



Article A Contribution to Improve Barrier Properties and Reduce Swelling Ratio of κ-Carrageenan Film from the Incorporation of Guar Gum or Locust Bean Gum

Ruixuan Wang, Song Zhang, Shuaichen Liu, Yuqi Sun and Hongyan Xu *💿

Department of Food Science and Engineering, College of Agriculture, Yanbian University, Yanji 133000, China * Correspondence: xuhongyan@ybu.edu.cn; Tel.: +86-139-4470-9169

Abstract: In this study, galactomannan (GM), including guar gum (GG) or locust bean gum (LG), was incorporated into a κ -carrageenan film to improve barrier properties and reduce the swelling ratio (SR). The effects of that with different concentrations on optical, mechanical, barrier, swelling and thermal properties of the κ -carrageenan-based film were researched. SEM and rheological results showed that both κ -carrageenan/GG and κ -carrageenan/LG had good compatibility and stability. FTIR results showed that LG was easier to form hydrogen bonds with κ -carrageenan. The KC-L exhibited excellent mechanical properties, barrier properties, and SR than KC-G. The film with 15% GM had good light transmittance. Moreover, the thermal stability of the film could be improved by adding GMs. This study reports that the κ -carrageenan–GM film has potential in packaging applications.

Keywords: k-carrageenan; galactomannan; composite film; physical properties



Citation: Wang, R.; Zhang, S.; Liu, S.; Sun, Y.; Xu, H. A Contribution to Improve Barrier Properties and Reduce Swelling Ratio of κ-Carrageenan Film from the Incorporation of Guar Gum or Locust Bean Gum. *Polymers* **2023**, *15*, 1751. https://doi.org/10.3390/ polym15071751

Academic Editor: Emiliano Bedini

Received: 17 February 2023 Revised: 28 March 2023 Accepted: 29 March 2023 Published: 31 March 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/).

1. Introduction

Plastic has caused a series of serious environmental issues. Every year, about 500 billion plastics bags were used, but only 3% of them could be recycled. The rest of those were discarded as solid waste, which is difficult to degrade since it causes white pollution to the ecological environment and landscape. In addition, plastics have a potential threat to human health because they may contain some chemical additives such as bisphenol A or phthalates. Thus, it is urgent to find a sustainable alternative [1]. Natural polysaccharides have received increased attention due to their renewability, biodegradability and filmforming properties [2]. Marques et al. prepared cassava starch-based films that can be used for packaging in the food industry [3]. The calcium alginate film loading ciprofloxacin potentially healed infected foot ulcers [4]. Chargot et al. fabricated a chitosan/carrot cellulose nanofibre film with antibacterial properties [5]. It is noted that polysaccharide films could effectively block the transport of CO_2 and O_2 , decreasing the quality loss of foods during storage. Carrageenan, a kind of linear polysaccharide with negative charges, is widely used in the field of food [6]. It has been used as the main film-forming substance, and has been turned into antimicrobial packaging film [7] and freshness monitoring labels [8]. However, carrageenan-based films swell easily in moist conditions [9] and have poor water vapor barrier properties [10,11]. Shahbazi et al. blended high-pressure homogenized starch into a carrageenan film to overcome those drawbacks. However, it is troublesome for starch to be modified physically [12]. Hanani et al. added various plant oils into k-carrageenan films to reduce WVP, while their mechanical properties decreased significantly [13]. Nanosilica was added to the κ -carrageenan film to improve the performances. However, dynamic oscillation measurements showed that the dissipation factor decreased significantly with the angular frequency from 0.1 to 100 rad/s, indicating that the stability of the film solution was poor [14].

Galactomannans consisting of a mannan (M) backbone with single galactose (G) are commonly found in the endosperm of legumes [15] and have been widely used for a vast range of foods [16], packaging [17] and pharmaceuticals [18]. Three major commercial galactomannans include guar gum, tara gum and locust bean gum, and the corresponding ratios of M/G were 2:1, 3:1 and 4:1, respectively. Figure 1 shows the typical structure of galactomannan (idealized) [19]. Wu et al. discovered that tara gum (10% to 60%) resulted in a higher gel strength of mixed gels than that of pure κ -carrageenan [20]. JT et al. prepared a clay/ κ -carrageenan/LG composite film with potential in food packaging and presenting good mechanical properties, lower WVP and better antibacterial activity for L. monocytogenes at the addition of 16 wt% clay [21]. As far as we know, the effects of galactomannan with different ratios of M/G and content on inherent properties of κ -carrageenan films have not been reported.

In this study, we prepared a composite films by mixing κ -carrageenan and GM solutions in which glycerin was used as a plasticizer to solve the drawback of brittleness. The effects of GM with two ratios of M/G (GG and LG) and the content on κ -carrageenan-based films were investigated. We used scanning electron microscopy (SEM) to observe the cross sections of the films and employed Fourier-transform infrared (FTIR) and thermogravimetric analysis (TGA) to research the functional groups and thermal stability. The light transmittance, mechanical properties, oxygen permeability, water vapor permeability, swelling, and rheological properties of κ -carrageenan–GM composite films were also investigated.



Figure 1. The typical structure of galactomannan.

2. Materials and Methods

2.1. Materials

Guar gum was bought from Fiyyed Biotech Co., Ltd. (Suzhou, China). Locust bean gum was purchased from Yaheng Biotech Co., Ltd. (Heze, China). κ-Carrageenan (food grade) was supplied by Bairen Biotech Co., Ltd. (Qingdao, China). Glycerol (AR) was offered by Yongda Chemical Reagent Co., Ltd. (Tianjin, China). Ethanol (AR) was obtained from Fuyu Chemical Co., Ltd. (Tianjin, China).

2.2. Preparation of Composite Film

GG or LG (5, 10, 15, 20, and 25%, w/w, based on κ -carrageenan and GM) was firstly dispersed in 10 mL of ethanol and then stirred in distilled water at 65 °C. At the same time, κ -carrageenan was dissolved in distilled water under stirring at 85 °C. After that, the solution was added into GM solution and stirred for 60 min. Subsequently, 30% (w/w, based on κ -carrageenan and GM) of glycerin was added and stirred for another 30 min. Finally, the resulting film-forming solution was cast into a mold (300 mm \times 290 mm) and dried for ~24 h at 50 °C. The corresponding films were labeled based on the concentrations of GG and LG. For example, the composite film consisting of 5% GG was recorded as KC-5G. The films prepared from 30% glycerol and GG or LG are noted as GGF and LGF, respectively.

2.3. Characterizations

The cross-sections of the composite films were observed using a Quanta 200 SEM (Philips-FEI Co., Eindhoven, The Netherlands). The functional groups were studied using a Nicolette 6700 spectrometer (Perkin Elmer, Frontier, MA, USA) with attenuated total reflection mode in the range of 600-4000 cm⁻¹ at a resolution of 4 cm⁻¹.

2.4. Film Properties

2.4.1. Light Transmittance

The light transmittance of the films (2.5 cm \times 4.0 cm) was measured using an UV-2600 spectrophotometer (Shimadzu, Kyoto, Japan) in the range of 200–800 nm.

2.4.2. Mechanical Properties

Thickness of the film was obtained by measuring 20 random points using a micrometer (ID-C112XBS, Mitutoyo Corp., Tokyo, Japan). An XLW-PC auto tensile tester (Labthink, Jinan, China) was used to measure tensile strength (TS) and the elongation at break (EB) of the films ($1.5 \text{ cm} \times 8.0 \text{ cm}$) with a strain rate of 300 mm/min.

2.4.3. Oxygen Permeability

The oxygen permeability (OP) of the film was measured using an OX/230 oxygen transmission rate tester (Labthink Instruments Co., Ltd., Jinan, China).

2.4.4. Swelling Property

The swelling property of the films was determined according to a modified method [22]. The films (15 mm \times 15 mm) were dried and the volume of them was measured, then they were placed in separate dryers with relative humidities of 53% and 75% for 24 h at room temperature. The swelling ratio of the films was calculated using the following formula:

$$SR = (v_s - v_e)/v_e \tag{1}$$

where v_s and v_e represent the volume of the films before and after swelling, respectively.

2.4.5. Water Vapor Permeability

The water vapor permeability (WVP) of the films was obtained through the gravimetric method. The weighing bottles containing 23 g of anhydrous calcium chloride were sealed with the film (~16.6 cm²) then conditioned in desiccators under 75% RH at 25 °C. The weight was weighed periodically, and the WVP was calculated according to the following equation:

$$WVP = (k \times d) / (s \times \Delta P)$$

where k is the weight of moisture gain per unit of time (g/s), d is the average film thickness (mm), s is the area of the exposed film surface (m²) and ΔP is the driving force (1753.55 Pa).

2.4.6. Thermogravimetric Analysis

TGA Q500 (TA Instruments, DE, USA) was used to characterize the thermal stability in a nitrogen atmosphere from room temperature to 600 °C with a heating rate of 10 °C/min. The derivative thermogravimetric analysis (DTG) was obtained via the first-order derivative of TGA.

2.4.7. Rheological Analysis

After the films were immersed in distilled water for 30 s, the rheological behavior of the films with different concentrations of GG and LG in gel state was measured by using a rotational rheometer (TAAR2000EX, Newcastle, DE, USA) with a parallel-plate geometry (diameter = 40 mm, gap = 1000 μ m) at a shear rate range of 0.1–100 s⁻¹ at 25 °C. The rheological properties of films in the gel state were studied by using the Ostwald–de Wale model, as below:

$$\eta_a = \mathbf{K} \times \gamma^{n-1} \tag{2}$$

where η_a is the viscosity (Pa•s), γ denotes the shear rate (s⁻¹), **n** represents the flow index and K is the parameter of the materials.

The viscoelasticity of the films in gel states was characterized by dynamic oscillation measurements in the frequency range of 0.1–100 rad/s. The viscoelasticity of the films in gel states can be described via the following equation:

$$\tan \delta = \mathbf{G}''(\boldsymbol{\omega}) / \mathbf{G}'(\boldsymbol{\omega}) \tag{3}$$

where $\tan \delta$, $G''(\omega)$ and $G'(\omega)$ are the dissipation factor, loss modulus and the storage modulus, respectively.

2.4.8. Statistics Analysis

Data were analyzed by SPSS Statistics (v17.0; SPSS Inc., Chicago, IL, USA). The differences among mean values were processed by using the Duncan multiple range tests (p < 0.05).

3. Results and Discussions

3.1. SEM Analysis

Micrographs of the cross-sections of κ -carrageenan-based films containing various concentrations of GG and LG are exhibited in Figure 2. An ordered and homogeneous stripy structure exhibited on the cross-section of the κ -carrageenan-based film can be seen due to the helical aggregation of κ -carrageenan chains; meanwhile, the entanglements among the κ -carrageenan and the GG/LG molecule chains hint at a good compatibility and interactions between GM and κ -carrageenan. In addition, it can be observed that the cross-section of the LG–carrageenan film was more compact than that of the GG–carrageenan film. This is because the branch units of the GG are twice that of the LG, thus the compatibility of the GG–carrageenan film is lower.



Figure 2. SEM images of carrageenan film containing GG and LG.

3.2. FTIR Spectroscopy

The FTIR spectra of KC, KC-25G and KC-25L are shown in Figure 3. The bands at 3301 cm^{-1} , 2925 cm⁻¹, 2884 cm⁻¹ and 1022 cm⁻¹ are assigned to O-H stretching vibration, C-H stretching vibration, and C-O (pyranose ring) stretching vibration, respectively [23,24]. For KC, the bands at 1239 cm⁻¹ (sulfate ester), 918 cm⁻¹ (3,6-anhydrogalactose) and 846 cm⁻¹ (galactose-4-sulfate ester) are the characteristic peaks of κ -carrageenan [25]. After the incorporation of GG and LG, the spectra of KC-25L and KC-25G shows the bands at 868 cm⁻¹ and 814 cm⁻¹, which are attributed to the β -linked D-mannopyranose units and α -linked D-galactopyranose units, respectively [26]. Meanwhile, the two spectra of KC-25L and KC-25G are similar. However, it is found that the intensity of the band at 3295 cm⁻¹ decreased and shifted to 3280 cm⁻¹, which is owing to the fact that with LG with fewer branched chains it is easier to form hydrogen bonding because of the lower steric hindrance. Martins et al. also found that carrageenan could interact with the unbranched smooth segments of LG via hydrogen bonds [26].



Figure 3. FTIR spectra of KC, KC-25G and KC-25L.

3.3. Optical Properties

The optical properties of the composite films are shown in Figure 4. With the increase in GM, the light transmittance of the composite films increased first and reached the maximum value with a 15% GM addition, then decreased. The regularity of KC-L was better than that of KC g due to the fewer branch chains of LG because the scattering and refraction of light decreased. When the GM content was less than 15%, the transmittance of KC-L was poor compared with that of KC-G. When the GM content further increased from 15% to 25%, the transmittance of KC-L and KC g decreased from 57.06% to 51.74% and 54.58% to 43.24%, respectively, showing that the light transmittance of KC-L was slightly higher than that of KC-G. This is mainly because the looser and more disordered network of the composite film formed after adding the GG with more branched chains, resulting in more reflection and refraction when light passed through the films.



Figure 4. Light transmittance of the composite film containing GG (a) and LG (b).

3.4. Mechanical Properties

The mechanical properties of the κ -carrageenan film and GM– κ -carrageenan film are displayed in Table 1. EB decreased when the GM content increased from 5% to 25% because the substituted and branched structure of the GM obstructed the formation of intermolecular hydrogen bonds, resulting in an easier breaking (lower EB). When the content of GG was in the range of 5–15%, the TS of KC g had no obvious change. This is because GG with a higher branching degree is difficult to form more hydrogen bonds with κ -carrageenan. It can be seen in Figure 5a that GG just interweaved randomly with κ -carrageenan. The TS of KC-5L and KC-10L was higher than that of KC due to the

formation of hydrogen bonds between the unsubstituted regions of the LG chain and κ -carrageenan (Figure 5b) [27]. In addition, the larger steric hindrance of GM tended to hinder the formation of intermolecular hydrogen bonds, so the TS value gradually decreased with the further increase in the content.

Table 1. TS and EB of KC, KC-G and KC-L films.

Sample	Thickness (mm)	TS (MPa)	EB (%)	$\begin{array}{c} OP \ (cm^3 \ mm \ m^{-2} \\ day^{-1} \ atm^{-1}) \end{array}$
КС	$0.107\pm0.003~\mathrm{cd}$	$21.775\pm0.556~\mathrm{ab}$	37.150 ± 1.161 ce	$3.247\pm0.490bd$
KC-5G	$0.100\pm0.007~\mathrm{bc}$	$21.380\pm1.536~\mathrm{ab}$	$32.300 \pm 2.598 \text{ d}$	$2.783\pm0.074~\mathrm{bcd}$
KC-10G	$0.105\pm0.008~\mathrm{cd}$	$21.298\pm0.870~\mathrm{ab}$	$23.900 \pm 1.723 \text{ c}$	$2.647\pm0.147~ m bc$
KC-15G	0.086 ± 0.000 a	$21.880 \pm 2.088 \ { m b}$	$21.050 \pm 1.143 \ { m b}$	$2.360\pm0.139~\mathrm{ab}$
KC-20G	$0.096\pm0.002~\mathrm{b}$	$19.973\pm1.013~\mathrm{ab}$	$19.200\pm0.849~\mathrm{ab}$	1.935 ± 0.074 a
KC-25G	$0.096\pm0.001~\mathrm{b}$	19.130 ± 1.179 a	18.150 ± 1.090 a	$2.919\pm0.017~\mathrm{cd}$
KC-5L	$0.100\pm0.002\mathrm{b}$	$22.933\pm0.564\mathrm{bc}$	$36.700 \pm 2.858 \text{ c}$	1.847 ± 0.070 a
KC-10L	$0.097\pm0.004~\mathrm{ab}$	$23.988 \pm 1.270 \text{ c}$	35.950 ± 1.873 c	1.799 ± 0.049 a
KC-15L	$0.101\pm0.003~{ m bc}$	$21.545\pm0.809~\mathrm{ab}$	$26.500 \pm 1.212 \text{ b}$	1.595 ± 0.061 a
KC-20L	$0.113 \pm 0.005 \ d$	$21.555\pm1.469~\mathrm{ab}$	21.000 ± 2.490 a	1.642 ± 0.196 a
KC-25L	$0.091\pm0.003~\mathrm{a}$	19.993 ± 0.851 a	20.600 ± 0.707 a	$1.853\pm0.145~\mathrm{a}$

Different lowercase letters in the same column indicate significant difference (p < 0.05).



κ-carrageenan the guar gum the locust bean gum

Figure 5. Binding mechanism diagram of KC g film (a) and KC-L film (b).

3.5. Oxygen Permeability

The OP of GM– κ -carrageenan films are shown in Table 1. The addition of GM could effectively reduce the gap through random interleaving and winding, resulting in a lower OP of KC-5G and KC-5L than that of the KC film (3.247 ± 0.490 cm³ mm m⁻² day⁻¹ atm⁻¹). The KC g with 20% GG and KC-L with 15% LG had the minimum value of OP. When the amount of GM continued to increase, the density of the composite film decreased again. A similar result was obtained with the chitosan and guar gum composite films [28]. Furthermore, the OP of KC-L was lower than that of KC g under the same addition because the fewer branch chains of LG made the film structure denser.

3.6. Swelling Property and WVP

The SR and WVP of the κ -carrageenan-based composite films are displayed in Table 2. The SR of the κ -carrageenan-based composite films decreased with the increased GM content and decreased relative humidity. This is because the wettability of κ -carrageenan decreased as the GM content increased, leading to the decrease in water transfer (WVP). A similar result was achieved when the WVP of the yellow tragacanth-LG composite film decreased with the increase in LG [29]. In addition, the SR and WVP of KC g were higher than that of KC-L with the same content because GG with a higher branching degree decreased the compactness of the composite films. Kurt and Kahyaoglu also reported that the WVP of GG films was higher than that of LG films [30].

Sample	SR	WVP (g mm s ^{-1} m ^{-2}	
	RH53%	RH75%	$Pa^{-1} \times 10^{-11}$)
KC	$14.67\pm2.08~\mathrm{b}$	$25.09\pm1.84~\mathrm{c}$	$4.44\pm0.09~\mathrm{ef}$
KC-5G	$11.55\pm1.47~\mathrm{ab}$	$19.13\pm0.07~\mathrm{b}$	$3.81\pm0.14~\mathrm{d}$
KC-10G	$9.70\pm0.44~\mathrm{ab}$	$18.83\pm0.92\mathrm{b}$	$3.68\pm0.11~{ m c}$
KC-15G	$9.20\pm1.02~\mathrm{ab}$	$17.35\pm1.17~\mathrm{ab}$	$3.60\pm0.10~{ m c}$
KC-20G	$7.44 \pm 1.27~\mathrm{a}$	$16.88\pm1.72~\mathrm{ab}$	$3.19\pm0.08~\mathrm{b}$
KC-25G	6.10 ± 2.44 a	$13.93\pm1.34~\mathrm{a}$	$3.08\pm0.07~\mathrm{a}$
KC-5L	9.72 ± 0.46 a	$18.73\pm1.19~\mathrm{b}$	$3.56\pm0.04~\mathrm{e}$
KC-10L	$8.21\pm1.17~\mathrm{a}$	$17.00\pm0.80~\mathrm{b}$	$3.36 \pm 0.07 \text{ d}$
KC-15L	7.40 ± 1.51 a	$16.70\pm1.55~\mathrm{b}$	$3.08\pm0.07~\mathrm{c}$
KC-20L	$6.61\pm1.16~\mathrm{a}$	$15.82\pm1.15~\mathrm{ab}$	$2.98\pm0.08~\mathrm{b}$
KC-25L	$5.94\pm1.23~\mathrm{a}$	$11.24\pm2.09~\mathrm{a}$	$2.35\pm0.06~\text{a}$

Table 2. SR and WVP of KC, KC-G and KC-L.

Different lowercase letters in the same column indicate significant difference (p < 0.05).

3.7. Thermal Stability

TGA and DTG curves of KC-GM films are shown in Figure 6. The decomposition parameters for GG, LG and all samples are listed in Table 3. For the KC films, the first stage was from 21.70 to 121.47 °C due to the evaporation of moisture. The second one from 121.47 to 227.43 °C was related to the pyrolysis of glycerol, and the third weight loss at 227.43~414.91 °C was attributed to the decomposition of polysaccharide. A similar thermal decomposition curve was observed with the cassia gum film added with a plasticizer [31]. The thermal decomposition temperature of the GM– κ -carrageenan composite films was higher than that of the polysaccharide with charges. The pyrolysis temperature of GGF was higher than that of LGF, which resulted in a lower thermal stability for KC-L.



Figure 6. TGA (a,c,e) and DTG (b,d,f) curves of KC, KC-G, KC-L, GGF and LGF.

Sample	To	T _m	T _E
КС	227.43	242.31	298.38
KC-5G	228.19	243.13	307.46
KC-10G	228.91	242.84	312.18
KC-15G	230.12	246.98	303.56
KC-20G	231.21	245.59	310.06
KC-25G	229.84	245.73	307.85
KC-5L	228.49	242.60	304.25
KC-10L	228.33	241.80	302.76
KC-15L	228.43	241.76	305.10
KC-20L	229.28	243.15	299.29
KC-25L	229.07	243.83	305.32
GGF	234.97	299.44	361.48
LGF	231.47	301.92	361.19

Table 3. Decomposition parameters of KC, KC-G, KC-L, GGF and LGF.

 T_O is the initial decomposition temperature of the polysaccharide. T_m is the temperature of maximum decomposition rate. T_E is the end temperature of the main decomposition of polysaccharide.

3.8. Rheological Analysis

Effects of galactomannan concentrations on steady-state shear properties of the films in gel states are shown in Figure 7a,b. The viscosity of the gel decreased with the shear rate range from 0.1 to 100 s^{-1} , which showed a pseudoplastic fluid. This shear-thinning behavior might be caused by the inability of the intermolecular entanglement structure to recombine under shear. The Ostwald–de Wale model was used to research the relationship between the shear rate and the apparent viscosity. As shown in Table 4, the value of R² was higher than 0.9, indicating that the Ostwald–de Wale model was suitable for evaluating the steady-state shear properties of the films in the gel state. All of the gels were pseudo-plastic fluids because the values of n were lower than 1.

The dynamic flow behaviors of gelled films were investigated at the linear viscoelastic region and the dissipation factor as a function of the angular frequency. Figure 7c,d shows that the dissipation factor (tan δ) of the films did not significantly change with the angular frequency from 0.1 to 100 rad/s, and its value was less than 1 (<0.15), indicating that the films in gel states had good long-term stability.



Figure 7. The steady-state shear properties (a,b) and the dissipation factor (c,d) of the films.

Sample	Ostwald-de Wale			
	K	n	R ²	
КС	197.353 ± 2.728	0.2956 ± 0.007	0.99939	
KC-5G	130.905 ± 1.146	0.297 ± 0.005	0.99975	
KC-10G	149.079 ± 0.860	0.326 ± 0.003	0.99988	
KC-15G	65.673 ± 1.319	0.462 ± 0.011	0.99717	
KC-20G	196.737 ± 2.272	0.277 ± 0.006	0.99961	
KC-25G	102.195 ± 2.143	0.515 ± 0.011	0.99578	
KC-5L	197.413 ± 2.318	0.254 ± 0.006	0.99963	
KC-10L	53.858 ± 1.240	0.277 ± 0.012	0.99846	
KC-15L	142.467 ± 11.551	0.578 ± 0.046	0.91772	
KC-20L	166.968 ± 10.988	0.614 ± 0.038	0.92924	
KC-25L	178.615 ± 1.934	0.246 ± 0.006	0.9997	

Table 4. The Ostwald-de Wale model parameters of the films.

4. Conclusions

In this research, the κ -carrageenan film properties were improved by the addition of galactomannan (guar gum or locust bean gum). The results showed that galactomannan (GM) has good compatibility with κ -carrageenan and could enhance the thermal stability of κ -carrageenan-based films. As the GM content increased, the TS of the composite films had little variation and exceeded 19 MPa; moreover, the barrier performance of the composite films also improved. The GM with the higher M/G ratio could have led to the lower OP, SR and WVP of the GM–carrageenan composite films. Therefore, green and safe GM–carrageenan films with certain strength and barrier properties have potential in the food packaging industry. This study will promote the edible film to be applied in the future.

Author Contributions: Conceptualization, Methodology, Investigation, Data curation, Writingoriginal draft, R.W.; Investigation, Methodology, S.Z.; Software, Validation, S.L.; Visualization, Y.S.; Conceptualization, Methodology, Supervision, Writing-review & editing, H.X. All authors have read and agreed to the published version of the manuscript.

Funding: This research was supported by Key scientific and technological projects of Jilin Provincial Science and technology development plan (20230202056NC).

Institutional Review Board Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

References

- Sajjan, A.M.; Naik, M.L.; Kulkarni, A.S.; Rudgi, U.F.-E.; Ashwini, M.; Shirnalli, G.G.; Sharanappa, A.; Kalahal, P.B. Preparation and characterization of PVA-Ge/PEG-400 biodegradable plastic blend films for packaging applications. *Chem. Data Collect.* 2020, 26, 100338. [CrossRef]
- Cazón, P.; Velazquez, G.; Ramírez, J.A.; Vázquez, M. Polysaccharide-based films and coatings for food packaging: A review. *Food Hydrocoll.* 2017, 68, 136–148. [CrossRef]
- Marques, G.S.; de Carvalho, G.R.; Marinho, N.P.; de Muniz, G.I.B.; Jorge, L.M.D.M.; Jorge, R.M.M. Production and characterization of starch-based films reinforced by ramie nanofibers (*Boehmeria nivea*). J. Appl. Polym. Sci. 2019, 136, 47919. [CrossRef]
- 4. Ahmed, A.; Boateng, J. Calcium alginate-based antimicrobial film dressings for potential healing of infected foot ulcers. *Ther. Deliv.* **2018**, *9*, 185–204. [CrossRef]
- Szymańska-Chargot, M.; Chylińska, M.; Pertile, G.; Pieczywek, P.M.; Cieślak, K.J.; Zdunek, A.; Frac, M. Influence of chitosan addition on the mechanical and antibacterial properties of carrot cellulose nanofibre film. *Cellulose* 2019, 26, 9613–9629. [CrossRef]
- Zarina, S.; Ahmad, I. Biodegradable Composite Films based on κ-carrageenan Reinforced by Cellulose Nano-crystal from Kenaf Fibers. *BioResources* 2014, 10, 258–271. [CrossRef]
- 7. Venkatesan, R.; Rajeswari, N.; Thendral Thiyagu, T. Preparation, characterization and mechanical properties of k-Carrageenan/SiO₂ nanocomposite films for antimicrobial food packaging. *Bull. Mater. Sci.* **2017**, *40*, 609–614. [CrossRef]
- 8. Liu, J.; Wang, H.; Wang, P.; Guo, M.; Jiang, S.; Li, X.; Jiang, S. Films based on κ-carrageenan incorporated with curcumin for freshness monitoring. *Food Hydrocoll.* **2018**, *83*, 134–142. [CrossRef]

- Sun, G.; Liang, T.; Tan, W.; Wang, L. Rheological behaviors and physical properties of plasticized hydrogel films developed from κ-carrageenan incorporating hydroxypropyl methylcellulose. *Food Hydrocoll.* 2018, 85, 61–68. [CrossRef]
- Bedane, A.H.; Eić, M.; Farmahini-Farahani, M.; Xiao, H. Water vapor transport properties of regenerated cellulose and nanofibrillated cellulose films. J. Membr. Sci. 2015, 493, 46–57. [CrossRef]
- 11. Aider, M. Chitosan application for active bio-based films production and potential in the food industry: Review. *LWT—Food Sci. Technol.* **2010**, *43*, 837–842. [CrossRef]
- 12. Shahbazi, M.; Majzoobi, M.; Farahnaky, A. Physical modification of starch by high-pressure homogenization for improving functional properties of κ-carrageenan/starch blend film. *Food Hydrocoll.* **2018**, *85*, 204–214. [CrossRef]
- 13. Nur Fatin Nazurah, R.; Nur Hanani, Z.A. Physicochemical characterization of kappa-carrageenan (*Euchema cottoni*) based films incorporated with various plant oils. *Carbohydr. Polym.* **2017**, *157*, 1479–1487. [CrossRef]
- Rane, L.R.; Savadekar, N.R.; Kadam, P.G.; Mhaske, S.T. Preparation and Characterization of K-Carrageenan/Nanosilica Biocomposite Film. J. Mater. 2014, 2014, 1–8. [CrossRef]
- Martins, J.T.; Cerqueira, M.A.; Souza, B.W.S.; Avides, M.D.C.; Vicente, A.A. Shelf Life Extension of Ricotta Cheese Using Coatings of Galactomannans from Nonconventional Sources Incorporating Nisin against *Listeria monocytogenes*. J. Agric. Food Chem. 2010, 58, 1884–1891. [CrossRef]
- 16. García-García, E.; Totosaus, A. Low-fat sodium-reduced sausages: Effect of the interaction between locust bean gum, potato starch and κ-carrageenan by a mixture design approach. *Meat Sci.* **2008**, *78*, 406–413. [CrossRef]
- 17. Zhao, N.; Chai, Y.; Wang, T.; Wang, K.; Jiang, J.; Yang, H.-Y. Preparation and physical/chemical modification of galactomannan film for food packaging. *Int. J. Biol. Macromol.* **2019**, *137*, 1060–1067. [CrossRef]
- Barddal, H.P.D.O.; Faria, F.A.M.; Nogueira, A.V.; Iacomini, M.; Cipriani, T.R. Anticoagulant and antithrombotic effects of chemically sulfated guar gum. *Int. J. Biol. Macromol.* 2019, 145, 604–610. [CrossRef]
- Dea, I.C.M.; McKinnon, A.A.; Rees, D.A. Tertiary and quaternary structure in aqueous polysaccharide systems which model cell wall cohesion: Reversible changes in conformation and association of agarose, carrageenan and galactomannans. *J. Mol. Biol.* 1972, 68, 153–172. [CrossRef]
- 20. Wu, Y.; Ding, W.; He, Q. The gelation properties of tara gum blended with κ-carrageenan or xanthan. *Food Hydrocoll.* **2018**, 77, 764–771. [CrossRef]
- Martins, J.T.; Bourbon, A.I.; Pinheiro, A.C.; Souza, B.W.S.; Cerqueira, M.; Vicente, A.A. Biocomposite Films Based on κ-Carrageenan/Locust Bean Gum Blends and Clays: Physical and Antimicrobial Properties. *Food Bioprocess Technol.* 2012, 6, 2081–2092. [CrossRef]
- 22. De Paola, M.G.; Paletta, R.; Lopresto, C.G.; Lio, G.E.; De Luca, A.; Chakraborty, S.; Calabrò, V. Stability of Film-Forming Dispersions: Affects the Morphology and Optical Properties of Polymeric Films. *Polymers* **2021**, *13*, 1464. [CrossRef]
- 23. Hou, X.; Xue, Z.; Liu, J.; Yan, M.; Xia, Y.; Ma, Z. Characterization and property investigation of novel eco-friendly agar/carrageenan/TiO₂ nano-composite films. *J. Appl. Polym. Sci.* **2019**, *136*, 47113. [CrossRef]
- 24. Kassab, Z.; Aziz, F.; Hannache, H.; Ben Youcef, H.; El Achaby, M. Improved mechanical properties of k-carrageenan-based nanocomposite films reinforced with cellulose nanocrystals. *Int. J. Biol. Macromol.* **2019**, *123*, 1248–1256. [CrossRef]
- Balqis, A.I.; Khaizura, M.N.; Russly, A.R.; Hanani, Z.N. Effects of plasticizers on the physicochemical properties of kappacarrageenan films extracted from *Eucheuma cottonii*. Int. J. Biol. Macromol. 2017, 103, 721–732. [CrossRef]
- Martins, J.T.; Cerqueira, M.A.; Bourbon, A.I.; Pinheiro, A.C.; Souza, B.W.; Vicente, A.A. Synergistic effects between κ-carrageenan and locust bean gum on physicochemical properties of edible films made thereof. *Food Hydrocoll.* 2012, 29, 280–289. [CrossRef]
- 27. Williams, P.A. Molecular interactions of plant and algal polysaccharides. *Struct. Chem.* 2009, 20, 299–308. [CrossRef]
- Rao, M.; Kanatt, S.; Chawla, S.; Sharma, A. Chitosan and guar gum composite films: Preparation, physical, mechanical and antimicrobial properties. *Carbohydr. Polym.* 2010, 82, 1243–1247. [CrossRef]
- 29. Mostafavi, F.S.; Kadkhodaee, R.; Emadzadeh, B.; Koocheki, A. Preparation and characterization of tragacanth–locust bean gum edible blend films. *Carbohydr. Polym.* **2016**, *139*, 20–27. [CrossRef]
- Kurt, A.; Kahyaoglu, T. Characterization of a new biodegradable edible film made from salep glucomannan. *Carbohydr. Polym.* 2014, 104, 50–58. [CrossRef]
- Cao, L.; Liu, W.; Wang, L. Developing a green and edible film from Cassia gum: The effects of glycerol and sorbitol. J. Clean. Prod. 2018, 175, 276–282. [CrossRef]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.