

Catalyst-Free Fabrication of emulsion Gels from Cellulose-Based Pickering Emulsions via Synergistic Esterification and Hydrogen Bonding

Pingping Gao , Xiaoyun Chen , Ruizhi Yang , Fang Tian ,
Qiaoling Zhao , Isabel Lim Fong , Soottawat Benjakul ,
Yadong Zhao

PII: S2772-5022(26)00886-3
DOI: <https://doi.org/10.1016/j.afres.2026.102548>
Reference: AFRES 102548



To appear in: *Applied Food Research*

Received date: 2 April 2026
Revised date: 30 July 2026
Accepted date: 26 August 2026

Please cite this article as: Pingping Gao , Xiaoyun Chen , Ruizhi Yang , Fang Tian , Qiaoling Zhao , Isabel Lim Fong , Soottawat Benjakul , Yadong Zhao , Catalyst-Free Fabrication of emulsion Gels from Cellulose-Based Pickering Emulsions via Synergistic Esterification and Hydrogen Bonding, *Applied Food Research* (2026), doi: <https://doi.org/10.1016/j.afres.2026.102548>

This is a PDF of an article that has undergone enhancements after acceptance, such as the addition of a cover page and metadata, and formatting for readability. This version will undergo additional copyediting, typesetting and review before it is published in its final form. As such, this version is no longer the Accepted Manuscript, but it is not yet the definitive Version of Record; we are providing this early version to give early visibility of the article. Please note that Elsevier's sharing policy for the Published Journal Article applies to this version, see: <https://www.elsevier.com/about/policies-and-standards/sharing#4-published-journal-article>. Please also note that, during the production process, errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

© 2026 Published by Elsevier B.V.
This is an open access article under the CC BY-NC-ND license
(<http://creativecommons.org/licenses/by-nc-nd/4.0/>)

Highlights:

- Pickering emulsions were converted into emulsion gels via a catalyst-free thermal strategy.
- . Interfacial crowding was revealed to act as a spatial microreactor.
- A dual-crosslinking network was established via covalent and hydrogen bonds.
- Superior potential as sustainable fat replacers was demonstrated.

Catalyst-Free Fabrication of emulsion Gels from Cellulose-Based Pickering Emulsions via Synergistic Esterification and Hydrogen Bonding

Pingping Gao^{a,1}, Xiaoyun Chen^{b,1}, Ruizhi Yang^{b,*}, Fang Tian^b, Qiaoling Zhao^c, Isabel Lim Fong^{a,*}, Soottawat Benjakul^d, Yadong Zhao^{b,*}

^a Department of Paraclinical Sciences, Faculty of Medicine and Health Sciences, Universiti Malaysia Sarawak (UNIMAS), Kota Samarahan, 94300, Malaysia

^b School of Food and Pharmacy, Zhejiang Ocean University, Zhoushan, 316022, China

^c Zhoushan Institute for Food and Drug Control, National Ocean Food Quality Supervision and Inspection Center, Zhoushan 316000, China

^d International Center of Excellence in Seafood Science and Innovation (ICE-SSI), Faculty of Agro-Industry, Prince of Songkla University, Hat Yai, Songkhla 90110, Thailand

¹ These authors contributed equally to this work.

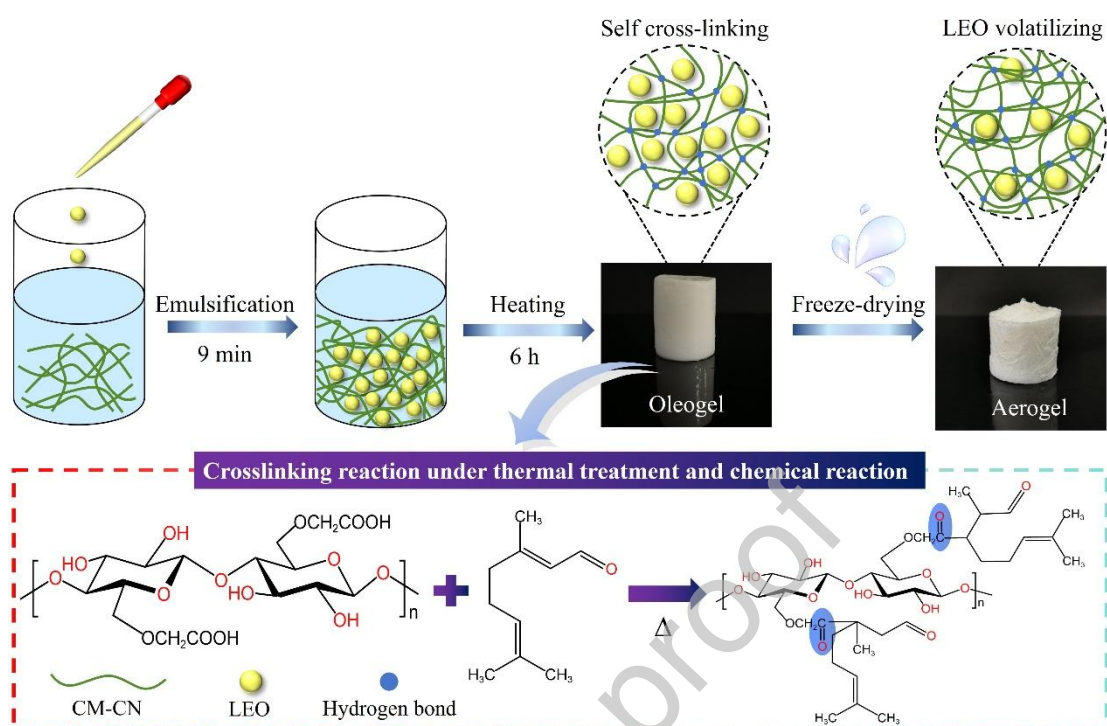
* Corresponding authors: ruizhiyang2001@163.com (R. Yang), flisabel@unimas.my (I. L. Fong) and zhaoyd@zjou.edu.cn (Y. Zhao)

Abstract:

In this study, emulsion-templated oleogels (emulsion gels) were prepared through simply heating Pickering emulsions composed of carboxymethylated cellulose nanofibrils (CM-CN) and lemon essential oil (LEO). Based on Fourier-transform infrared spectroscopy (FTIR) and microstructural characterizations, a dual-crosslinking mechanism is proposed: thermal treatment is hypothesized to induce esterification between the carboxyl groups of CM-CN and the aldehyde groups of LEO, supplemented by enhanced multiple hydrogen-bonding interactions. The prepared emulsion gels exhibited a dense network microstructure with quantitatively improved physical properties. Under optimized conditions (80 °C, O/W ratio of 1:5), the emulsion gels achieved a maximum hardness of 107.66 g and a chewiness of 54.99 g. Dynamic rheological measurements confirmed a robust solid-like behavior with the storage modulus (G') consistently dominating the loss modulus (G''). Furthermore, low-field NMR analysis indicated a significant reduction in transverse relaxation time (T_2), reflecting restricted water mobility and excellent water-holding capacity, while the resulting aerogels demonstrated a compressive strength of up to 0.1121 MPa. This study presents a facile, catalyst-free heating strategy to fabricate emulsion gels using Pickering emulsion templates, offering a highly promising alternative to saturated and trans fats in the food industry.

Keywords: Emulsion gels; Pickering emulsion template; Carboxymethyl cellulose; Lemon essential oil; Catalyst-free heat-driven.

TOC:



1. Introduction

Trans and saturated fats have a beneficial impact on certain food characteristics, such as texture, flavor, and shelf stability. Nevertheless, consuming high amounts of trans and saturated fatty acids is linked to a heightened risk of several health issues, including cardiovascular disease, Type II diabetes, stroke, and metabolic syndrome. Emulsion-templated oleogel (emulsion gel) is a gel-like substance that is either semisolid or has a solid-like consistency, created by blending edible liquid oil into a well-structured three-dimensional framework through various physical techniques, enzymatic approaches, chemical methods, or a mixture of these strategies (Zhi et al., 2023). To address these critical public health concerns, emulsion gels have emerged as an innovative, healthier alternative to solid fats (Hu et al., 2023; Zhao et al., 2021). For food-grade applications, the development of all-natural, "clean label" emulsion gels utilizing edible polysaccharides as gelators has become a prominent research focus. This strategy effectively avoids the cytotoxicity associated with synthetic polymer precursors or toxic chemical crosslinkers, while simultaneously improving the nutritional profile by substituting saturated fatty acids with polyunsaturated alternatives (Pintado et al., 2018).

To fabricate these polysaccharide-based emulsion gels, the emulsion-templating method is widely recognized as a highly efficient structural approach (Pan, Tang, Dong, Li, & Zhang, 2021). However, a critical technical bottleneck persists: systems with low oil fractions exhibit weak mechanical stability and are highly susceptible to flocculation

when relying exclusively on physical interactions, such as hydrogen bonding or chain entanglements (Farjami & Madadlou, 2019). To fortify these fragile networks, covalent crosslinking is often required. This creates a structural paradox in food science: introducing robust covalent bonds typically necessitates toxic chemical crosslinkers or external catalysts, which are strictly prohibited in food systems. Therefore, developing a robust, completely catalyst-free covalent crosslinking strategy within a natural emulsion template represents a formidable but essential challenge.

To overcome this limitation, natural nanocellulose emerges as an ideal structural building block due to its excellent biocompatibility, intrinsic mechanical strength, and abundance of reactive sites (Shao et al., 2020; Zhang et al., 2020). Specifically, carboxymethylated cellulose nanofibrils (CM-CN) feature a long, flexible backbone rich in anionic carboxymethyl ($-\text{CH}_2\text{COOH}$) and hydroxyl ($-\text{OH}$) groups (Kim, Kim, Park, & Hong, 2023). As demonstrated in our previous research, this unique structure grants CM-CN exceptional interfacial activity, allowing it to effectively stabilize oil-water interfaces to form robust Pickering emulsions (Chen et al., 2025). To complement this continuous phase, lemon essential oil (LEO) is selected not merely as a passive, flavor-enhancing oil phase, but as an active component. Its specific chemical constituents possess the chemical potential to engage in complex interfacial interactions with the functional groups of the CM-CN matrix, presenting a unique opportunity for synergistic network formation (Cebi, Taylan, Abusurrah, & Sagdic, 2021). Recently, magnetic aerogels were synthesized by Wang et al., utilizing sodium

carboxymethylcellulose (CMC) hydrogel and employing citric acid (CA) as a crosslinking agent, which was then processed using a vacuum freeze-drying method (Wang et al., 2021). Nevertheless, the synthesis of these emulsion gels occurred with crosslinking agents, and the investigation focused solely on the structure, morphology, thermal sensitivity, and swelling behavior of the emulsion gels.

Building upon these material properties, we hypothesize that a CM-CN-stabilized Pickering emulsion can overcome the energy barriers of traditional chemical reactions by serving as an effective spatial microreactor. At the oil-water interface, the high local concentration of CM-CN and LEO, combined with their initial pre-organization via hydrogen bonding, strategically aligns their reactive groups. This unique interfacial confinement facilitates a completely catalyst-free, one-step thermal gelation process. Driven solely by a 6-hour heating treatment, spontaneous ester linkage formation occurs between the active groups of CM-CN and LEO. This innovative approach leverages the synergy of permanent covalent esterification and reversible hydrogen bonding to construct a highly robust, dual-crosslinked network. To comprehensively elucidate this crosslinking mechanism and evaluate its efficacy, we systematically investigated the impact of different oil phases, LEO-to-water ratios, and heating temperatures on the mechanical strength, water-retention capacity, and microstructural characteristics of the resulting composite emulsion gels.

2. Materials and methods

2.1. Materials and chemicals

Softwood pulps (SW) were sourced from Jining Nantian Nongke Chemical Co., Ltd. (located in Jining, China). A lemon essential oil, with a purity level of 98%, was acquired from Macklin Biochemical Technology Co., Ltd (based in Shanghai, China). Fish oil was obtained from ARCTIC BLUE OMEGA B.V. (Utrecht, the Netherlands), while dodecane (with a purity of $\geq 99\%$) was procured from Aladdin Reagent Co., Ltd. (Shanghai, China). Chemicals, including NaOH, anhydrous ethanol, along with various other reagents, were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). All materials and reagents utilized in the present study met analytical grade standards. Deionized water was employed throughout the experimental procedures.

2.2. Preparation of Pickering emulsion

The carboxymethylated cellulose nanofibrils (CM-CN) used in this work were prepared according to our previous methods (Chen et al., 2025). Lemon essential oil, fish oil, dodecane, and CM-CN were individually combined in various proportions (1:3, 1:5, and 1:7 w/w), while maintaining the cellulose concentration in the emulsion at 0.8 wt%. The blend was homogenized for 9 min at a speed of 15,000 rpm with the aid of a high-speed homogenizer (RCD-1A, China). During homogenization, the emulsion was placed in ice water and the temperature of the samples was maintained at about 10°C.

2.3. Preparation of emulsion gels and aerogels

The newly created emulsion underwent heating in a water bath at varying temperatures of 70°C, 80°C, and 90°C for a duration of 6 h to facilitate the development of the Pickering emulsion gels. Afterward, the resulting Pickering emulsion gels was transferred to a vacuum freeze dryer (LGJ-18, China) and subjected to drying to produce aerogels. To systematically verify the gelation mechanism and structural changes, necessary control groups were established. Specifically, unheated fresh emulsions (0 h) were prepared and kept at room temperature. In addition, pure CM-CN suspensions (without the oil phase) and emulsions prepared with alternative oil phases (dodecane and fish oil) were subjected to the exact same heating procedure (80 °C for 6 h) and subsequent freeze-drying to serve as experimental controls.

2.4. Characterizations of emulsion gels and aerogels

2.4.1. Rheological tests

Rheological measurements were measured by using a previously described method with some modifications (Piao et al., 2023). The rheological characteristics of emulsion gels containing various oils and oil-to-water ratios were evaluated using an AR2000ex dynamic rheometer (TA Instrument Ltd, New Castle, Delaware, USA), which was fitted with a 40 mm parallel plate and a 1 mm gap size. An oscillatory temperature sweep was carried out from 25°C to 80°C at a strain of 0.1%, a frequency of 1 Hz, and a heating rate of 5°C/min to monitor real-time thermal gelation, whereas samples treated at 70 °C, 80 °C, and 90 °C in water baths were subsequently evaluated at 25 °C. These

measurements were strictly performed within the linear viscoelastic region (LVR) of the emulsion gels. These measurements were strictly performed within the linear viscoelastic region of the emulsion gels. The 0.1% strain parameter was carefully predetermined and selected based on established, highly conservative protocols for biopolymer-based emulsion gels (Wijarnprecha et al., 2018), which guarantees that the structural integrity of the delicate micro-network remains intact and non-disrupted during the dynamic data acquisition process. The duration of the experiment was 3600s.

2.4.2. Texture profile analysis (TPA)

The previously outlined procedure was followed to cut the emulsion gels into cylinders measuring 25 mm × 25 mm. Subsequently, TPA was evaluated using a cylindrical P/36R probe at a speed of 2 mm/s, applying a compression distance of 5 mm with a diameter of 36 mm, employing a TA.XT Plus texture analyzer from Stable Micro Systems Company, UK (Zhao et al., 2023).

2.4.3. Fourier-transform infrared spectroscopy (FTIR)

An FTIR spectrometer (IRTracer-100, Shimadzu, Japan) was used to scan the samples at 25°C with wavenumbers ranging from 500 and 4000 cm^{-1} with a resolution of 4 cm^{-1} (16 scans were obtained) based on the method reported by the literature (Feng et al., 2024).

2.4.4. X-ray diffraction (XRD)

The aerogel's crystal structure was investigated using an XRD diffractometer (Ultima IV, Rigaku, Japan). The diffraction patterns were observed at 40 mA and 40 kV, with a scanning range of 5° to 90°. The crystallinity index was calculated employing the traditional Segal method (French & Santiago Cintrón, 2013).

2.4.5. Cryo-scanning electron microscopy (Cryo-SEM)

The microstructure of newly prepared Pickering emulsion and emulsion gels (heating 80°C for 0.5 h and 6 h respectively) were observed using cryo-scanning electron microscopy (Helis G4 UC, USA) with a 5 kV acceleration voltage. Firstly, the samples were frozen, followed by 20-min sublimation at -90°C. Subsequently, the samples were sputter-coated with a gold layer, which was less than 20 nm thick (Yang et al., 2025a).

2.4.6. Low-field nuclear magnetic resonance (LF-NMR)

A gel emulsion created from 1g was performed and wrapped in polyethylene film to avoid moisture evaporation. Subsequently, it was positioned in a LF-NMR glass tube (30mm in diameter) and placed into a LF-NMR analyzer (Niumag Co., Shanghai, China). The radio frequency pulse sequence utilized was the Carre-Purcell-Meiboom-Gill (CPMG). The curve for T₂ relaxation time was generated using multi-exponential inversion software (Yang et al., 2024).

2.4.7. Brunauer-Emmett-Teller (BET) surface area analysis

The nitrogen adsorption (BET) method was utilized to access the pore profiles of aerogels using a Micromeritics ASAP 2460 analyzer (Micromeritics-3Flex, USA). Initially, the samples underwent degassing at 110 °C for a duration of 3h before being positioned in the measuring cell for analysis. Following this, it was possible to determine the BET surface area, total pore volume, and average pore diameter (Piao et al., 2022).

2.4.8. Compressive properties testing

The compressive characteristics of the aerogels were assessed utilizing an electronic universal testing machine (UTM-4103, China). For the compression tests, cylindrical samples with a height of 25 mm and a diameter of 20 mm were employed, conducting the experiments at a compression speed of 5 mm/min and at a temperature of 25 °C until a strain of 98% was achieved (Xiao, Tang, Huang, Kang, & Zhou, 2023).

2.4.9. Scanning electron microscope (SEM)

The aerogel's microstructure, which developed following freeze-drying, was analyzed through scanning electron microscopy (SEM) employing a Sigma-500 instrument. To enhance SEM imaging quality, the samples were coated with a 5 nm layer of gold utilizing a Cressington 208HR high-resolution sputter coater.

2.5. Statistical analysis

The statistical evaluation was performed utilizing IBM SPSS software, specifically version 22.0. In order to determine notable differences among the samples ($p < 0.05$), an analysis of variance (ANOVA) combined with Duncan's multiple range test was employed.

3. Results and discussion

3.1. Characterization of the emulsion gels (EG)

3.1.1. Morphological observation and rheological characterization of emulsion gels

In this study, three nanocelluloses that proved to possess good emulsifying properties were selected to prepare Pickering emulsion gels, namely TCN40, TCN80 and CM-CN (Zhao et al., 2024). Cellulose was obtained by incrementally introducing varying amounts of NaClO solutions (40 mL for TCN40 and 80 mL for TCN80) to start the TEMPO-mediated oxidation reaction. On the other hand, CM-CN was created via the mechanical fibrillation of raw cellulose materials after undergoing carboxymethylation, a process that included both alkalization and etherification. **Fig. S1** illustrates the visual appearance of the emulsion gels made from the three celluloses, which were subjected to heating for durations ranging from 0 to 6 hours. A notable decrease in deformability and flowability of the emulsion was observed, with the state of the emulsion transitioning from a cream-like consistency to a self-supporting gel upon heating. Notably, the emulsion gels stabilized by CM-CN distinguished itself by forming a self-supporting gel after just one hour of heating. The remarkable characteristics of CM-CN can explain this, as it possesses long, flexible, and twisted fibrils that often cluster and intertwine, which results in the creation of a densely interwoven fibrillar structure that efficiently stabilizes the emulsion (Wei et al., 2019). Therefore, CM-CN was selected for emulsion gels stabilization in this study.

Oil plays a crucial role in the composition of emulsions, potentially impacting their durability, as well as the flavor and texture of emulsion-based products (Taha et al., 2018). This study aimed to examine how various oil types influence the characteristics of emulsion gels. Three distinct oil phases were chosen for the preparation of these gels: lemon essential oil (LEO), fish oil, and dodecane (**Fig. S2**). Notably, both LEO and dodecane are volatile and exhibit strong emulsifying properties (Perdones, Escriche, Chiralt, & Vargas, 2016). However, in our prepared system, the dense CM-CN interfacial layer firmly wrapping the oil droplets serves as a robust physical barrier during the mild 80 °C thermal treatment. This specific Pickering stabilization effectively encapsulates the essential oil, significantly mitigating the rapid volatilization and severe thermal oxidation of its primary active components, such as citral. Moreover, preparations containing fish oil are abundant in omega-3 unsaturated fatty acids, which are vital compounds for human well-being. A recent study conducted in the United States revealed that among American citizens, fish oil is the most frequently taken specialty supplement (Huang et al., 2023). In contemporary times, these materials are also suitable for the creation of emulsions, particularly considering that the study of emulsions containing fish oil has become a prominent area of research within food science (Xu et al., 2021). We selected the aforementioned oil for the preparation of emulsion gels. As illustrated in **Fig. S2**, all three newly prepared Pickering emulsions exhibited good fluidity. However, only the emulsion containing LEO was able to form a stable and more elastic oleogel after 6 hours of heating. This phenomenon may be attributed to covalent cross-linking polymerization between LEO

and CM-CN. The formation of emulsion gels was confirmed through texture profile analysis (TPA). As presented in **Table 1**, the emulsion gels using LEO as the oil phase demonstrated the highest values across all TPA testing indices (hardness, springiness, cohesiveness, chewiness, gumminess, and resilience), followed by dodecane, while fish oil exhibited the lowest values. This suggests that LEO-based emulsion gels facilitated greater interaction among CM-CN molecular chains, resulting in increased structural rigidity after gelation, which corresponds to the highest levels of hardness and consistency.

Table 1. TPA parameters of Pickering emulsion gels with different factors (oil phase, oil/water ratio, temperature).

	Hardness (g)	Springiness	Cohesiveness (g·s)	Chewiness (g)	Gumminess (g·cm)	Resilience
LEO	105.85±18.06 ^a	0.78±0.01 ^a	0.58±0.09 ^a	48.72±14.87 ^{ab}	62.14±18.61 ^{ab}	2.99±0.14 ^a
Fish oil	50.78±5.23 ^c	0.26±0.02 ^c	0.46±0.03 ^a	4.31±1.48 ^c	16.26±4.39 ^{cd}	1.24±0.33 ^c
Dodecane	35.47±4.07 ^c	0.43±0.12 ^a	0.57±0.19 ^a	8.4±3.25 ^c	20.74±8.42 ^d	2.04±0.24 ^{bc}
O/W=1/3	79.99±7.36 ^b	0.66±0.03 ^b	0.56±0.06 ^a	29.55±4.14 ^b	44.58±6.43 ^{bc}	2.71±0.35 ^{abc}

O/W=1	99.19±10.0	0.75±0.0	0.63±0.06	47.31±12.	62.37±11.	3.3±0.35 ^a
/5	9 ^a	7 ^a	a	01 ^{ab}	81 ^{ab}	
O/W=1	68.86±1.45	0.75±0.1	0.62±0.07	32.57±10.	42.65±4.8	3.01±0.5
/7	b	9 ^a	a	05 ^b	5 ^{bc}	6 ^a
70°C	75.19±4.49	0.74±0.0	0.64±0.10	35.41±6.9	47.74±5.9	3.37±0.3
	b	9 ^a	a	2 ^{ab}	4 ^{bc}	9 ^a
80°C	107.66±5.0	0.79±0.1	0.63±0.11	54.99±16.	68.46±13.	2.83±0.6
	9 ^a	1 ^a	a	03 ^a	84 ^a	6 ^{ab}
90°C	105.14±10.	0.71±0.0	0.6±0.06 ^a	44.52±9.9	62.58±9.1	2.63±0.4
	57 ^a	6 ^a		2 ^{ab}	5 ^{ab}	3 ^{abc}

*Different superscripts in the same column represent significant differences at $p < 0.05$.

To further investigate the cross-linking and mutual interactions during the thermal gelation of Pickering emulsions, dynamic viscoelastic tests were conducted at a constant strain of 0.1% within the linear viscoelastic region. **Fig. 1a, b, and c** illustrate the storage modulus (G') and loss modulus (G'') of emulsion gels during heating from 25°C to 80°C. It is observed that the G' modulus of the fish oil-based emulsion gels exhibits a slight decrease at the initial stage of gelation. According to previous studies, excessive temperature treatment can lead to the disruption of hydrogen bonds, ionic bonds, and electrostatic interactions, thereby weakening intermolecular interactions (Yao et al., 2024). Research indicates that hydrophobic interactions are effectively preserved at 60°C but are disrupted beyond 80°C, further validating the breakdown of the gel network structure (Zheng, Zhou, Zhang, Lv, & Zhao, 2019). The slight decrease

in the G' value of the fish oil-based emulsion gels can be attributed to the disruption of the gel network structure formed by the emulsion itself due to high-temperature treatment. This disruption reduces the number of hydrogen bonds and weakens electrostatic interactions. Generally, the G' moduli of the three emulsions exhibited an upward trend during the heating process, confirming an increase in the degree of cross-linking and a stronger three-dimensional gel network structure. Although the G' and G'' moduli of the fish oil-based emulsion gels were higher than those of LEO and dodecane at certain stages due to droplet packing and physical chain entanglements, the gel network remained relatively soft and fluid-like (Fig. S2). This occurs because high viscoelastic resistance in unlinked concentrated emulsions reflects localized droplet friction rather than a permanent volume-spanning network; thus, indicating less stable aggregation and insufficient re-bonding of the cellulose chains, which made it challenging to define its true macroscopic gel state without covalent reinforcement. In comparison, LEO-based emulsion gels demonstrated a continuous increase in gel strength during heating. Unlike typical physical gels dominated purely by reversible hydrogen bonds, which tend to melt and exhibit a decrease in storage modulus (G') at elevated temperatures, the thermo-irreversible increase in G' observed here strongly suggests the formation of a permanent covalent network. This indicates that the esterification provides an irreversible structural backbone that dominates the gel's high-temperature stability.

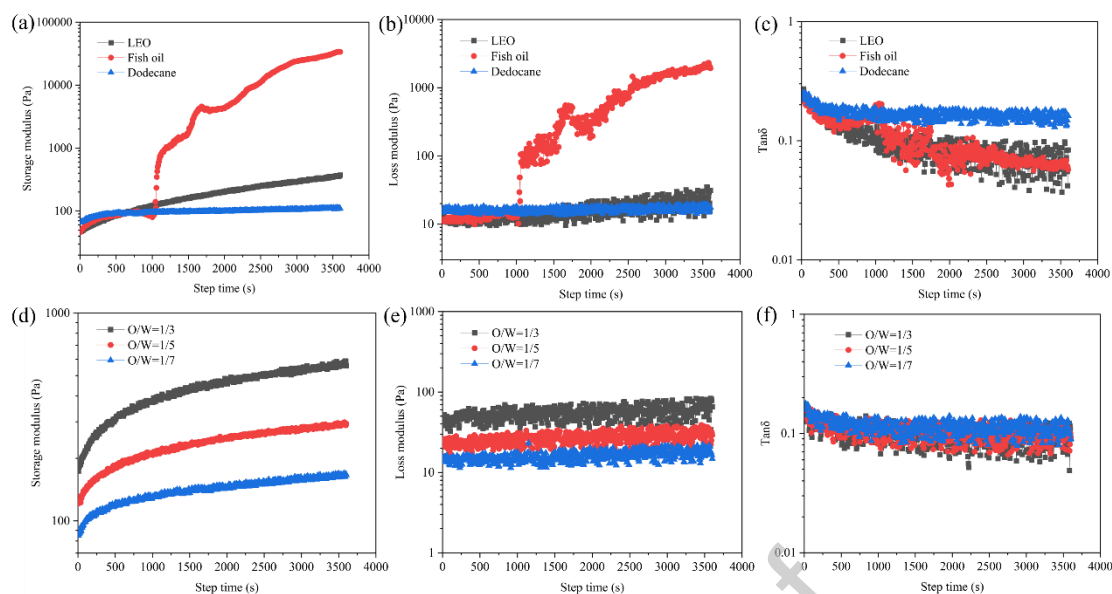


Fig. 1. Time sweep test for emulsion gels in various oil phases (LEO, fish oil, dodecane) and oil/water ratios ($O/W=1/3$, $1/5$, $1/7$) during the entire gelation process.

To gain insight into the formation of emulsion gels, we studied various oil-water ratios ($O/W = 1/3$, $1/5$, $1/7$) at different heating temperatures (70°C , 80°C , 90°C) (**Fig. 2** and **Fig. 3**). The emulsion gels with an O/W ratio of $1/3$ exhibited a slight yellow hue and numerous small bubbles on its surface, whereas the gels with O/W ratios of $1/5$ and $1/7$ appeared smooth and white. Furthermore, an increase in the LEO content of the gel (oil-water ratio of $1/3$) resulted in higher G' and G'' values at the same temperature, indicating a higher bulk viscoelastic resistance due to increased dispersed phase packing and a relatively stronger network structure within the system. Higher water/oil ratios contributed to the increased G' and G'' values and facilitated the formation of a rigid network. These results demonstrate that LEO plays an important role as a multifunctional cross-linker, bridging CM-CN. Notably, even at the lowest oil-water

ratio, the G' modulus exceeded the G'' modulus, and $\tan \delta$ was less than 1, which indicates elastic solid-like behavior and confirms the occurrence of gel formation (Wijarnprecha et al., 2018). This sol-gel transition can be rigorously explained by the Percolation Theory. In this framework, the CM-CN fibrils and LEO droplets act as interactive building blocks that interconnect. Once the concentration of these nodes surpasses a critical percolation threshold, they form an infinite, volume-spanning hybrid polymeric cluster, leading to a macroscopic solid-like state. Given that all investigated O/W ratios at the constant 0.8 wt% CM-CN concentration rapidly developed a self-supporting structure with dominant elastic properties ($G' > G''$), it is evident that the spatial density of the dispersed phase and the cross-linked fibrils in our system is well above the percolation threshold required to form a continuous three-dimensional network. However, while higher bulk moduli were observed at O/W = 1/3 due to enhanced droplet packing, the textural parameters reached their maximum values at an optimal O/W ratio of 1/5. This occurs because an excessively high oil fraction (O/W = 1/3) introduces dense droplet crowding that slightly disrupts the spatial continuity of the continuous nanofibrillar network, leading to minor structural imperfections, whereas a reduction to 1/7 diminishes active cross-linking bridges. Thus, an O/W ratio of 1/5 balances droplet reinforcement and matrix continuity. In general, an increase in the LEO/water ratio resulted in higher values of G' and G'' . This behavior can be explained by a rise in the density of cross-linking within the nanofibril network structure. The key factors affecting the textural characteristics of nanofibril gels were

found to be hardness and elasticity, which strongly relate to the interactions between the gel matrix and the oil droplets contained within.



Fig. 2. Visual appearances of emulsion gels prepared by different oil/water ratios ($O/W=1/3$, $1/5$, $1/7$) under heating conditions of 0-6 h, respectively.

Table 1 indicates that, as anticipated, a rise in the LEO/water ratio resulted in a notable enhancement in hardness, chewiness, and gumminess values, implying a stronger gel structure within the continuous network. Earlier studies indicate that the relationship between emulsion droplets and the gel matrix in polysaccharide-based emulsion gels is often weak, regardless of the presence of synthetic emulsifiers. Enhancing the polysaccharide content can strengthen the gel, primarily due to a reduction in void spaces, which in turn increases the overall strength of the gel matrix (Li et al., 2025). However, the aforementioned indices of emulsion gels reached their maximum value at an LEO-water ratio of 1:5, subsequently decreasing when the oil-water ratio was

reduced to 1:7, which hindered the effective maintenance of the network. This indicates that an appropriate oil-water ratio in emulsion gels can lead to stronger gel strength. This result aligns with earlier research suggesting that oleogel networks containing a high proportion of oil phase demonstrate an increased ability to deform the oil droplets, which in turn improves the gel network's resistance to collapse.

To further elucidate the differences in thermal gelation of the oleogel network, various heating temperatures (70°C, 80°C, 90°C) were investigated. As shown in **Fig. 3**, appropriate temperatures (70°C and 80°C) contributed to the formation of a more homogeneous and smoother oleogel compared to other treatment temperatures. In contrast, excessive heating at 90°C significantly promoted textural deterioration of the gels, resulting in a color change from milky white to yellow in the macroscopic appearance of the gel. This phenomenon can be explained by the formation of an active-filled oleogel network at 70°C and 80°C, which was established through the connection of CM-CN and LEO droplets. Increasing the temperature to 90°C initially enhanced gel strength related to the CM-CN filling volume. However, at this higher temperature, strength decreased due to granule disintegration, corresponding to the observed textural deterioration (**Table 1**). The results indicated that gel strength increased and then decreased with rising temperature, reaching maximum values of hardness (107.66 g), chewiness (54.99 g), and gumminess (68.46 g·cm) at 80°C. This could be due to a decrease in noncovalent cross-linking involving LEO and CM-CN, leading to a network that is looser and rougher with comparatively larger water channels (Wang et al., 2020).

Consequently, an optimal temperature of 80°C was identified for enhancing gelation. It is noteworthy that while the wet gel optimized at 80 °C exhibited peak TPA hardness and elasticity, the corresponding freeze-dried aerogel (**Table 2**) showed a modulated compressive strength compared to the 70 °C and pure cellulose controls. This discrepancy arises because the optimal thermal treatment promotes more extensive structural relaxation and flexible pore-wall rearrangement during lyophilization, yielding a resilient, compliant porous matrix rather than a rigid, brittle framework.

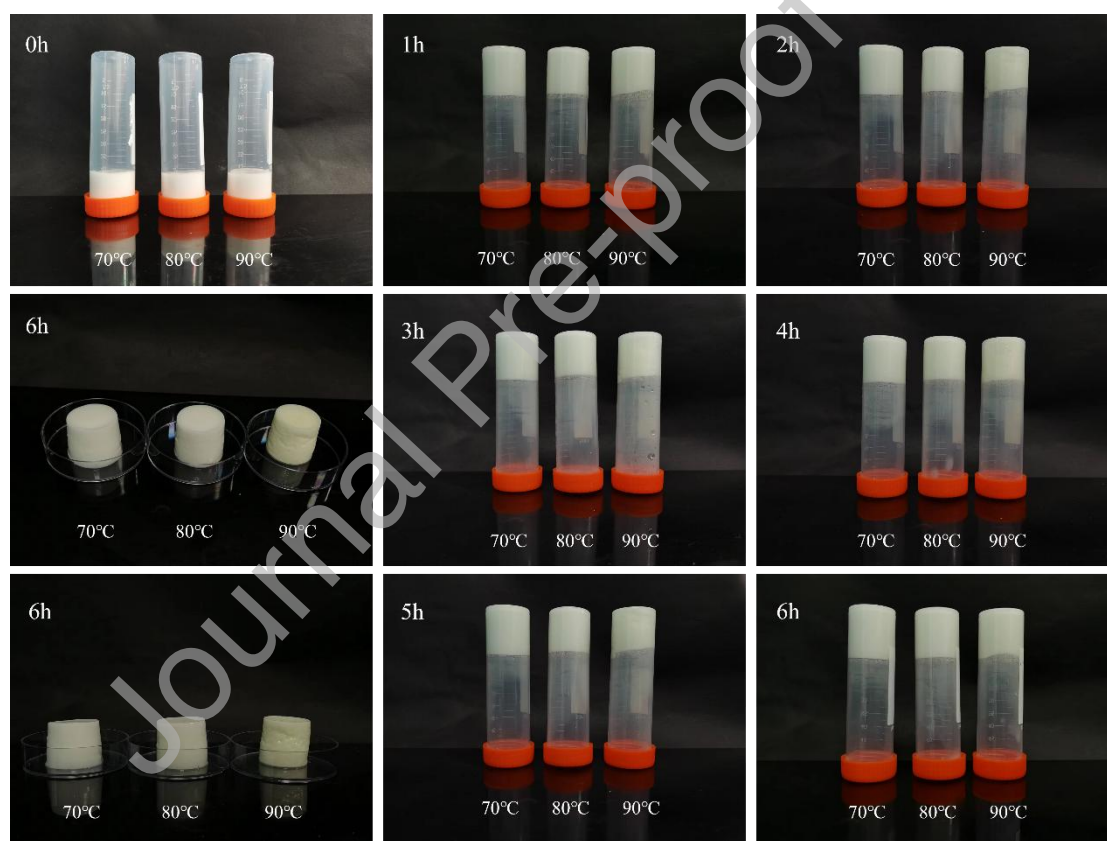


Fig. 3. Visual appearances of emulsion gels prepared at different temperatures (70°C, 80°C, 90°C) under heating conditions of 0-6 h, respectively.

3.1.2. Chemical and crystalline structure characterization of the emulsion gels

The FTIR spectra of freeze-dried emulsion gels, cellulose aerogel, and pure LEO were depicted in **Fig. 4a, b, c**. The distinctive peaks of CM-CN were identified, including 3337 cm^{-1} corresponding to O-H stretching vibration, 2900 cm^{-1} related to C-H stretching vibration, 1589 cm^{-1} associated with the -CO stretching of the ionized COO^- , and 1028 cm^{-1} which represents C-O-C asymmetric stretching (Chen, et al., 2025). LEO represented a multifaceted mixed system, wherein citral served as the principal terpenoid and limonene acted as the predominant terpene; the absorption peaks corresponding to each functional group within the compound can be identified through the infrared spectrum. According to a previous study that LEO was analyzed by GC/MS in detail, and it was found that citral as a major component, accounted for 74.58% (Bhatnagar, 2019).

Based on the FTIR results, the peak corresponding to the bending vibration of =C-H is observed at 879 cm^{-1} , while the peaks for C-O-C stretching, C=C stretching, and C-H stretching occur at 1372 , 1673 , and 2958 cm^{-1} , respectively (Jiang et al., 2021). It can be proved that citral in LEO played a crucial role in emulsion gels crosslinking. The absorption peak intensity around 3343 cm^{-1} rose as the oil content diminished, which is primarily attributed to heightened intermolecular or intramolecular hydrogen bonding within the emulsion gels (Zhang et al., 2024) (**Fig. 4b**). In the case of LEO-based emulsion gels, a distinct new absorption band at 1715 cm^{-1} emerged, which is assigned to the C=O stretching vibration of ester groups formed via cross-linking between LEO and CM-CN (Kumar, Sauraj, & Negi, 2019). It is crucial to note that this specific peak

originates from the targeted esterification between the reactive groups of CM-CN and LEO, rather than the thermal degradation of the essential oil. The gelation process was conducted under a mild heating condition of 80 °C for 6 h. Under such moderate thermal treatment, the primary active constituents of LEO are structurally preserved. Unlike random thermal degradation, which typically yields heterogeneous oxidation products with broad and overlapping spectral bands, the highly specific and sharp nature of the 1715 cm^{-1} peak perfectly aligns with the uniform formation of ester carbonyl linkages. Notably, this characteristic ester peak is well-resolved from the ionized carboxylate peak at 1589 cm^{-1} and the C=C stretching peak at 1673 cm^{-1} , avoiding potential spectral overlap and ambiguity. Furthermore, the significant alteration in the peak shape at 1673 cm^{-1} , corresponding to C=C stretching, further confirms the active participation of LEO functional groups in the crosslinking reaction. Therefore, the interaction between LEO and CM-CN under thermal treatment was essentially mediated by the C=O stretching and hydrogen bond, which is conducive to crosslinking to form emulsion gels.

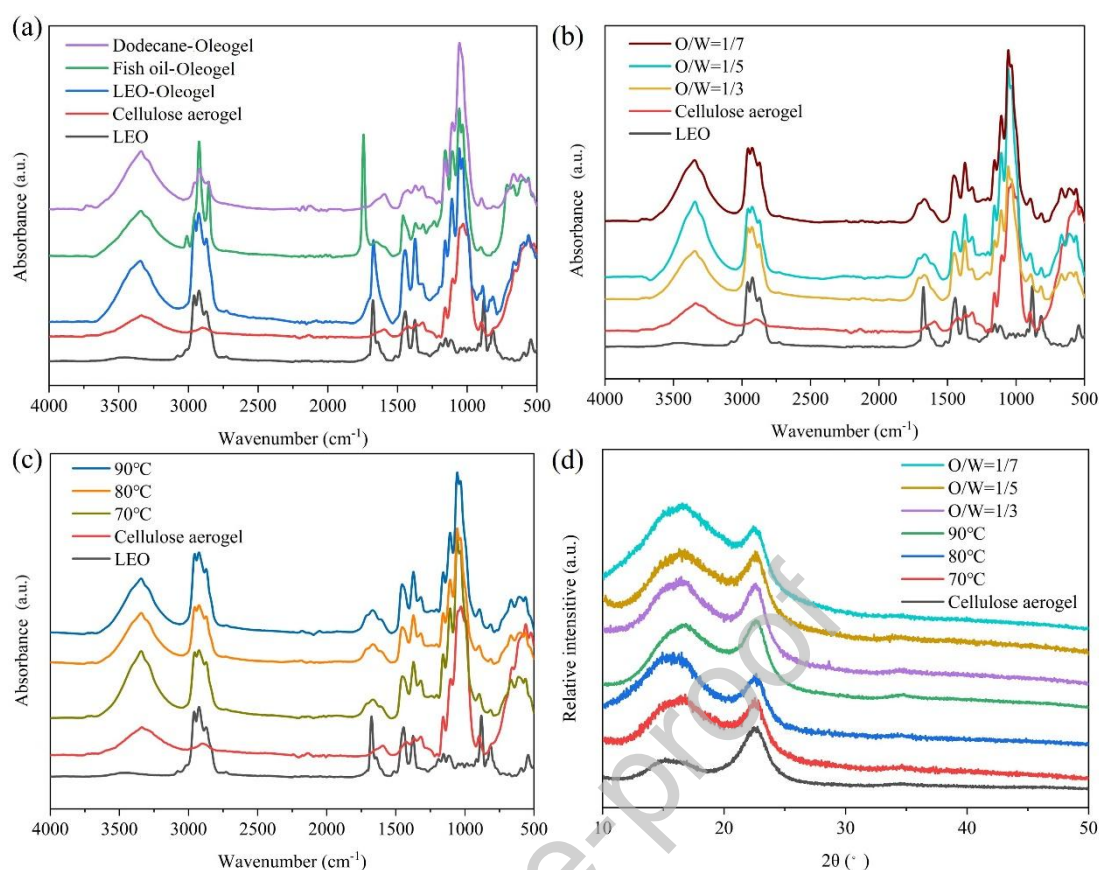


Fig. 4. FTIR spectra of different oil phases (a), oil/water ratio (b), heating temperatures (c) and X-ray diffraction patterns (d) of aerogels and emulsion gels.

The XRD pattern provided information on the crystallinity and internal structure of the emulsion gels samples (**Fig. 4d**). For all samples, three characteristic peaks at $2\theta = 16^\circ$, 22.5° , and 34.5° were observed, indicating the 110, 200 and 004 planes of emulsion gels and cellulose, respectively. LEO addition, in turn, narrowed such a diffraction peak in the peak of 22.5° . It shows that the typical crystal structure has not altered after heating treatment. The crystal structure of emulsion gels has a sharp peak at $2\theta = 22.5^\circ$, which indicates its crystallization behavior (Yang et al., 2025a). CM-CN had a crystallinity index of 62%, which decreased to 13%, 23%, 16%, 11%, 7% and 35% for

emulsion gels with different temperatures (70°C、80°C、90°C) and oil-water ratio (O/W=1/3, 1/5, 1/7), respectively.

3.2. Microstructure characterization of the emulsion gels and aerogels

Cryo-SEM images (**Fig. 5a**) provided additional evidence that CM-CN aggregated and formed cross-linked three-dimensional network structures containing oil droplets. In a freshly prepared emulsion, oil droplets were interconnected by filament-like cellulose. Nonetheless, the construction of the pore wall structure in the CM-CN emulsion gels occurred due to the growth of ice crystals during the freeze-drying phase, with the filamentous CNF fibers being flattened into sheets (Li et al., 2020). Following a heating treatment, fiber-like CM-CN interlinked, and the honeycomb structure present on the droplet surfaces played a vital role in connecting disparate droplets, ultimately leading to the establishment of a network structure, thereby reinforcing the existence of heat-induced emulsion gels. As illustrated in **Fig. 5b**, the CM-CN aerogel was found to be porous and featured a honeycomb network structure; however, some variations were noted between the freshly prepared emulsion and the emulsion gels.

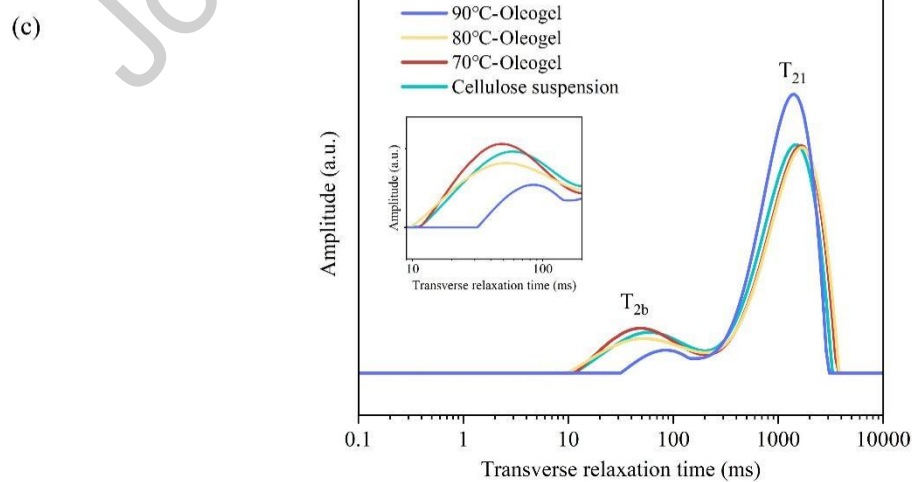
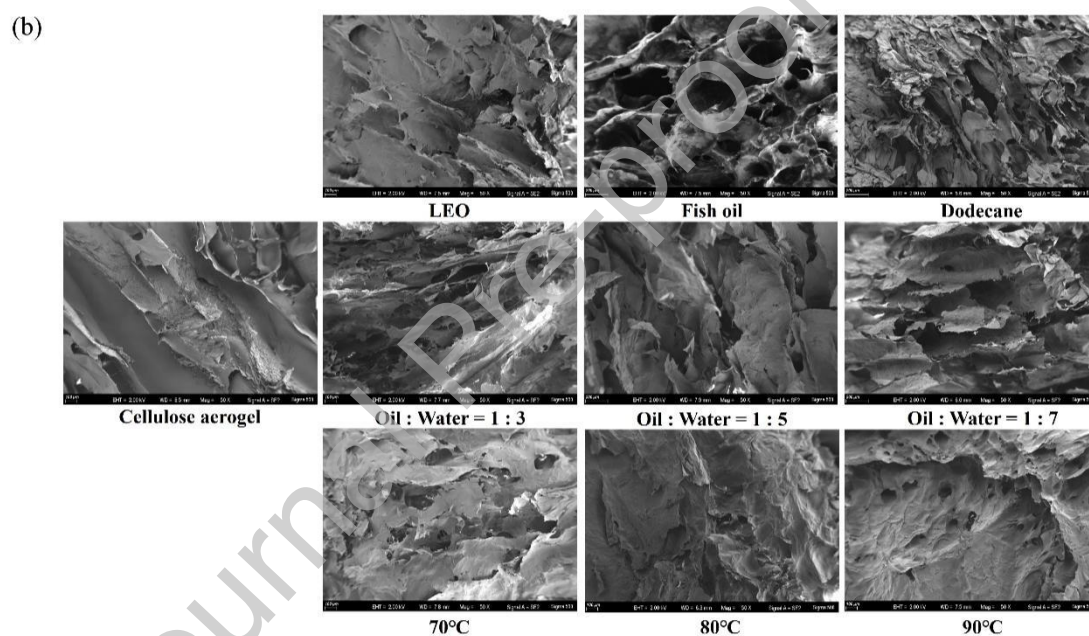
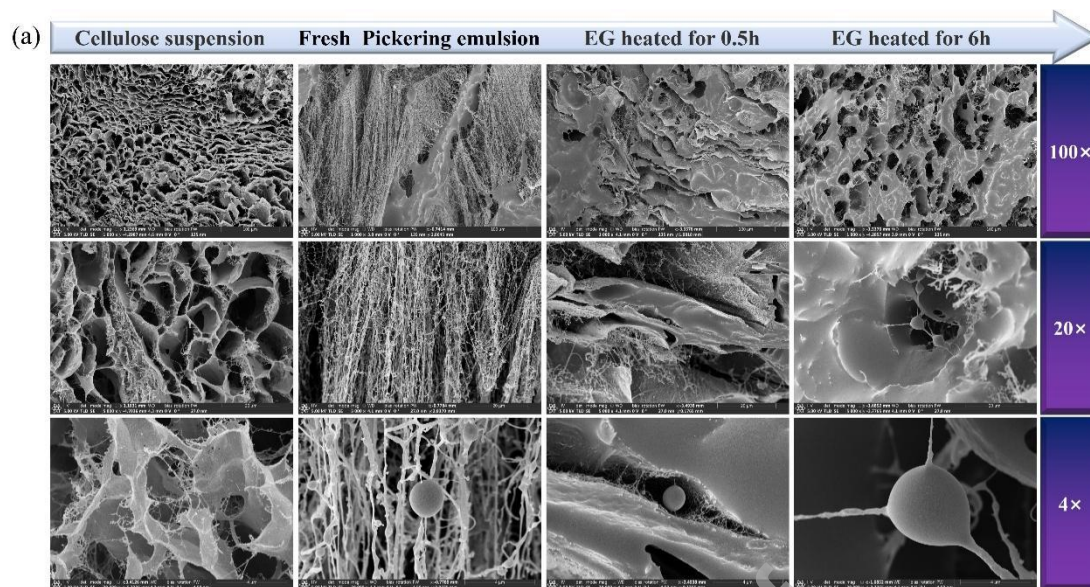


Fig. 5. (a) Microstructures of cellulose suspension newly-prepared emulsion and emulsion gels heated for 0.5 h and 6h as observed by cryo-SEM. (b) Microstructures of aerogels with different oil phases (LEO, fish oil, dodecane), different oil/water ratios ($O/W=1/3, 1/5, 1/7$), and different temperatures ($70^{\circ}\text{C}, 80^{\circ}\text{C}, 90^{\circ}\text{C}$) as observed by SEM. (c) LF-NMR spectra of cellulose suspension and emulsion gels under different heating temperatures ($70^{\circ}\text{C}, 80^{\circ}\text{C}, 90^{\circ}\text{C}$).

3.3. LF-NMR of the emulsion gels

Considering the essential role of water in the formation of gels, its movement and distribution acted as indirect indicators of important gel characteristics (Nie, Du, Xu, & Wu, 2022). Low-field NMR analysis illustrated the distribution of water molecules within the emulsion gels and cellulose suspension (**Fig. 5c**). All samples examined in this research exhibited two distinctive peaks corresponding to bound water (T_{2b} , 10-200 ms) and immobilized water (T_{2i} , 200-2000 ms). In contrast to cellulose suspension, varying heating temperatures had a significant impact on the relaxation time (T_2) of the emulsion gels. The T_2 value is indicative of the quantity of free water contained in the gel, with a lower T_2 suggesting a reduced free water content and a slower migration rate (Liu et al., 2021). When comparing the cellulose suspension, the T_2 of the emulsion gels showed a notable decrease with the rise in heating temperature. This indicated that the thermal process might enhance the interaction between the emulsion gels and water, leading to a reduction in the free water content. A prior study (Zhang et al., 2023) also

reported that Carboxymethylated CNFs could facilitate the transformation of free water into bound water.

3.4. Porous structure and mechanical property analysis

The specifics regarding the BET surface area (SSA) and the mean pore diameter for cellulose aerogel alongside various emulsion aerogels can be found in **Table 2**. Notably, the emulsion aerogel exhibited a significantly smaller mean pore diameter compared to the cellulose aerogel (11.02 nm), aligning with findings from a prior study (Mubarak, Kim, Elsayed, & Hassan, 2023). The smaller mean pore diameter of the CM-CN-LEO aerogels, which arises from the Pickering emulsion template, may contribute to the hierarchical pore arrangement, as illustrated by the Cryo-SEM and SEM images in **Fig. 5a** and **5b**. Furthermore, it is highly plausible that the N₂ adsorption measurements did not account for larger pores. Altogether, the significant variances in SSA and mean pore diameter indicate that the microporous structure of the CM-CN-LEO aerogel is markedly distinct from that of the cellulose aerogel. Theoretically, this dense, hierarchical pore architecture is directly responsible for the satisfactory liquid-holding capacity of the emulsion gels. According to the Young-Laplace equation, the capillary pressure that retains the liquid within the porous matrix is inversely proportional to the pore radius. The remarkably small average pore diameters of the CM-CN-LEO aerogels theoretically generate immense internal capillary forces. These strong capillary forces firmly lock the trapped water and liquid oil within the densely interwoven CM-CN fibrillar network, effectively preventing liquid migration or phase separation, thereby

providing a robust physical basis for the superior macroscopic stability and retention performance observed.

Table 2. Porosity and mechanical properties of cellulose aerogel and emulsion gels.

	Specific surface area (m ² /g)	Cumulative volume of pores (cm ³ /g)	Average pore diameter (nm)	98% compressive strength /MPa	98% compressive modulus /MPa
Cellulose aerogel	0.7856	0.0022	11.0243	0.2300	1.2239
70°C	0.7830	0.0016	8.1588	0.1768	0.7438
80°C	0.7401	0.0015	7.8440	0.1121	0.5150
90°C	0.8031	0.0014	7.1589	0.0627	0.2388
O/W=1/3	0.8580	0.0015	7.1269	0.0853	0.4195
O/W=1/5	0.6074	0.0009	6.1568	0.1057	0.6470
O/W=1/7	0.7463	0.0015	8.0769	0.2916	1.5542

3.5. Proposed mechanism of CM-CN-LEO emulsion gels formation

This study investigates the stabilization mechanism of Pickering emulsion gels stabilized by CM-CN and LEO. A schematic model illustrating the formation of CM-CN-LEO emulsion gels is proposed based on the analyses presented (**Fig. 6**). The CM-CN-LEO emulsion gels were cross-linked through a facile thermal reaction at 80 °C for 6 hours, followed by freeze-drying to create an aerogel. Thermal-induced covalent

cross-linking, particularly via catalyst-free esterification, serves as an effective approach for producing stable hybrid networks. Initially, during the emulsification stage, CM-CN, being polysaccharides with negative charges and an abundance of hydroxyl groups, can influence the electrostatic interactions with LEO, effectively stabilizing the oil-water interface.

Subsequently, upon thermal treatment, physical crosslinking initiates the preliminary gelation process. CM-CN enhances the stiffness of the network through intermolecular interactions and hydrogen bonding. The hydroxyl groups on the glucopyranosyl ring of CM-CN are critical for facilitating both intermolecular and intramolecular hydrogen bonds, thereby enhancing the overall structural stability and properties of the emulsion gels (Selvaraj et al., 2024). Moreover, the hydroxyl groups found in CM-CN are capable of creating hydrogen bonds with water molecules. This interaction restricts the mobility of free water, thus improving the water retention ability of the emulsion gels. In addition, CM-CN acts as effective fillers that compact the network structure, aiding in the development of a denser and more uniform three-dimensional gel network. Throughout the process of heat-induced gelation, physical crosslinking reveals specific hydrophobic groups that further promote the development of the emulsion gels.

As the thermal reaction progresses, chemical crosslinking occurs to further lock the matrix, and CM-CN significantly contributes to the formation of a hybrid polymeric network. Spectroscopic characterizations (**Fig. 4**) suggest that the crosslinking

mechanism of the Pickering emulsion gels is predominantly linked to the chemical reaction involving ester linkages between CM-CN and citral, a key component of LEO. From a theoretical chemistry perspective, while the esterification between carboxyl and aldehyde groups typically requires specific catalysts, the unique microenvironment of the Pickering emulsion interface facilitates this reaction under catalyst-free conditions. The dense accumulation of CM-CN at the oil-water interface acts as a spatial microreactor, significantly increasing the local concentration of both carboxyl groups from CM-CN and aldehyde groups from citral. This interfacial crowding, synergistically coupled with the extensive hydrogen-bonding network of the cellulose backbone, pre-organizes the reacting groups into close spatial proximity, thereby lowering the activation entropy required for the reaction. Consequently, the prolonged thermal treatment (80°C for 6 hours) provides sufficient activation energy to drive the esterification process, which is empirically supported by the emergence of the ester bond absorption band at 1715 cm^{-1} .

Consequently, the emulsion gels structures consist of chemically and physically crosslinked networks. Theoretically, this dual-crosslinking architecture provides distinct structural advantages. The esterification provides a 'permanent', irreversible backbone that maintains the overall geometric integrity and prevents structural collapse, whereas the hydrogen bonds serve as dynamic, reversible interactions that can potentially dissipate energy under mechanical stress. Together, this permanent covalent backbone and the reversible intramolecular and intermolecular hydrogen bonding

contribute to their relatively orderly structure, effectively trapping liquid oil within. Building upon this secure entrapment, the highly compacted network inherently endows the emulsion gels with a robust oil-binding capacity by severely restricting the mobility of the internal oil phase. Furthermore, the dense CM-CN interfacial layer firmly wrapping the oil droplets acts as a formidable physical barrier against oxygen permeation. These distinctive structural and rheological attributes provide a compelling theoretical basis for their anticipated lipid oxidation stability and long-term oil retention. Furthermore, because this emulsion-templated network effectively immobilizes both the internal oil droplets and the substantial continuous aqueous phase through hierarchical capillary forces and hydrogen bonding, it functions exceptionally well as a structured fat replacer for high-moisture or emulsified food matrices rather than anhydrous shortenings.

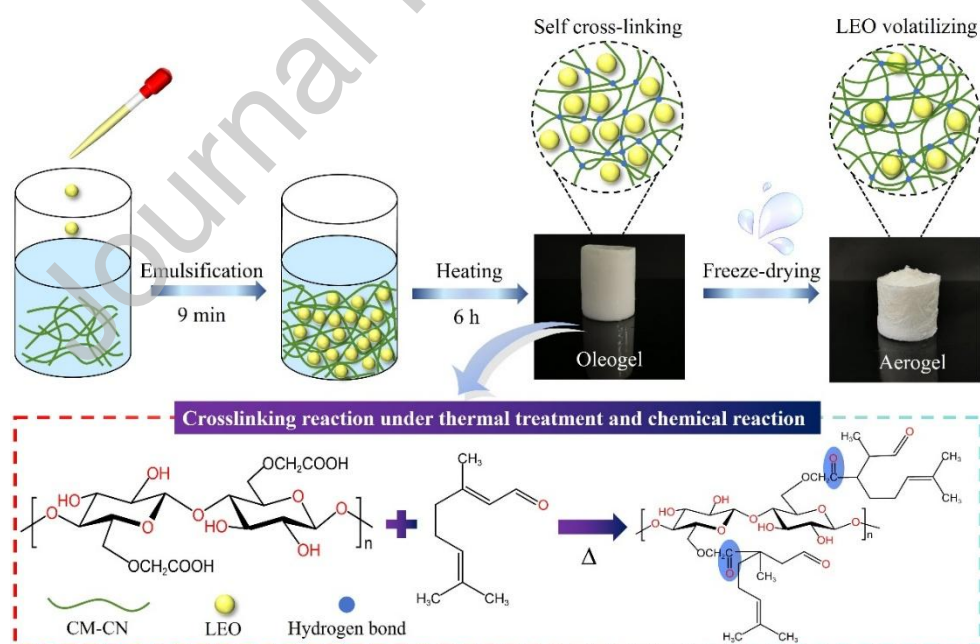


Fig. 6. Schematic representations for the preparation of CM-CN-LEO-based emulsion gels and possible mechanism of stabilization of emulsion gels.

4. Conclusions

This research successfully produced carboxymethylated cellulose-lemon essential oil-based emulsion-templated Pickering emulsion gels and aerogels without the use of extra cross-linking agents or catalysts, employing a facile, thermally activated cross-linking strategy. The structure of gel formed by emulsion is a change in the formation of chemical and physical cross-linked networks. The investigation focused on the oil-to-water ratio alongside the heating temperature of CM-CN/LEO emulsion gels and aerogels. The findings revealed that CM-CN enhances the gels both chemically, through covalent crosslinks formed between their carboxymethyl groups and citral present in LEO, and physically, due to their capability for hydrogen bonding that interacts with adjacent CM-CN molecules. These characteristics are primarily linked to the dense and organized gel structure produced by the reinforced CM-CN-LEO nanofibrillar networks, which arise from numerous hydrogen-bonding interactions within both the continuous phase and the droplets. This research paves the way for creating structured soft materials that hold significant promise as fat replacers in high-moisture or emulsified food systems.

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.

CRediT authorship contribution statement

Pingping Gao: Writing - original draft, Investigation, Methodology, Validation, Writing - review & editing. **Xiaoyun Chen:** Writing - original draft, Formal analysis, Investigation, Methodology, Data curation. **Ruizhi Yang:** Writing - original draft, Formal analysis, Investigation, Methodology, Visualization, Data curation, Writing - review & editing. **Fang Tian:** Supervision, Project administration, Writing - review & editing. **Qiaoling Zhao:** Supervision, Project administration, Writing - review & editing. **Isabel Lim Fong:** Conceptualization, Supervision, Writing - review & editing. **Soottawat Benjakul:** Formal analysis, Investigation, Writing - review & editing. **Yadong Zhao:** Conceptualization, Supervision, Project administration, Funding acquisition, Writing - review & editing.

Acknowledgements

We are grateful for the financial support from the Zhejiang Provincial Natural Science Foundation of China (LR23C160001) and Key Fishing and Agricultural Science and Technology Project in Dongtou District, Wenzhou (N2025Y11).

References

- Abitbol, T., Rivkin, A., Cao, Y., Nevo, Y., Abraham, E., Ben-Shalom, T., Lapidot, S., & Shoseyov, O. (2016). Nanocellulose, a tiny fiber with huge applications. *Current Opinion in Biotechnology*, 39, 76-88.
- Bhatnagar, A. (2019). Altitudinal variation in the volatile constituents of *Cymbopogon flexuosus* from Uttarakhand region of Himalaya. *Journal of Applied and Natural Science*, 11(2), 263-266.
- Cebi, N., Taylan, O., Abusurrah, M., & Sagdic, O. (2021). Detection of Orange Essential Oil, Isopropyl Myristate, and Benzyl Alcohol in Lemon Essential Oil by FTIR Spectroscopy Combined with Chemometrics. *Foods*, 10(1), 27.
- Chen, X., Zhang, F., Yang, Q., Zhou, R., Xu, Y., Gao, P., & Zhao, Y. (2025). Carboxylated celluloses as effective stabilizers for super-stable Pickering emulsions: Effects of different carboxyl moieties and particle morphologies on performance. *Food Hydrocolloids*, 160, 110735.
- Farjami, T., & Madadlou, A. (2019). An overview on preparation of emulsion-filled gels and emulsion particulate gels. *Trends in Food Science & Technology*, 86, 85-94.
- Feng, X., Wu, X., Gao, T., Geng, M., Teng, F., & Li, Y. (2024). Revealing the interaction mechanism and emulsion properties of carboxymethyl cellulose on soy protein isolate at different pH. *Food Hydrocolloids*, 150, 109739.
- French, A. D., & Santiago Cintrón, M. (2013). Cellulose polymorphy, crystallite size, and the Segal Crystallinity Index. *Cellulose*, 20(1), 583-588.

- Hu, X., Jiang, Q., Du, L., & Meng, Z. (2023). Edible polysaccharide-based oleogels and novel oleogels as fat analogues: A review. *Carbohydrate Polymers*, 322, 121328.
- Huang, S., Peng, J., Zi, Y., Zheng, Y., Xu, J., Gong, H., Kan, G., Wang, X., & Zhong, J. (2023). Regenerated silk fibroin for the stabilization of fish oil-loaded Pickering emulsions. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 678, 132523.
- Jiang, T., Wang, C., Liu, W., Li, Y., Luan, Y., & Liu, P. (2021). Optimization and characterization of lemon essential oil entrapped from chitosan/cellulose nanocrystals microcapsules. *Journal of Applied Polymer Science*, 138(43).
- Jorfi, M., & Foster, E. J. (2015). Recent advances in nanocellulose for biomedical applications. *Journal of Applied Polymer Science*, 132(14).
- Kim, H. S., Kim, D.-W., Park, I.-S., & Hong, H.-J. (2023). Adsorptive recovery of rare earth elements from aqueous solution by citric acid crosslinked carboxymethylated cellulose nanofibril aerogel. *Journal of Cleaner Production*, 418, 138189.
- Kumar, B., Sauraj, & Negi, Y. S. (2019). To investigate the effect of ester-linkage on the properties of polyvinyl alcohol/carboxymethyl cellulose based hydrogel. *Materials Letters*, 252, 308-312.
- Li, J., Ye, G., Wang, J., Gong, T., Wang, J., Zeng, D., Cifuentes, A., Ibañez, E., Zhao, H., Lu, W., Recent advances in pressurized hot water extraction/modification of polysaccharides: Structure, physicochemical properties, bioactivities, and

- applications, *Comprehensive Reviews in Food Science and Food Safety*, 24 (2025) e70104.
- Li, S., Zhou, C., He, Y., Liu, H., Zhou, L., Yu, C., Wei, C., & Wang, C. (2020). Novel Nanocellulose/Polymer Composite Aerogel as Solid-State Fluorescence Probe by Pickering Emulsion Route. *Macromolecular Materials and Engineering*, 305(11), 2000467.
- Liu, W., Wang, R., Li, J., Xiao, W., Rong, L., Yang, J., Wen, H., & Xie, J. (2021). Effects of different hydrocolloids on gelatinization and gels structure of chestnut starch. *Food Hydrocolloids*, 120, 106925.
- Mubarak, S. A., Kim, Y., Elsayed, I., & Hassan, E. B. (2023). Cellulose Nanofibril Stabilized Pickering Emulsion Templated Aerogel with High Oil Absorption Capacity. *ACS Omega*, 8(40), 36856-36867.
- Nazik, E. M. M. (2015). Citrus Essential Oils: Current and Prospective Uses in the Food Industry. *Recent Patents on Food, Nutrition & Agriculture*, 7(2), 115-127.
- Nie, B., Du, Y.-N., Xu, S.-Q., & Wu, H.-T. (2022). Improvement of low-acyl gellan gum on gelation and microstructural properties of protein hydrolysates from male gonad of scallop (*Patinopecten yessoensis*). *Food Chemistry*, 371, 131114.
- Pan, J., Tang, L., Dong, Q., Li, Y., & Zhang, H. (2021). Effect of oleogelation on physical properties and oxidative stability of camellia oil-based oleogels and oleogel emulsions. *Food Research International*, 140, 110057.

- Perdones, Á., Escriche, I., Chiralt, A., & Vargas, M. (2016). Effect of chitosan–lemon essential oil coatings on volatile profile of strawberries during storage. *Food Chemistry*, 197, 979-986.
- Piao, X., Huang, J., Sun, Y., Zhao, Y., Zheng, B., Zhou, Y., Yu, H., Zhou, R., & Cullen, P. J. (2023). Inulin for surimi gel fortification: Performance and molecular weight-dependent effects. *Carbohydrate Polymers*, 305, 120550.
- Piao, X., Li, J., Zhao, Y., Guo, L., Zheng, B., Zhou, R., & Ostrikov, K. (2022). Oxidized cellulose nanofibrils-based surimi gel enhancing additives: Interactions, performance and mechanisms. *Food Hydrocolloids*, 133, 107893.
- Pintado, T., Herrero, A. M., Jiménez-Colmenero, F., Pasqualin Cavalheiro, C., & Ruiz-Capillas, C. (2018). Chia and oat oleogels as new animal fat replacers and healthy bioactive sources in fresh sausage formulation. *Meat Science*, 135, 6-13.
- Selvaraj, S., Chauhan, A., Dutta, V., Verma, R., Rao, S. K., Radhakrishnan, A., & Ghotekar, S. (2024). A state-of-the-art review on plant-derived cellulose-based green hydrogels and their multifunctional role in advanced biomedical applications. *International Journal of Biological Macromolecules*, 265, 130991.
- Shao, P., Feng, J., Sun, P., Xiang, N., Lu, B., & Qiu, D. (2020). Recent advances in improving stability of food emulsion by plant polysaccharides. *Food Research International*, 137, 109376.
- Taha, A., Hu, T., Zhang, Z., Bakry, A. M., Khalifa, I., Pan, S., & Hu, H. (2018). Effect of different oils and ultrasound emulsification conditions on the

physicochemical properties of emulsions stabilized by soy protein isolate.

Ultrasonics Sonochemistry, 49, 283-293.

Wang, C., Ma, R., Huang, Z., Liu, X., Wang, T., & Chen, K. (2021). Preparation and characterization of carboxymethylcellulose based citric acid cross-linked magnetic aerogel as an efficient dye adsorbent. *International Journal of Biological Macromolecules*, 181, 1030-1038.

Wang, Y., Zhou, Y., Wang, X.-x., Ma, F., Xu, B.-c., Li, P.-j., & Chen, C.-g. (2020). Origin of high-pressure induced changes in the properties of reduced-sodium chicken myofibrillar protein gels containing CaCl_2 : Physicochemical and molecular modification perspectives. *Food Chemistry*, 319, 126535.

Wei, J., Zhou, Y., Lv, Y., Wang, J., Jia, C., Liu, J., Zhang, X., Sun, J., & Shao, Z. (2019). Carboxymethyl Cellulose Nanofibrils with a Treelike Matrix: Preparation and Behavior of Pickering Emulsions Stabilization. *ACS Sustainable Chemistry & Engineering*, 7(15), 12887-12896.

Wijarnprecha, K., Aryusuk, K., Santiwattana, P., Sonwai, S., & Rousseau, D. (2018). Structure and rheology of oleogels made from rice bran wax and rice bran oil. *Food Research International*, 112, 199-208.

Xiao, F., Tang, J., Huang, X., Kang, W., & Zhou, G. (2023). A robust, low swelling, and lipid-lubricated hydrogel for bionic articular cartilage substitute. *Journal of Colloid and Interface Science*, 629, 467-477.

- Xu, J., Zhang, T., Zhang, Y., Yang, L., Nie, Y., Tao, N., Wang, X., & Zhong, J. (2021). Silver carp scale gelatins for the stabilization of fish oil-loaded emulsions. *International Journal of Biological Macromolecules*, 186, 145-154.
- Yao, W., Zhao, Z., Zhang, J., Kong, B., Sun, F., Liu, Q., & Cao, C. (2024). Revealing the deterioration mechanism in gelling properties of pork myofibrillar protein gel induced by high-temperature treatments: Perspective on the protein aggregation and conformation. *Meat Science*, 217, 109595.
- Yang, R., Bao, L., Liu, Y., Liang, J., Zheng, B., Miao, W., Shi, X., Gao, P., Zhou, R., & Zhao, Y. (2024). Plant oil body as an effective improver for surimi-based 3D printing. *Additive Manufacturing*, 93, 104422.
- Yang, R., Cheng, Z., Zhao, Y., Song, Y., Shi, X., Yu, H., & Zhao, L. (2025a). Peanut oil body as a food-grade ink for 3D printing: Preparation, characterization and performance. *Food Research International*, 212, 116486.
- Yang, R., Deng, H., Bao, L., Tian, F., Li, J., Zhao, L., Liu, L., & Zhao, Y. (2025b). Non-covalent binding of cyanidin-3-O-glucoside onto peanut oil body for improved stability: Molecular interactions and properties. *Food Chemistry*, 484, 144384.
- Yang, R., Deng, H., Zhao, Y., Lin, H., Song, Y., Zhao, L., Miao, W., & Zheng, B. (2025c). Interface engineering of plant oil body for an innovative food ingredient: A review. *Trends in Food Science & Technology*, 159, 104954.

- Zhang, H., Jiang, Q., Li, J., Sun, Y., Zhang, R., Zhang, L., & Zhang, H. (2024). Oil-droplet anchors accelerate the gelation of regenerated silk fibroin-based oleogels. *International Journal of Biological Macromolecules*, 278, 134579.
- Zhang, K., Tian, X., Shen, R., Zhao, K., Wang, Y., Zhang, Y., & Wang, W. (2023). Delaying In vitro gastric digestion of myofibrillar protein gel using carboxymethylated cellulose nanofibrils: Forming a compact and uniform microstructure. *Food Hydrocolloids*, 140, 108661.
- Zhang, R., Belwal, T., Li, L., Lin, X., Xu, Y., & Luo, Z. (2020). Recent advances in polysaccharides stabilized emulsions for encapsulation and delivery of bioactive food ingredients: A review. *Carbohydrate Polymers*, 242, 116388.
- Zhao, W., Wei, Z., & Xue, C. (2021). Recent advances on food-grade oleogels: Fabrication, application and research trends. *Critical Reviews in Food Science and Nutrition*, 62(27), 7659-7676.
- Zhao, Y., Lu, K., Piao, X., Song, Y., Wang, L., Zhou, R., Gao, P., & Khong, H. Y. (2023). Collagens for surimi gel fortification: Type-dependent effects and the difference between type I and type II. *Food Chemistry*, 407, 135157.
- Zhao, Y., Zhang, F., Chen, M., Liu, F., Zheng, B., Miao, W., Gao, H., & Zhou, R. (2024). Cellulose nanofibrils-stabilized food-grade Pickering emulsions: Clarifying surface charge's contribution and advancing stabilization mechanism understanding. *Food Hydrocolloids*, 152, 109920.

Zheng, B., Zhou, K., Zhang, T., Lv, C., & Zhao, G. (2019). Designed Two- and Three-Dimensional Protein Nanocage Networks Driven by Hydrophobic Interactions Contributed by Amyloidogenic Motifs. *Nano Letters*, 19(6), 4023-4028.

Zhi, L., Liu, Z., Wu, C., Ma, X., Hu, H., Liu, H., Adhikari, B., Wang, Q., & Shi, A. (2023). Advances in preparation and application of food-grade oleogels. *Food Chemistry*, 424, 136399.

This article does not involve content related to human health or animal ethics.

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.