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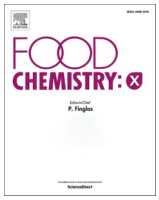

Effect of cellulose on gel properties of heat-induced low-salt surimi gels: Physicochemical characteristics, water distribution and microstructure

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ABSTRACT

The processing of surimi products requires the addition of high levels of salt, which makes it a high-salt food that poses a risk to human health. The search for exogenous additives to reduce the salt content of surimi products while ensuring their quality characteristics is crucial. Therefore, the effect of different species of cellulose on enhancing the quality characteristics of low-salt surimi gels was investigated and the best-modified cellulose was identified. Carboxymethyl cellulose (CMC), hydroxypropyl methylcellulose (HPMC), and microcrystalline cellulose (MCC) were selected for this study to compare with high-salt control and low-salt control. The results showed that cellulose could induce conformational transitions of proteins and promote the formation of an ordered and dense surimi gel network and the minimum porosity of 15.935% was obtained in the MCC-treated group. The cellulose-treated group conferred good textural properties to the surimi gels, significantly improved gel strength and water retention capacity ($p < 0.05$), and reduced the amount of water lost after cooking treatment ($p < 0.05$). Low-field NMR results showed that cellulose reduced the release of water, converting more free water to immobile water, thus increasing the water proton density. The higher energy storage modulus G' in the presence of cellulose indicated a more stable surimi gel system dominated by springiness. In summary, cellulose could confer better quality characteristics to low-salt surimi gels and MCC performance was superior to other cellulose species. This study helps the understanding of the mechanism of cellulose-surimi action on the development of high-quality low-salt surimi gels.

Introduction

Surimi is a concentrated myogenic fibrous protein by fine filtration, rinsing, and dehydration of raw fish muscle tissue (Mi et al., 2021a). Surimi products, including fish balls, fish intestines, and fish tofu, are popular among consumers (Yingchutrakul et al., 2022; Zhao et al., 2019). Golden threadfin bream (*Nemipterus virgatus*) is an important marine economic fish in China, with an expected catch of approximately in 2021 is expected to be about 32,200 tons in 2021 (Bureau of Fisheries Management Chinese Ministry of Agriculture, 2022, pp. 39). With its tender meat, high protein content, and high yield, the golden threadfin bream has become an essential raw material for producing surimi and surimi products (Fang et al., 2021; Nadeem Muhammad et al., 2018a; Mi et al., 2021b).

Myofibrillar protein is a class of salt-soluble proteins. Therefore, in the processing of surimi products, it is typically necessary to add 2–3% salt during the chopping stage of surimi to precipitate and unfold myofibrillar proteins. This process is crucial for fully cross-linking them during the subsequent gelation to obtain surimi products with desirable gelation properties (Núñez-Flores et al., 2018; Nadeem Muhammad et al., 2021; Tahergorabi et al., 2012). However, consumers have become increasingly health-conscious about healthy foods recently, and excessive sodium intake could cause diseases such as hypertension and heart disease. As a result, food salt reduction has been advocated (Gao et al., 2023). Unfortunately, the reduction of salt concentration usually hurts the functional properties of surimi products (Zhu et al., 2021). To

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address this issue, researchers have found that the use of gel-enhancing additives can be effective in compensating for the loss of quality in surimi products due to salt reduction (Monto et al., 2022). For example. Xiong et al. (2021) found that low-salt (0.5 g/100 g) surimi gels with 0.1 g/100 g of L-arginine and 0.5 g/100 g of oxidized caffeic acid (0.5 g/100 g) surimi gels were able to improve the gel strength by 1.3-fold. Similarly, Cando et al. (2016) found that the addition of microbial glutamine aminotransferase with lysine (0.1 g/100 g) or cystine (0.1 g/100 g) also significantly improved the properties of low-salt (0.5 g/100 g) surimi gels.

Dietary fiber has several benefits, including preventing cardiovascular disease, diabetes, obesity, and intestinal disorders, as it is resistant to human digestion and absorption. Additionally, it has hydrocolloid properties and is often used as an improver in food development and production (Debusca et al., 2014). Sánchez-González et al. (2009) suggested that cellulose could act as an active dehydrating agent, resulting in the exposure of surimi sols to hydrophobic side chains while promoting the increase of β-sheet structure. The application of modified cellulose in surimi products has received much attention. For instance, Piao et al. (2022) prepared functionalized cellulose nanofibrils with high surface hydrophilicity and activated –coo- groups and used them at a very low concentration (0.1 g/100 g surimi) by matrix enhancement, water binding, and encapsulation. Zhang et al. (2023) found that 1.5 g/100 g of CMC improved the quality characteristics of surimi gel. Chen (2006) found that although Hydroxypropyl methylcellulose (HPMC) did not strongly cross-link with other components during the gelation of surimi, the HPMC gel occupied a certain space at high temperature and formed a gel with higher rigidity and gel strength of the combined gels.

However, more discussions have focused on the effect of cellulose on normal high-salt surimi products, and there are few reports on the improvement of cellulose on the quality of low-salt surimi products. Therefore, this study investigated the effects of three modified cellulose (Carboxymethyl cellulose, Hydroxypropyl methylcellulose, and Microcrystalline cellulose) on the gelation properties of low-salt (1 g/100 g) surimi products. Furthermore, the mechanisms of their effects in combination with the moisture distribution state and rheological properties were analyzed. This study aims to provide theoretical references for improving the gel characteristics of low-salt surimi products, reducing the salt content of surimi products, and developing healthy surimi products.

Materials and methods

Materials

The raw material fish (*Nemipterus virgatus*) was purchased from Zhanjiang Aquatic Market (Zhanjiang, Guangdong, China), and weighed 100±10 g each. The three celluloses used in this research: Carboxymethyl cellulose (CMC), Hydroxypropyl methylcellulose (HPMC), and Microcrystalline cellulose (MCC) for food grade were obtained from Henan Wanbang Industrial Co., Ltd. (Zhengzhou, Henan, China). All other chemical reagents were of analytical grade and were supplied by Sinopharm Chemical Reagent Company Limited (Shanghai, China).

Preparation of surimi

First removed the head, scales, and internal organs of the chilled golden threadfin bream. Then, after washing off the blood, the golden threadfin bream was removed from the skin and spines. Further, connective tissue and other impurities were removed with a fine filter, and then antifreeze was added to obtain surimi. Precisely weighed 3 g of surimi sample and dried it at 105℃ to a constant weight and then determined its moisture content of 75.23 g/100 g (water/surimi). Finally, the surimi sample was stored at -20℃.

Preparation of surimi gels

The prepared surimi was chopped for 3 min in a blender, 2 g/100 g (salt/surimi) salt was added and chopped for 3 min, finally the water content of the surimi was adjusted to 78 g/100 g (water/surimi) with chilling deionized water and chopped for 1 min. This sample was used as a control group for high-salt content surimi gel labeled as H-Salt. low-salt (L-Salt) content group of surimi gel was prepared with the same process but the salt supplementary amount was reduced to 1 g/100 g (salt/surimi). Based on low-salt surimi gel (1 g/100 g), 0.4 g/100 g (cellulose/surimi) CMC, HPMC, and MCC three kinds of cellulose were added at the end as the treatment group respectively. And the whole process temperature was controlled below 10 °C. Two-stage heating (30 min at 40 °C and 20 min at 90 °C) was used to obtain surimi gel samples and stored overnight at 4 °C.

Gel strength

Gel strength measurements were performed using a TA-XT Plus Texture Analyzer (Stabilized Microsystems, Godalming, UK) equipped with a P/0.5S probe model. The deformation rate was 75%, the compression distance was 10 mm and the trigger force was 10 g. Six replicates were performed for each group of samples.

Texture profile analysis (TPA)

Texture analysis using TA-XT Plus Texture Analyzer (Stabilized Microsystems, Godalming, UK) equipped with a P/0.5 probe model. The deformation rate was 40%, the speed of the test was 1 mm/s and the trigger force was 10 g. Each group of samples was tested in 10 replicates to ensure accuracy and reliability.

Color

The color of the surimi gel was measured with reference to the method described by Yi et al. (2020a). The gel samples were prepared as cylinders with a height of 20 mm and the brightness (L*), red and green values (a*), and yellow and blue values (b*) were recorded with a Chroma Meter CR 400 Colorimeter (Konica-Minol-ta, Japan). Whiteness calculation based on Equation (3). Six samples were prepared in parallel for each group, ensuring accuracy and consistency in the results.

$$\text{Whiteness} = 100 - \left[(100 - L^*)^2 + a^{*2} + b^{*2}\right]^{1/2} \quad (1)$$

Water-holding capacity (WHC)

Determination of water holding capacity (WHC) and weight loss (WL) of surimi gel samples were referred to the method described by Zhang et al. (2023). The WHC was assessed using the following equation (2), and three replicates of each gel sample ensured accurate results. W1 and W2 represent the weight of the sample before and after centrifugation, respectively.

$$\text{WHC}(\%) = W2 \div W1 \times 100 \quad (2)$$

The Weight loss (WL) was calculated using equation (3), and three replicates of each gel sample ensured accurate results. G1 and G2 represent the weight of the samples before and after steaming treatment, respectively.

$$\text{WL}(\%) = (G1 - G2) \div G1 \times 100 \quad (3)$$

Low-field nuclear magnetic resonance (LF-NMR)

Surimi gel samples with a height of 20 mm were prepared and subjected to 90° and 180° CPMG sequence pulse measurements in the sample chamber with NMR pulse analyzer (MicroMR, Niumag Co.,

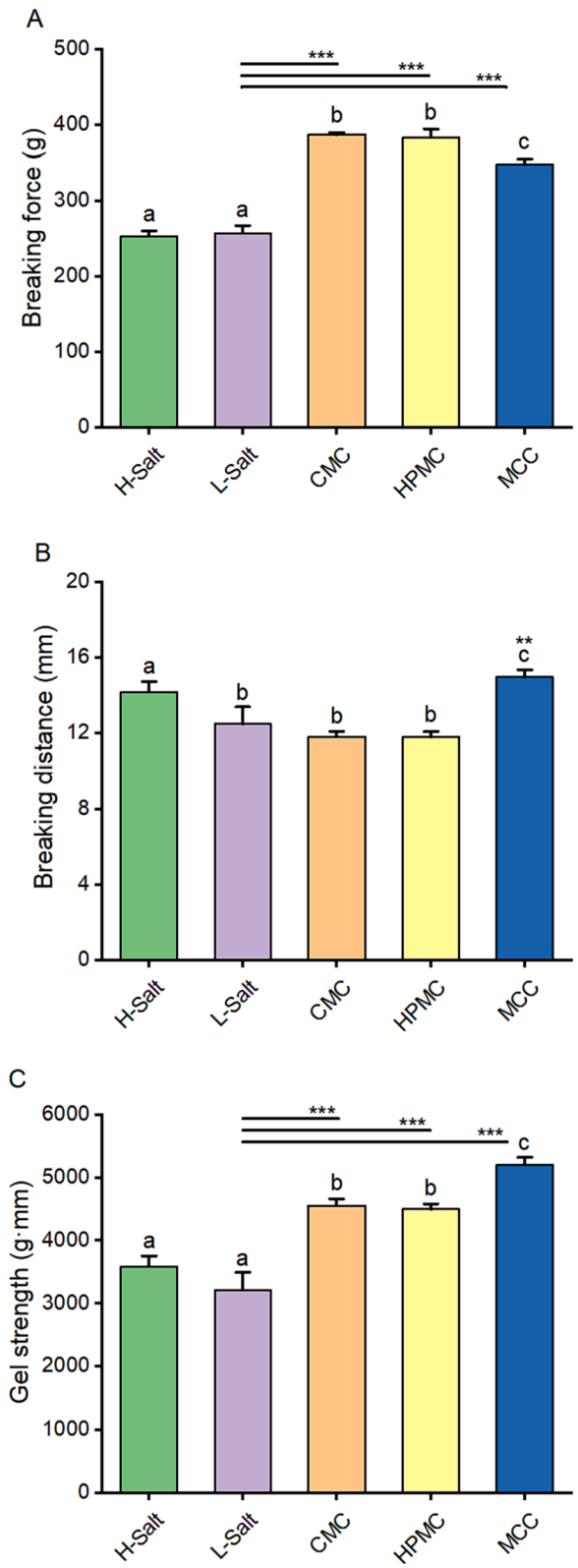

Fig. 1. Effect of different cellulose on the Breaking force (A), Breaking distance (B) and Gel strength (C) of surimi gels. Different letters in the figure indicate significant differences among different cellulose species ($p < 0.05$). The asterisk indicates the difference between the treatment group and the 1 g/100 g low-salt group (*$p < 0.05$, **$p < 0.01$, ***$p < 0.001$). H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.

Shanghai, China), and the data were inverted to acquire the water composition and distribution in the surimi gel samples.

Magnetic resonance imaging (MRI)

The proton density assessment of surimi gels was conducted using Magnetic Resonance Imaging (MRI) with the same LF-NMR analyzer mentioned earlier. The grayscale images obtained were transformed into proton density-weighted color images using NMR image processing software V3.0 with uniform pseudo-color processing.

Dynamic rheology

The dynamic rheology was performed by referring to the method of Zhang et al. (2023) using a temperature scan mode with a temperature range of 20–90 °C. The values of storage and loss modulus (G' and G'') during the heating process were recorded.

Fourier transform-infrared spectroscopy (FT-IR)

The freeze-dried surimi gel samples were mixed with potassium bromide (1:100, g/g) and the infrared spectra were determined using a Nicolet iS-5 FT-IR spectrometer (Thermo Fisher Scientific, USA). The analytical parameters of the FT-IR spectra were as follows: wave number range of 400–4000 cm^{-1}, scanning frequency of 64 times, and resolution of 4 cm^{-1}.

Scanning electron microscopy (SEM)

Surimi gel samples (5 mm × 5 mm × 10 mm cubes) were prepared, immersed in 2.5% glutaraldehyde solution for 24 h, dehydrated in anhydrous ethanol, and finally freeze-dried before observation using a scanning electron microscope (SEM, Regulus 8100, Hitachi, Tokyo, Japan).

Scanning electron microscopy images of MP-CUR gels at a magnification of 3 k were selected for analysis. The porosity of the gel samples was obtained by ImageJ software (National Institutes of Health, Bethesda, MD, USA) processing, and the porosity was calculated by referring to the following Equation (4).

$$\text{Porosity}\ (\%) = (\text{Pore area}/\text{Total image aera}) \times 100\% \quad (4)$$

Statistical analysis

Data were statistically analyzed using SPSS 19.0 software (SPSS Institute Inc., Chicago, USA) and figures were created using Origins version 2021 (Origins Laboratories, Northampton, USA). Experimental results were replicated at least three times, and mean comparisons were assessed using Duncan's multiple range test and expressed as mean ± standard error (SE) (Nadeem Muhammad et al., 2018b).

Results and discussion

Gel strength

The gel strength is usually used to evaluate the quality characteristics of surimi products (Walayat et al., 2022). Effect of cellulose on gel strength of surimi refers to Figure 1, where the change was mainly influenced by the breaking force (Figure 1A, $p < 0.05$). As shown in Figure 1C, the gel strength of 2 g/100 g high-salt surimi (3584.55 g·mm) was slightly higher than that of 1 g/100 g low-salt surimi gel (3210.72 g·mm). The addition of salt promoted the solubilization of salt-soluble proteins in surimi, forming a more dense three-dimensional gel network structure. However, the enhancement effect was not significant ($p > 0.05$). In contrast to the 1 g/100 g low-salt control group, the addition of cellulose very significantly enhanced the strength of surimi

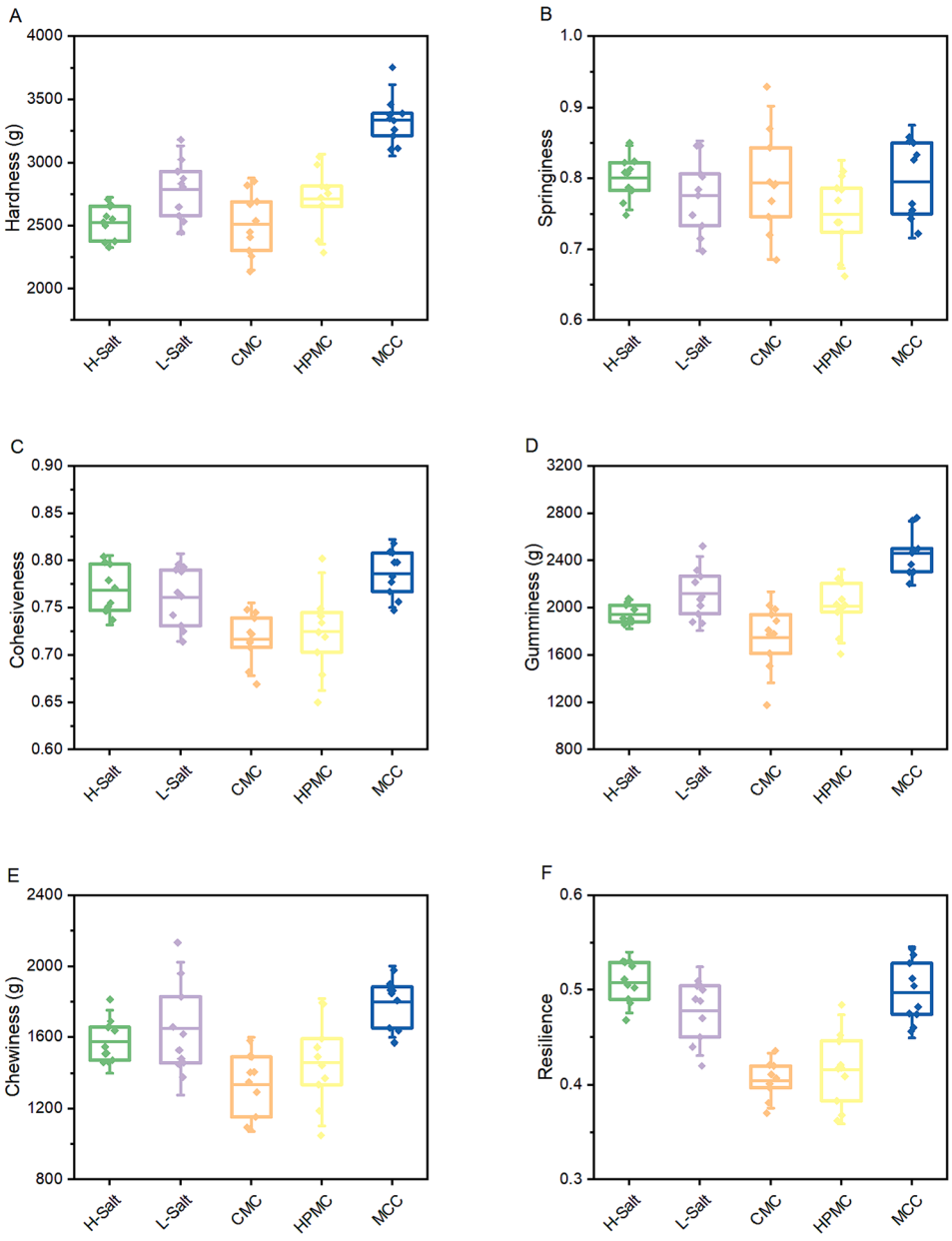

Fig. 2. Effect of different cellulose on the texture profile of surimi gels. Where A-F stand for Hardness, Springiness, Cohesiveness, Gumminess, Chewiness, Resilience. The box represents the 25%-75% confidence interval of the data and the horizontal line represents the mean. H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.

gels ($p < 0.001$), and the effect was higher than that of the 2 g/100 g high-salt control group ($p < 0.05$). Among the three cellulose species, MCC showed the highest gel strength. When MCC was added, the gel strength of low-salt surimi increased from 3210.72 g·mm to 5201.21 g·mm (Figure 1C). We could speculate that the physical filling of the gel network by the MCC after water absorption and expansion could cause this experimental result. Piao et al. (2022) found that oxidized cellulose nanofibers combined with surimi of striped bass could act as physical fillers to enhance protein-cellulose interactions and thus enhance gel properties. The effect of CMC and HPMC on low-salt surimi gels was primarily observed in the breaking force, rather than the breaking distance. The addition of CMC and HPMC significantly increased the breaking force of the gel system, whereas the deformation variable exhibited a decrease in results ($p < 0.05$). This suggested that CMC and HPMC have the ability to change the hardness but do not affect the springiness of surimi products.

Appropriate amounts of carboxymethylated cellulose nanofibers (cCNF) could interact with cow myofibrillar proteins (CCMP) via hydrophobic interaction leading to a decrease in surface hydrophobicity (Zhang et al., 2022). Consequently, CMC and HPMC have the potential to enhance the stability of the gel system by influencing the chemical forces and the formation of protein secondary structure following water absorption and swelling. Comparable findings were obtained from Fourier infrared spectroscopy.

Texture profile analysis (TPA)

TPA is used as a key indicator to assess the structural properties of solid or semi-solid foods, providing information about the mechanical properties of foods based on the method of two compressions. This method has been universally used in the quality evaluation system of surimi products (Yi et al., 2020b). Figure 2 illustrates the effect of different types of cellulose on the textural properties of surimi gels. Six parameters of surimi gel textural properties were obtained by TPA.

Table 1
Color changes of surimi gel under different cellulose treatments.

Sample	Color			
	L*	a*	b*	Whiteness
H-Salt	84.77±0.20^{ab}	0.93±0.71^{a}	10.83±0.08^{a}	81.28±0.15^{ab}
L-Salt	83.92±0.24^{a}	0.38±0.48^{b}	10.10±0.12^{b}	81.00±0.16^{ab}
CMC	84.25±0.87^{a}	1.28±0.61^{c}	11.07±0.15^{a}	80.68±0.77^{a}
HPMC	86.48±0.34^{b}	1.62±0.31^{d}	11.07±0.15^{a}	82.45±0.24^{b}
MCC	85.42±0.24^{ab}	0.93±0.21^{a}	12.81±0.05^{c}	80.57±0.21^{a}

Note: Different letters in the same column mean significant differences at different cellulose species ($p < 0.05$). Data are shown as means ± SE (n = 6). H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.
Note: kindly check footnote 1 text (need or not).

Compared to the 1 g/100 g salt addition group, the 2 g/100 g group had higher springiness, cohesiveness, and resilience and lower hardness, gumminess, and chewiness. This is because the addition of more salt fully dissolved the salt-soluble proteins in the surimi, forming a more elastic and compact network after gelation. These results are consistent with the breaking force and breaking distance in gel strength. When cellulose was added, the addition of CMC improved the springiness of the low-salt surimi gels, and the springiness results were better than those of the 2 g/100 g high-salt gel group. However, performances for other physical properties were poor, which were even lower than that of the 1 g/100 g low-salt gel group. Similar results were obtained in a study by Zhang et al. (2022). The addition of a moderate amount of cellulose increased the springiness of the gel network, which is primarily dominated by particle–particle interactions, resulting in better gel springiness. MCC was the best-performing cellulose among the three types of cellulose, and the addition of MCC improved all six physical properties in Figure 2A-F and improved the qualitative properties of the low-salt

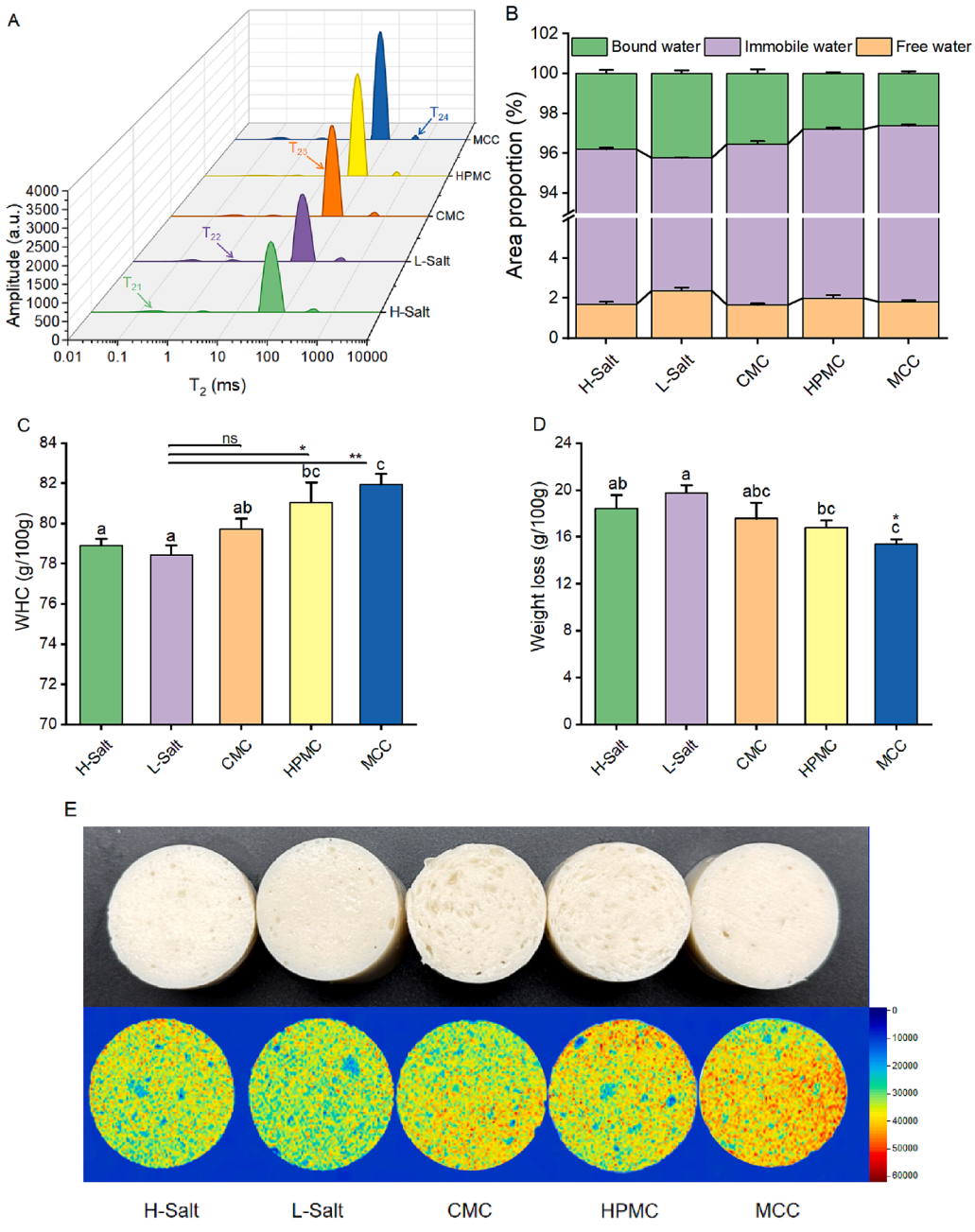

Fig. 3. Effect of different cellulose on the spin–spin relaxation times T_2 (A), relative area percentages of $T_{21} + T_{22}$, T_{23} and T_{24} peaks (B), water holding capacity (C), weight loss (D) and hydrogen proton density images (E) of surimi gels. Different letters in the figure indicate significant differences among different cellulose species ($p < 0.05$). The asterisk indicates the difference between the treatment group and the 1 g/100 g low-salt group (*$p < 0.05$, **$p < 0.01$, ***$p < 0.001$). H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.

surimi gels better than the 2 g/100 g high-salt gels. The MCC showed better compatibility in the surimi system. MCC was filled into the gel structure of surimi by heating gelation to form a more elastic and reversible network structure. When the surimi gel is cooled to room temperature, the MCC formed a sticky paste to increase the stiffness of the surimi gel (Gibis et al., 2015), which is responsible for the higher gel strength of the MCC.

Color

The color of surimi products is a crucial judging criteria for consumers when selecting meat products, and it can be evaluated by measuring L*, a*, b*, and whiteness (W) to reflect the color variation of meat products. Usually, surimi gels with higher L* and whiteness values indicate better quality (Chen et al., 2020). The presence of cellulose promotes an ordered and dense gel network structure, where the pores inside the gel network were filled with dissolved cellulose. The smooth cross-section is more favorable for light reflection, and a large amount of light being reflected can promote whiteness. (Buda et al., 2021). Table 1 shows that the different cellulose treatment did not significantly affect the brightness and whiteness of the surimi gels ($p > 0.05$). This suggests that the addition of cellulose altered the moisture content and substrate of the system, leading to fluctuations in light reflection, but did not result in significant differences. This outcome may also be attributed to the type, color, and concentration of the cellulose polysaccharide used (Oujifard et al., 2012).

Water holding capacity (WHC)

The moisture content of the surimi sample which we prepared was 75.23 g/100 g. The water holding capacity (WHC) of surimi gels refers to the water holding rate of the gel system under external forces, which represents the binding of the gel structure to water. Weight loss of surimi gels after heating is dominated by water during the cooking process. WHC and cooking loss have a negative correlation and are often used to evaluate the organoleptic properties of surimi products (Ma et al., 2015). As depicted in Figure 3C, the WHC of the 2 g/100 g high-salt gel group (78.89 g/100 g) was higher than that of the 1 g/100 g low-salt gel group (78.44 g/100 g). The high-salt environment can enhance the interaction between protein and water molecules through hydrogen bonding, resulting in the retention of more water molecules in the gel network. This is because the dominant factors affecting water holding capacity (WHC) are the electrostatic interactions and hydrogen bonding between water and protein (He et al, 2022). Compared to the 1 g/100 g low-salt control (WHC 78.44 g/100 g), the addition of carboxymethyl cellulose (CMC) to surimi gels increased the water holding capacity (WHC) to 79.72 g/100 g, although this difference was not statistically significant ($p > 0.05$). CMC is a naturally occurring cellulose that has been modified by carboxymethylation to make it water-soluble. During this process, anions are introduced to the cellulose, which prevents aggregation and increases the interaction between the cellulose and water molecules. As a result, more water molecules are trapped, leading to improved WHC (Pematilleke et al., 2021). Compared to the 1 g/100 g low-salt control group, the MCC group obtained highly significant ($p < 0.01$) improvement in WHC performance with a maximum value of 81 g/100 g. The smaller size of cellulose allowed for the full exposure of hydrophilic groups and easier binding to water molecules, resulting in a better WHC performance. Zhang et al. (2018) also obtained similar results when investigating the effect of cNFC on TGase induced salt-soluble proteins in chicken, and found nanosized cellulose polysaccharides could improve the water retention of salt-soluble proteins.

The cooking loss is an indicator of the ability of the surimi system to retain water during the middle and late stages of the protein thermal aggregation process caused by denaturation. As shown in Figure 3D, the cooking loss of the MCC-treated group (15.37 g/100 g) was significantly lower than that of the 1 g/100 g low-salt gel group (19.73 g/100 g) and 2 g/100 g high-salt gel group (18.44 g/100 g). This finding suggested that the MCC acted as an effective thickening agent in the surimi gel system. The hydrophilic groups of MCC facilitated the absorption of water molecules, which was highly correlated (Vasquez Mejia et al., 2018). Additionally, the pasted MCC polysaccharide absorbed more water and filled the internal pore structure of the surimi gel matrix, thereby enhancing water binding.

Water distribution of different surimi gels

The spin–spin T_2 relaxation times of the surimi gels are presented in Figure 3A. The components (T_{21}, T_{22}, T_{23}, T_{24}) were isolated and observed in the continuous plots of all samples. The accumulation of T_{21} (0–1 ms) and T_{22} (1–10 ms) accounted for 2.63%-4.23% of the total signal and was attributed to the tight binding of water to the protein molecules through the functional groups. This was the first water component of the system to pass through the plots and was defined as bound water. T_{23} (40–160 ms) was the most dominant water component accounting for>93.41% of the total signal, representing immobilized water and considered as the main water component in the gel network. The water T_{24} (300–650 ms), which appears most slowly in the plot at a rate of 1.64–2.36%, corresponds to water free from myogenic fibronectin, or free water trapped in the macropores of the gel network. During high-temperature gelation, cellulose promoted protein–protein and protein-water interactions in the surimi gel system, enhancing protein cross-linking and aggregation, prompting the formation of a regular and ordered gel network, weakening the mobility of water, and allowing more water to be trapped and immobilized in the surimi gel network (Zhang et al., 2023). As a result, the cellulose group in the figure has a higher T_{23} peak. The variation of the T_{23} relaxation curve was generally consistent with the results of WHC.

The percentage of the water presence state in the samples is shown in Figure 3B. The 1 g/100 g low-salt gels bound less immobilized water in the gel network (93.41%), and the presence of cellulose allowed most of the free water to bind to proteins to form immobilized water and be encapsulated in the gel network. Meanwhile, the MCC obtained an immobilized water maximum of 94%, which was attributed to the micron size of cellulose, leading to the decrease of T_{24} and the increase of T_{23} in the gel system (Xiao et al., 2021). These findings all indicated a facilitative effect of cellulose on the transfer of free water to the immobilized water state, which well supports the results of WHC and steaming losses.

Appearance and MRI image of different surimi gels

The distribution of water molecules in surimi gels can be visualized more clearly using MRI techniques, as demonstrated in Figure 3E, which displays the MRI images and apparent cross-sectional maps of the corresponding groups after proton density weighting (Liu et al., 2021). MRI technique could demonstrate the distribution of water molecules in surimi gels more visually, and Figure 3E exhibited the MRI images and apparent cross-sectional maps of the corresponding groups after proton density weighting (Yi et al., 2023).

The MRI images reveal that immobilized water ($T_{23} > 93.41\%$) is more prevalent due to the low proportion of bound water ($T_{21} + T_{22} < 4.23\%$) and free water ($T_{24} < 2.36\%$). The differences between the sample groups were referenced by color bars, which ranged from red (high-density protons) to blue (low-density protons). Samples containing cellulose exhibited higher red and yellow pixels, indicating a higher density of water protons. MCC-bound surimi gel samples had the largest proportion of red pixels, also suggesting that they contain the largest number of water molecules of all the groups. Furthermore, the apparent cross-sectional maps of the MCC samples also showed tight, smooth cross-sections. It was speculated that the reason for the excellent performance of MCC was due to the fact that some of the water molecules of the tiny size cellulose formed a complex, while the rest were tightly

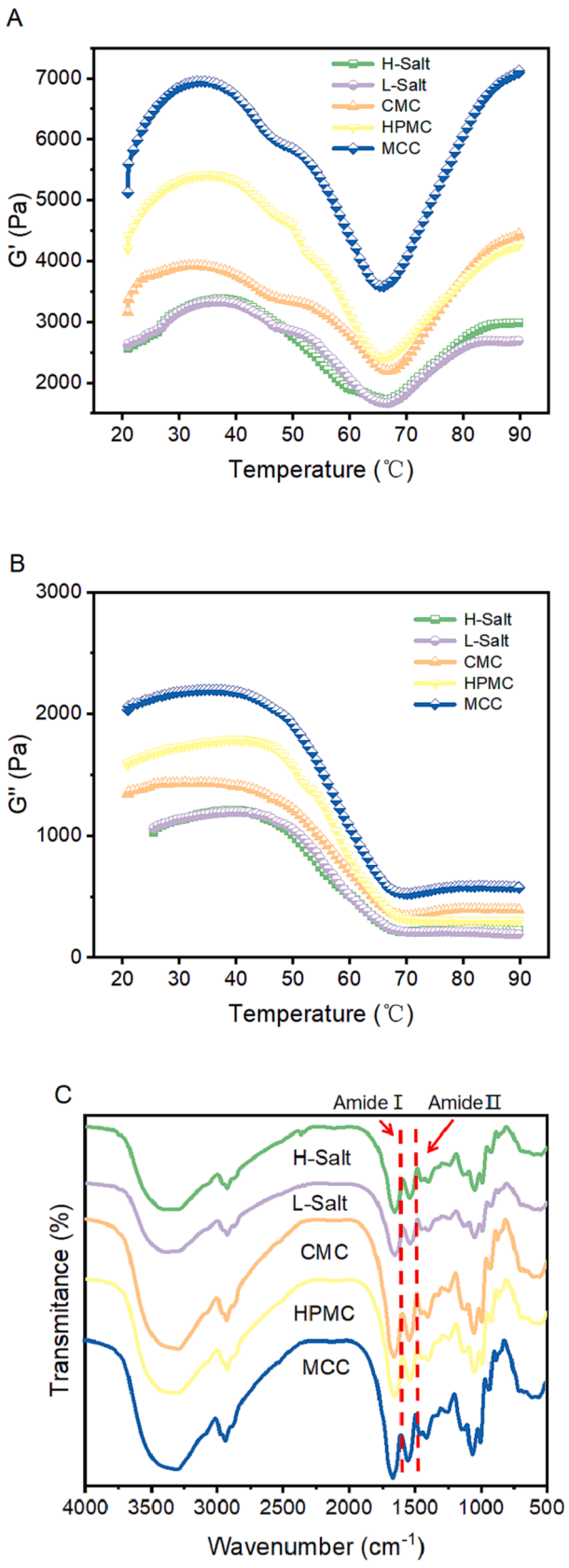

Fig. 4. Changes in storage modulus (A), loss modulus (B) and fourier infrared spectra (C) of surimi gels after treatment with different species of cellulose. H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.

cross-linked with the protein, thereby locking more water to stay in the homogeneous gel network and adsorbing it inside the gel (Zhang et al., 2023). This finding was consistent with the trend of T_2 relaxation time and WHC.

Rheological properties

Dynamic rheology in the surimi gel system is used to describe the viscoelastic behavior of surimi due to denaturation and aggregation of myosin during heating. The energy storage modulus (G') represents the elastic characteristic of the fluid, while the loss modulus (G'') represents the viscous characteristic of the fluid. Typically, the value of G' was greater than the G'' throughout the heating process, indicating that the entire surimi gel system was dominated by elastic characteristics. Figure 4A and Figure 4B illustrate the dynamic modulus of each sample throughout the heating process. The cellulose treatment group had higher G' and G'' than the salt control group, with the 1 g/100 g low-salt gel group exhibiting the smallest G' and G'' values. Throughout the heating process, the G' and G'' values of each treatment group followed the same trend: G' increased slightly at 30-35℃ due to protein–protein interactions at low temperatures and the initial formation of a looser gel network structure. As the temperature increased, G' decreased sharply and reached a minimum at around 65℃. The results suggest that cellulose treatment can enhance the viscoelastic properties of surimi gel. The endogenous proteins of surimi undergo thermally induced hydrogen bond breakage and proteolytic dissociation at this temperature. Afterward, G' rises sharply along with the temperature increase, leading to the formation of more denatured proteins were formed in the gel network. Eventually, G' decreases to a minimum of around 65℃ (Zhang et al., 2022).

Afterward, G' rises sharply along with the temperature increase, more denatured proteins are deposited faster in the gel network, and the number of protein crosslinks increases sharply. However, Wang et al. (2016) demonstrated that the anions and polar hydroxyl groups present in insoluble cellulose could promote protein interactions. The electrostatic interactions generated between cellulose and myofibrillar protein induce more protein unfolding, thus exposing more hydrophobic groups. The use of MCC, which has a micron size that facilitated protein interactions, resulted in superior G' values.

Fourier transform spectroscopy analysis of different surimi gels

The FT-IR spectra of surimi gels treated with different salt concentrations (1 g/100 g and 2 g/100 g) and various species of cellulose (CMC, HPMC, MCC) are shown in Figure 4C. No new peak was observed in the five groups, indicating that the alteration of salt concentration and cellulose did not result in the formation of new chemical covalent bonds. The two absorption peaks at 1612 cm^{-1} and 1504 cm^{-1} correspond to the amide I band (α-helix, α-helix, β-sheet, β-turn, and random coil, 1600–1700 cm^{-1}) and the amide II band (C-N and N-H stretching vibrations, 1500–1600 cm^{-1}), which are commonly used to evaluate the spectral characteristics of proteins (Chen et al., 2020). The increased signal intensity of the absorption peaks in the amide I and amide II bands in the cellulose-treated group was attributed to the presence of cellulose, which involves the formation of new compounds (C═N, C-O, C-N). In the amide I band, the protein secondary structure of surimi gels is mainly influenced by the α-helix, β-sheet, β-turn, and random coil structure contents. In this region, the enhancement of signal intensity in the amide I region after cellulose addition was attributed to the change in motif content, probably due to more α-helix structure unraveling and conversion to β-sheet and β-turn structure by intermolecular interactions (Piao et al., 2022). The formation of an ordered surimi gel system dominated by β-sheet and β-turn structure, gel strength, and microstructure could verify this speculation.

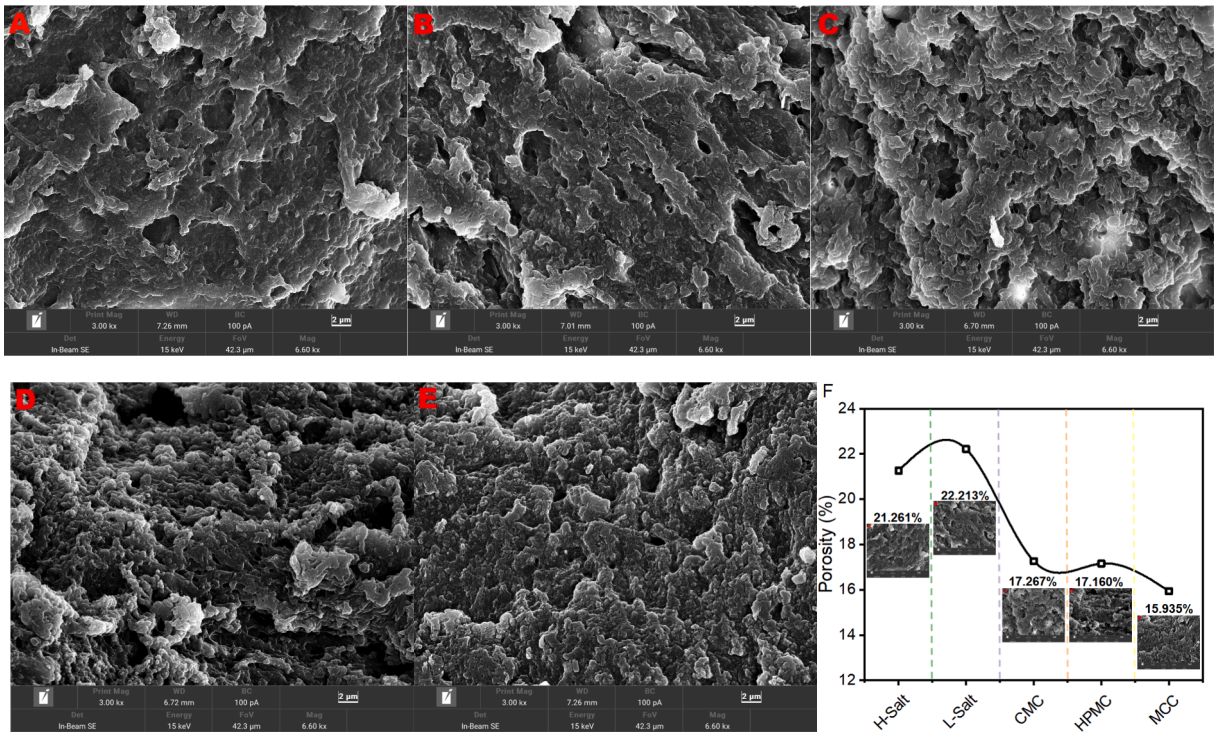

Fig. 5. Microstructure of surimi gels under different species of cellulose treatment (Magnification 3 k×). Note A-E in the figure: H-Salt, L-Salt, CMC, HPMC, MCC. Note F in the figure: Porosity of surimi gel in each treatment group. H-Salt stands for 2 g/100 g salt concentration, L-Salt stands for 1 g/100 g salt concentration.

Microstructure of different surimi gels

The microstructure of surimi gels is dependent on the degree and rate of protein aggregation, primarily myofibrillar proteins. As shown in Figure 5B, the microstructure of the 1 g/100 g low-salt surimi gel control sample exhibited a much coarser appearance with a loose, fractured, and disordered gel matrix. The porosity results obtained according to Equation (4) showed that the low-salt gel sample group had the highest porosity of 22.213%. This poor gel structure resulted in a large amount of water escaping from the gel interior, generating larger-sized pores, and reducing the water-holding capacity and cohesiveness of the gel structure (Shi et al., 2022). The 2 g/100 g high-salt gel control sample performed slightly better than the 1 g/100 g low-salt sample with 21.261% porosity but still had large cavities and internal pores. However, the addition of cellulose improved the poor condition and formed a uniformly aggregated, smooth, and tight structure in the gel matrix. Cellulose induced the proteins to clump tightly together, resulting in uniformly dense cavities inside the gel. HPMC-carrying cations appear as salt bridges between the negatively charged carboxyl groups of two neighboring protein molecules and consequently reduce the electrostatic repulsion between proteins and enhance protein cross-linking (Hu et al., 2017). On the other hand, MCC excelled in surimi gel structure due to its smaller micro size, which facilitated better protein cross-linking. And the lowest porosity of 15.935% was obtained for the MCC treated group of surimi gel.

Conclusion

This study investigated the effect of different species of modified cellulose on the improvement of gel quality of low-salt surimi. The addition of cellulose had a positive impact on the quality of poor low-salt surimi gels. Carboxymethyl cellulose (CMC), hydroxypropyl methylcellulose (HPMC), and microcrystalline cellulose (MCC) formed interpenetrating structures with the surimi protein system, promoting surimi gels with better three-dimensional gel networks and also significantly improving the strength of the gels. Among them, microcrystalline cellulose (MCC) represented a superior improvement with its micron-level size and better protein compatibility promoting gel strength from 3210.72 g·mm to 5201.21 g·mm compared to low salt surimi gel. And, the MCC treatment group had the lowest microstructural porosity of 15.935%. Additionally, the tightly ordered network structure could bind more immobilized water in the surimi gel, thereby inhibiting the migration of immobilized water to free water, altering the water distribution and state, and significantly improving the water-holding capacity. In summary, the cellulose improved the poor quality of the low-salt surimi gel, and the MCC effect was the most prominent. This study not only contributes to the understanding of the role and mechanism of cellulose in surimi gels but also to the development of high-quality low-salt surimi products.

CRediT authorship contribution statement

Chang Zhang: Conceptualization, Methodology, Investigation, Writing – original draft. **Lei Chen:** Conceptualization, Methodology, Investigation, Writing – original draft. **Minxin Lu:** Investigation, Validation. **Chao Ai:** Writing – review & editing. **Hui Cao:** Investigation, Validation. **Jianbo Xiao:** Investigation, Validation. **Saiyi Zhong:** Writing – review & editing. **Hui Teng:** Supervision, Writing – review & editing, Project administration, Funding acquisition.

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

The data that has been used is confidential.

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