Food Chemistry: X 22 (2024) 101497

Contents lists available at ScienceDirect

Food Chemistry: X

journal homepage: www.sciencedirect.com/journal/food-chemistry-x

Effect of κ-carrageenan on the quality of crayfish surimi gels

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ARTICLE INFO

Keywords:
κ-Carrageenan
Composite gel
Crayfish
Myofibrillar protein
Microstructure

ABSTRACT

The demand for crayfish surimi products has grown recently due to its high protein content. This study examined the effects of varying κ-carrageenan (CAR) and crayfish surimi (CSM) concentrations on the gelling properties of CAR-CSM composite gel and its intrinsic formation process. Our findings demonstrated that with the increasing concentration of carrageenan, the quality of CAR-CSM exhibited rising trend followed by subsequently fall. Based on the textural qualities, the highest quality CAR-CSM was achieved at 0.3% carrageenan addition. With the exception of chewiness, and the cooking loss of the gel system was 1.62%, whiteness was 82.35%, and the percentage of β-sheets increased to 57.18%. Further increase in CAR (0.4–0.5%) addition resulted in internal build-up of LCAR-CSM, conversion of intermolecular forces into disulfide bonds and gel breakage. This study exudes timely recommendations for extending the CAR application for the continuous development of crayfish surimi and its derivatives and its overall economic worth.

1. Introduction

Crayfish are a type of crustacean decapod that are mostly farmed in freshwater regions of Hubei, Anhui, Hunan, Jiangsu, Jiangxi, and other provinces along the middle and lower sections of the Yangtze River. Due to their softness and a high economic value, they have garnered a lot of interest as a biological resource with potential applications because of its high protein and selenium content (Gómez-Estaca, Montero, & Gómez-Guillén, 2018; Lu et al., 2022). Food products manufactured from shrimp meat are becoming more and more popular both domestically and internationally as consumers are becoming more discerned for their diets. However, because shrimp have very active enzymes, proteins tend to self-melt which causes a number of issues during the development and manufacturing of products made from minced shrimp, leading to poor texture, low gel strength, and poor water retention (Zheng et al., 2022). In addition, gel properties are crucial for assessing the quality of minced shrimp products, and the incorporation of biomolecules such as starch and polysaccharides can significantly improve the flexibility of the gel to a certain extent (Zhu et al., 2023; Liu et al., 2021). The inclusion of such ingredients can improve the gelation, water holding and texture of meat products. Carrageenan, a linear anionic gel polysaccharide extracted from red algae, has found its application in the gelation process of surimi meat products and for the improvement of the texture and water holding capacity of the gel. It is also reported to maintain the network scaffolding of protein gels through filling action and produce thermos-reversible gels through a double helix cross-linking process. It has been shown that 1.0% κ−/ι-CAR significantly improved the gel strength, water retention and viscoelasticity of MP gels extracted from South American shrimp (Li et al., 2023). The addition of ι-carrageenan (2%) or κ-carrageenan (1%–2%) was reported to improve the textural properties of Antarctic krill surimi (Li et al., 2022). However, no study has explored the potential role of carrageenan for the modification of the crayfish surimi gel. Based on the current limitation of data, we used crayfish as the raw material, κ-carrageenan (CAR) and crayfish shrimp mince (CSM) were included at varying mass ratios of 0%–0.5% to prepare the composite gel system. The effect of CAR incorporation on the properties and quality of the prepared composite gels of minced shrimp was determined by comparing several

Abbreviations: CAR, κ-carrageenan; CSM, Crayfish Minced Shrimp; CAR-CSM, κ-carrageenan and crayfish surimi composite gel; L-NMR, A low-frequency nuclear magnetic resonance device.

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https://doi.org/10.1016/j.fochx.2024.101497
Received 9 April 2024; Received in revised form 5 May 2024; Accepted 19 May 2024
Available online 21 May 2024

experimental parameters such as cooking loss rate, whiteness value, gel strength, textural properties, moisture distribution (low-frequency nuclear magnetic resonance), dynamic rheology, infrared spectroscopy, intermolecular forces, and gel microstructure of minced shrimp gels. Herein, we intend to provide appropriate data for the practical use of CAR to improve the production and economic value of shrimp surimi and its derivatives.

2. Materials and methods

2.1. Experimental materials

Crayfish (Huaxiang Food Company Limited, Hefei, Anhui Province). CAR (Beijing Solebo Technology Co. Ltd.) and sodium chloride (Sinopharm Chemical Reagent Co.). The primary tools and instruments utilized in this investigation were the NMI20-015 V-I magnetic resonance imaging analyzer (Suzhou Newmax Electronic Technology Co., Ltd.), the Jiuyang cooking crusher (Jiuyang Co., Ltd.), the differential scanning calorimeter (Q2000), the physical property tester (TA-XTPLUS), and the 10 N freeze dryer (Beijing Songyuan Huaxing Science and Technology Development Co., Ltd.).

2.2. Preparation of minced shrimp gel

The preparation of CAR-CSM was carried out according to previously reported method (Htwe et al., 2023; Man, Sun, Lin, Ren, & Li, 2024). Firstly, the frozen crayfish samples were thawed at room temperature for 2 h, and they were immersed in running water for 10 min. The samples were washed to remove the intestinal threads and crustaceans and other impurities, and then rinsed three times with water, drained and divided into a number of portions according to the quality of the mixture, to which refined salt with a mass fraction of 3% (in terms of total weight of shrimp) was added per portion followed by grounding at high-speed using a food processor. Further, CAR (0.1% - 0.5% by mass) was added to each portion and grounded twice at high speed in a food processor to obtain a paste. CAR was replaced by 3% of refined salt in the blank control group. The obtained paste was packed in plastic watch bags and filled into 24 mm rubber square molding boxes. Finally, the samples were heated in a shrimp water bath at 30 °C for 90 min and immediately cooled with crushed ice to obtain the CAR-CSM samples. All the testing samples were set up in triplicates and CAR-CSM was stored at 4 °C.

2.3. Test method

2.3.1. Rate of cooking loss

The cooking loss rate of mince gel samples was determined using the previous method with slight modifications (Diao, Tao, Chen, Zhang, & Wang, 2022). A specific quantity of ground gel was precisely weighed (the mass is m1) and sealed in a bag, submerged in water at 40 °C for 40 min, and swiftly moved to an 80 °C water bath for 30 min. Finally, filter paper was used to absorb surface moisture, followed by accurately weighing (the mass is m_2), and the cooking loss rate was computed as follows: $CI(\%) = \left[1 - \frac{m_2}{m_1}\right]*100$.

2.3.2. Whiteness value

Firstly, a portable high-precision colorimeter was used to evaluate the brightness, redness, and yellowness values of the ground shrimp gel sample after the calibration of the balance for 20 min at room temperature. Based on the previously reported method, whiteness values were computed (Wijayanti, Singh, Benjakul, & Sookchoo, 2021).

2.3.3. Gel strength and texture characteristics

According to previous studies (Zou et al., 2022; Hu et al., 2022), the determination of gel strength and textural qualities were evaluated. Firstly, the samples with varying degrees of carrageenan in mince gel were sliced into 25 mm-tall cylinders and incubated for 12 h at 20 °C with constant humidity and temperature. Secondly, the physical property tester and P/5S probe were used to set the pre-test rate of 1.00 mm/s, the rate of the test in 1.10 mm/s, the rate of the test after 10.00 mm /s, the displacement of 15 mm, and the trigger force of 10 g. The texture property testing result was automatically determined by the software reading, and the product of the breaking strength of the first peak on the gel curve and the breaking distance was selected.

2.3.4. Protein secondary structure

After following the previous method by Guo et al. (2017), FT-IR was used to determine secondary structure for each sample. For this, freeze-dried ground gel powder was first combined with potassium bromide in a 1:100 ratio, and then grounded. A total of 64 scans with a resolution of 4 cm^{-1} were carried out in the 600–4000 cm^{-1} range. Using peak fit software, the amido zone (1600–1700 cm^{-1}) in the atlas was examined to determine the relative secondary structure richness of each sample.

2.3.5. Water distribution

A low-frequency nuclear magnetic resonance device (L-NMR) was used to determine the water distribution according to previously reported method (Han, Wang, Chu, & Bao, 2022). The T_2 map was obtained by inversion of the exponential attenuation curve by measuring the attenuation rate curve of the index peak obtained by pulse train CPMG. The minced shrimp gel sample was divided into squares measuring 14 mm by 14 mm by 14 mm and covered with plastic wrap and allowed to stand for some time. After that, a L-NMR probe was introduced into a 15 cm L-NMR tube containing the minced shrimp samples. Using the CPMG sequence, T2 values were calculated at the proton resonance frequency of 22.5 MHz. The τ value was set to 100 μs, and the scanning was repeated 16 times to stabilize the wave curve at a scanning time of 2 s/time. A total of 6000 echo data were collected, and each sample was repeated for three times.

2.3.6. Rheological property test

Determination of the rheological properties of the prepared CAR-CSM samples was carried out by rheometer (Min et al., 2023). A 2 mL sample was taken and placed on the measuring platform. The temperature ramp parameters were: a PP50 parallel aluminum plate fixture with 1 mm spacing, a temperature range of 20 to 80 °C, a temperature increase rate of 2 °C/min, an oscillation frequency of 0.1 Hz and a strain of 0.5%. Frequency scanning of minced shrimp samples was carried out at 25 °C: PP50 parallel aluminum fixture with a spacing of 1 mm, oscillation frequency range of 0.01–1 Hz, and strain of 0.5%.

2.3.7. Chemical forces

The determination of the internalization power of shrimp gel was based on the previous method with slight modifications (Huang et al., 2022). Accurately weighed 4 g of minced shrimp gel was homogenized with 20 mL dissolved solution for 30 s. The dissolved solution consisted of 0.05 mol/L NaCl (A), 0.6 mol/L NaCl (B), B + 1.5 mol/L urea (C), C + 8 mol/L urea (D), and D + 0.25% β-mercaptoethanol (E) solution. After heating at 80 °C for 30 min and then cooling, mixture was centrifuged for 15 min (10,000 g/min), and the amount of protein extracted after treatment with different solution buffers was used to represent the main force in the gel.

2.3.8. Microstructure determination

The microstructure of ground shrimp gel was observed by scanning electron microscopy (SEM) in accordance with previous research method (Shao et al., 2017). Firstly, the ground shrimp gel was cut into 2 mm × 2 mm × 2 mm chunks, fixed with glutaraldehyde solution (2.5%) for overnight, washed with phosphoric acid buffer for 4 times, and gradient dehydration was carried out by ethanol solution. Then dehydrated gel sample was vacuum freeze-dried and gold sprayed prior to the final observation.

2.3.9. Sensory assessment

The evaluation team consisted of 12 evaluators with professional sensory training, with a balanced ratio of men and women. CAR was added in varying quantities to the minced shrimp samples, which were then cooked, organized, and presented using the random number approach. The evaluation team members used a nine-point scoring system and conducted a sensory evaluation based on the properties of minced shrimp in five dimensions: acceptability, elasticity, color, flavor, and taste. This study was approved by the ethics committee of Hefei University of Technology. Appropriate protocols were used to protect the rights and privacy of all the participants throughout the study. The study was performed in accordance with the 1964 declaration of HELSINKI and later amendments. Written informed consent was obtained from all the participants prior to the enrollment of this study.

2.4. Data processing

In this study, all the data were presented as mean ± standard deviation and analyses were conducted in triplicates to ensure the accuracy of the experimental results. Data was analyzed using ANOVA, IBM SPSS Statistics 20 and graphs were plotted using Origin 2021.

3. Results and discussion

3.1. Specific characteristics of the gel

Fig. 1A shows the effect of different CAR additions on the cooking loss rate of CAR-CSM system. The cooking loss rate of CAR-CSM system was reported to decrease at first and then increase with the increasing concentration of CAR (0.1%–0.5%). The lowest cooking loss of the gel system was achieved at 0.3% CAR. Whereas, no significant difference ($p < 0.05$) was reported between the cooking loss rate of the CAR-CSM system at 0.4% CAR and 0.3% CAR. In conclusion, the addition of CAR could effectively reduce the cooking loss of CAR-CSM. This is due to the fact that CAR is a hydrophilic colloid, which undergoes physical rearrangement in the gel system and fills in the pores of the gel. Subsequently, it forms gel fragments that can bind water when cooled, which allows more water molecules to be retained in the gel, thus reducing the rate of cooking loss of the samples to be tested (Saengsuk et al., 2022). Meanwhile, during heating, myofibrillar fibrillar proteins (with a unique linear sulfated polysaccharide structure) in CAR and CSM were fully opened up and the intermolecular interactions were enhanced to form a cross-linked structure, which strengthened the stability of the gel, leading to the reduction of the cooking loss after the incorporation of CAR (Xia et al., 2023).

The change in the whiteness value of the gel is an extrinsic characterization feature of the protein structure alterations. Fig. 1B showed

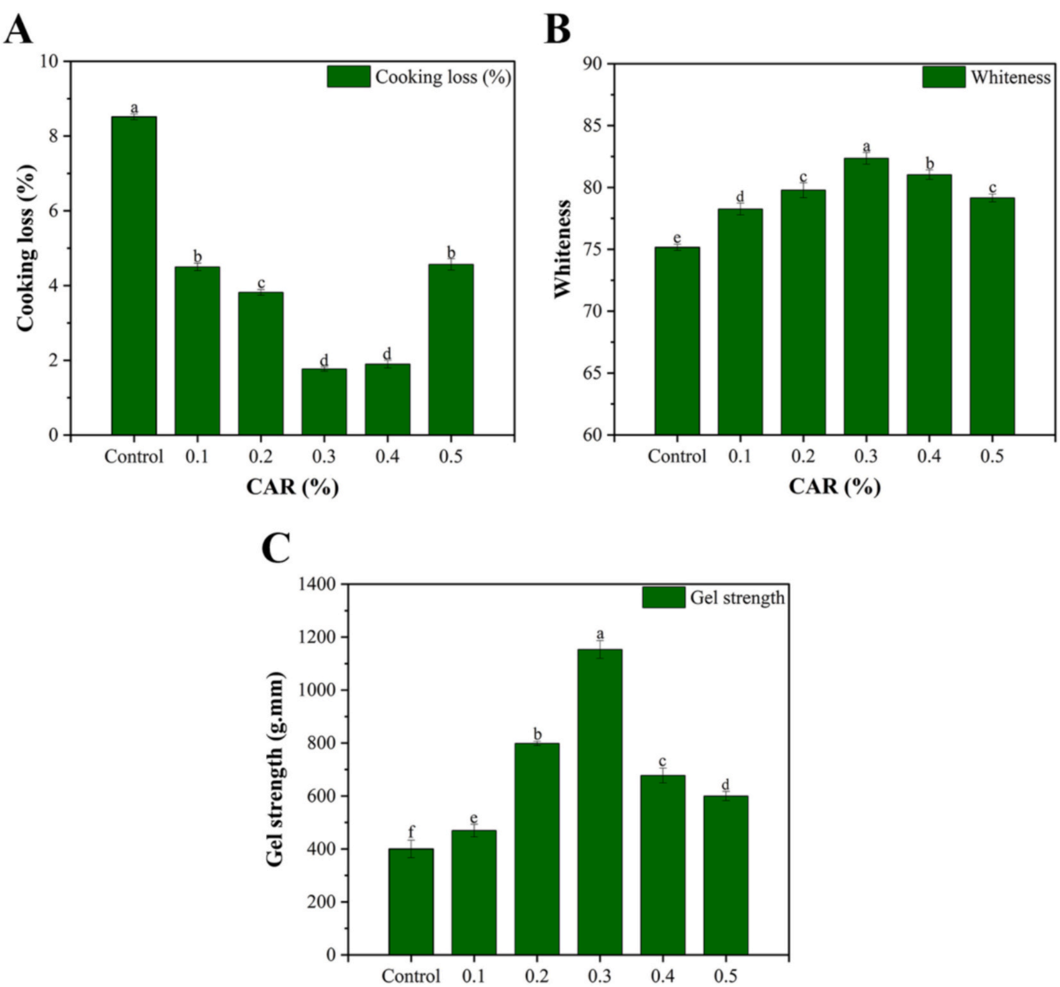

Fig. 1. Specific characteristics of κ-carrageenan and crayfish surimi composite gel (CAR-CSM). Cooking loss rate (A), whiteness value (B), gel strength (C). The means are significantly different ($p < 0.05$) according to the multiple range of least significant difference (LSD), indicated by letters (a-f).

that whiteness value of the CAR-CSM gel system had a tendency to increase and then decrease with the increase of CAR addition. The maximum whiteness value of 82.35 was observed at 0.3% carrageenan addition. Subsequently, the whiteness value of the gel started to decrease as the CAR concentration continued to increase. The whiteness value of the CAR-CSM system increased by 9.57% compared to the control. In summary, the addition of CAR resulted in a significant increase in the whiteness values of all the systems ($p < 0.5$). This may be due to the fact that CAR was filled into the gaps of the gel, which made the network structure of the gel denser, and the refraction phenomenon of light when passing through the gel increased the whiteness value of the gel (Zhang, Fang, Hao, & Zhang, 2018). Interestingly, the whiteness value of the CAR-CSM system showed a decreasing trend when the addition of CAR exceeded 0.3%. It may be due to the fact that a large number of hydroxyl structures in the carrageenan structure combine with the polar residues present in the gel system to further form hydrogen-bonded structures. The presence of a large number of hydrogen-bonded structures could compete with myofibrillar proteins in the CSM for water molecules, which resulted in a decrease in the amount of bound water in the CAR-CSM gel system, a decrease in the refractive index of the measured light, and a subsequent decrease in the whiteness value (Chen, Luo, Wang, Chen, & Zhuang, 2023).

Gel strength is an important parameter for evaluating the quality of minced shrimp gel, which can reflect the water retention force of minced shrimp myofibrillar fibrous protein in the formation of gel to a certain extent. As shown in Fig. 1C, the gel strength of minced shrimp gel was significantly increased to different degrees ($p < 0.05$) after the addition of CAR. With the increase of CAR concentration, the gel strength showed an increasing trend which was followed by gradual decrease. At 0.3%, the gel strength of minced shrimp reached a maximum value of 1153 g. mm which could be attributed to the fact that CAR at a low concentration could bind well with MPs and absorb the free water in the system, thus enhancing the cross-linking density of the gel network and forming an ordered and stable three-dimensional gel structure. At higher concentrations, CAR may produce thermodynamic incompatibility in the system, leading to phase separation of the minced shrimp gel, and resulting in less rigid the minced shrimp gel (Pei et al., 2023).

3.2. Textural characterization

Textural properties are important parameters for evaluating the quality of minced shrimp gels, which can characterize the integrity of the protein matrix structure and the tight binding status with related components (Zhang et al., 2021). Based on this, this study investigated the effects of different concentrations of CAR on the elasticity, cohesiveness, hardness and other textural properties of minced shrimp gel (Table 1). Compared with the control, the addition of different concentrations of CAR significantly improved the hardness, cohesiveness, chewiness, and reparability of minced shrimp gel ($p < 0.05$); the elasticity of minced shrimp gel had a significant effect ($p < 0.05$) at 0.1%–0.3% of CAR, but the change in the elasticity of minced shrimp gel was insignificant after 0.3% CAR. Interestingly, the hardness, elasticity, cohesiveness, chewiness, and repulsiveness of the minced shrimp gel showed an upward trend which was then reversed with the increasing CAR concentration. At 0.3% CAR, all the parameters related to textural properties except chewiness reached their maximum values. This may be due to the fact that at lower concentration of CAR (0.1%–0.3%), it exerts a synergistic effect on the gel, and the addition of CAR can immobilize more water, promote gel formation, and thus improve the strength of the gel (Li et al., 2023). However, as the concentration of CAR is further increased, the system may flocculate due to the repulsive effect, resulting in the gradual decrease of the hardness, elasticity, and other parameters related to the textural properties of the gel of minced shrimp (Li et al., 2023). The above results indicate that the increase of the appropriate amount of CAR resulted in an increase in the stress generated during the same deformation of the minced shrimp, which in turn maintained the tight connectivity of the gel structure and improved the texture of the gel.

Table 1
Texture properties of κ-carrageenan and crayfish surimi composite gel (CAR-CSM).

CAR (%)	Hardness	Elasticity	Cohesiveness	Chewiness	Resilience
Control	943.31 ± 13.02^{d}	0.55 ± 0.01^{c}	0.36 ± 0.01^{e}	611.45 ± 20.18^{e}	0.12 ± 0.02^{e}
0.1	1510.95 ± 21.79^{c}	0.60 ± 0.02^{b}	0.44 ± 0.02^{d}	828.70 ± 26.90^{d}	0.25 ± 0.03^{d}
0.2	2112.57 ± 55.89^{b}	0.62 ± 0.04^{b}	0.53 ± 0.03^{c}	1160.11 ± 67.69^{c}	0.45 ± 0.03^{b}
0.3	2359.31 ± 27.06^{a}	0.85 ± 0.05^{a}	0.68 ± 0.02^{a}	1556.28 ± 48.12^{b}	0.56 ± 0.02^{a}
0.4	2165.29 ± 67.21^{b}	0.55 ± 0.02^{c}	0.58 ± 0.03^{b}	1679.23 ± 25.83^{a}	0.41 ± 0.02^{c}
0.5	1478.35 ± 29.98^{c}	0.56 ± 0.03^{c}	0.47 ± 0.03^{d}	1545.49 ± 45.05^{b}	0.37 ± 0.01^{c}

Note: The amount of κ-carrageenan added in the experimental group was 0.1–0.5% in order, and no κ-carrageenan was added in the control group. a-e sequentially indicate the cases of significant differences between the groups ($p < 0.05$).

3.3. Specific properties of the secondary structure

Infrared spectroscopy is one of the most important means to determine the relative content of protein secondary structure. The characteristic absorption band (1600–1700 cm) of proteins in infrared spectra contains rich secondary structure information of β-sheet, random coil, α-helix, β-turn and so on. Based on this, the relative content of secondary structure of shrimp surimi myofibrillar fibrillar protein gel was analyzed by baseline correction, deconvolution and peak fitting of amide I band using Peak Fit software. As shown in Supplemental Table 1, the addition of CAR substantially decreased the structural composition ratio of Random coil in MP gels, while the content of β-sheet structure increased substantially. At 0.3% CAR addition, the percentage of β-sheet in the samples reached a peak of 57.18%. The conversion of α-helix and Random coil structure to ordered β-sheet in proteins is more favorable for the formation of regular and ordered gel network structure (Liu et al., 2021). This may be due to the fact that the addition of CAR caused more protein molecules to cross-link with each other by hydrogen bonding, and the three-dimensional network structure of proteins became more uniform. However, excessive CAR addition may cause the internal gel structure of minced shrimp to be too tight, which may have some effect on the formation of β-sheet. This may be due to the fact that the presence of excess CAR enhanced the interaction force between molecules inside the gel of minced shrimp, which impeded its spatial arrangement and regular movement to a certain extent, and thus had an effect on the formation of secondary structure (Hu et al., 2022). Interestingly, the relative contents of α-helix and β-turn structures showed irregular variations, with the relative contents of both increasing at 0.1%, 0.4%, and 0.5% additions, and the opposite trend was noticed at the rest of the additions, which suggests that the structural properties of the minced shrimp gel are not only related to the β-sheet structure among myofibrillar fibrillar proteins, but are also influenced to a certain extent by the α-helix and β-turn structures in the system.

3.4. Moisture distribution (low-frequency NMR)

The distribution of water molecules in different states in the gel of minced shrimp can directly reflect the state of the three-dimensional mesh structure of the gel, which is an important indicator to characterize the quality of minced shrimp. As shown in Fig. 2A, minced shrimp showed a total of three peaks in the L-NMR curve. Among them, the peak appearing at 0.01–10 ms T_2 indicated the bound water in the system; the

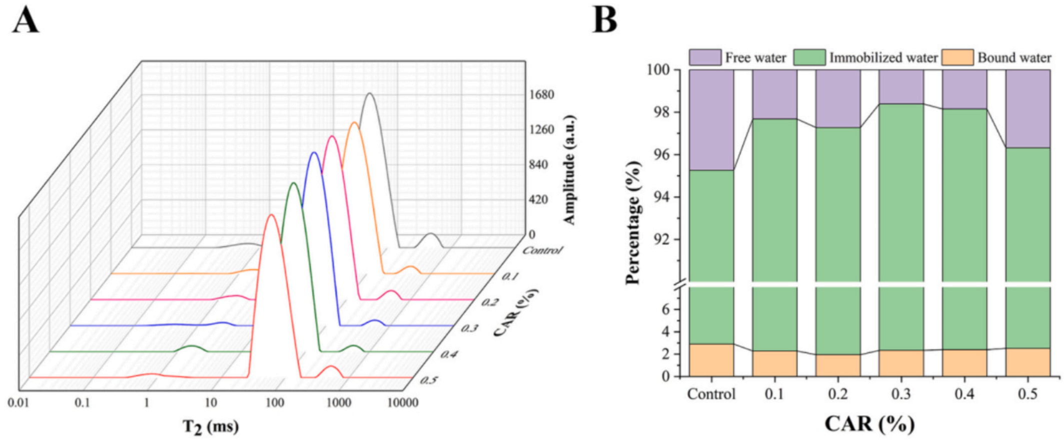

Fig. 2. Moisture distribution of of κ-carrageenan and crayfish surimi composite gel (CAR-CSM). Low-frequency NMR T_2 relaxation time spectrum T_2 (A), and moisture distribution stacking diagram (B).

peak appearing in the T_2 range of 10–100 ms was the wrapped water in the three-dimensional mesh structure inside the minced shrimp gel. The peak in the 100–1000 ms T_2 range represented free-flowing water that was not bound to the matrix CAR-CSM (Liang, Lin, Chen, & Sun, 2022). With the increase in relaxation time T_2, the water gradually transitioned from bound form to free water, and the water molecules turned more mobile. In this study, the state of water molecules in each experimental group was further visualized by stacking diagrams (Fig. 2B). The results showed that the water that was not easily flowable is the main state of water in shrimp surimi gels. Compared with the control group, the addition of different concentrations of CAR increased the content of immobile water in minced shrimp. At 0.3% CAR addition, the highest value of immobile water content in minced shrimp was 96.05%, which was increased by 3.69% compared with the control group. Meanwhile, the free water content decreased from 4.74% to 1.61%. Interestingly, up to 0.5% CAR, the immobile water content in CAR-CSM system decreased

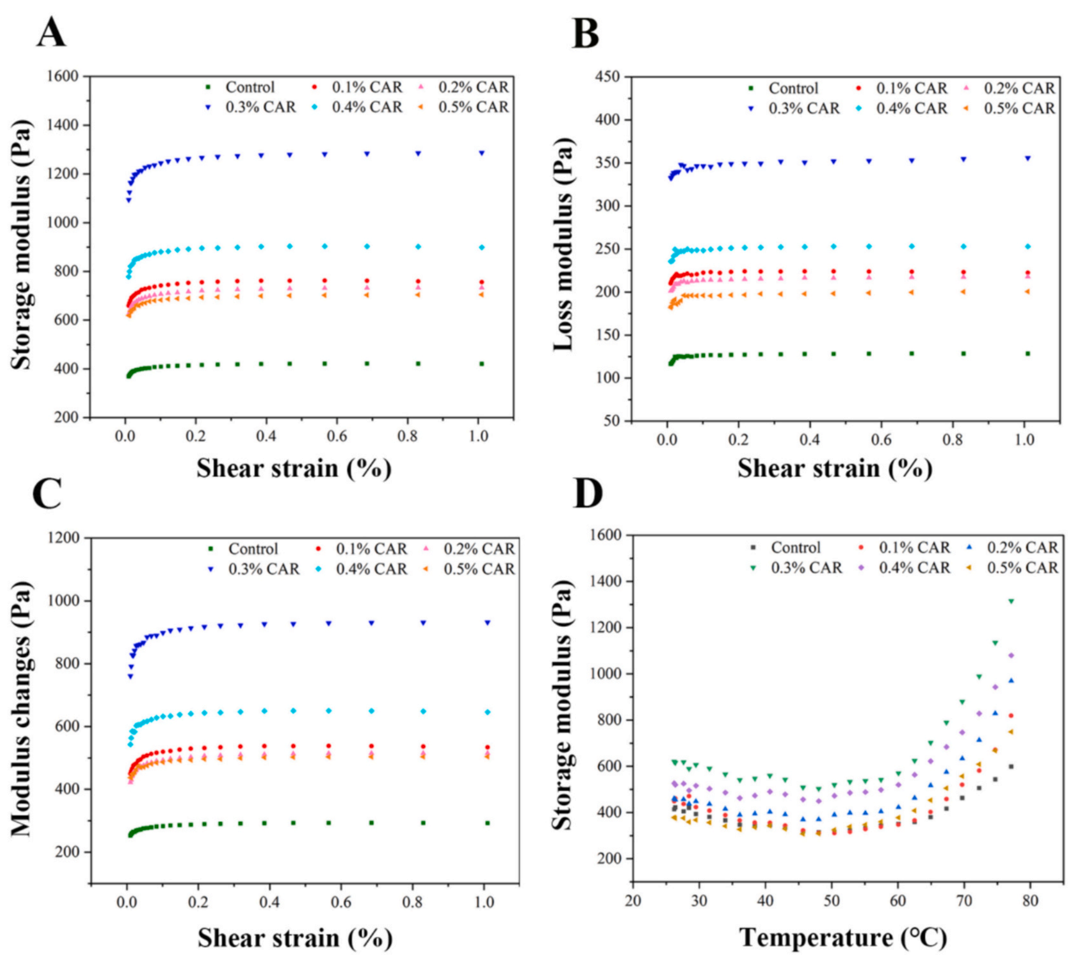

Fig. 3. Rheological properties of κ-carrageenan and crayfish surimi composite gel (CAR-CSM). Change in storage modulus (A), change in loss modulus (B), change in difference between storage and loss modulus (C), and effect of temperature on the change in storage modulus of the gel (D).

to some extent, the free water content increased, and the bound water content remained essentially unchanged, which may be attributed to the fact that the excess CAR caused fragmentation of the gel network, and the free water was released as a result (Gao, Xiong, & Fu, 2019).

The above experimental results reveal the conversion of free water into fixed water after the appropriate addition of CAR. This is due to the fact that the addition of CAR allowed the network structure of the prepared gel system to appear denser and more compact, with the property of trapping more water molecules with water absorption (Zhou et al., 2014).

3.5. Rheological characterization (viscoelasticity)

In this study, the energy storage modulus and loss modulus of minced shrimp gel samples before and after CAR incorporation as well as the difference of their modulus change with the change of shear strain rate were investigated under the same vibration frequency conditions. As shown in Fig. 3A, the energy storage modulus of the minced shrimp gel was significantly higher than that of the control after the addition of different concentrations of CAR. The loss modulus also showed a similar distribution pattern (Fig. 3B). It is worth noting that the difference between the loss modulus and the energy storage modulus of all minced shrimp gel samples was >0, indicating that the samples showed elasticity characteristics and the formation of a more stable reticulated gel structure. The difference between the loss modulus and energy storage modulus showed an increasing and then decreasing trend with the addition of CAR (Fig. 3C). At 0.3% CAR addition, the difference between loss modulus and energy storage modulus was significantly higher than the rest of the samples, indicating the best gel elasticity. With the increasing CAR addition, the excess CAR could easily form a continuous network structure, which in turn hindered the formation of the gel network of κ-carrageenan and crayfish surimi composite gel, decreasing the corresponding difference between the modulus of loss and energy storage modulus. In addition, we further explored the gelation process of shrimp surimi with CAR by comparing the effects of different concentrations of CAR and temperature on the energy storage modulus (Fig. 3D). The trends of gel energy storage modulus of minced shrimp samples in the control group and the experimental group with different CAR concentrations were basically the same. At 0.3% CAR addition, a wave peak appeared at 40 °C, which may be a signal of early gel formation of minced shrimp (Zhang et al., 2017). With the continuous increase in temperature (40–50 °C), the energy storage modulus showed a slightly decreasing trend. This may be due to the fact that the internal myosin was subjected to endogenous MP hydrolase at this stage, which led to the enhancement of the internal fluidity of the protein, which in turn led to a certain degree of denaturation in this temperature interval (Krop, Hetherington, Holmes, Miquel, & Sarkar, 2019). When the temperature was changed from 60 °C to 80 °C, the energy storage modulus showed a sharp increase. In the control group, the energy storage modulus of minced shrimp gel was about 598 Pa, which was significantly lower than the rest of the experimental groups. The modulus of energy storage of the gel samples with 0.3% CAR was 1316 Pa. The modulus of energy storage of the gel samples with 0.5% CAR was 749 Pa, which could be attributed to the fact that a certain concentration of CAR filled up the structure of the gel of minced shrimp and strengthened the gel, whereas further CAR addition might lead to gelation, which was not favorable to the formation of gel of minced shrimp (Borsani et al., 2021).

3.6. Chemical forces

The chemical bonding that maintains the network structure of the gel of minced shrimp mainly includes hydrogen bonding, ionic bonding, disulfide bonding, and hydrophobic interactions, which interact with each other, thus affecting the structure of the gel of minced shrimp myofibrillar fibrillar proteins and their gel properties (Cao et al., 2023). The effects of different concentrations of CAR on the chemical bonding of the gel of minced shrimp are shown in Fig. 4. CAR concentration is an important factor affecting the composition of the main chemical bonds within the shrimp surimi. The main chemical force in the control group was hydrophobic interaction, which was 0.274 mg/mL. This is mainly because the heating treatment during the preparation of shrimp surimi gel caused the unfolding of peptide chains in MP, breaking of the internal disulfide bonds, and the formation of inhomogeneous gel network, resulting into poor gel properties (Chen et al., 2023). After the addition of different concentrations of CAR, the content of chemical force inside the gel showed different degrees of significant increase ($p < 0.05$). When the concentration of CAR reached 0.3%, the overall strength of chemical forces in the system reached the highest value, and the relative contents of ionic bonding, hydrogen bonding, hydrophobic interactions, and disulfide bonding were 0.201, 0.153, 0.417, and 0.386 mg/mL, respectively. Among them, hydrogen bonding can make the state of bound water in the system more stable, and thus help to maintain the stability of the secondary structure of proteins. Disulfide bonding and hydrophobic interactions constitute the main chemical forces in the gel of shrimp surimi. When CAR was added at a concentration lower than 0.3%, hydrophobic interactions were the main force maintaining the gel network; on the contrary, the main force maintaining the gel network shifted to disulfide bonds (Fig. 4C). This may be due to the fact that with the initial addition of CAR (0.1%–0.3%), the exposed hydrophobic groups in the structure were masked, and the hydrophilicity of MPs was further enhanced, which attenuated the possibility of rapid protein intermolecular connections and the generation of related macromolecular aggregates, thus favoring the improvement of the gel network densification (Walayat et al., 2022). When the concentration of CAR in the system continued to increase (0.4%–0.5%), the excess CAR then produced further accumulation, which hindered the inter-cross-linking of hydrophobic groups between protein molecules, and the chemical force in the system was gradually shifted from hydrophobic interactions to disulfide bonds (Fig. 4D). Interestingly, the concentration of ionic bonds in both control and different concentration treatment groups was significantly lower than the rest of the chemical bonds, which may be due to the fact that CAR-CSM did not use ionic bonding as the main chemical force to maintain the stability of the gel network at the intense hydrophobic interactions, disulfide bonding, and hydrogen bonding.

3.7. Microstructural characterization

In this study, the microstructure of shrimp surimi gel was analyzed using scanning electron microscopy (Fig. 5). The control shrimp surimi gel was mainly characterized by large protein aggregates, rough network, and a large number of voids or cavities which coincides well with the conformational features of its most abundant free water. This is because the MPs in the shrimp surimi were unfolded by heat denaturation and then polymerized on cooling (Cen et al., 2022). With the addition of CAR, the pore size of protein cavities in the gel of minced shrimp became smaller and the number decreased, and at 0.3% CAR, the distribution of the particle matrix was more uniform, and the network structure was smoother and firmer, which improved the denseness of the gel network structure and maintained the homogeneity and stability of the gel texture. It showed that the addition of CAR can repair the large number of voids in the gel produced by thermal deformation, and therefore, presenting a more regular and orderly gel network structure. However, with the continuous increase of CAR addition (0.5%), the gel network structure inside the gel of minced shrimp gradually changed into a hierarchical network, which may be due to the addition of too high concentration of CAR which changed the surface hydrophobicity of myofibrillar fibrous protein particles and zeta potential and other particles properties (Ouyang et al., 2021).

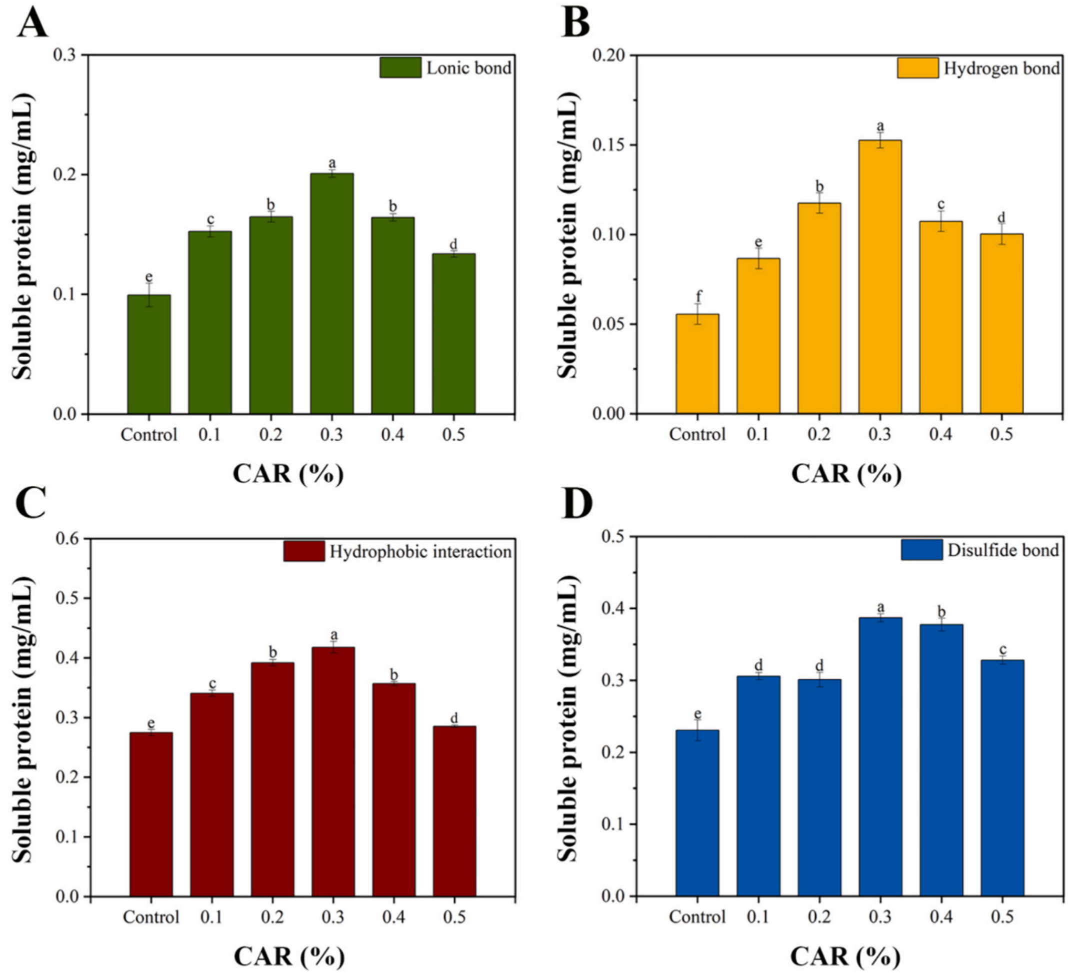

Fig. 4. Variation of intermolecular forces in κ-carrageenan and crayfish surimi composite gel (CAR-CSM). Ionic bonding (A), Hydrogen bonding (B), Hydrophobic interactions (C), Disulfide bonding (D).

3.8. Sensory evaluation

Sensory evaluation is an important index for evaluating food quality. Based on this, we comprehensively investigated the quality of minced shrimp from multiple dimensions, such as acceptability, elasticity, color, flavor, and texture. As shown in the Fig. S1, the overall acceptability score of minced shrimps showed an increasing and then decreasing trend with the increasing concentration of CAR in the system. The highest acceptability score of 8.4 was obtained at a CAR concentration of 0.3%, while the elasticity and flavor scores of the minced shrimp showed a similar trend. This was attributed to the fact that the myogenic fiber proteins in the minced shrimp combined with CAR to form a solid three-dimensional network structure, which led to a significant improvement in the water-holding property, elasticity, and homogeneity of texture of the minced shrimp samples. However, with the increase of CAR concentration, the flavor, elasticity, and overall acceptability of minced shrimp showed a decreasing trend, which might be due to the fact that excessive CAR addition in the minced shrimp system can result into compact whole gel network structure, harder texture, and profound gelatinous feature.

4. Conclusion

Based on the special structural signs of CAR, a series of CAR-CSM systems with different concentrations were constructed for improving the quality of minces shrimp gel. The results showed that 0.3% CAR enhanced the cooking loss rate, gel strength, and whiteness of CAR-CSM to some extent. With the increase of CAR addition, the hardness, elasticity, cohesivity, chewability, and recovery of minced shrimp gel were increased at first and then decreased, with a more ordered secondary structure of CAR-CSM. At 0.3% of CAR, the ratio of β-sheet reached the peak of 57.18, the overall chemical strength of the gel system reached the highest value, the pores of the gel network structure decreased, and the network structure became denser and smoother. Excessive CAR could produce accumulation effect in the gel system, preventing the cross-linking of hydrophobic groups between protein molecules, and the chemical force in the system were gradually changed from hydrophobic interaction to disulfide bond, the gel network was broken, the hydration ability of proteins was weakened, and the overall quality of CAR-CSM gel was declined. In this study, the quality difference of the gelling properties of crayfish myofibrillar protein prepared by different CAR concentrations can provide the basic framework for the widespread application of CAR in a variety of seafood products.

CRediT authorship contribution statement

Qing-Jun Wei: Resources, Methodology, Investigation. **Wang-Wei Zhang:** Writing – original draft, Methodology, Investigation. **Jing-Jing Wang:** Visualization, Software, Resources. **Kiran Thakur:** Visualization, Validation, Software. **Fei Hu:** Software, Resources. **Mohammad Rizwan Khan:** Validation, Project administration, Funding acquisition. **Jian-Guo Zhang:** Writing – review & editing, Supervision, Project administration, Conceptualization. **Zhao-Jun Wei:** Writing – review & editing, Supervision, Project administration, Funding acquisition,

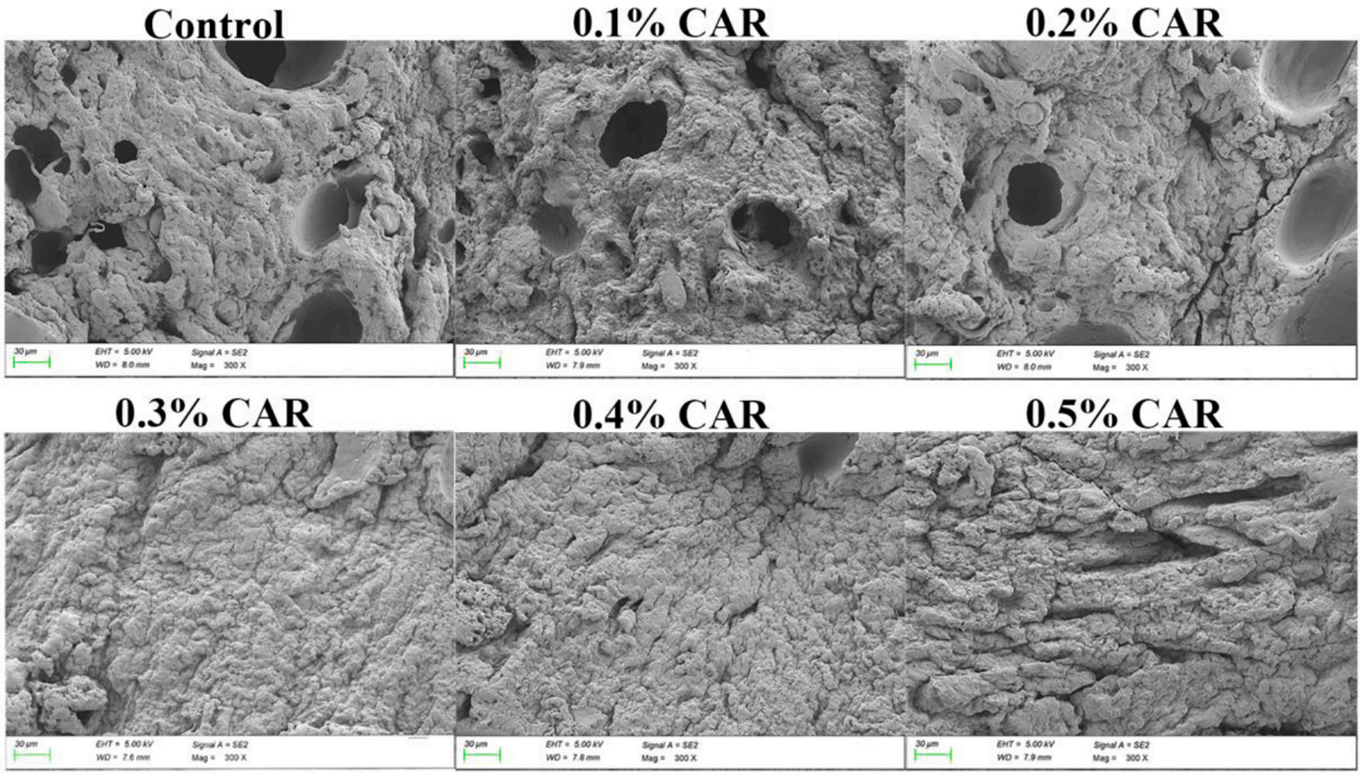

Fig. 5. Microstructure of κ-carrageenan and crayfish surimi composite gel (CAR-CSM).

Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

Acknowledgements

This study was supported by the Major Projects of Science and Technology in Anhui Province (202203a06020009, 202203a06020028, 2023n06020054), the Natural Science Foundation of Ningxia province (2022AAC03262, 2023AAC05043), and the Researchers Supporting Project number (RSP2024R138) King Saud University, Riyadh, Saudi Arabia.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2024.101497.

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