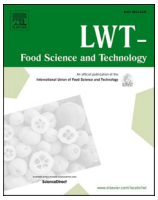
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Development and characterization of food-grade oleogel-in-hydrogel bigels as an animal fat substitute

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ABSTRACT

In response to growing consumer demand for healthier and more sustainable fat alternatives, this study developed and characterized food-grade oleogel-in-hydrogel bigels as potential substitutes for animal fat. The biphasic bigels were formulated by dispersing ethyl cellulose (EC)-based medium-chain triglyceride (MCT) oleogels into a konjac glucomannan-xanthan gum (KGM-XG) hydrogel matrix at varying hydrogel-to-oleogel ratios (9:1 to 6:4). Microstructural analysis revealed that higher hydrogel content promoted finer, more uniform oleogel droplet dispersion, enhancing overall structural stability. All bigels exhibited high liquid binding capacity (>99.8 %) and self-supporting behavior without phase separation. Fourier Transform Infrared (FT-IR) spectroscopy confirmed the coexistence of the two phases without formation of new chemical bonds. Rheological assessments, including frequency sweep and three-interval thixotropy tests, demonstrated solid-like gel characteristics with excellent recovery properties, particularly at higher hydrogel contents. Temperature sweep analysis indicated that the bigels exhibited thermo-responsive viscoelasticity comparable to porcine back fat, especially near 62.8 °C, the USDA's minimum temperature for consumable meat. Texture profile analysis further showed springiness values similar to those of animal fat. These findings underscore the potential of EC-based oleogel-in-KGM-XG hydrogel bigels as structurally stable, thermo-responsive fat replacers for processed food applications.

1. Introduction

As global dietary patterns continue to evolve, there has been a notable increase in diet-related health issues, including obesity and cardiovascular disease (Ismail et al., 2020). A major contributing factor is excessive intake of saturated fats, primarily from animal-based food products. In parallel, the conventional production of meat and animal fat raises significant environmental concerns, such as greenhouse gas emissions and resource-intensive agricultural practices (Willett et al., 2019). These growing health and sustainability challenges have fueled consumer interest and industry demand for plant-based alternatives to conventional meat and fat products (Wild et al., 2014).

In meat products, fat plays a critical role not only in delivering flavor and mouthfeel but also in providing texture and thermal stability. However, replacing animal fats remains particularly challenging due to their complex hierarchical structure and thermo-responsive behavior (Smith-Ryan et al., 2019). The viscoelastic properties of animal fat arise from a semi-solid lipid matrix and connective tissue components, such as collagen, which together impart gel-like behavior during heating and cooling (Gómez-Estaca et al., 2019). Therefore, any viable fat substitute must closely mimic these structural and rheological characteristics.

In recent years, structured gels such as oleogels, hydrogels, and emulsion gels have been extensively investigated as potential fat replacers in food systems (Badar et al., 2023; Siachou et al., 2023; Silva et al., 2023). Oleogels are semi-solid systems formed by immobilizing liquid oil within a three-dimensional gelator network (Li et al., 2022; Marangoni & Garti, 2018). Among various gelators, ethyl cellulose (EC) is the only polymer approved by the Food and Drug Administration (FDA) for food-grade oleogelation (FAO & WHO, 2012). EC-based oleogels are typically prepared by dissolving the polymer in oil at elevated temperatures (>130 °C), followed by cooling to induce gelation. Medium-chain triglyceride (MCT) oil, widely recognized for its oxidative stability and health benefits, serves as a suitable oil phase for EC-based oleogels (Lee et al., 2022). However, its liquid state at room temperature limits its direct use in solid or semi-solid food applications. This limitation can be overcome through oleogelation, where MCT oil is

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structured into a solid-like form, broadening its functional applicability in food systems.

Hydrogels (HG), in contrast, are typically formed by dispersing hydrophilic polymer networks in water (Bahram et al., 2011). Due to their water-holding capacity, biocompatibility, and biodegradability, hydrogels are widely applied in pharmaceutical, cosmetic, and food industries (Bueno et al., 2013; Doan et al., 2015; Li et al., 2021). Natural polysaccharides, including konjac glucomannan (KGM) and xanthan gum (XG), are commonly employed as hydrogelators in food systems owing to their excellent gelation ability and biodegradability (Zhu et al., 2022). However, KGM alone exhibits poor mechanical strength. Blending with XG has been shown to improve the network structure through synergistic hydrogen bonding and polymer entanglement (Dea et al., 1977).

To address the limitations of individual gel systems, biphasic bigels—comprising both oleogel and hydrogel phases—have emerged as promising fat mimetics. These systems offer several advantages, including the ability to encapsulate both hydrophilic and lipophilic compounds, tunable rheological behavior, and improved physical stability (Almeida et al., 2008). Based on their internal phase distribution, bigels can be categorized as oleogel-in-hydrogel, hydrogel-in-oleogel, or bicontinuous structures (Liu et al., 2024). Among these, oleogel-in-hydrogel systems are particularly promising for food applications due to their continuous aqueous phase, which promotes compatibility with high-moisture food matrices.

While bigel systems have been studied using a variety of gelator combinations, investigations into EC-based oleogel-in-KGM-XG hydrogel systems remain scarce. In particular, direct comparisons of their thermo-rheological properties with those of animal fats under realistic cooking conditions—such as 62.8 °C, the United States Department of Agriculture (USDA)-recommended minimum cooking temperature for meat—remain insufficiently explored.

Therefore, the present study aims to develop and characterize food-grade oleogel-in-hydrogel bigels using EC-based MCT oleogels dispersed within a KGM-XG hydrogel matrix. The microstructural, physicochemical, and thermo-responsive rheological properties of bigels with varying hydrogel-to-oleogel ratios were systematically investigated. Furthermore, the thermal behavior and texture profile of the bigels were compared with those of porcine back fat to assess their feasibility as functional fat substitutes in processed food formulations.

2. Material and methods

2.1. Material

MCT oil (Earthborn, Thailand) was purchased from a local supermarket, and ethyl cellulose (viscosity 50 cP) was obtained from Daejung Chemical & Metals Co., Ltd (Republic of Korea). The KGM was procured from Haena Food (Republic of Korea), and XG was purchased from ES Food (Republic of Korea). Porcine back fat, used as the control, was purchased from a local meat market. Nile Red, used as the fluorescent dye, was obtained from Tokyo Chemical Industry (Tokyo, Japan).

2.2. Methods

In this study, the hydrogel-to-oleogel ratio (H9O1, H8O2, H7O3, and H6O4) was set as the independent variable. The dependent variables were the measured outcomes associated with bigel characterization, including (i) microsctructure (optical and confocal microscopy), (ii) physical stability (liquid binding capacity), (iii) molecular interactions (FT-IR spectra), (iv) rheological performance (amplitude sweep, frequency sweep, thixotropic recovery, temperature sweep), and (v) textural parameters (hardness, adhesiveness, and springiness).

2.2.1. Bigel preparation

In the preliminary study, EC-based oleogels were produced at concentrations of 3, 5, 7, 9, and 11 wt%. The minimum concentration required for gelation, as determined by the inversion test (Fig. S1), was 5 wt%. Based on the oil binding capacity results of oleogels at 5, 7, 9, and 11 wt% (Fig. S2), the 11 wt% formulation was selected for subsequent experiments. For oleogel preparation, ethylcellulose (11 wt% of the oleogel phase) was dissolved in MCT oil at 140 °C and the mixture was then cooled to room temperature. To prepare the hydrogel, solutions of XG and KGM were dissolved in distilled water at room temperature. The total polysaccharide concentration in the hydrogel was fixed at 2 wt%, consisting of 1 wt% KGM and 1 wt% XG (i.e., a 1:1 ratio within the total 2 % concentration). The mixed solution was heated to 90 °C with stirring and then cooled at 4 °C to obtain a homogeneous hydrogel. The oleogel phase was added to the hydrogel phase at various oleogel-to-hydrogel ratios, ranging from 1:9 to 4:6, and homogenized at 11,000 rpm using a homogenizer (Polytron PT2100, Kinematica, Switzerland) to form bigels. After homogenization, all samples were stored at 4 °C. The samples were labeled using the convention 'H#O#,' where 'H' represents the hydrogel component and 'O' represents the oleogel component. The numbers following each letter indicate the percentage ratio of each component in the bigel formulation. For example, in the sample 'H4O6,' the hydrogel accounts for 40 % and the oleogel accounts for 60 % of the total weight of the sample.

2.2.2. Morphology

The microstructural properties of bigels were investigated using optical microscopy (ECLIPSE Ys2 inverted microscope, Nikon Corporation, Tokyo, Japan) and confocal laser scanning microscopy (LSM 880, Carl Zeiss AG, Oberkochen, Germany). Digital imaging was performed with optical microscopy fitted with a 5-megapixel cooled digital color microphotography camera (DS-5Mc, Nikon Corporation). The images obtained from optical microscopy were analyzed using Nis-Elements Br 5.0 Software (Nikon Corporation). For confocal imaging analysis, the oleogel phase was stained with a fluorescent dye solution. A 0.1 wt% Nile red solution was used to stain the oleogel phase before bigel preparation. The wavelengths used for image acquisition were 561 nm for excitation and 595 nm for emission of Nile red. All samples were observed using a 40 × objective lens, and the captured images were processed using Zen software.

2.2.3. Bigel stability

Samples of 10 g were placed in 15 mL centrifuge tubes and tested after cooling to 4 °C and conditioning at room temperature for 1 h. The samples were centrifuged at 11,000 rpm and 20 °C for 10 min using a Labogene centrifuge (1696 R; Daejeon, Republic of Korea). After centrifugation, the degree of liquid separation was determined by inverting the tubes onto Whatman^{tm} filter paper to separate the liquid from the bigels. The liquid binding capacity (LBC) was calculated as follows:

$$\text{LBC}\ (\%) = \left(1 - \frac{Wi - Wf}{Wi}\right) \times 100 \quad (1)$$

where W_i is the weight of the tube before centrifugation, and W_f is the weight of the tube after centrifugation and liquid separation. The values were calculated three times for repeatability.

2.2.4. FT-IR spectra

FT-IR spectra were recorded using a Cary 630 FTIR Spectrometer with a diamond ATR accessory (Agilent Technologies, Santa Clara, CA, USA) under conditions (4000 to 650 cm^{-1}, 4 cm^{-1} resolution) reported previously (Saffold & Acevedo, 2022). Data were collected using MicroLab PC software.

2.2.5. Rheology analysis

The rheological properties of the samples were evaluated using a rheometer (MCR102, Anton Paar, Austria) with a 25 mm parallel plate and a gap of 2 mm. A small quantity of silicon oil was applied to cover

the boundary of the probe to prevent evaporation before measurement. After covering the boundary of the samples with silicon oil, a rest time of 10 min was included. Amplitude sweep tests (strain ranged from 0.01 % to 100 %) and frequency sweep tests (frequency ranged from 0.01 to 10 Hz) were performed at 20 °C. The frequency sweep tests were conducted at a strain of 0.1 %, which is within the linear viscoelastic region of the bigels. The recovery of the bigels was evaluated using a 3-interval thixotropic test (3ITT). This test setup included 2 min of low strain (0.01 %), 2 min of high strain (10 %), and 2 min of low strain (0.01 %) to assess bigel recovery ability. The bigel recovery was calculated as the ratio of the G' value at the end of the third interval to that at the end of the first interval (Wijarnprecha et al., 2019). Temperature sweep tests were performed at a strain of 0.1 % and a frequency of 1 Hz to measure the temperature-dependent viscoelastic properties. The test setup included a cooling process (from 80 °C to 4 °C) followed by a heating process (from 4 °C to 80 °C). Rheometry data were collected using Anton Rheo Compass V1.22 software.

2.2.6. Texture analysis

The texture properties of the bigels were measured using a texture analyzer (TA.XT2i Plus, Stable Microsystems, Godalming, UK) equipped with a 5 kg load cell. The samples were prepared as 20 × 20 × 10 mm blocks. A double-compression texture profile analysis (TPA) test was conducted with a relative deformation of 50 % of the sample's initial height at a test speed of 5 mm/s, using a 25 × 25 mm square probe (Dreher et al., 2020). The interval between compressions was set to 5 s, after which the probe was retracted to its original position at a post-test speed of 5 mm/s. Hardness, adhesiveness, and springiness were recorded from the test curve.

2.2.7. Statistical analysis

All analyses were performed in triplicate. One-way Analysis of Variance (ANOVA) followed by Duncan's test was conducted using SPSS 12.0 (SPSS Inc., Chicago, IL, USA). All graphs were created using Sigma Plot software (Systat Software, Chicago, IL, USA).

3. Results and discussion

3.1. Physicochemical analysis of bigels with varying hydrogel-to-oleogel ratios

3.1.1. Microstructural properties

The microstructural properties of bigels were analyzed using optical microscopy and confocal scanning laser microscopy (CLSM) (Fig. 1). As the hydrogel ratio increased from H6O4 to H9O1, smaller and more uniformly dispersed oleogel droplets were observed, indicating improved homogeneity and enhanced mechanical integrity (Fig. 1A). CLSM analysis, with the oleogel phase stained using Nile Red, confirmed the internal droplet morphology and dispersion pattern (Fig. 1B). These results suggest that higher hydrogel content facilitates better droplet dispersion due to increased viscosity and steric stabilization, forming a more interconnected network that enhances structural robustness. These observations align with previous findings that attribute enhanced stability to continuous-phase dominance by the hydrogel (Zheng et al., 2020; Zhu et al., 2021).

3.1.2. Physical stability

The formation of bigels depends on the self-assembly and molecular properties of each gelator. Specifically, the physical stability of bigels is closely tied to the oleogel to hydrogel ratio (Singh et al., 2014). The physical stability of the bigels was assessed through vial inversion and liquid binding capacity (LBC) measurements. All formulations demonstrated self-supporting behavior with no visible phase separation (Fig. 2). LBC values exceeded 99.8 % across all samples (Fig. 3), indicating minimal oil release. Notably, despite the absence of emulsifiers, biphasic systems remained highly stable, likely due to the synergistic

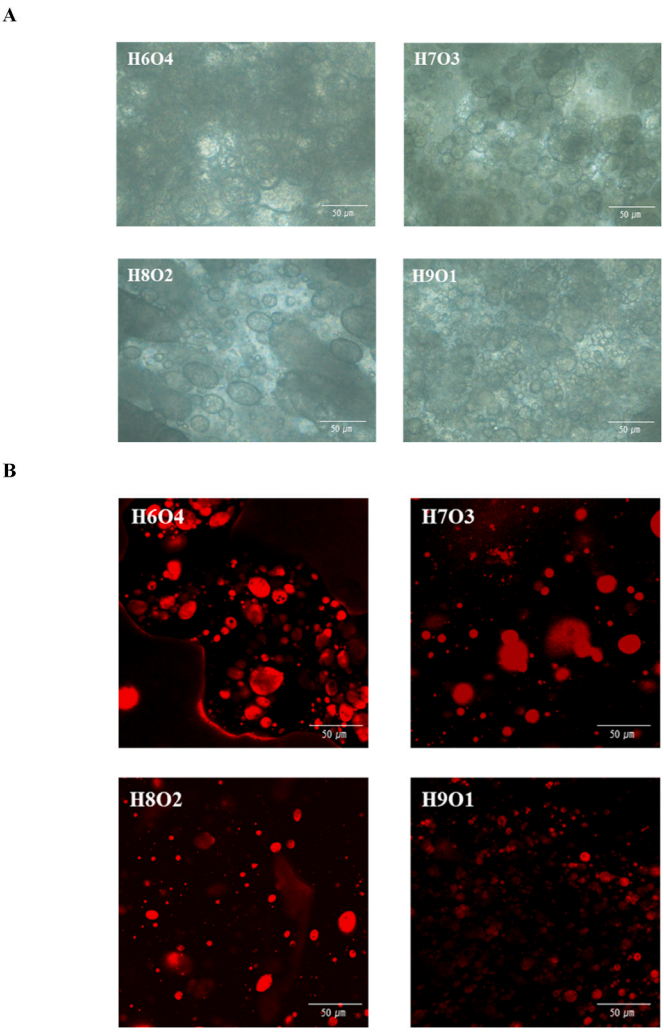


Fig. 1. Micrographs of bigels with different hydrogel-to-oleogel ratios. A. Optical microscopy; B. Confocal scanning laser microscopy.

interaction between the EC oleogel network and the KGM-XG hydrogel matrix. The ordered dispersion of oleogel droplets in the high-hydrogel formulations may contribute to capillary force entrapment and matrix entanglement, preventing oil leakage even under stress.

3.1.3. FT-IR properties

FT-IR analysis was conducted to examine the intermolecular interactions between the oleogel and hydrogel components in the bigel samples (Fasolin et al., 2021; Martins et al., 2023). FT-IR spectra confirmed the coexistence of the two components without the formation of new chemical bonds (Fig. 4). Characteristic peaks corresponding to EC-based oleogels were observed at 2927, 2858, and 1738 cm^{-1}, while the KGM-XG hydrogel exhibited broad absorption bands in the range of 3300–3200 cm^{-1}, attributed to hydrogen bonding, and a peak at 1638 cm^{-1} associated with acetyl groups (Ogbu & Ajiwe, 2016). Importantly, these peaks were retained in the bigel spectra, indicating that the two phases were physically integrated rather than chemically bonded (Alves et al., 2020). As the hydrogel content increased, the intensity of hydrogen-bond-related peaks became more pronounced, supporting the presence of a physically interpenetrated network (Saffold & Acevedo, 2021). Overall, these findings validate the formation of a true biphasic bigel system in which both the oleogel and hydrogel maintain their structural identity while coexisting in a unified matrix.

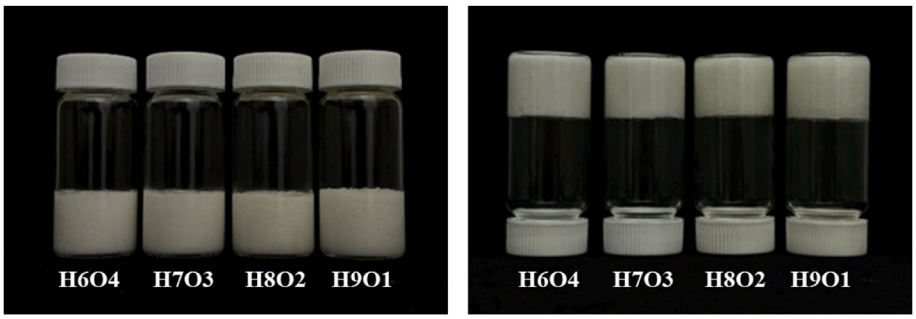


Fig. 2. Photographs of the prepared bigels with various hydrogel-to-oleogel ratios.

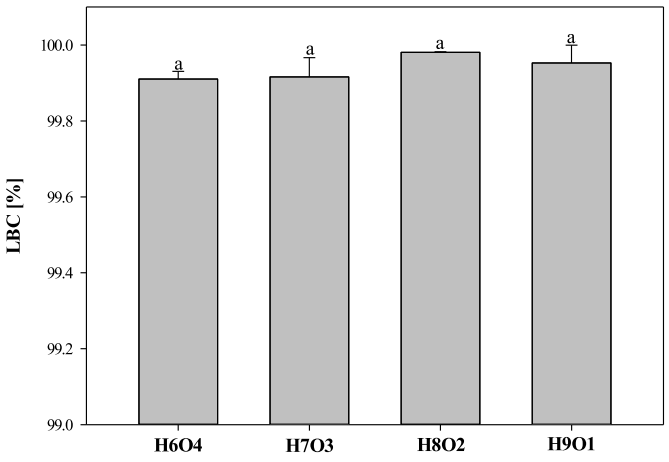


Fig. 3. LBC properties of bigels with different hydrogel-to-oleogel ratios.

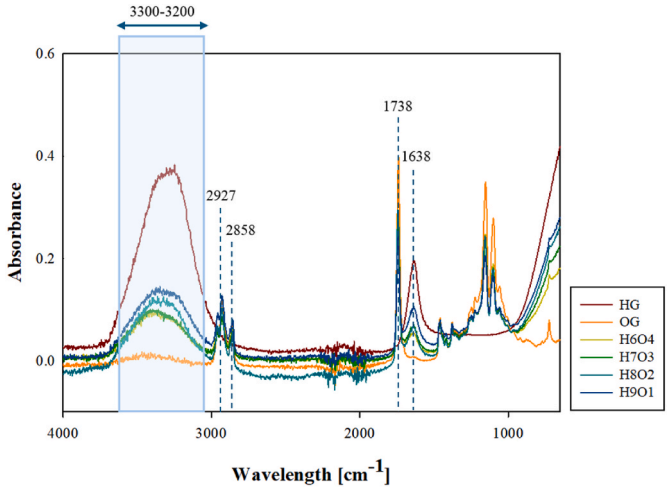


Fig. 4. FT-IR spectra of bigels with various hydrogel-to-oleogel ratios.

3.1.4. *Rheological analysis: amplitude sweep, frequency sweep, and 3ITT tests*

The rheological properties of bigels with various oleogel-to-hydrogel ratios were assessed through amplitude sweep, frequency sweep, and 3ITT analysis (Figs. 5 and 6). In the amplitude sweep test, all samples exhibited a stable storage modulus (G′) at low strain, indicating a well-maintained gel structure within the linear viscoelastic region (LVR). As strain increased beyond this region, the gel network began to break down, as evidenced by a decrease in both G′ and G″ (Fig. 5A). Based on these results, a 0.1 % strain—within the LVR—was selected for subsequent tests to ensure structural integrity during measurement.

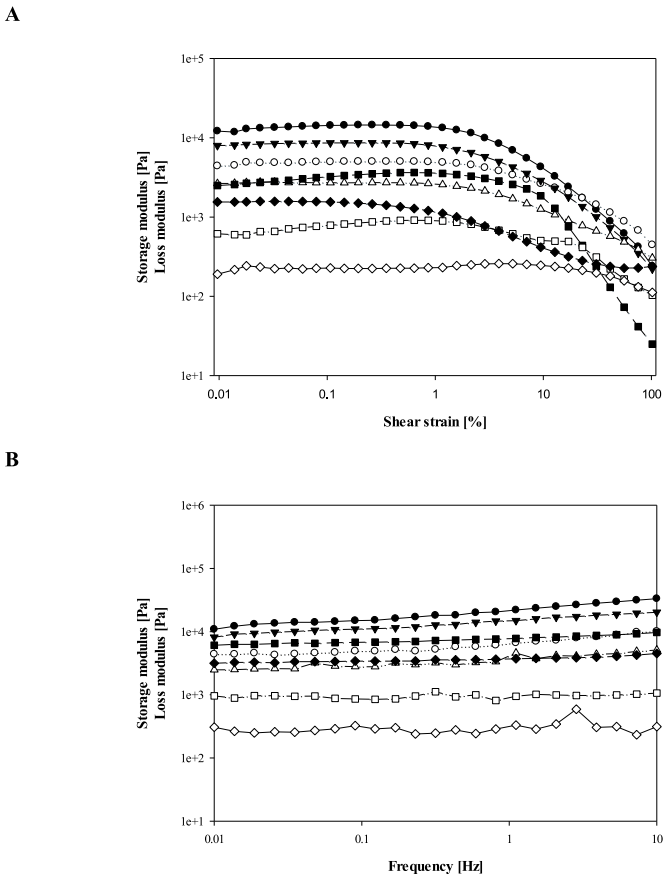


Fig. 5. Rheological properties of bigels with various hydrogel-to-oleogel ratios: H6O4 (○, ●); H7O3 (△, ▲); H8O2 (□, ■); H9O1 (◇, ◆). Closed markers represent the storage modulus (G′), and open markers represent the loss modulus (G″). A. Amplitude sweep test; B. Frequency sweep test.

In the frequency sweep test (Fig. 5B), all bigel samples exhibited solid-like viscoelastic behavior, with G′ consistently exceeding G″ across the tested frequency range (0.01–10 Hz). The absence of a crossover point indicated no transition to viscous dominance within the LVR. Additionally, as the oleogel content increased from 10 % to 40 %, both G′ and G″ values increased, suggesting that the dispersed oleogel phase contributed significantly to the mechanical rigidity of the bigel matrix. This trend aligns with the observed microstructure (Fig. 1) and is consistent with previous reports highlighting the dominant influence of oleogel content on rheological properties (Kaimal & Singhal, 2023; Zhu et al., 2021).

To evaluate the structural recovery capacity of bigels following deformation—an important criterion for food processing—3ITT tests were performed (Fig. 6). High-hydrogel-content samples (H9O1, H8O2)

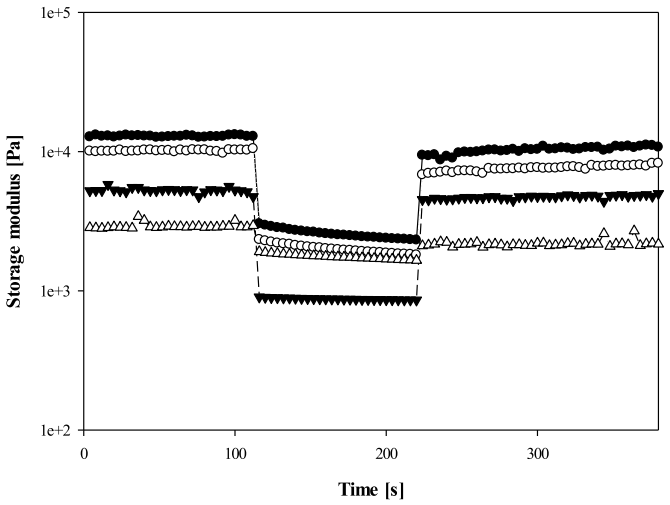


Fig. 6. Thixotropic properties of bigels with various hydrogel-to-oleogel ratios: H6O4 (●), H7O3 (○), H8O2 (▼), H9O1 (△).

exhibited recovery ratios close to 1.0, indicating strong elasticity and thixotropic behavior (Table 1). Conversely, formulations with lower hydrogel content (e.g., H6O4) showed significantly reduced recovery (0.84), reflecting poor internal structure regeneration. These results suggest that the hydrogel phase, as the continuous matrix, plays a pivotal role in elasticity and structural integrity after shear deformation.

3.2. Evaluation of bigel as a fat substitute: texture and thermal properties

3.2.1. Texture analysis

Texture profile analysis provided information on properties such as hardness and springiness, offering insight into micro- and macro-structural characteristics of food. The texture properties of animal fats in meat products contribute to tenderness and mouthfeel. Therefore, it is important for plant-based meat analogs to exhibit texture properties that resemble those of animal fat. The texture properties of the oleogel-in-hydrogel system bigels are presented in Table 2. The hardness of the bigels increased as the oleogel phase fraction rose from 10 % to 40 %, aligning with the bigel's flow behavior (Fig. 5). In contrast, springiness increased as the oleogel phase in bigels decreased (Table 2). This springiness is related to the elastic recovery of the bigels, which can be attributed to the synergistic gelation of the KGM and XG-based hydrogel, potentially enhancing its elasticity (Qiao et al., 2024). The hardness and springiness of pork fat were approximately 13,445 g and 82 %, respectively. Although the hardness of the bigels showed a significant difference compared to lard, the elasticity showed no significant difference in H7O3, confirming its potential for replacement of animal fat.

3.2.2. Rheological analysis: temperature sweep

Replicating the sensory qualities and functionality of fat in meat alternatives requires an in-depth understanding of the thermo-mechanical properties of fat mimetics in comparison to animal fat (Soleimanian et al., 2024). To achieve this, a temperature sweep test was conducted to analyze the thermo-responsive rheological properties of bigels across a temperature range of 4 °C–80 °C. The complex shear modulus and loss factors were recorded during the temperature scan (Figs. 7 and 8). During heating, all bigels, along with pork fat as a control, showed a decrease in complex shear modulus but maintained a solid-like gel state (Fig. 7). This was indicated by the loss factor, which is the ratio of the viscous to elastic components (Rai et al., 2016). The intersection of the storage modulus and loss modulus, at a loss factor of 1, was the gel point of the material (Tan et al., 2011). Pork fat experienced a significant decrease at the beginning of the heating stage because crystallinity in fat cells weakened its structure, leading to substantially reduced rigidity. After this steep decrease in modulus, no further decrease occurred during heating, but a slight increase was followed by a plateau. This may be attributed to factors such as tightening of the collagen network and water loss (Wijarnprecha et al., 2022). During the cooling stage, from 80 °C to 4 °C, the modulus of all bigels

Table 1
Recovery value of bigels at various ratios of hydrogel and oleogel.

H6O4	H7O3	H8O2	H9O1
0.84 ± 0.13[b]	0.85 ± 0.00[b]	0.95 ± .085[a]	1.03 ± 0.06[a]

*All processes were repeated three times. Values are expressed as mean ± SD (n = 3), and values with different letters are significantly different at $p < 0.05$ (ANOVA with post-hoc Duncan's test).

Table 2
Texture properties of bigels with different ratios of hydrogel to oleogel.

Sample[a]	Hardness (g)	Springiness (%)
Pork fat	13,445.23 ± 3876.62[a]	81.90 ± 4.89[b]
H6O4	1134.09 ± 37.84[b]	67.15 ± 4.56[c]
H7O3	882.55 ± 71.18[b]	79.77 ± 0.58[b]
H8O2	510.30 ± 19.63[b]	94.39 ± 0.53[a]
H9O1	55.21 ± 38.91[b]	95.04 ± 1.17[a]

[a] All processes were repeated three times. Values are expressed as mean ± SD (n = 3), and values with different letters are significantly different at $p < 0.05$ (ANOVA with post-hoc Duncan's test).

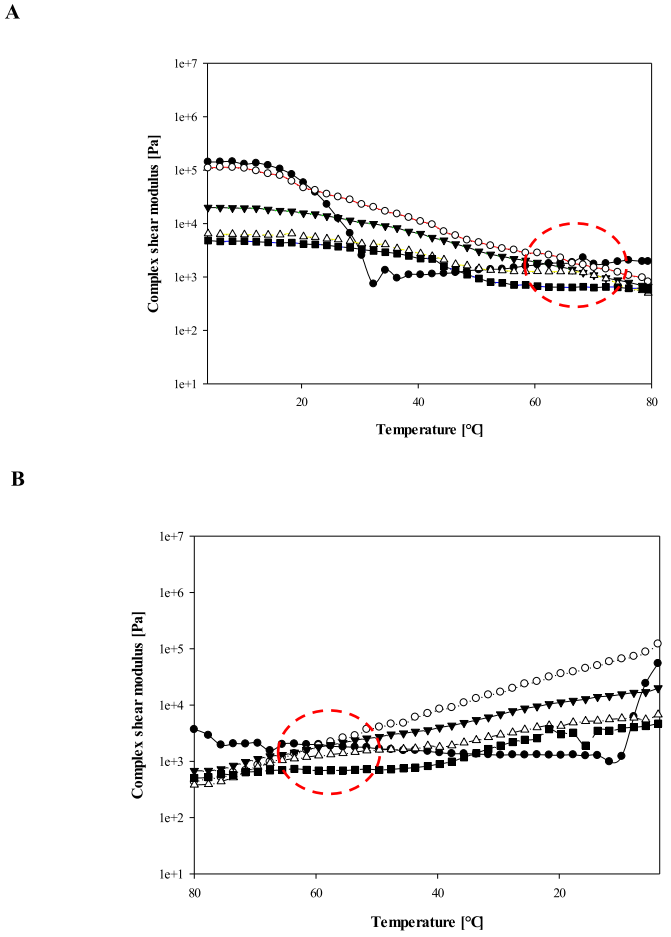


Fig. 7. Temperature-dependent complex shear modulus of pork fat and bigels with various hydrogel-to-oleogel ratios under heating and cooling conditions: pork fat (●), H6O4 (○), H7O3 (▼), H8O2 (△), H9O1 (■). A. Heating lamp; B. Cooling lamp.

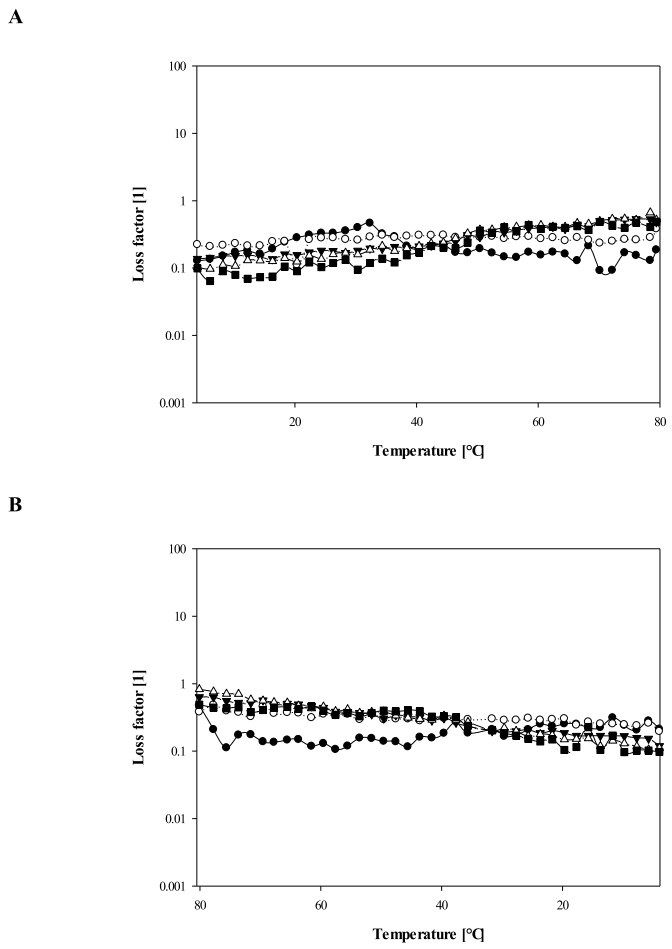


Fig. 8. Temperature-dependent loss factor of pork fat and bigels at various hydrogel-to-oleogel ratios under heating and cooling conditions: pork fat (●), H6O4 (○), H7O3 (▼), H8O2 (△), H9O1 (■). A. Heating lamp; B. Cooling lamp.

and pork fat increased, contrasting with the heating stage, while still retaining a solid-like gel state. Around 62.8 °C during cooling, the complex shear modulus of bigels was similar to that of pork fat. This similarity suggests that the bigels exhibit comparable rheological properties to pork fat at 62.8 °C, the USDA's minimum cooking standard for meat. In conclusion, these findings indicate that bigels can display thermo-dependent behavior similar to animal fat and are promising candidates as animal fat substitutes.

4. Conclusion

This study developed food-grade oleogel-in hydrogel bigels by incorporating EC-based oleogels into KGM-XG hydrogels, aiming to mimic the structural and thermo-mechanical behavior of animal fat. The resulting bigels exhibited stable biphasic structures, with higher hydrogel content promoting uniform droplet dispersion and improved elasticity. FT-IR analysis confirmed the coexistence of both phases without new chemical bonds, while rheological and thermal tests demonstrated solid-like viscoelasticity and thermo-responsive properties comparable to pork fat near 62.8 °C.

These outcomes suggest that bigels can provide desirable textural and thermal functions in processed food applications. At the same time, the study has inherent limitations. The formulations were prepared at laboratory scale, and their scalability to industrial processing has not yet been demonstrated. Storage stability, oxidative resistance, and consumer acceptance were also beyond the scope of this work but represented critical factors for practical applications. Such considerations highlight that, while bigels offer technical feasibility, further validation is needed before commercial deployment.

Nevertheless, the findings carry broader implications. Oleogel-in-hydrogel bigels may serve not only as animal fat replacers in meat analogs but also as versatile carriers for bioactive compounds or flavor systems. Future research should therefore focus on process optimization for scale-up, evaluation of long-term stability, and sensory studies to bridge laboratory innovation with industrial implementation.

CRediT authorship contribution statement

Yehyun Ham: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Yourim Oh:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Mina Kim:** Formal analysis, Data curation. **Jin-Kyu Rhee:** Validation, Supervision, Project administration.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.lwt.2025.118743.

Data availability

I have shared the link to my data at the attach file step.

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