

Development and Characterization of Bioactive Edible Films Based on Semi-Refined Kappa Carrageenan Incorporated with Honey and *Kaempferia galanga* L. Essential Oil

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Abstract

This paper emphasized the innovation of commercial semi-refined kappa carrageenan-based (SRKC) films incorporated with *Ceiba pentandra* honey and *Kaempferia galanga* L. essential oil to develop bioactive edible film. The influence of honey and essential oil concentrations on the mechanical, physical, moisture barrier, and sensory properties has been investigated. It was found that the thickness, solubility, elongation at break of the edible film with honey addition (50, 100, 150, and 200 % w/w carrageenan) were increased ($p < 0.05$), whereas the tensile strength was decreased ($p < 0.05$), and there was no significant difference in the water vapor transmission rate of all the films ($p > 0.05$). In the next stage, increment of essential oil concentration (0.25, 0.5, and 1 % w/v) in SRKC film with 200 % honey increased the thickness, but reduced the solubility, tensile strength, and elongation at break, and had no significant influence on the water vapor transmission rate ($p > 0.05$). The edible film incorporated with 200 % honey and 0.25 % *Kaempferia galanga* L. essential oil demonstrated a significant effect on the sensory evaluation test on aroma, taste, and overall attributes ($p < 0.05$). Furthermore, the edible films also encompassed many bioactive compounds. In summary, the present research findings confirmed the potentials of SRKC edible film enriched with 200 % honey and 0.25 % *Kaempferia galanga* L. essential oil as a novel edible food packaging with enhanced bioactive characteristics.

Keywords: Edible film, Kappa carrageenan, *Kaempferia galanga* L. essential oil, Honey, Characterization

Introduction

Edible films serve as potential alternatives to petrochemical-based packaging responsible for instigating severe environmental challenges, specifically in terms of pollution, therefore many researchers have extensively studied this promising topic [1]. They function as a protective barrier that able to maintain food stability by reducing the transfer of water vapor, flavor volatiles, gasses, lipid, and preventing food contamination [1,2]. Several biopolymer types derived commonly from proteins, polysaccharides, and lipids have been developed as the primary film matrix [3]. Among these film-forming materials, semi-refined kappa carrageenan (SRKC) possessed the capacity to form a matrix with useful properties [2,4,5]. Unlike the refined form of kappa carrageenan, semi-refined kappa carrageenan is produced through shorter extraction steps making it available at a lower price [2]. The cellulose residue remaining in SRKC makes the solution and gel less clear. Nonetheless, it may enhance the strength of the gel produced [6]. However, as the term edible film refers to a ready-to-consume packaging material alongside the actual food [7], the constituents have recently become a significant consideration, not only in terms of performance but also in safety and functional properties.

The incorporation of certain substances with functional properties into edible films tends to enhance their properties and enrich their characteristics with bioactive functionality [8]. Conversely, the direct application of volatile and active compounds, e.g. essential oils, into a food product appears challenging and ineffective, due to particular attributes, including water-insolubility, volatility, and specific odor. Therefore, incorporating these compounds into edible films is possibly an effective application in preserving food products as well as exhibiting bioactivities [9].

Several researchers have incorporated the essential oils as natural antimicrobial carriers in biopolymer-based films for improved antimicrobial activity and physical characteristics [10-12]. For instance, a particular essential oil offering high potentials is the *Kaempferia galanga* L. essential oil. This species originated from the *Zingiberaceae* family, known to demonstrate vital medicinal properties for treating rheumatism, dyspepsia, hypertension, toothache, abdominal and pectoral pains. It is predominantly cultivated in Asia's tropics and subtropics, including Indonesia, Thailand, Malaysia, India, and China [13,14]. In addition, the active component, ethyl-p-methoxycinnamate is characterized with various bioactivities, termed anti-inflammatory and anti-angiogenic effects [15], vasorelaxation [16], and sedative activity [17], while the rhizome performs several health impacts, including hypocholesterolemia and antitussive conditions [18]. Furthermore, the essential oil showed antimicrobial [19] and antioxidant activities [20]. Despite these possibilities, the incorporation of *Kaempferia galanga* L. essential oil into biopolymer-based film has not been previously conducted.

Honey, another potential substance, is also possible to introduce, based on the rich functional properties. Honey has been employed for centuries as a natural sweetener with a variety of health benefits, due to its high nutritional value and bioactive compounds [21,22]. In addition, further investigations revealed antimicrobial [23] and antioxidant activities [24], as well as anti-inflammatory impacts [25]. Honey collected from the kapok tree (*Ceiba pentandra*) has been reported to have high phenolic content (926.98 mg gallic acid/kg) and also exhibit beneficial antimicrobial and antioxidant properties [26]. Moreover, honey is also applied as a potential plasticizer for edible film, as demonstrated in whey protein films [27]. On the other hand, the high sugar content in honey [23] will contribute to giving a fine taste to edible film.

By considering the above conditions, the present study was aimed to determine the effects of incorporating *Ceiba pentandra* honey and *Kaempferia galanga* L. essential oil in various concentrations on mechanical, physical, chemical, and sensory characteristics of the semi-refined kappa carrageenan edible film in order to develop a bioactive edible film which possesses not only better mechanical and physical properties but also functional properties from the loaded bioactive compounds.

Materials and methods

Materials

The materials used for film preparation were commercial food-grade semi-refined kappa carrageenan (SRKC) powder from Galic Artabahari, Co., Ltd. (Bekasi, Indonesia) with the following specifications: derived from *Kappaphycus alvarezii*, water-soluble at 60 °C or more, boiling point at 85 °C, has a maximum moisture content of 12 %, a maximum ash content of 35 %, and a cream to light brown color; plasticizer sorbitol (Brataco, Co., Ltd., Bekasi, Indonesia); *Ceiba pentandra* honey in its pure form with dark amber color obtained from Perum Perhutani Forest Management Unit (Surakarta, Indonesia); and *Kaempferia galanga* L. essential oil from Orizho Indonesia, Ltd. (Bantul, Indonesia) which extracted from fresh *K. galanga* L. rhizome through water distillation technic and produced oil with a clear yellow color and has a specific aroma. All the chemicals and solvents used for analysis were of analytical grade.

Edible film preparations

Edible film with honey incorporation

The edible film was prepared, using the modified technique of Praseptiangga *et al.* [2]. 2 g of SRKC powder was dissolved in distilled water (less than 100 mL) and heated with continuous stirring at 60 °C until totally dissolved. Subsequently, sorbitol (30 % w/w carrageenan), honey (50, 100, 150, and 200 % w/w carrageenan), and distilled water (to reach a total volume of 100 mL) were consecutively mixed into the film solution. The temperature was elevated to 80 °C and maintained for 5 min. After that, the film solution was poured into a 15×23×2 cm plastic mold and left for about 20 min before drying in a drying oven (type MOV-112, SANYO, Japan) at 50 °C for 12 h. The dried film was finally detached for characterizations to determine the film with the best honey concentration.

Edible film with honey and Kaempferia galanga L. essential oil incorporation

A 2 g SRKC powder was dissolved in distilled water (less than 100 mL) and heated using a hot plate under continuous stirring at 60 °C until totally dissolved. Subsequently, sorbitol (30 % w/w carrageenan), and honey (best concentration) were consecutively mixed with the film solution. Consequently, the temperature was extended to 80 °C, while the *Kaempferia galanga* L. (0.25, 0.5, and 1 % w/v) and distilled water (to reach a total volume of 100 mL) were mixed at a constant temperature (80

°C) for 5 min, followed by pouring the film solution into a 15×23×2 cm plastic mold and left for approximately 20 min before drying in a drying oven (type MOV-112, SANYO, Japan) at 50 °C for 12 h. The dried film was finally peeled from the mold for characterizations to determine the film with the best *Kaempferia galanga* L. essential oil concentration.

Film characterizations

Thickness

Film thickness was measured by a digital micrometer (type KW06-85, KRISBOW, Indonesia) at 5 distinct positions with 0.001 mm accuracy according to Manuhara *et al.* [5].

Water solubility

The film solubility in water test was conducted according to a modified method of Farhan and Hani [6]. Edible film samples were finely divided to a size of 2×2 cm, and dried at 105 °C for 6 h before weighing (w_1). Subsequently, the individual component was added into a 100 mL Erlenmeyer flask comprising 10 mL distilled water, followed by storing for 6 h at room temperature (25 °C) and stirred periodically using an orbital shaker (type KS 130 basic, IKA, Germany) at 240 rpm. The solution was filtered and the filter paper ($d = 15$ cm) (Grade 41, Whatman) was oven-dried at 105 °C for 6 h. This paper was then weighed to ascertain the water-insoluble dry matter (w_2). Furthermore, the solubility was calculated using the Eq. (1);

$$\% \text{ Solubility} = \frac{w_1 - w_2}{w_1} \times 100 \quad (1)$$

Mechanical properties

The mechanical properties of the edible films were examined on the modified method of ASTM D 882-02 standard [28]. These features comprised tensile strength (TS) and the elongation at break (EAB). The ends of the specimen (10×2.5 cm) were grasped with the grips of the testing machine (type Z0.5, zwickiLine, Germany) which was run at a constant rate of separation (10 mm/min) to attain breaking point. Subsequently, TS and EAB were determined using the Eqs. (2) - (3);

$$\text{TS (MPa)} = \frac{\text{maximum load (N)}}{\text{initial cross sectional area (m}^2\text{)}} \quad (2)$$

$$\text{EAB (\%)} = \frac{\text{elongated length at break}}{\text{initial length}} \times 100 \quad (3)$$

Water vapor transmission rate (WVTR)

The film sample with a diameter of 7.5 cm was sealed on the test cup surface which contained 10 g of silica gels. The test cup was then put into a desiccator comprising a saturated NaCl solution to maintain the RH at 75 %, with a temperature of 28 ± 2 °C. The test cup was weighed every hour for the subsequent 8 h and the weight gained over time was plotted using linear regression analysis to determine the slope (g/h). WVTR was calculated by dividing the slope (g/h) with the film surface area (m^2) [5].

Determination of the best honey and essential oil concentration

A compensatory approach was used to determine the honey or essential oil concentration which produced the best film properties. This study employed a non-dimensional scaling model by Sullivan *et al.* [29], where each parameter was assigned a variable weight (VW) between 0 - 1, depending on the contribution value to film quality. Subsequently, normalization weight (NW) and non-dimensional value (NV) were calculated, using the Eqs. (4) - (5);

$$\text{NW} = \frac{\text{VW}}{\sum \text{VW}} \quad (4)$$

$$\text{NV} = \frac{\text{value} - \text{worst value}}{\text{best value} - \text{worst value}} \quad (5)$$

The variable scores in each film type were calculated by multiplying NW with NV, followed by the summation of the resulting values to obtain the total score of individual edible film. In addition, the film with the highest total score was determined as the best film type (selected concentration).

Sensory evaluation

The difference-from-control (preference) test was used in the sensory evaluation, referring to Meilgaard *et al.* [30], to determine the influence of adding honey and essential oil on the color, aroma, solubility in the mouth, taste, and overall attributes of the edible film. The SRKC film (without honey and essential oil addition) was designed as the “reference/control” and the 2 other samples (SRKC film with the best honey concentration and SRKC film with the best honey and essential oil concentration) were evaluated with respect to how different each was from that control. The difference was expressed in preference, whether a sample was preferable or not to the control. Thirty untrained panelists were involved which were selected based on their good health condition and their preference for herbal products.

Gas chromatography-mass spectrometry (GC-MS)

The chemical composition of essential oil and an edible film containing essential oil was analyzed by the GC-MS method according to the modified method of Praseptiangga *et al.* [31]. The edible film was initially prepared into the supernatant by the consecutive steps: 1 g of finely divided samples were placed in a closed test tube containing 5 mL distilled water and heated in a water bath (110 °C) for 30 min. After cooling, 5 mL of n-hexane was added and vortexed to form the supernatant (n-hexane phase). The GC-MS test was performed using Agilent 6890N GC (United States) equipped with Agilent 5973 inert MSD detector and HP-5 fused-silica capillary column. The oven temperature was set from 50 to 280 °C at a rate of 10 °C/min with an initial hold time of 5 min at 50 °C and a final hold time of 5 min at 280 °C. The inlet temperature was 290 °C and the flow rate of carrier gas (helium) was 1.5 mL/min (constant). While the MS Quad and MS Source temperatures were set to 150 and 230 °C, respectively. The mass scan range was 20 - 800 amu. Furthermore, 2 µL of the supernatant was injected into the GC-MS device. Meanwhile, for the analysis of essential oil components, 2 µL of essential oil sample was injected directly without being prepared into a supernatant. The chemical composition of each sample was identified by matching the mass spectra with the GC-MS databases recorded in the Wiley Spectral Libraries.

Data analysis

The overall test results were statistically examined, using the one-way analysis of variance (ANOVA) method at the significance level of $\alpha = 5\%$, and if there was a difference, Duncan’s multiple range test (DMRT) was then applied. The statistical analyses were run using SPSS 16.0 software.

Results and discussion

Edible films with honey incorporation

Thickness

Table 1 shows the addition of honey with an increased concentration between 50 - 200 %, significantly enhanced the film thickness from 0.060 to 0.109 mm ($p < 0.05$). This circumstance indicates a more intense solute portion as the inclusion also elevated the total dissolved solids, resulting in additional thickness. Based on Farhan and Hani [6], the distance between polymer chains in the matrix was possibly extended as a result of dispersed plasticizer molecules believed to contribute to the increased thickness. In addition, the production process was also an influencing factor. However, a previous study revealed a similar trend in SRKC film thickness by increasing the amount of plasticizer [2].

Water solubility

In terms of edible packaging film, solubility appears very necessary for consumption purposes [32]. Soluble materials are essential to employ in ready-to-eat products for easy melting and dissolution in the mouth or when boiled in water [33]. In this study, the film solubility in water was enhanced by the increment in honey concentration (**Table 1**). In addition, substantial improvement ($p < 0.05$) occurred as the honey concentration was increased from 50 to 100 and then 150 %. Meanwhile, no significant difference ($p > 0.05$) was observed in the solubility at 150 and 200 %. This phenomenon is due to the honey’s plasticizing effect, where the increase in the plasticizer was known to reduce the intermolecular bonds between the polymer bonds, and therefore promoted the interaction of hydrophilic chain and plasticizer with water molecules, leading to higher film solubility and permeability [32]. The extended intermolecular spacing between the polymers due to the presence of honey molecules which was expressed by increasing thickness may also explain this phenomenon as it offers more available hydrophilic molecules in the matrix to interact with the water [6].

Mechanical properties

The film's tensile strength (TS) was significantly reduced along with increasing honey concentration, while the flexibility presented in elongation at break (EAB) tended to improve ($p > 0.05$) (Table 1). These conditions indicate that the presence of honey probably reduces the polymer chain intermolecular interactions which on the 1 side decreases the hardness, deformation tension, and density of the matrix, while on the other side expands the chain flexibility [34]. Another study also reported a similar trend, where TS of whey protein films declined and the elongation became extensive. The glucose and fructose in honey were assumed to play a major role in the plasticizing effect [27].

Water vapor transmission rate (WVTR)

Table 1 shows the water vapor transmission rate (WVTR) of the edible film fairly increased with added honey. Despite that the increment was generally insignificant, the film with 150 % honey content resulted in a significantly higher WVTR, compared to 50 % honey content ($p < 0.05$). As a consequence, the barrier properties were declined as against water vapor molecules. However, the hydrophilic nature of either SRKC or honey was probably responsible for the increase in water vapor permeability. This circumstance was possibly described by the reduction in structural density of polymer network with increasing hydrophilic materials. Also, the presence of honey modified the intermolecular interactions between the polymer chains by extending the molecular network distance to allow water molecules to diffuse more easily, resulting in a higher WVTR [6]. Consequently, the insignificant difference of WVTR between the entire films with different honey concentrations matched the results in another investigation, where a stable moisture permeability of whey protein films was reported, despite increasing honey intensity [27].

Table 1 The properties of SRKC-based edible film incorporated with various concentrations of honey.

Honey concentration (%)	Thickness (mm)	Water solubility (%)	Tensile strength (MPa)	Elongation at break (%)	WVTR (g/h.m ²)
50	0.060 ± 0.000 ^a	56.55 ± 3.34 ^a	45.14 ± 3.65 ^d	5.54 ± 1.96 ^a	16.56 ± 0.32 ^a
100	0.076 ± 0.000 ^b	67.51 ± 3.03 ^b	32.16 ± 1.18 ^c	15.69 ± 3.94 ^b	16.88 ± 0.47 ^{ab}
150	0.091 ± 0.000 ^c	72.72 ± 2.28 ^c	21.64 ± 0.57 ^b	17.63 ± 2.06 ^b	17.15 ± 0.48 ^b
200	0.109 ± 0.004 ^d	75.82 ± 2.52 ^c	17.68 ± 0.61 ^a	25.73 ± 3.47 ^c	17.09 ± 0.36 ^b

Note: Values are presented as mean ± SD (n = 6). Different letters in the same column indicate significantly different ($p < 0.05$).

Determination of the best honey concentration

The determination of honey concentration used to produce the best edible film was based on 4 parameters/variables, termed tensile strength, water solubility, elongation at break, and water vapor transmission rate. In this study, all the parameters were assigned a variable weight (VW) of 1, indicating equal importance to the film quality. Under this condition, an effective edible film showed high solubility, tensile strength, and elongation at break values, but a low water vapor transmission rate. Subsequently, the total score on each edible film was calculated on the basis of these parameters. The total scores collected for the film in 50, 100, 150, and 200 % honey were estimated at 0.500, 0.514, 0.396, and 0.525, respectively (Table 2). According to these calculations, the edible film with the highest score and the best honey concentration was 200 % (4 g). This concentration was applied in subsequent stages involving edible film preparation with various essential oil concentrations.

Table 2 Determination of the best honey concentration using non-dimensional scaling model.

Honey concentration (%)	Variable score				Total score
	Water solubility	Tensile strength	Elongation at break	WVTR	
50	0.000	0.250	0.000	0.250	0.500
100	0.142	0.132	0.126	0.114	0.514
150	0.210	0.036	0.150	0.000	0.396
200	0.250	0.000	0.250	0.025	0.525

Edible films with honey and *Kaempferia galanga* L. essential oil

Thickness

The edible film thickness showed a tendency to incline as *Kaempferia galanga* L. essential oil concentration becomes more intense. **Table 3** specified the variation between 0.109 - 0.130 mm which was significantly different toward each of the film samples ($p < 0.05$). This occurrence is associated with an improvement in the total solid content, due to essential oil addition [35]. In addition, the thickness was also dependent on certain drying conditions, the total volume of solution, and applied mold size [2]. Other studies also suggested a similar trend, where Praseptiangga *et al.* [2] reported a significant increase in the thickness of SRKC edible film comprising cinnamon essential oil, while Shojaee-Aliabadi *et al.* [36] observed greater thickness in kappa carrageenan films with increased concentration of *Satureja hortensis* essential oil.

Water solubility

Based on **Table 3**, enhancing the concentration of *Kaempferia galanga* L. essential oil also influenced the solubility of SRKC films. The addition of the oil from 0.25 - 1 % significantly reduced ($p < 0.05$) the water solubility, attributed to the hydrophobic nature [37]. Moreover, the hydrophilic component contributed to the solubility increase, while the hydrophobic factor was responsible for the decline [38]. Therefore, incorporating additional hydrophobic substances into the hydrophilic biopolymer matrix, e.g., SRKC, tends to limit the interactions between hydroxyl groups and water molecules, thus resulting in a more water-resistant film [36]. A similar finding was also observed in other studies, including kappa carrageenan-based film incorporated with *Zataria multiflora* Boiss and *Mentha pulegium* essential oils [39], *Satureja hortensis* essential oil [36], as well as with oregano and thyme essential oils [37].

Mechanical properties

Table 3 shows the effects of incorporating various concentrations of *Kaempferia galanga* L. essential oil on the mechanical characteristics, with the range of 0.25 - 1 % significantly decreasing the TS of the edible films ($p < 0.05$). This decline is probably correlated with the robust electrolyte property of carrageenan permitting it to bind with other molecules, e.g. essential oil, creating less resilient interaction, compare to polymer-polymer interaction, which may cause discontinuities within the film matrix, and therefore, resulting in a vulnerable network structure [39]. The occupancy of the essential oil inside the film matrix may also contribute to loosening the internal structure and reducing its cohesiveness which consequently lowering the TS [35,40]. Furthermore, the decline in TS due to the essential oil addition also occurred in kappa carrageenan-based film with *Zataria multiflora* Boiss and *Mentha pulegium* essential oils [39], *Satureja hortensis* essential oil [36], as well as with oregano and thyme essential oils [37]. However, after the reduction in TS occurred in the addition of the essential oil from 0.25 to 0.5 %, the increasing amount of the *K. galanga* essential oil from 0.5 to 1 % was followed by an improvement of the TS. This may be associated with the particular interaction between the polymers and the essential oil that possibly occurred at a certain ratio. This molecular interaction could strengthen the structural bond within the network resulting in a higher TS value [41].

Conversely, the EAB of the SRKC film with 0.25 % added *Kaempferia galanga* L. essential oil (29.12 %) was higher, compared to the film without the essential oil (25.73 %). However, the increase to 0.5 % was known to decrease the EAB to 23.04 % but subsequently recovered to 26.17 % as the oil expanded to 1 %. This circumstance assumes the essential oil incorporation at low concentration into biopolymer film tends to enhance film elongation, but at a higher amount, the reduction was observed. In addition, the plasticizing effect of the essential oil was probably responsible for the cumulative elongation. Also, the interaction between the essential oil and carrageenan tends to reduce the polymer cohesion, allowing easier chain movement during film stretching [39]. Meanwhile, the decline of EAB was probably associated with a higher amount of essential oil, leading to several discontinuities in the film matrix [42]. However, the increase in EAB was investigated in kappa carrageenan-based film with *Zataria multiflora* Boiss and *Mentha pulegium* [39] and *Satureja hortensis* essential oil additions [36]. Moreover, other studies revealed the elongation of biopolymer film decreased with the incorporation of essential oil as reported in SRKC edible film containing cinnamon essential oil [2], a carrageenan-based edible film with oregano and thyme essential oils [37], as well as a chitosan-based edible film containing bergamot essential oil [42].

Water vapor transmission rate (WVTR)

Table 3 shows the incorporation of *Kaempferia galanga* L. essential oil from 0.25 - 1 % reduced the WVTR of the edible films, although not significantly different ($p > 0.05$). This decrease was possibly attributed to the essential oil's hydrophobic state. Water vapor transmission depends on the ratio of hydrophobic-hydrophilic portion influencing the diffusion of water vapor molecules. The hydrophobic or nonpolar substances are evenly distributed across the film matrix to enhance the hydrophobicity and consequently lower the diffusion and adsorption of water vapor molecules [35]. Also, the presence of lipid globules plays an important role in developing a tortuous diffusion pathway [43]. Sufficient water vapor barrier observed due to the incorporation of essential oil into biopolymer films was consistent with other studies, including carboxymethyl cellulose/chitosan-based films with frankincense essential oil [12], kappa-carrageenan films with *Satureja hortensis* essential oil [36], pullulan-based films with cinnamon essential oil addition [40], and pectin films with copaiba oil nanoemulsions [44].

Table 3 The properties of SRKC-based edible film incorporated with 200 % honey and various concentrations of *Kaempferia galanga* L. essential oil.

Essential Oil concentration (%)	Thickness (mm)	Water solubility (%)	Tensile strength (MPa)	Elongation at break (%)	WVTR (g/h.m ²)
0.25	0.109 ± 0.001 ^a	96.98 ± 1.53 ^c	14.70 ± 0.71 ^c	29.12 ± 1.71 ^c	12.99 ± 0.35 ^a
0.5	0.113 ± 0.001 ^b	93.18 ± 1.76 ^b	11.46 ± 0.46 ^a	23.04 ± 2.33 ^a	12.92 ± 0.64 ^a
1	0.130 ± 0.003 ^c	89.45 ± 0.42 ^a	13.13 ± 0.79 ^b	26.17 ± 1.38 ^b	12.88 ± 0.69 ^a

Note: Values are presented as mean ± SD (n = 6). Different letters in the same column indicate significantly different ($p < 0.05$).

Determination of the best essential oil concentration

Determining the concentration of essential oil responsible for the best produced edible film was also based on 4 parameters (variables), termed tensile strength, water solubility, elongation at break, and water vapor transmission rate, with variable weight (VW) of 1, indicating mutual relevance to film quality. Subsequently, the overall score on each edible film was calculated in terms of these parameters. The total scores collected for the film with 0.25, 0.5, and 0.75 % essential oil were estimated at 0.750, 0.278, and 0.508 respectively (**Table 4**). Based on these calculations, the 0.25 % essential oil was the best concentration due to the highest score obtained.

Table 4 Determination of the best essential oil concentration using non-dimensional scaling model.

Essential oil concentration (%)	Variable score				Total score
	Water solubility	Tensile strength	Elongation at break	WVTR	
0.25	0.250	0.250	0.250	0.000	0.750
0.5	0.124	0.000	0.000	0.154	0.278
1	0.000	0.129	0.129	0.250	0.508

Sensory evaluation test

Sensory evaluation was conducted to evaluate the organoleptic properties (sensory attributes) of the edible films based on the perception of panelists. The sensory properties of the edible film become an important consideration as it served along with the food product [45]. **Table 5** represents the results of the sensory evaluation test. Based on the analysis of variance, the sample films (SRKC film with honey and SRKC film with honey and *Kaempferia galanga* L. essential oil) demonstrated a significant difference (in panelist preference) from the reference film (SRKC film without honey and essential oil) on the attributes of aroma, taste, and overall performance ($p < 0.05$). Meanwhile, no major difference was observed in the color and solubility attributes.

Table 5 The sensory properties of SRKC-based edible film incorporated with honey and *Kaempferia galanga* L. essential oil.

Tested film	Sensory attributes				
	Color	Aroma	Solubility	Taste	Overall
H ₂₀₀ K ₀	3.43 ± 1.48 ^a	2.87 ± 0.90 ^a	3.33 ± 1.18 ^a	2.87 ± 0.73 ^a	2.93 ± 0.91 ^a
H ₂₀₀ K _{0.25}	3.30 ± 1.86 ^a	1.73 ± 1.20 ^b	2.80 ± 1.42 ^a	1.97 ± 1.27 ^b	2.07 ± 0.87 ^b

Notes: (n = 30)

- 1) H₂₀₀K₀ = SRKC-based edible film with 200 % honey.
- 2) H₂₀₀K_{0.25} = SRKC-based edible film with 200 % honey and 0.25 % *Kaempferia galanga* L. essential oil.
- 3) Score 1 = highly preferable; 2 = preferable; 3 = slightly preferable; 4 = equal to R; 5 = slightly not preferable; 6 = not preferable; 7 = highly not preferable.
- 4) Different letters within a column indicates significantly different from R (in preference) ($p < 0.05$).
- 5) R (Reference) = SRKC-based edible film without honey and *Kaempferia galanga* L. essential oil.

On the aroma attribute, the edible film with added essential oil was preferred to edible film without the addition of essential oil. The addition of honey and the pleasant aromatic odor emitted from the essential oil tend to conceal the fishy semi-refined carrageenan odor, in a bid to increase the possibility of the panelists' acceptance of the aroma attribute. In terms of taste, the edible film with the addition of honey and essential oil was preferable, compared to edible film without the addition of essential oil, due to the sweet taste of honey from its high fructose content [23] and slightly spicy, warm taste from the *K. galanga* L. essential oil [19]. On the overall attribute, the edible film with additional essential oil exhibited substantial preference, compared to the edible film without the addition of essential oil.

Chemical composition of *Kaempferia galanga* L. essential oil and edible film

The refractive index and density of the *Kaempferia galanga* L. essential oil in this paper were analyzed at 1.47 and 0.9230 g/mL, respectively. **Table 6** represents the chemical composition of the essential oil and the edible film comprising 200 % honey and 0.25 % essential oil.

Table 6 Chemical composition of *Kaempferia galanga* L. essential oil and SRKC-based edible film with 200 % honey and 0.25 % *Kaempferia galanga* L. essential oil.

No.	Chemical composition	Essential oil (%)	Edible film (%)
1	2-Ethyl cyclopentanone	29.55	-
2	Cinnamaldehyde	17.84	-
3	Cyclopropane	11.68	-
4	Ethyl p-methoxycinnamate	10.32	13.29
5	1,5-cyclodecadiene	3.64	-
6	8-Heptadecene	2.18	-
7	Cyperene	2.03	2.17
8	Heptadecane	1.42	2.12
9	Champene	1.14	1.03
10	α -pinene	1.02	1.67
11	δ -cadinene	1.02	-
12	α -gurjunene	0.93	0.92
13	Germacrene	0.81	0.99
14	α -humulene	0.74	0.69
15	Limonene	0.67	0.71
16	<i>Trans</i> -caryophyllene	0.66	1.42
17	δ -3-carene	0.08	12.88
19	Pentadecane	-	42.96
20	Ethyl-cinnamate	-	9.45
21	1,6-cyclodecadiene	-	2.48
22	γ -cadinene	-	2.20
23	β -cadinene	-	1.84
24	Cyclododecane	-	1.04

Note: Component profiling was using a similarity index of at least 50 %

Major constituents of the essential oil were 2-ethyl cyclopentanone, cinnamaldehyde, cyclopropane, and ethyl p-methoxycinnamate, while for the edible film, pentadecane, ethyl p-methoxycinnamate, delta-3-carene, ethyl-cinnamate were observed. The compositional difference between the essential oil and the edible film was probably associated with the presence of other materials, including honey, carrageenan, sorbitol, possible chemical interaction between the entire constituent film materials, and also the applied treatment process.

Meanwhile, the majority of other studies revealed ethyl p-methoxycinnamate, ethyl-cinnamate, and pentadecane as the primary chemical components in the *Kaempferia galanga* L. essential oil [19]. Several factors were believed to influence the chemical composition of essential oil, including growth location, soil quality, climate, cultivar, post-harvest, treatment process e.g drying, storage conditions, and extraction methods [9,14]. The incorporation of this bioactive substance promotes additional values in the resulting film due to its bioactive components. Chemical compounds loaded in the *K. galanga* L. essential oil are closely related to its functional properties. Those major components have been reported in exhibiting various bioactivities including antimicrobial activities, pharmacological properties, and antioxidant activities [19]. Its flavor and aromatic characteristic also offered the edible film with preferable sensory properties, as evaluated in the sensory test above.

Conclusions

This research investigated the influence of honey and *Kaempferia galanga* L. essential oil, at various concentrations, on SRKC-based edible film properties. Based on the results and discussions, the increase in honey concentration generated a corresponding improvement in thickness, solubility, and elongation of the edible film, but showed a decline in tensile strength. Subsequently, the increment in essential oil concentration in SRKC film with 200 % honey also increased the thickness but reduced the tensile strength, solubility, and elongation at break. However, no significant effect on water vapor transmission rate was reported. Therefore, 200 % honey and 0.25 % *Kaempferia galanga* L. essential oil were selected as the best concentration to incorporate into the SRKC film. Furthermore, the sensory evaluation revealed the preference of SRKC film with the *Kaempferia galanga* L. essential oil addition over the film without the essential oil on aroma, taste, and overall attributes. The edible film was also confirmed to contain several bioactive compounds with pentadecane, ethyl p-methoxycinnamate, delta-3-carene, ethyl-cinnamate as the major constituents. Based on these findings, SRKC edible film enriched with honey and *Kaempferia galanga* L. essential oil showed significant potential as a new edible food packaging material offering enhanced bioactive characteristics. In addition, further study regarding the functional properties and bioactivity of this *K. galanga* essential oil-loaded film such as antimicrobial and antioxidant activity should be carried out.

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References

- [1] SAA Mohamed, M El-Sakhawy and MAM El-Sakhawy. Polysaccharides, protein and lipid -based natural edible films in food packaging: A review. *Carbohydr. Polymer*. 2020; **238**, 116178.
- [2] D Praseptianga, N Fatmala, GJ Manuhara, R Utami and LU Khasanah. Preparation and preliminary characterization of semi refined kappa carrageenan-based edible film incorporated with cinnamon essential oil. *AIP Conf. Proc.* 2016; **1746**, 020036.
- [3] HPSA Khalil, CK Saurabh, YY Tye, TK Lai, AM Easa, E Rosamah, MRN Fazita, MI Syakir, AS Adnan, HM Fizree, NAS Aprilia and A Banerjee. Seaweed based sustainable films and composites for food and pharmaceutical applications: A review. *Renew. Sustain. Energ. Rev.* 2017; **77**, 353-62.
- [4] AE Saputri, D Praseptianga, E Rochima, C Panatarani and IM Joni. Mechanical and solubility properties of bio-nanocomposite film of semi refined kappa carrageenan/ZnO nanoparticles. *AIP Conf. Proc.* 2018; **1927**, 030040.
- [5] GJ Manuhara, D Praseptianga, DRA Muhammad and BH Maimuni. Preparation and characterization of semi-refined kappa carrageenan-based edible film for nano coating application on minimally processed food. *AIP Conf. Proc.* 2016; **1710**, 030043.
- [6] A Farhan and NM Hani. Characterization of edible packaging films based on semi-refined kappa-

- carrageenan plasticized with glycerol and sorbitol. *Food Hydrocolloids* 2017; **64**, 48-58.
- [7] D Praseptianga. Development of seaweed-based biopolymers for edible films and lectins. *IOP Conf. Series Mater. Sci. Eng.* 2017; **193**, 012003.
- [8] S Khedri, E Sadeghi, M Rouhi, Z Delshadian, AM Mortazavian, JDT Guimarães, M Fallah and R Mohammadi. Bioactive edible films: Development and characterization of gelatin edible films incorporated with casein phosphopeptides. *LWT* 2021; **138**, 110649.
- [9] S Ebrahimzadeh, MR Bari, H Hamishehkar, HS Kafil and L Loong-Tak. Essential oils-loaded electrospun chitosan-poly(vinyl alcohol) nonwovens laminated on chitosan film as bilayer bioactive edible films. *LWT* 2021; **144**, 111217.
- [10] W Zhou, Y He, F Liu, L Liao, X Huang, R Li, Y Zou, L Zhou, L Zou, Y Liu, R Ruan and J Li. Carboxymethyl chitosan-pullulan edible films enriched with galangal essential oil: Characterization and application in mango preservation. *Carbohydr. Polym.* 2021; **256**, 117579.
- [11] J Cai, J Xiao, X Chen and H Liu. Essential oil loaded edible films prepared by continuous casting method: Effects of casting cycle and loading position on the release properties. *Food Packag. Shelf Life* 2020; **26**, 100555.
- [12] HE Salama, MSA Aziz and MW Sabaa. Development of antibacterial carboxymethyl cellulose/chitosan biguanidine hydrochloride edible films activated with frankincense essential oil. *Int. J. Biol. Macromol.* 2019; **139**, 1162-7.
- [13] Q Ma, F Xiao-Dan, L Xiao-Cao, Q Tai-Qiu and J Jian-Guo. Ultrasound-enhanced subcritical water extraction of essential oils from *Kaempferia galangal* L. and their comparative antioxidant activities. *Separ. Purif. Tech.* 2015; **150**, 73-9.
- [14] N Srivastava, Ranjana, S Singh, AC Gupta, K Shanker, DU Bawankule and S Luqman. Aromatic ginger (*Kaempferia galanga* L.) extracts with ameliorative and protective potential as a functional food, beyond its flavor and nutritional benefits. *Toxicol. Rep.* 2019; **6**, 521-8.
- [15] MI Umar, MZ Asmawi, A Sadikun, AMSA Majid, FSR Al-Suede, LEA Hassan, R Altaf and MBK Ahamed. Ethyl-p-methoxycinnamate isolated from *kaempferia galanga* inhibits inflammation by suppressing interleukin-1, tumor necrosis factor- α , and angiogenesis by blocking endothelial functions. *Clinics* 2014; **69**, 134-44.
- [16] N Srivastava, S Mishra, H Iqbal, D Chanda and K Shanker. Standardization of *kaempferia galanga* L. rhizome and vasorelaxation effect of its key metabolite ethyl p-methoxycinnamate. *J. Ethnopharmacol.* 2021; **271**, 113911.
- [17] MS Ali, PR Dash and M Nasrin. Study of sedative activity of different extracts of *kaempferia galanga* in swiss albino mice. *BMC Compl. Alternative Med.* 2015; **15**, 158.
- [18] YD Handiati, D Praseptianga, GJ Manuhara and LU Khasanah. Effects of *kaempferia galanga* L. essential oil incorporation on sensory and physical properties of dark chocolate bar. *IOP Conf. Series Mater. Sci. Eng.* 2019; **633**, 012036.
- [19] S Munda, P Saikia and M Lal. Chemical composition and biological activity of essential oil of *kaempferia galanga*: A review. *J. Essent. Oil Res.* 2018; **30**, 1486240.
- [20] S Sahoo, R Parida, S Singh, RN Padhy and S Nayak. Evaluation of yield, quality and antioxidant activity of essential oil of *in vitro* propagated *kaempferia galanga* linn. *J. Acute Dis.* 2014; **3**, 124-30.
- [21] SKT Seraglio, M Schulz, P Brugnerotto, B Silva, LV Gonzaga, R Fett and ACO Costa. Quality, composition and health-protective properties of citrus honey: A review. *Food Res. Int.* 2021; **143**, 110268.
- [22] SK Yap and NL Chin. Identification of distinctive properties of common Malaysian honeys. *Mater. Today Proc.* 2021; **42**, 115-8.
- [23] S Almasaudi. The antibacterial activities of honey. *Saudi J. Biol. Sci.* 2021; **28**, 2188-96.
- [24] NMDMW Nayaka, I Fidrianny, Sukrasno, R Hartati and M Singgih. Antioxidant and antibacterial activities of multiflora honey extracts from the Indonesian *Apis cerana* bee. *J Taibah Univ. Med. Sci.* 2020; **15**, 211-7.
- [25] B Silva, FC Biluca, LV Gonzaga, R Fett, EM Dalmarco, T Caon and ACO Costa. *In vitro* anti-inflammatory properties of honey flavonoids: A review. *Food Res. Int.* 2021; **141**, 110086.
- [26] M Srisayam and P Chantawannakul. Antimicrobial and antioxidant properties of honeys produced by *Apis mellifera* in Thailand. *J. ApiProduct ApiMedical Sci.* 2010; **2**, 77-83.
- [27] JT Soinenen, J Heinämäki and J Yliruusi. From acacia honey monosaccharide content to a new external binary plasticizer applicable in aqueous whey protein films. *Food Bioprocesses* 2013; **91**, 440-6.
- [28] ASTM. *Standard Test Method for Tensile Properties of Thin Plastic Sheeting*. ASTM International,

- Pennsylvania, 2010.
- [29] WG Sullivan, EM Wicks and CP Koelling. *Engineering Economy, 16th Edition*. Pearson, London, 2015.
- [30] M Meilgaard, GV Civille and BT Carr. *Sensory Evaluation Techniques, 4th Edition*. CRC Press, Florida, 2006.
- [31] D Praseptiangga, AR Qomaruzzaman and GJ Manuhara. The effect of clove leaves essential oil addition on physicochemical and sensory characteristics of milk chocolate bar. *Int. J. Adv. Sci. Eng. Inform. Tech.* 2021; **11**, 165-71.
- [32] M Beigomi, M Mohsenzadeh and A Salari. Characterization of a novel biodegradable edible film obtained from dracocephalum moldavica seed mucilage. *Int. J. Biol. Macromol.* 2018; **108**, 874-83.
- [33] R Arham, MT Mulyati, M Metusalach and S Salengke. Physical and mechanical properties of agar based edible film with glycerol plasticizer. *Int. Food Res. J.* 2016; **23**, 1669-75.
- [34] T Wittaya. *Protein-based edible films: Characteristics and improvement of properties*. In: AA Eissa (Ed.). Structure and Function of Food Engineering. IntechOpen, London, 2012.
- [35] H Seyedeh-Maryam and J Dehghannya. Development and characterization of novel edible films based on Cordia dichotoma gum incorporated with Salvia mirzayanii essential oil nanoemulsion. *Carbohydr. Polymer.* 2021; **257**, 117606.
- [36] S Shojae-Aliabadi, H Hosseini, MA Mohammadifar, A Mohammadi, M Ghasemlou, SM Ojagh, SM Hosseini and R Khaksar. Characterization of antioxidant-antimicrobial κ -carrageenan films containing Satureja hortensis essential oil. *Int. J. Biol. Macromol.* 2013; **52**, 116-24.
- [37] A Soni, G Kandeepan, SK Mendiratta, V Shukla and A Kumar. Development and characterization of essential oils incorporated carrageenan based edible film for packaging of chicken patties. *Nutr. Food Sci.* 2016; **46**, 82-95.
- [38] D Veena, EN Mallika, GV Reddy and K Sudheer. Quality of edible polymer films incorporated with plant essential oils. *Int. J. New Tech. Sci. Eng.* 2015; **2**, 43-7.
- [39] S Shojae-Aliabadi, H Hosseini, MA Mohammadifar, A Mohammadi, M Ghasemlou, SM Hosseini and R Khaksar. Characterization of κ -carrageenan films incorporated plant essential oils with improved antimicrobial activity. *Carbohydr. Polymer.* 2014; **101**, 582-91.
- [40] Y Chu, T Xu, CC Gao, X Liu, N Zhang, X Feng, X Liu, X Shen and X Tang. Evaluations of physicochemical and biological properties of pullulan-based films incorporated with cinnamon essential oil and Tween 80. *Int. J. Biol. Macromol.* 2019; **122**, 388-94.
- [41] M Vargas, A Albors, A Chiralt and C González-Martínez. Characterization of chitosan-oleic acid composite films. *Food Hydrocolloids* 2009; **23**, 536-47.
- [42] L Sánchez-González, M Cháfer, A Chiralt and C González-Martínez. Physical properties of edible chitosan films containing bergamot essential oil and their inhibitory action on penicillium italicum. *Carbohydr. Polymer.* 2010; **82**, 277-83.
- [43] Y Zhang, L Zhou, C Zhang, PLShow, A Du, JC Fu and V Ashokkumar. Preparation and characterization of curdlan/polyvinyl alcohol/ thyme essential oil blending film and its application to chilled meat preservation. *Carbohydr. Polymer.* 2020; **247**, 116670.
- [44] LB Norcino, JF Mendes, CVL Ntarelli, A Manrich, JE Oliveira and LHC Mattoso. Pectin films loaded with copaiba oil nanoemulsions for potential use as bio-based active packaging. *Food Hydrocolloids* 2020; **106**, 105862.
- [45] K Candogan, GV Barbosa-Cánovas and E Çarkcioglu. *Edible films and coatings: Sensory aspects*. In: MPM García, MC Gómez-Guillén, ME López-Caballero and GV Barbosa-Cánovas (Eds.). Edible Films and Coatings: Fundamentals and Applications. CRC Press, Florida, 2016, p. 497-518.