPE film (without plasma treatment) with that of the LDPE film with plasma treatment. Wherein, the control film has a uniform and smooth surface. Meanwhile, the film with plasma treatment had an increase in roughness due to the plasma treatment producing etched characters with irregularly shaped textures. In another study by Theapsak et al. [30] plasma treatment can modify the surface of the PE film by producing

29**oxygen containing polar** functional **groups** (OH, **C−O, and C=O**) and increasing **the surface** roughness. **Plasma treatment** can increase **the**

binding affinity of MFEO with LDPE film. This is because plasma treatment can cause conformational changes. Based on a previous study, through exposure to hydrophilic groups, plasma treatment can increase the

1**affinity of LDPE film for chitosan**, hence boosting **the** effect **of**

chitosan and gallic acid on LDPE [26,43]. In another study by Loke et al. [27], plasma treatment of LDPE film which was then coated with gallic acid and chitosan showed that the structure of LDPE could not be damaged by plasma, but there was an increase in the

1**affinity of LDPE film for chitosan through exposure** to **hydrophilic**

groups. In addition, collagen has a polarity and function closer to cinnamaldehyde, which has better affinity, and more essential oils are retained in the film. In addition, with the use of Pickering emulsion film, there was a decrease in the hydrophobicity of EO, which led to an increase in its affinity (as a hydrophilic material) with the film [44]. 3.3. Physical and Mechanical Properties The physical and mechanical properties of the LDPE films prepared in this study are presented in

31**Table 3. The physical properties of the**

film examined were shear strength (SS), tensile strength (TS), thickness, WVP, and OP. The SS and TS of the film increased as the MFEO concentration increased; a significant difference was found for PE ($p < 0.05$), greater than the increase observed for the CE and NE films. The SS and TS parameters have the same pattern, where the PE 6% treatment has the highest value and is significantly different from the other treatments. Based on previous studies, plasma treatment can reduce TS in the LDPE films [27]. The TS value in the LDPE film was 18.19 MPa, but after plasma treatment, the TS value decreased to 13.71 MPa. In addition, the increase in plasma treatment power of LDPE film had no effect on TS compared with LDPE film without plasma treatment. However, increasing plasma treatment time had a significant effect on decreasing TS in LDPE films. This phenomenon is caused by increased roughness and the formation of polar groups. In addition, prolonged exposure to plasma causes slower aging effects in samples. Increased surface roughness makes preventive coating easier [43]. In Table 3, it can be seen that CE and NE treatment can reduce the TS of LDPE film, although the NE 6% treatment has an increase in TS. This is consistent with previous studies in which the addition of NE to WPI-based films reduces the tensile strength of the films caused by the plasticizing effect of NE droplets so that it can weaken the intermolecular interactions between polymer chains [1]. In another study, the addition of rosemary essential oil to carboxymethyl cellulose-polyvinyl alcohol blend films resulted in a film with a decrease in film strength. This was attributed to the effect of EO plasticization on the film structure [45]. Table 3.

31**Physical and mechanical properties of** LDPE film-**treated**

cold plasma-stabilized MFEO. Emulsion SS (Mpa) TS (Mpa) Thickness

35**WVP OP** (mm) (×**10−7 g·m**−1·s−**1·Pa−1) (×10−12 g·m·m−2·s−1·Pa−1**

) Control CE 1% CE 3% CE 6% NE 1% NE 3% NE 6% PE 1% PE 3% PE 6% 20.0 ± 0.26 a 15.5 ± 0.26 d 15.5 ± 0.28 d 14.5 ± 0.28 e 14.3 ± 0.01 e 14.9 ± 0.26 d 15.1 ± 0.05 d 17.0 ± 0.28 c 19.4 ± 0.23 b 23.3 ± 0.40 a 18.3 ± 0.25 a 18.6 ± 0.34 a 12.3 ± 0.25 d 11.5 ± 0.28 d 11.3 ± 0.01 d 11.8 ± 0.01 d 18.9 ± 0.05 a 13.4 ± 0.12 c 15.3 ± 0.01 b 18.4 ± 0.02

24a 0.034 ± 0.03 b 0.034 ± 0.01 b 0.034 ± 0.00 b 0.034 ± 0.10 b 0.034 ± 0.01 b 0.035 ± 0.00 b 0.038 ± 0.02 a 0.035 ± 0.01 b 0.039 ± 0.11 a

0.039 ± 0.01 a 2.45 ± 0.01 a 6.16 ± 0.01 a 1.47 ± 0.01 b 4.50 ± 0.01 b 1.41 ± 0.06 c 4.37 ± 0.01 b 1.39 ± 0.01 c 4.42 ±

41 0.01 b 1.46 ± 0.01 b 4.98 ± 0.01 b 1

.45 ± 0.01 b 3.18 ± 0.01 c 1.20 ± 0.01 c 2.37 ± 0.01 d 1.46 ± 0.01 b 3.32 ± 0.01 c 1.52 ± 0.09 b 3.04 ± 0.01 c 1.59 ± 0.20 b 3.05 ± 0.01 c a–e The values

25 in the table are the average ± standard error of n = 3 samples and the

14 different lowercase letter in each column indicate significant differences ($p < 0.05$

). CE: coarse emulsion; NE: nanoemulsion; PE: Pickering emulsion; SS: Shearing strength; TS: Tensile strength; WVP:

33 Water vapor permeability; OP: Oxygen permeability. The LDPE film with PE treatment had a higher

TS value than the CE and NE treatment, but even at PE 6% treatment, it was not significantly different from the control film. Based on previous studies, the addition of PE from SiO_2 nanoparticles and functional oil phase resulted in an increase in the tensile strength of the epoxy composite compared with the reference sample [46]. The addition of cellulose nanofiber to sandalwood oil Pickering emulsion can increase the tensile strength of the film. It is

30 associated with the formation of a rigid continuous network of cellulose nanofibers linked through hydrogen

bonds, and is also attributed

30 to the geometry and rigidity of the nano-filler [47]. The

thickness parameter for the PE treatment shows a significant increase (

50 $p < 0.05$) compared with the CE and NE treatments

(Table 3). The thickness parameter for CE and NE films was not significantly different except for NE 6% films. In the study of Fasihi et al. [45], the increase in thickness after the addition of a PE was proposed to be due to the increased solid material content of the film. This was in accordance with the results of our study, the CE and NE treatments did not have a significant effect on thickness. As

48 shown in Table 3, increasing the concentration of

MFEO reduced the WVP of film ($p < 0.05$). Treatment of CE, NE, and PE films for all MFEO concentrations had lower WVP values than control films. According to Ghadetaj et al. [1] the hydrophobic nature of lipid compounds from MFEO can reduce WVP, where the film with CE treatment absorbs less than the film with NE. This is related to the nano size which can limit the droplet size and reduce its plasticizing effect on the film. Different types of emulsion type result in different WVP values; the CE treatment resulted in a significantly lower value than the NE or PE treatments ($p < 0.05$). The study by Fasihi et al. [45] showed that the addition of 5% PE from rosemary essential oil, can reduce WVP in the carboxymethyl cellulose- polyvinyl alcohol blend film. This is attributed to the PE providing decreased water vapor diffusion due to increased tortuous paths. In a previous study, the use of PE in the chitosan matrix interfered with the formation of hydrogen bonds between chitosan molecules and weakened the chitosan network in the film, which facilitated the migration of water vapor molecules [13]. In Table 3, the control group had a

29**significantly (p < 0.05) higher** OP **than the** treated film. **The**

addition of MFEO decreased the OP of the LDPE film. OP parameters can be seen NE 6% is the lowest OP value. . In another study, cinnamaldehyde increased the oxygen barrier, resulting in a decrease in OP [48], which was possibly a result of the antioxidant properties of cinnamaldehyde, which conferred the oxygen-capturing capacity [49]. The PE film can reduce the OP parameter, it can be seen that PE film has a lower OP than the control film. In a previous study, the loading of zein/chitosan-stabilized PEs increased the oxygen barrier properties of the films. This is because the zein/chitosan-stabilized PEs film has a thick and firm network so that it can act as a natural barrier to oxygen. In addition, the increase in the barrier properties of the chitosan film was related

28**to the** delicate **interactions between** the **Pickering emulsion and** the chitosan **matrix** [13]. **The** infrared spectra **of the films**

shown in Figure 1 indicate the specific functional groups and their vibrational modifications after cold plasma treatment. This analysis was performed only on films treated with 3% MFEO. FTIR showed that there were differences between control films and plasma-treated films including films that were coated with CE, NE, and PE. In the film, with the addition of MFEO, new peaks ap- pear and can be seen in the band between 800 and 1800 cm−1. The CE and NE films have quite similar trends because the two emulsions only have size differences, while for PE films there are significant differences (Figure 1). The film with MFEO coating shows the addition of a peak, thus indicating that the active compound from MFEO was successfully added to the LDPE film. The MFEO spectrum contained a high num- ber of peaks, indicating the existence of a variety of volatile compounds and functional groups. In a previous study, Silva-Damasceno et al. [50] found that

4**sharp peaks of MFEO** consisted of peaks **at 775**

and 824 cm−1 (

4**aromatic–CH bending), 922** cm−1 (–**OH** bend- ing), **1037**

cm−1 (–

4**CH2 group vibration), 1238** cm−1 (**carbonyl vibration), 1507** cm−1 (–**CH bending), 1715** cm−1 (**C=C stretching at aromatic groups), 2920** cm−1 (–**CH** stretch- ing), **and 3455** cm−1 (–**OH stretching**). The **incorporation** of MFEO **into** the **matrix led to minor changes in** the wavenumber **of different peaks, as well as increased peak** in- tensity **at 1107** cm−1 (**P=O), 1418** cm−1 (–CH **stretching), 1554** cm−1 (–NH **bending), and 2920–3411** cm−1 (**broadening of–CH stretching**

). The flavonoid functional groups corre- spond to aromatic ring vibrations [51]. Functional groups of flavonoids

23**are located in the range of 1610–1600 cm−1 and 1480–1450 cm−1**

[52]. Based on previous studies, the plasma-treated gallic acid film resulted in hydroxyl stretching of the film where the –OH bond showed a radical scavenging capacity which may have antioxidant properties [26].
PoPloylmymeresr2s022022,21,41,4x, 1F6O1R8 PEER REVIEW FiFgiugruer1e.1. ATR–FTIRofoLfDLDPEPEfilfimlmtrteraetaetdedwwithithcoclodldplasma containingCEC-E,-N,NE-E,-a,nadnd PE-stabilized MMFEFOEO.. InInaa previous study related to the FT-IR spectrum ofopfolyvinyl alcaolhcoohloalndancdar- carboxymethyclellulose filmfislmcosmcboimnebdinwedithwdiitfhferdeniftfecroennctenctornactieonntsraotfiornossemofarryoseesmseanrtyial esosieln,ttihael oIiRl,
tshpeecIRtruspmecptrauttmerpnawttearsnnwoatssnigontisfiigcanniftilcyandtilfyfedriefnfetr,einntd,iicnadtiicnagtinthgatthdautdriunrgintghe thiencorporation of roofsemary esseensstieanltoiaill ionitloinththoe fithlmefmilmatrmixa,ttrhixe,stthruecstutrruecotufrteheofifltmhemfialmtrix mdaitdrixnodtidchnaontgceh[a4n5g].eW[4i5th]. tWheitahdtdhietiaodndoiftiPonE otof tPhEe tfiolmth,ethfielmre,wthaesrae swhaifst aatshthifet paetatkheof p1ea0k37ofcm10−317 ccomm−1pcaormedpawreitdhwCiEthaCndE aNnEd fiNlmEfsi.lmBsa.sBedasoend opnrevious studies, increasing the thceontent ofocfhitosan anadndbabsailsiolioliilninthtehPeoly (L-Lactic Acid) film resulted ininaadecrease in inthe characteristic peak of the FT-IR plot, possibly due to the film's low transmittanceinin thtehe wave numberranragnegoef of 1050–13c0m0−c1m.−1. FurthermnooreP,onlyo(PLo-Llyac(Lti-

cLAaccitdic) Apecaidk)shpiefatk wsahsifotbwsearsvoebdseinrvPedoliyn(PLo-Llyac(Lti-cLAacctiidc)A-cchiidto)-scahnitoorsaPnoolyr P(Lo-lLya(cLt-iLcactic Acid)-chitosan-boaislil films. This suggests that the chemical groups PLLA and CS have no interaction [53].

> 32**Polymers 2022, 14, x FOR PEER REVIEW** 12 **of 22** oil films. **This**

suggests that the chemical groups PLLA and CS have no interaction [53]. Figure 1 shows
thatthtehePEPEfilmfilmhashdasiffdeirfefnertepnetapkseafrkosmfrComEaCnEd
NanEdfiNlmEs,fwilmhesr,ewnheewrepenaekws papepakeasraopnpePaEr fiolnmPsEatfi1l5m4s8
aatn1d514082a8ncdm1−012.8Tchmis−i1s. TrehlaisteidsrteolaWtePdI,twohWicPhI,iswuhsiecdhaiss
tuhseedcoares tohfePcEo,rwehoefrPeEp,ewakhe1r5e4p8ecamk−1154in8dcimca−t1eisnNdi–cHatesstrNet–
cHhinsgtreotfcahminigdeofIIaamnidde10II28ancdm1−012s8hcomw−s1 sCh–oNwsstrCe–
tNchisntrgetocfhainmginoefsa.mThinisesi.sTinhiascicsoirndaancccoerwdaitnhcethweiAthlmthaesiAeltmaal.si[1e6t]aslt.u[1d6y],
wstuhdicyh, wshhoiwchedshthoweeemdetrhgeenemceeorfgennecwe poefankeswinpfielmakss
usinfiglmPEs.uTshinugs,PthEe. pTrheusse,nctheeofparedsdeinticoenoafl
padeadkitsioansawlepIleaasksanaisncwreeallseaisnatnheisntcrreenagsteh ionf cthhoesesntrepnegakths
aotfdcifhfoerseennt wpeaavkesnuamtbdeifrfserweanst
wanavinedniucmatiboenrsthwaatsMaFnEiOndwicaastieofnfetchtaivteMlyFlEoOadwedasinetfofetchtievmelaytlroixa.ded
into the matrix. The characteristic of the LDPE film after cold plasma treatment are presented in Figure 2.
This analysis was performed only on films treated with 6% MFEO to determine the differences resulting from
the highest concentration of MFEO. The control films had a smooth surface and the LDPE films treated with
a CCEE aanndd NNEE hhaadd rough surface, whereas tthhee
ffiiIlmmtrtreeaatteeddwwitihthPEPEapappepaeraerde‘dsa‘nsadnyd’,yw’,itwhiftihnefignreanguralensu(lFesig(uFrieg2uare–
d2)a.–Sdu)r.faScuesrfwacitehs
wgriathnuglerasncuanleisnccraenasinecthreearsoeutghhenreosusgohftnheessfilomf.tIhneafiplrmev.ioInusastpurdeyv,iothuesussteudofy,ptehcetin
wpeicthtina ficlomncsewntirtahtiaoncoonfce7n.5t%ratPioEn
ionfcr7e.5a%sedPEthiencrroeuagshednetshseorfouthgehnfielmss.oTfhtihseinficlmre.asTehiins
rinoucrgehanseesinsirsorueglahtneedstsoisthreelmatiegdrattoiothneomfaigggraretigoanteosfoargdgrreogpaletetss
toortdhreotpolpetosfttohtehfeiltmopdoufritnhge ffiiIlmm
ddurryiinnggfi,ltmhedrerybiyngc,atuhseirnegbyscuarufascinegirsruergfauclaeriirtrieesgu[l1a6r]i.tieIsn[1a6].pIrnevaiopursevsiotuudsys,tutdhye,
athpepalipcaptliiocantioofncoolfdcoplldaspmlaasmanadacnidnncainmnaalmdeahldydehe-ycdoen-
tcaoinntinaigniCnMgCCMcoCatcinoagtirnegsurletesudltinedthine
ftohremfoartimonatioofnmoficmroipcroorpeosraensdancdavciatvieistieosnonthtehefifilmlm,,wwhhicichhaalllloowweedd
cciinnnnaammaalldddeehhyyddee ttoo evaporate

> 11**during the drying process. The** surface **of the** modified **film**

when combined evaporate

> 11**during the drying process. The** surface **of the** modified **film**

when combined with polymer compounds can increase EO loading [27]. Plasma treatment can improve with
polymer compounds can increase EO loading [27]. Plasma treatment can improve interfacial adhesion and
polymer matrix compatibility [26]. interfacial adhesion and polymer matrix compatibility [26]. Figure 2. Cont.
PPoollyymmeerrss 22002222,, 1144,, x16F1O8R PEER REVIEW 1312ooff2221 Figure 2. Cont.
PPoollyymmeerrss 22002222,, 1144,, 1x6F1O8R PEER REVIEW Figure 2. Surface
characterizFaigtiuorneo2f. pSularsfmacae-ctrheaartaecdteLriDzaPtEiofnilomfpcloaastmeda-
wtreitahteddifLfeDrPenEtfitylmpecooafteemduwlistihondi(fafe)rCenotnttyrpole(obf)eCmEulsion 6% (c) NE
6% and (d) PE 6%(ao)bCsoernvtreodl u(bn)dCerE160%,00(c0)×NmEa6g%nifaincadti(odn).PE 6% observed
under 10,000× magnification. 3.4. Antioxidant Properties As shown in Figure 3, increasing the MFEO
concentration significantly ($p < 0.05$) increased the total phenolic content (TPC) and antioxidant activity
(DPPH scavenging activity) of the films. TPC and antioxidant activity have a close relationship with EO
concentration, and the phenolic group has an important role in antioxidant activity. MFEO contains

> 23**sabinene, α-pinene, β-pinene**, limonene [6], **myristicin, and safrole** and **it**

contains high phenolic content [7]. Nutmeg essential oil was analyzed using GC-MS and revealed 27
components. myris- ticin,

> 15**terpineol-4, alpha-terpinol, dodecanoic acid, torreyol, palmitin**, and **safrol**

are some of the compounds found in seeds. Whereas the IC50 value of the

15**essential oil of fuli and fruit**

is higher than that of

15**other nutmeg parts such as seeds, roots, and bark. The IC50**

values for mace and fruit essential oils are

15**185,943 ppm and 221,036 ppm, respectively** [54]. **The**

antiox- idant activity of CE incorporated film samples was not significant different ($p < 0.05$) with NE and PE in the same concentration. In this phenomenon, phenolic acids and terpenoids were found to be responsible for the emulsion, including the film's DPPH scavenging properties [16]. The antioxidant activity in this study is stronger than in Shokri et al. [37], maximum antioxidant activity was only 30.17% on film containing 3% CE or NE of Ferulago angulata EO. The NE films displayed

22**fast and efficient free radical absorption**

because the formation of NE results in an increase in the specific surface area [25]. In addition, PE films result in the low mobility of the loaded compounds [55] and their slower release is associated with lower antioxidant activity [1]. The antioxidant properties of EO related to the redactors contained in EO can stop and stabilize radical chain reactions.

47**However, it is difficult to** trace **the antioxidant** activity **of** the whole **EO to one or a few active** molecules **because**

16**both minor and major constituents** must **be taken into** account **to account for its biological**

action [56].

32**Polymers 2022, 14, x FOR PEER REVIEW 15 of 22**

Total phenolic content (m g G AE /100 g) 14 12 10 8 6 4 2 0

42**c c c b b** b **a a a**

(aa)) 100 Control CE1% NE1% PE1% CE3% NE3% PE3% CE6% NE6% PE6%

21**D P P H S ca ve ng**

i ng (%) 80 60 40 (

42**b) a b b** b **a a c c c**

20 0 Control CE1% NE1% PE1% CE3% NE3% PE3% CE6% NE6% PE6% Figure 3. Effectiveness of LDPE film treated with cold plasma containing CE-, NE-, and PE-stabilized superscripts are signifaicsasnaytl.ya–dcifTfehreevnatlauteps <w0it.0h5d.iTffheereenrrtosrupbaerrsscrreipprtsesaernetstihgenisfitacanndtalyrdddifefevrieantitonsp(n<=0.30)5.. The error bars Fpihgeunroeli3c. cEofnfetcetnivt;en(be)ssMaonFftEiLoOxD.idP(aaE)nfTtilomatcatltirvpeihatyteendtohlrwiociutchgohnctotelhdnet;pD(labPs)PmaHnatcirooaxnditidatg,yiNntghEr-a,osuasngadhy.PthaE–ec-sTDthaPbePilHvizaelrudaedMsicFwaElitOshc.ad(vaie)fnfTegroientnagtI represent the standard deviations (n = 3). 3.5. Anti-Bacterial Assay As shown in Figure 4, the MFEO film caused a reduction in E. coli and S. aureus of up to 3.25−4.01 log CFU/mL. In general, there were slight differences in the bacterial inhibition of the CE, NE, and PE treatments, and the reduction in E. coli was greater than that in S. aureus ($p < 0.05$). An increase in the concentration of MFEO led to an increase in bacterial inhibition, which was indicated by the

lower number of bacteria. In addition, MFEO has bacteriostatic properties that can inhibit bacteria and yeasts, including Arizona, Salmonella, Enterobacter, E. coli, Klebsiella pseudomonas, S. aureus, and Aspergillus flavus, although the highest inhibitory effectiveness is against E. coli [8]. In a previous study, S. aureus and E. coli were reduced

1**to 4.61–5.14 log CFU/mL**

after plasma treatment of a cinnamaldehyde coating on LDPE [27]. The inhibition of microbial growth by NE occurs in a variety of ways depending on the encapsulated antimicrobial agents (e.g., EOs, proteins, and surfactants) and the structure of the NE droplets (e.g., composition, charge, and size) [57]. In a previous study, the antibacterial activities of Origanum majorana EO were attributed to α-pinene, γ-terpinene, and sabinene [58]. These compounds improved the permeability and fluidity of fungal cells. Terpenes are considered to cause changes

16**in cell permeability by** penetrating **the fatty acyl chains that** comprise **the membrane lipid bilayers**, altering **lipid packing and** inducing **changes** in the **membrane** functions **and**

properties [1,16]. The increase in anti-bacterial activity depends on the concentration of EO. In addition, the nanoemulsification process in EO can also increase anti-bacterial activity, where the smaller droplet size provides

40**faster penetration of antimicrobial compounds through the** bacterial cell **membrane** [37]. **The**

inhibitory effects of CE and NE treatment were slightly different but were signifi- cantly different from the PE film ($p < 0.05$).

40**Nanoemulsification increased the antibacterial activity of coating solutions**

[37]. The PE films resulted in a significantly slower release than NE, which was the cause of the low antibacterial activity [16]. However, within a short storage time, controlled release, such as NE, may reduce their functional activity [1].

44**Due to the protective** effects **of the stable interfacial** film, **the**

PE exhibited greater antibacterial activity [45] and could release the active compounds for a longer time. 3.6. Release Properties In order to determine the film's possible application, the

2**release capacity and** cor- responding **release mechanism of active** substances **loaded in the film matrix were in**

- vestigated. As shown in Figure 5, there was a slight significant difference in the release properties for all films ($p < 0.05$) after 30 h. In general, the release rate of PE films was lower than that of CE and NE films. After 2 h, similar trends were found for all film samples. This is similar to Ghadetaj et al. [1], who reported that WPI-based films containing NE-loaded EOs had antioxidant activity that was not significantly different from films containing free EOs. The release of compounds loaded in PE films is slower than NE films, resulting in the reduced mobility of loaded compounds [55]. However, over a long storage time, the release of PE is enhanced and causes the retention of their functional activity in a variety of environments [2]. The PE-treated film had lower antioxidant activity than the NE-treated film owing

5**to the slower release of** the **encapsulated MEO** from **the film matrix**

[16].

18**The PE** is **stabilized by the WPI-inulin complex**, which is **absorbed at the oil-water** phase **interface**

, thereby shielding MEO from external factors and inhibiting the coalescence of EO [1]. The decrease in the release rate and anti-bacterial activity in the PE film in the short term is possible due to the decrease in the diffusivity of the MFEO in the film matrix [44]. Polymers 2022, 14, x FOR PEER REVIEW 17 of 22 8 a(a)) 6 Vi abi l i ty (Log CFU/m L)

21**a b b c c** 4 **d d** d **e e**

2 0 Control CE1% CE3% CE6% NE1% NE3% NE6% PE1% PE3% PE6% 8 6 Vi abi l i ty (Log CFU/m L) 4 2 b(

21**b)) a b c** d **c d d d e e**

0 Control CE1% CE3% CE6% NE1% NE3% NE6% PE1% PE3% PE6% Figure 4. EffectivenessFiogfuLreD4P.EEfffielcmtivterneeastesdofwLDithPEcfiolldm ptrleaastmeda wcoitnhtcaoinldinpglaCsmE-a, cNonEt-a,inainndg CPEE--,sNtaEb-i,liaznedd PME-FsEtaOb.ili(zae)d antimicrobial assay agaMinFstESO..a(uar)eaunst,i(mbi)caronbtiimaliacrsosabyiaalgaasisnasyt Sag.aauinresutsE,.(bco)lai.nat–iemTihcerovbaialuleassswayithagdaiifnfestreEn.tcoslui.pae–rescTrhipetvsaalurees significantly different atwpit<h0d.0if5fe.rTehnet seurrpoerrsbcarripstrseaprreesseignntitfihceansttalynddaifrfderdenevtiatpio<ns0.(0n5=. T3h).e

1**error bars represent the standard deviations (n = 3**

). PPoollyymmeerrss 22002222,, 1144,, x16F1O8R PEER REVIEW 100 90 80 70 Rel ese rate (%) 60 50 40 30 20 10 0 CE 1% a CE 3% a CE 6% a NE 1% b NE 3% b NE 6% b b PE 1% c PE 3% c PE 6% a a 0 2 4 6 8 10 12 14 16 18 20 22 24 26 28 30 32 34 Time (hours) Figure 5. Release rate of LDPE film treated with cold plasma containing CE-, NE-, and PE-stabilized MFEO in 95% alcohol.

1**Values with different superscripts are significantly different at p < 0.05. The error bars represent the standard** deviation (**n = 3**

). Higuchi and Korsmeyer–Peppas models were utilized

2**to further** define **the in vitro release** properties **of the active** compounds **in loaded films**

. The correspondence between the actual active compounds released is indicated by the R value close to 1. The previous study by Ritger and Peppas [59], showed that

2**when n > 1** the **composite Case-II transport mechanism**

was valid;

2**when 0.5 < n < 1** the **diffusion behavior** followed **non-Fickian diffusion**; and **when**

nn<<0.50.t5hethdeiffusion behbaevhioarvoiofrthoef atchteivaecitnivgreediniegnretdietnhte ifinlmthmeafitlrmix mfoalltorwixsfoFlilcokwiasnFdicifkfiuasniodni.ffusion. The correlation coefficients of all films generated in this investigation were desirable when applying the Higuchi equation (R2 = 0.9687–0.9912) (Table 4). The release of active compounds

2**in the Higuchi model is based on Fick's laws of diffusion**, with the assumption **that** the matrix's **swelling and dissolution are** minor **or** non-existent, **and** that **the matrix** exhibits **square root time** dependency. **The Korsmeyer–Peppas equation was** also utilized **to** confirm **and explain the**

aforesaid findings [24].

38**Based on the** values **in Table** 4, **it shows that** the opening **of**

all

2films was based on the Fickian diffusion pattern. In addition, the release of

active compounds from the film structure can be encouraged due to the larger concentration difference between the inner and outer environment [24].

11Table 4. Parameters of Higuchi/Korsmeyer–Peppas model

for release properties of LDPE film treated with cold plasma containing CE-, NE-, and PE-stabilized MFEO. Emulsion Higuchi K1 R2 CE 1% 2.5627 0.9727 CE 3% 2.5829 0.9890 CE 6% 2.6465 0.9843 NE 1% 2.8933 0.9809 NE 3% 2.8512 0.9687 NE 6% 2.8522 0.9844 Korsmeyer–Peppas K2 n 1.1136 0.1334 1.0464 0.2295 1.0669 0.1872 1.1831 0.0707 1.2052 0.0236 1.0586 0.2529 R2 0.9805 0.9816 0.9721 0.9643 0.9638 0.9659

11Table 4. Parameters of Higuchi/Korsmeyer–Peppas model

for release properties of LDPE film treated with cold plasma containing CE-, NE-, and PE-stabilized MFEO. Emulsion

2Higuchi Korsmeyer–Peppas K1 R2 K2 n R2

CE 1% 2.5627 0.9727 1.1136 CE 3% 2.5829 0.989 1.0464 CE 6% 2.6465 0.9843 1.0669 NE 1% 2.8933 0.9809 1.1831 NE 3% 2.8512 0.9687 1.2052 NE 6% 2.8522 0.9844 1.0586 PE 1% 3.1184 0.9894 1.0212 PE 3% 2.9334 0.9862 1.1089 PE 6% 2.49 0.9912 1.0225 0.1334 0.2295 0.1872 0.0707 0.0236 0.2529 0.0348 0.2674 0.4515 0.9805 0.9816 0.9721 0.9643 0.9638 0.9659 0.9968 0.9859 0.9746 CE: coarse emulsion; NE: nanoemulsion; PE: Pickering emulsion. 4. Conclusions Cold

31plasma treatment can improve the properties of LDPE films

by facilitating MFEO coating, it is supported by the FTIR results that showed the differences between the control film and plasma-treated film and showed the presence of a new group of active compounds from MFEO on the LDPE coating. The use of different types of emulsion causes different characteristics of the film; in general, the use of CE and NE results in better optical characteristics than PE.

48Increasing the concentration of MFEO provides increased antioxidant activity and inhibition of

bacteria. However, PE has more stability and improved controlled release, where PE can inhibit coalescence of MFEO. PE is suitable, especially over long-term storage. There needs to be more research on how to use emulsions in cold plasma treatment protocols and how to apply them to food.

46Author Contributions: Conceptualization, B.Y. and C.-W.H.; methodology, B.Y., C.-K.C. and C.-W.H.; software, B.Y., A.S.S. and

M.G.; validation, B.Y., C.-K.C. and C.-W.H.; formal analysis, B.Y., C.-K.C. and F.P.; investigation, B.Y., C.-T.L. and A.S.S.; resources, C.-W.H.;

27data curation, B.Y., S.P.S. and C.-T.L.; writing—original draft preparation, B.Y., F.P., C.-K.C. and C.-W.H.; writing—review and editing, B.Y., A

.S.S., F.P., C.-K.C., M.G., S.P.S. and

37C.-W.H.; visualization, B.Y. and C.-T.L.; supervision, C.-W.H

.;

37**project administration, C**.-K.C. **and** C.-**T**.L.; **funding acquisition, C.-W.H**

.

3**All authors have read and agreed to the published version of the manuscript. Funding: This research** is **supported by the Ministry of Science and Technology, Taiwan** (grant no. **110-2221-E**-005 -012 -**MY3). Institutional Review Board Statement: Not applicable. Informed Consent Statement: Not applicable. Data Availability Statement**: All the **data** are available within **the** manuscript. Acknowledgments: The **authors** wish to **thank the**

Ministry of Science and Technology, Taiwan and i-Center for Advanced Science and Technology, National Chung Hsing University, Jiann-Yeu Chen. Conflicts of Interest: The authors declare no conflict of interest. References 1. Ghadetaj, A.; Almasi, H.; Mehryar, L. Development and characterization of whey protein isolate active films containing nanoemulsions of Grammosciadium ptrocarpum Bioss. essential oil. Food Packag. Shelf Life 2018, 16, 31–40. [CrossRef] 2. Li, J.; Xu, X.; Chen, Z.; Wang, T.; Lu, Z.; Hu, W.; Wang, L. Zein/gum Arabic nanoparticle-stabilized Pickering emulsion with thymol as an antibacterial delivery system. Carbohydr. Polym. 2018, 200, 416–426. [CrossRef] [PubMed] 3. Gavahian, M.; Farahnaky, A.; Javidnia, K.; Majzoobi, M. Comparison of ohmic-assisted hydrodistillation with traditional hydrodistillation for the extraction of essential oils from Thymus vulgaris L. Innov. Food Sci Emerg. Technol. 2012, 14, 85–91. [CrossRef] 4. Atarés, L.; Chiralt, A. Essential oils as additives in biodegradable films and coatings for active food packaging. Trends Food Sci. Technol. 2016, 48, 51–62. [CrossRef] 5. D'Souza, S.P.; Chavannavar, S.V.; Kanchanashri, B.; Niveditha, S.B. Pharmaceutical Perspectives of Spices and Condiments as Alternative Antimicrobial Remedy. J. Evid.-Based Complement. Altern. Med. 2017, 22, 1002–1010. [CrossRef] [PubMed] 6. Matulyte, I.; Marksa, M.; Ivanauskas, L.; Kalvenien, Z.; Lazauskas, R.; Bernatoniene, J. GC-MS analysis of the composition of the extracts and essential Oil from Myristica fragrans Seeds Using Magnesium Aluminometasilicate as Excipient. Molecules 2019, 24, 1062. [CrossRef] 7. El-Alfy, A.T.; Abourashed, E.A.; Patel, C.; Mazhari, N.; An, H.R.; Jeon, A. Phenolic compounds from nutmeg (Myristica fragrans Houtt.) inhibit the endocannabinoid-modulating enzyme fatty acid amide hydrolase. J. Pharm. Pharmacol. 2019, 71, 1879–1889. [CrossRef] 8. Mousavi, S.M.; Hashemi, S.A.; Ramakrishna, S.; Esmaeili, H.; Bahrani, S.; Koosha, M.; Babapoor, A. Green synthesis of supermagnetic Fe3O4–MgO nanoparticles via Nutmeg essential oil toward superior anti-bacterial and anti-fungal performance. J. Drug Deliv. Sci. Technol. 2019, 54, 101352. [CrossRef] 9. Balakrishnan, S.; Sivaji, I.; Kandasamy, S.; Duraisamy, S.; Kumar, N.S.; Gurusubramanian, G. Biosynthesis of silver nanoparticles using Myristica fragrans seed (nutmeg) extract and its antibacterial activity against multidrug-resistant (MDR) Salmonella enterica serovar Typhi isolates. Environ. Sci. Pollut. Res. 2017, 24, 14758–14769. [CrossRef] 10. Galeano, L.Y.; Torres, V.O.; García, S.Á. Evaluation of nutmeg (Myristica fragrans Houtt) as active component during storage of bovine loins. Rev. Cienc. Agrícolas 2018, 35, 48–57. [CrossRef] 11. Liu, Q.R.; Wang, W.; Qi, J.; Huang, Q.; Xiao, J. Oregano essential oil loaded soybean polysaccharide films: Effect of Pickering type immobilization on physical and antimicrobial properties. Food Hydrocoll. 2019, 87, 165–172. [CrossRef] 12. Burgos, N.; Mellinas, A.C.; García-serna, E. Nanoencapsulation of Flavor and Aromas in Food Packaging. In Food Packaging; Elsevier Inc.: Philadelphia, PA, USA, 2017; pp. 567–601. 13. Shi, W.J.; Tang, C.H.; Yin, S.W.; Yin, Y.; Yang, X.Q.; Wu, L.Y.; Zhao, Z.G. Development and characterization of novel chitosan emulsion films via pickering emulsions incorporation approach. Food Hydrocoll. 2016, 52, 253–264. [CrossRef] 14. Cossu, A.; Wang, M.S.; Chaudhari, A.; Nitin, N. Antifungal activity against Candida albicans of starch Pickering emulsion with thymol or amphotericin B in suspension and calcium alginate films. Int. J. Pharm. 2015, 493, 233–242. [CrossRef] [PubMed] 15. Hosseinnia, M.; Khaledabad, M.A.; Almasi, H. Optimization of Ziziphora clinopodiodes essential oil microencapsulation by whey protein isolate and pectin: A comparative study. Int. J. Biol. Macromol. 2017, 101, 958–966. [CrossRef] 16. Almasi, H.; Azizi, S.; Amjadi, S. Development and characterization of pectin films activated by nanoemulsion and Pickering emulsion stabilized marjoram (Origanum majorana L.) essential oil. Food Hydrocoll. 2020, 99, 105338. [CrossRef] 17. Gavahian, M.; Meng-Jen, T.; Khaneghah, A.M. Emerging techniques in food science: The resistance of chlorpyrifos pesticide pollution against arc and dielectric barrier discharge plasma. Qual. Assur. Saf. Crops Foods 2020, 12, 9–17. [CrossRef] 18. Pankaj, S.K.; Bueno-Ferrer, C.; Misra, N.N.; Milosavljević, V.; O'Donnell, C.P.; Bourke, P.; Keener, K.M.; Cullen, P.J. Applications of cold plasma technology in food packaging. Trends Food Sci. Technol. 2014, 35, 5–17. [CrossRef] 19. Mendes, J.F.; Norcino, L.B.; Martins, H.H.A.; Manrich, A.; Otoni, C.G.; Carvalho, E.E.N.; Piccoli, R.H.; Oliveira, J.E.; Pinheiro, A.C.M.; Mattoso, L.H.C. Correlating emulsion characteristics with the properties of active starch films loaded with lemongrass essential oil. Food Hydrocoll. 2020, 100, 105428. [CrossRef] 20. González, A.; Gastelú, G.; Barrera, G.N.; Ribotta, P.D.; Álvarez Igarzabal, C.I. Preparation and characterization of soy protein films reinforced with cellulose nanofibers obtained from soybean by-products. Food Hydrocoll. 2019, 89, 758–764. [CrossRef] 21. Hu, Y.; Shi, L.; Ren, Z.; Hao, G.; Chen, J.; Weng, W. Characterization of emulsion films prepared from soy protein isolate at different preheating temperatures. J. Food Eng. 2021, 309, 110697. [CrossRef] 22. Wang, Q.; Liu, W.; Tian, B.; Li, D.; Liu, C.; Jiang, B.; Feng, Z. Preparation and characterization of coating based on protein nanofibers and polyphenol and application for salted duck egg yolks. Foods 2020, 9, 449. [CrossRef] [PubMed] 23. Jiang, B.; Wang, L.; Zhu, M.; Wu, S.; Wang, X.; Li, D.; Liu, C.; Feng, Z.; Tian, C. Separation,

structural characteristics and biological activity of lactic acid bacteria exopolysaccharides separated by aqueous two-phase system. LWT 2021, 147, 111617. [CrossRef] 24. Tian, B.; Cheng, J.; Zhang, T.; Liu, Y.; Chen, D. Multifunctional chitosan-based film loaded with hops β-acids: Preparation, characterization, controlled release and antibacterial mechanism. Food Hydrocoll. 2022, 124, 107337. [CrossRef] 25. Noori, S.; Zeynali, F.; Almasi, H. Antimicrobial and antioxidant efficiency of nanoemulsion-based edible coating containing ginger (Zingiber officinale) essential oil and its effect on safety and quality attributes of chicken breast fillets. Food Control 2018, 84, 312–320. [CrossRef] 26. Wong, L.W.; Loke, X.J.; Chang, C.K.; Ko, W.C.; Hou, C.Y.; Hsieh, C.W. Use of the plasma-treated and chitosan/gallic acid-coated polyethylene film for the preservation of tilapia (Orechromis niloticus) fillets. Food Chem. 2020, 329, 126989. [CrossRef] 27. Loke, X.J.; Chang, C.K.; Hou, C.Y.; Cheng, K.C.; Hsieh, C.W. Plasma-treated polyethylene coated with polysaccharide and protein containing cinnamaldehyde for active packaging films and applications on tilapia (Orechromis niloticus) fillet preservation. Food Control 2021, 125, 1–10. [CrossRef] 28. Han, H.S.; Song, K.B. Noni (Morinda citrifolia) fruit polysaccharide films containing blueberry (Vaccinium corymbosum) leaf extract as an antioxidant packaging material. Food Hydrocoll. 2021, 112, 1–8. [CrossRef] 29. Grzegorzewski, F.; Rohn, S.; Kroh, L.W.; Geyer, M.; Schlüter, O. Surface morphology and chemical composition of lamb's lettuce (Valerianella locusta) after exposure to a low-pressure oxygen plasma. Food Chem. 2010, 122, 1145–1152. [CrossRef] 30. Theapsak, S.; Watthanaphanit, A.; Rujiravanit, R. Preparation of chitosan-coated polyethylene packaging films by DBD plasma treatment. ACS Appl. Mater. Interfaces 2012, 4, 2474–2482. [CrossRef] 31. Dammak, I.; de Carvalho, R.A.; Trindade, C.S.F.; Lourenço, R.V.; do Amaral, S.P.J. Properties of active gelatin films incorporated with rutin-loaded nanoemulsions. Int. J. Biol. Macromol. 2017, 98, 39–49. [CrossRef] 32. Zhang, L.; Liu, Z.; Wang, X.; Dong, S.; Sun, Y.; Zhao, Z. The properties of chitosan/zein blend film and effect of film on quality of mushroom (Agaricus bisporus). Postharvest Biol. Technol. 2019, 155, 47–56. [CrossRef] 33. Blois, M.S. Antioxidant Determinations by the Use of a Stable Free Radical. Nature 1985, 181, 1199–1200. [CrossRef] 34. Piñeros-Hernandez, D.; Medina-Jaramillo, C.; López-Córdoba, A.; Goyanes, S. Edible cassava starch films carrying rosemary antioxidant extracts for potential use as active food packaging. Food Hydrocoll. 2017, 63, 488–495. [CrossRef] 35. Nurjanah, N.; Jacoeb, A.M.; Asmara, D.A.; Hidayat, T. Phenol Component of Fresh and Boiled Sea Grapes (Caulerpa sp.) From Tual, Maluku. Food Sci. J. 2019, 1, 31. [CrossRef] 36. Qin, Y.; Yang, J.; Xue, J. Characterization of antimicrobial poly (lactic acid)/poly(trimethylene carbonate) films with cinnamalde- hyde. J. Mater. Sci. 2015, 50, 1150. [CrossRef] 37. Shokri, S.; Parastouei, K.; Taghdir, M.; Abbaszadeh, S. Application an edible active coating based on chitosan- Ferulago angulata essential oil nanoemulsion to shelf life extension of Rainbow trout fillets stored at 4 ◦C. Int. J. Biol. Macromol. 2020, 153, 846–854. [CrossRef] [PubMed] 38. Söğüt, E. Properties of Solvent Cast Polycaprolactone Films Containing Pomegranate Seed Oil Stabilized with Nanocellulose. Turk. J. Agric.-Food Sci. Technol. 2019, 7, 67. [CrossRef] 39. Du, M.; Sun, Z.; Liu, Z.; Yang, Y.; Liu, Z.; Wang, Y.; Jiang, B.; Feng, Z.; Liu, C. High efficiency desalination of wasted salted duck egg white and processing into food-grade pickering emulsion stabilizer. LWT 2022, 161, 113337. [CrossRef] 40. Yang, Y.; Jiao, Q.; Wang, L.; Zhang, Y.; Jiang, B.; Li, D.; Feng, Z.; Liu, C. Preparation and evaluation of a novel high internal phase Pickering emulsion based on whey protein isolate nanofibrils derived by hydrothermal method. Food Hydrocoll. 2022, 123, 107180. [CrossRef] 41. Galus, S. Functional properties of soy protein isolate edible films as affected by rapeseed oil concentration. Food Hydrocoll. 2018, 85, 233–241. [CrossRef] 42. Dong, S.; Gao, A.; Zhao, Y.; Li, Y.T.; Chen, Y. Characterization of physicochemical and structural properties of atmospheric cold plasma (ACP) modified zein. Food Bioprod. Process. 2017, 106, 65–74. [CrossRef] 43. Wong, L.W.; Hou, C.Y.; Hsieh, C.C.; Chang, C.K.; Wu, Y.S.; Hsieh, C.W. Preparation of antimicrobial active packaging film by capacitively coupled plasma treatment. LWT 2020, 117, 108612. [CrossRef] 44. Almasi, H.; Zandi, M.; Beigzadeh, S.; Haghju, S.; Mehrnow, N. Chitosan films incorporated with nettle (Urtica dioica L.) extract- loaded nanoliposomes: II. Antioxidant activity and release properties. J. Microencapsul. 2016, 33, 449–459. [CrossRef] [PubMed] 45. Fasihi, H.; Fazilati, M.; Hashemi, M.; Noshirvani, N. Novel carboxymethyl cellulose-polyvinyl alcohol blend films stabilized by Pickering emulsion incorporation method. Carbohydr. Polym. 2017, 167, 79–89. [CrossRef] 46. Li, S. Effect of Pickering emulsion on the mechanical performances and fracture toughness of epoxy composites. Polym. Adv. Technol. 2020, 31, 722–730. [CrossRef] 47. Wardana, A.A.; Koga, A.; Tanaka, F.; Tanaka, F. Antifungal features and properties of chitosan/sandalwood oil Pickering emulsion coating stabilized by appropriate cellulose nanofiber dosage for fresh fruit application. Sci. Rep. 2021, 11, 1–15. [CrossRef] 48. Muller, J.; González-Martínez, C.; Chiralt, A. Poly(lactic) acid (PLA) and starch bilayer films, containing cinnamaldehyde, obtained by compression moulding. Eur. Polym. J. 2017, 95, 56–70. [CrossRef] 49. Bonilla, J.; Talón, E.; Atarés, L.; Vargas, M.; Chiralt, A. Effect of the incorporation of antioxidants on physicochemical and antioxidant properties of wheat starch-chitosan films. J. Food Eng. 2013, 118, 271–278. [CrossRef] 50. Silva Damasceno, E.T.; Almeida, R.R.; de Carvalho, S.Y.B.; de Carvalho, G.S.G.; Mano, V.; Pereira, A.C.; Guimaraes, L.G. Lippia origanoides Kunth. essential oil loaded in nanogel based on the chitosan and ϱ-coumaric acid: Encapsulation efficiency and antioxidant activity. Ind. Crops Prod. 2018, 125, 85–94. [CrossRef] 51. Yallapu, M.M.; Jaggi, M.; Chauhan, S.C. β-Cyclodextrin-curcumin self-assembly enhances curcumin delivery in prostate cancer cells. Colloids Surf. B Biointerfaces 2010, 79, 113–125. [CrossRef] 52. Członka, S.; Stra˛kowska, A.; Kairytė, A.; Kremensas, A. Nutmeg filler as a natural compound for the production of polyurethane composite foams with antibacterial and anti-aging properties. Polym. Test. 2020, 86, 1–13. [CrossRef] 53. Salmas, C.E.; Giannakas, A.E.; Baikousi, M.; Leontiou, A.; Siasou, Z.; Karakassides, M.A. Development of Poly (L-Lactic Acid)/Chitosan/Basil Oil Active Packaging Films via a Melt-Extrusion Process Using Novel Chitosan/Basil Oil Blends. Processes 2021, 9, 88. [CrossRef] 54. Ginting, B.; Maira, R.M.; Helwati, H.; Desiyana, L.S.; Mujahid, R. Isolation of essensial oil of nutmeg (Myristica fragrans Houtt) and antioxidant activity test with DPPH. J. Nat. 2018, 18, 11–17. [CrossRef] 55. Dammak, I.; Lourenço, R.V.; Sobral, P.J.A. Active gelatin films incorporated with Pickering

emulsions encapsulating hesperidin: Preparation and physicochemical characterization. J. Food Eng. 2019, 240, 9–20. [CrossRef] 56. Wang, W.; Wu, N.; Zu, Y.G.; Fu, Y.J. Antioxidative activity of Rosmarinus officinalis L. essential oil compared to its main components. Food Chem. 2008, 108, 1019–1022. [CrossRef] [PubMed] 57. Li, J.; Mcclements, D.J.; Mclandsborough, L.A. Interaction between emulsion droplets and Escherichia coli cells. J. Food Sci. 2001, 66, 570–657. [CrossRef] 58. Burt, S. Essential oils: Their antibacterial properties and potential applications in foods—A review. Int. J. Food Microbiol. 2004, 94, 223–253. [CrossRef] 59. Ritger, P.L.; Peppas, N.A. A simple equation for description of solute release II. Fickian and anomalous release from swellable devices. J. Control. Release 1987, 5, 37–42. [CrossRef]

11 1o0f o2f221