Characterization of mixed-component oleogels: Beeswax and glycerol monostearate interactions towards *Tenebrio Molitor* larvae oil

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ABSTRACT

Edible insects are attracting attention as an alternative food due to their excellent production efficiency, lower carbon consumption, and containing high protein. *Tenebrio Molitor* larvae (TM), one of the approved edible insects worldwide, contain more than 30 % fat content consisting of 70 % unsaturated fatty acids, and particularly high phospholipids. Most of the research has focused on the utilization of proteins, and there are few studies using oils from TM. Therefore, in this study, to expand the utilization of TM oil in food applications, the oleogel was prepared with TM oil fortified by the incorporation of beeswax (BSW) and glycerol monostearate (GMS), and their structure, rheological and thermal properties were evaluated. The interaction between BSW and GMS contributed to the strength of the oleogel structure. The addition of GMS or the increase of the gelator concentrations resulted in increasing the melting point, which is consistent with the observed increase in viscoelasticity. As the temperature increased, the solid fat content decreased. The result of FT-IR suggests that TM oil is physically solidified without changing chemical composition through oleogelation. This study suggests a new processing direction for edible insects by confirming the rheological, thermal, and physicochemical characteristics of TM oil-based oleogel.

1. Introduction

The consumption of saturated fat and *trans*-fat is widely recognized as a major contributor to non-communicable diseases such as type 2 diabetes and cardiovascular disease. Despite the essential role of fats as nutrients for our body, there are global recommendations to limit total fat intake. The World Health Organization (WHO) recommends limiting total fat intake to less than 30 % of energy intake, and saturated and trans fatty acids to less than 10 % and 1 %, respectively (Ghebreyesus and Frieden, 2018). The European Commission (EU) has also imposed a limitation on the use of *trans*-fats to 2 g per 100 g of total fat (Puşcaş et al., 2020). In response to health concerns, numerous studies have been conducted to substitute saturated fat with healthier alternatives, particularly polyunsaturated fat. Oleogelation, using liquid oil rich in polyunsaturated fat, stands out as one of the techniques to replace saturated fat. The oleogel is a lipophilic mixture structured by a three-dimensional network of gelator molecules without altering the chemical composition of liquid oil (da Silva et al., 2018). The physicochemical and functional characteristics of oleogel are determined by the types of oil used (fatty acid composition, saturated and unsaturated

ratio, source) and types of gelator. Most previous studies about oleogels focused on the utilization of different types of vegetable oils (Bodennec et al., 2016; Li et al., 2022; Pang et al., 2020), thus, there is a need for research exploring the physicochemical and functional properties of oleogel using new oils, including edible insect oil, in a food application.

In the midst of recent environmental issues, edible insects are intensively focused as alternative and sustainable future food materials. Researchers have explored various functionalities such as antioxidant, anti-collagenase (Yeerong et al., 2021), anti-inflammatory properties (Zielińska et al., 2017), and platelet aggregation activities (Pyo et al., 2020) in extracts derived from edible insects. However, most studies have focused on the extraction efficiency and functional properties of edible insect extracts or protein sources, few studies have been conducted on the use of insect fat (or oil) in foodstuffs. In general, the lipid content of edible insects is 10–50 % on a dry basis and is relatively high in unsaturated fatty acids (Sosa and Fogliano, 2017). Especially, *Tenebrio Molitor* larvae (TM) contains 13–35 % lipids and 61–78 % unsaturated fatty acids (Tzompa-Sosa et al., 2014; Paul et al., 2017), but there has been limited research on applying its oil in food applications. Moreover, to enhance the utilization of TM oil, which is rich in

unsaturated fatty acids, it is crucial to incorporate TM oil into food and develop oleogels as a replacement for saturated fats.

The blend of oleogelators is well known to one of the ways to manipulate the structural arrangement, enhance the properties of oleogels, and alleviate the disadvantages of each gelator (Barroso et al., 2020). In a recent study, a synergistic effect enhancing the mechanical strength was observed at a specific mixing ratio when sunflower oil was gelled with lecithin and sorbitan tri-stearate (Pernetti et al., 2007). Pérez-Monterroza et al. (2014) reported that the combination of gelators containing beeswax, mono-, diglycerides of fatty acids, and sorbitan monostearate affected the crystallization temperature of avocado oil oleogel. And the changed phase behaviors of binary blends of bees, paraffin, sunflower, and rice bran wax were investigated (Jana and Martini, 2016). Beeswax (BW) is among the most common structure-forming gelators for the production of oleogels, and an animal-based wax that is produced from the genus *Apis mellifera* L. of bee (Moghtadaei et al., 2018). Glycerol monostearate (GMS), which is a hydrophobic, non-ionic, and low molecular weight surfactant, acts as a lipid structuring gelator at relatively low concentrations (Co and Marangoni, 2012; Singh et al., 2015). Although many studies report that the blend of gelators affects the physicochemical properties such as melting point and hardness, few studies on the combination of insect oil with the blend of BW and GMS have been reported.

Therefore, in this study, TM oil oleogels were prepared using a blend of BW and GMS at the different concentrations (5 % and 10 %), and their structural, thermal, and rheological properties were investigated to explore the new application for TM oil.

2. Material and methods

2.1. Oil extraction from Tenebrio Molitor larvae

According to the methods of Kim and Oh (2022), *Tenebrio Molitor* larvae (TM), which were fasted for 48 h to clear their gastrointestinal tract of any residual food, were purchased from Myungpyum Co. Ltd. (Jangseong-gun, Korea). First, TM larvae were washed with water and dried by using a microwave oven for 11 min. Dried TM larvae were physically pressed at a low temperature using a compressor (Poongjin, Korea) with a pressure level of 700 kg/cm^2 for 25 min without using organic solvent. The extraction yield of TM oil was 31.8 % of the total weight. The TM oil used in this study is composed of 23.8 % saturated fatty acids and 73.6 % unsaturated fatty acids. Among them, oleic acid (C18:1) constitutes the largest portion at 46.1 %, followed by linoleic acid (C18:2) at 25.1 %, and palmitic acid (C16:0) at 17.3 % (Kim and Oh, 2022).

2.2. Preparation of oleogels

Beeswax (BW) was supplied by Hooper pharm GmbH Co. (Hamburg, Germany) and glycerol monostearate (GMS) was obtained from Ilshinwells Co., Ltd. (Seoul, Korea). The oleogelators (BW and GMS) were dispersed in TM oil at different concentrations of 5 % and 10 % (w/w), and the ratio of BW and GMS was 3: 1, which was determined according to preliminary study and the reference (Choi et al., 2020; Kim and Oh, 2022). The dispersions were heated at 90 °C until completely dissolved, cooled to room temperature for 30 min, and solidified in the freezer (−21 °C) for 1 h resulting in the formation of oleogels. The samples were named TM oil-oleogel with 5 % BW (TBO5 %), TM oil-oleogel with 5 % BW and GMS (TBGO5 %), TM oil-oleogel with 10 % BW (TBO10 %), and TM oil-oleogel with 10 % BW and GMS (TBGO10 %), respectively.

2.3. Texture analysis

The textural properties of oleogels were investigated using a texture analyzer with a spreadability fixture (TA-XT plus, Stable Micro Systems, Surrey, UK). Spreadability fixture consists of accurately paired male and female Perspex cones set at 90° angle. Before measurement, the oleogels were stored at room temperature for 30 min. Samples (4 g) were placed inside the female cone, and pressed down to leave a flat test surface. As reported by Oh et al. (2019), the male cone penetrated the oleogel at 100 mm/min and continued to a depth of 2 mm. The textural parameters, including firmness and work of shear, were determined by analyzing the force-time deformation curves. Firmness was indicated by the force at maximum penetration depth, while the total force required for the shearing process was represented by the area under the curve, denoting the work of shear.

2.4. Oil binding capacity (OBC)

Oil binding capacity was measured according to the centrifuge method reported by Meng et al. (2019a). Briefly, 1 g of oleogels was weighed in a 2 mL centrifuge tube and the sample was centrifuged (M13, Hanil Scientific Inc., Gimpo, Korea) at 12,000 X *g* for 15 min. The tube was inverted on filter paper for 10 min to remove the expressed oil. The OBC was calculated using the following formulas:

$$\% \text{ Released oil} = \frac{\text{Mass of expressed oil (g)}}{\text{Total mass of sample (g)}} \times 100 \tag{1}$$

$$\% \text{ OBC} = 100 - \% \text{ Released oil} \tag{2}$$

2.5. Rheological measurements

Dynamic viscoelastic properties of the oleogel samples were investigated using a rheometer (DHR-10, TA instrument, New Castle, USA) with a 40 mm parallel plate. The frequency sweep test was conducted within a frequency range of 0.1–100 Hz, employing a strain of 0.1 %, which remained within the linear viscoelastic limit. Furthermore, a dynamic temperature ramp test was executed, spanning from 30 to 60 °C with a heating rate of 5 °C/min. In both rheological assessments, the parameters under observation included the storage modulus (G′) and the loss modulus (G″).

2.6. Thermal analysis

Thermal properties of the TM oil-oleogel samples were investigated by differential scanning calorimetry (Q200, TA instrument, News Castle, USA). The oleogel samples (approximately 10 mg) were sealed into an aluminum pan and heated to 90 °C, cooled down to −20 °C at 10 °C/min, and then heated 100 °C at 10 °C/min.

2.7. Solid fat content (SFC)

Based on the approved methods of AOCS (Moon et al., 2021), the solid fat content (SFC) of oleogels was measured using time-domain nuclear magnetic resonance (TD-NMR) (MQC+, Oxford Instruments, Oxon, UK). The oleogels filled in an NMR tube (10 mm diameter) were placed at 90 °C for 30 min. Afterward, the tube was maintained for 30 min at each measurement temperature (from 10 °C to 90 °C) and then loaded into the NMR.

2.8. FT-IR spectroscopy analysis

The oleogels stored in a refrigerator (2 °C) were taken directly to the FT-IR test one by one in sequence. The infrared spectra of oleogels were recorded by using a Fourier transform infrared (FT-IR) spectrometer (Spectrum Two, PerkinElmer Inc., Waltham, USA) at room temperature (18 °C). The scanning range was 4000–1000 cm^{-1} and the peaks were analyzed to determine the interactions between the oleogel components.

2.9. X-ray diffraction analysis

The XRD patterns of oleogels were recorded using the X-ray diffractometer (XRD-7000, Shimadzu, Kyoto, Japan) with Cu-Kα operating at 40 kV and 30 mA. The oleogels were smoothed by loading onto glass slide wells at room temperature. The angular scans were performed at a diffraction angle range of 10°–60° with a scan speed. XRD data were analyzed with PCXRD software (Shimadzu, Kyoto, Japan).

2.10. Microscopy

Morphology of TM oil-oleogel samples were analyzed through optical microscopy (NB-2000T, Mightex, Toronto, Canada). The TM-oil oleogels were placed on a microscope slide glass, and a cover slip glass was placed over the sample and gently pressed until clear crystal structures could be shown. The slide was then mounted under the microscope and visualized using $100 \times$ lens. Finally, micrographs were analyzed using image processing software associated with the camera (Zen version 1.0, Carl Zeiss GMBH).

2.11. Statistical analysis

All experiments were performed in triplicate, and the obtained results were statistically analyzed with SPSS software (IBM SPSS Statistics 26, IBM Corp., Armonk, NY, USA). Duncan's multiple range tests were performed to identify any significant difference among the samples with a confidence level set at 95 %

3. Results and discussion

The visual appearance of TM oil oleogels with BW and GMS was presented in Fig. 1. All samples were successfully converted into self-standing solid-like oleogels without significantly visual difference. The textural properties of oleogels were investigated in terms of firmness and work of shear (Table 1- (a)). In particular, the firmness of TBGO10 % was significantly higher than that of other oleogels ($p < 0.05$), which was related to the crystal structure and intermolecular forces inside oleogels (Sun et al., 2021). And the firmness was significantly increased with increasing the oleogelators concentration. These results suggest that the higher the concentration of oleogelator, the more hydrogen bonds are formed between the chains, which strengthens the oleogelator chains (Moghtadaei et al., 2018). And the oleogel containing with GMS exhibited higher hardness than the oleogel without GMS, which indicated that the interaction between BW and GMS contributed to enhance the oleogel structure. This tendency was also observed in the result of Kim et al. (2022), and Choi et al. (2020). The work of shear represents the total amount of force required to perform the shearing process. A lower value of work of shear indicates a softer spread, ie higher spreadability (Shakerardekani et al., 2013). In this study, values of work
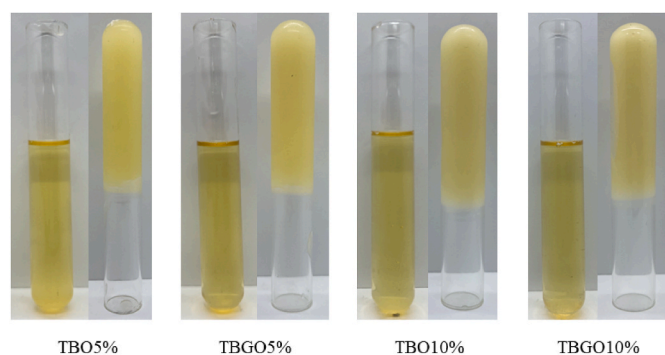

Fig. 1. Visual appearances of TM oil-oleogels prepared from TM oil with BW and GMS.

Table 1
Textural properties (a) and oil binding capacity (b) of TM oil-oleogels.

		TBO5 %	TBGO5 %	TBO10 %	TBGO10 %
(a)	Firmness (N)	0.19 ± 0.00 d	0.56 ± 0.01c	1.83 ± 0.03 b	4.56 ± 0.24a
	Work of shear (N·s)	0.23 ± 0.00c	0.76 ± 0.00c	2.27 ± 0.19 b	6.69 ± 0.55a
(b)	OBC (%)	22.62 ± 0.87 d	42.38 ± 0.84c	77.35 ± 0.68 b	90.88 ± 0.57a

Values are means ± standard deviation (n = 3).
Means with different small letters in the same column differ significantly by Duncan test ($p < 0.05$).

of shear also showed the same tendency as firmness. TBGO10 % showed the highest work of shear value, which exhibited significantly the lowest spreadability, but TBO5 % and TBGO5 % had the highest spreadability. Also, there was no significant difference at the level of 5 % oleogelator concentration (TBO5 % and TBGO5 %), the addition of GMS at the level of 10 % significantly increased the work of shear of oleogel.

The oil binding capacity (OBC) is an important criterion that reflects the degree of oil entrapment in the network by the gelator (Thakur et al., 2022) and the higher OBC indicates the superiority in terms of transportation and food applications (Zhang et al., 2020). The OBC values of the samples ranged from 22.62 to 90.88 %. The lowest value was shown in TBO5 % sample, on the other hand, the highest value was observed at TBGO10 %. The OBC values were increased as the concentration of wax increased and as the GMS was added. This tendency was consistent with the result of Thakur et al. (2022), which reported an increase in OBC with an increasing level of gelator. Choi et al. (2020) also reported that the lowest value of oiling-off was observed when candelilla wax and GMS were blended at the ratio of 3 and 1, and the harder texture of oleogel contributed to preventing the oil separation.

The rheological properties of oleogel-samples were investigated in terms of dynamic viscoelasticity. The storage (G′) and loss (G″) moduli of the oleogels represent their elastic and viscous behaviors, respectively (Oh et al., 2019). As shown in Fig. 2, all oleogel samples show the gel-like structure with elastic component (G′) prevailing over the viscous component (G″). In particular, no intersection occurs over the entire measured frequency sweep range, indication that all samples formed stable gel systems (Zhang et al., 2021). The oleogel samples containing GMS exhibited higher values of G′ and G″, remained almost unchanged during the increase of the frequency. This result was indicated that the oleogel had the ability to resist the high-frequency oscillation, and the semi-solid property of oleogels could not be broken by the high-frequency oscillation (Jiang et al., 2022). The tan delta (G″/G′) is presented in Fig. 2(B). The TBO5 % had the highest tan values with TBGO 10 % had the lowest tan value. Tan delta values of these oleogel samples were lower than 1, indicated that the oleogels demonstrated gel-like consistency and elastic dominant properties at room temperature.

The rheological behaviors as a function of temperature of oleogels were investigated over 30–60 °C (Fig. 3). It was noted that the oleogel sample prepared only with BW 5 % (TBO5 %) exhibited the lowest values of viscoelastic parameters at the temperatures before melting, followed by TBGO5 % < TBO10 % < TBGO10 %. Thus, although moduli had an overall tendency to increase with increasing levels of gelator, the highest G′ and G″ were clearly observed at the TBGO10 %, demonstrating that the TBGO10 % was harder than the other oleogel samples. These tendencies were in great agreement with the frequency results. All oleogels showed a sharply decline in G′ and G″ in temperature range of approximately above 35 °C. The cross-over points for G′ and G″ were also observed in all samples. The cross-over points are interpreted as an increase in the flowability of oleogel due to structural breakdown at high temperatures (above 40 °C). The cross-over points of TBO5 % and TBGO5 % were observed at around 40 °C, and TBG10 % at around 45 °C, and TBGO10 % at above 50 °C. In the presence of GMS at the 10 %
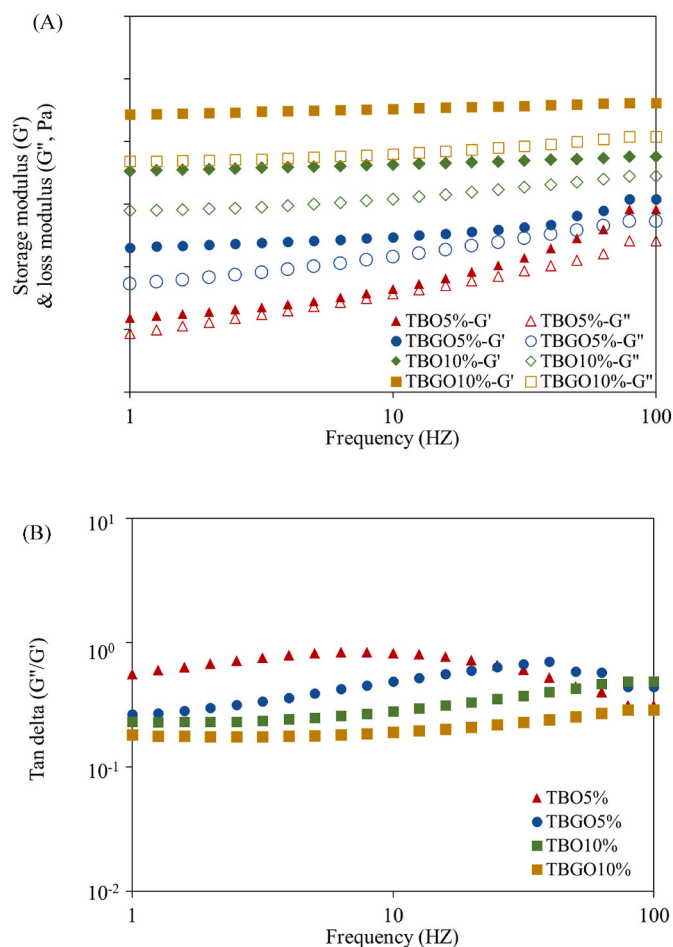
(A)

(B)

Fig. 2. Dynamic viscoelastic properties of TM oil-oleogels as a function of frequency: storage moduli & loss modulus (A), tan delta (B).
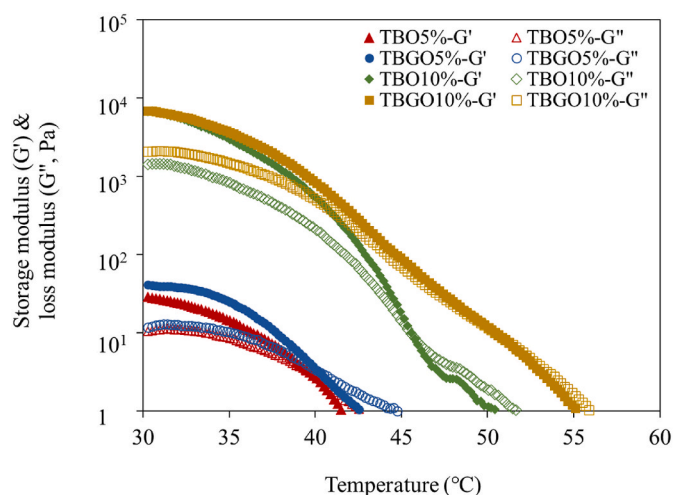

Fig. 3. The viscoelastic change of TM oil-oleogels on temperature.

oleogelator concentration, the cross-over point (temperature) and the strength of the crystalline network structure could be increased. BW and GMS consist of mixed crystal system, which shows the higher melting temperature and the wider range of melting temperature in the thermogram. Also, these viscoelastic properties of oleogels as a function of temperature is similar with the results of the DSC analysis.

The thermal properties (onset and peak crystallization and peak

melting temperatures) of oleogels were characterized by DSC. The melting and crystallization behavior of oleogels are presented in Table 2. The crystallization transitions of oleogels occur at 12.47–21.22 °C. Especially, as the concentration of oleogelator increased, the peak temperature increased, however, the oleogels containing the binary gelators were lower peak temperature than single gelators. The mixtures (BW and GMS) appeared to be eutectic because the crystallization T_{on} and T_{peak} was lower than single gelator (as seen with the only BW) and was the lowest for TBGO5 % oleogels. TBO5 % and TBO10 % both showed a single melting peak within the experimental temperature range. On the other hands, the melting peak temperature was shifted from 38.75 °C to 42.46 °C when increasing the oleogelator concentration from 5 % to 10 %, respectively. These results were consistent with the temperature sweep that as the concentration of oleogelator increased, the more stable structure during heating. Enthalpy change (ΔH) as calculated from the peak area was 2.29 J/g (TBO5 %), 1.41 J/g (TBGO5 %), 6.28 J/g (TBO10 %), and 3.52 J/g (TBGO10 %) respectively. The enthalpy was decreased with the addition of GMS at the same gelator concentrations.

The SFC value indicates the percentage of the solid phase of fats at a certain temperature and shows the changes in the consistency and plasticity of food products at different temperatures (Li et al., 2021). The solid/liquid ratio of fat affects the quality of baked goods (Demirkesen and Mert, 2020). Therefore, since a certain level of SFC value is related to the aeration of the batter, it is one of the important factors for obtaining a softer and uniform structure in bakery products (Ghotra et al., 2002). Fig. 4 demonstrates the SFC values of the oleogels. All oleogels were decreased with increasing temperature. The decrease in SFC is attributed to the destruction of the network structure by melting the oleogel when the temperature rises (Wang et al., 2021). The SFC value increased as the gelator concentration increased, and these results were consistent with Liu et al. (2021). In contrast to rheological and thermal properties, the addition of GMS incorporated to the oleogel did not significant impact the SFC results. At 10 °C, the SFC content of single gelator was lower than a binary gelator (i.e., TBO10 % > TBGO10 %, TBO5 % > TBGO5 %), suggesting that a single gelator may form a crystal network structure more rapidly than a binary gelator system. However, there was no significant differences among the samples above 50 °C. In addition, this result indicates that the degree of gel formation may not necessarily correlate with the strength of the formed gel.

FTIR can be used to understand the structural interactions of a molecule. The FTIR spectra of liquid oil and oleogels are presented in Fig. 5. FTIR can analyze structural information about molecular interactions in oleogel systems based on the molecular characteristic absorption peaks (Meng et al., 2019a). The peak observed at 2920 cm^{-1} and 1464 cm^{-1} indicated C–H stretching of CH_3 (Meng et al., 2018b). The peaks observed at 2852 cm^{-1} indicated C–H stretching of CH_2 (Meng et al., 2018a). The peak at 1743 cm^{-1} confirmed the presence of C=O stretching and reflected the contribution of high-molecular esters from TM oil and BW (Trujillo-Ramírez et al., 2022). The band around 1160 cm^{-1} corresponded to C–O stretching vibration (Meng et al.,

Table 2
Thermal properties of TM oil-oleogels.

		TBO5 %	TBGO5 %	TBO10 %	TBGO10 %
Crystallization	T_{on} (°C)	18.49 ± 0.40c	15.22 ± 0.59 d	22.57 ± 0.15 b	20.96 ± 0.24a
	T_{peak} (°C)	16.50 ± 0.53c	12.47 ± 0.78 d	21.22 ± 0.15a	19.61 ± 0.22 b
	ΔH (J/g)	2.99 ± 0.07c	1.60 ± 0.06 d	5.42 ± 0.14a	4.08 ± 0.10 b
Melting	T_{on} (°C)	29.76 ± 0.52 b	29.18 ± 0.31 b	32.72 ± 1.22a	31.93 ± 0.17a
	T_{peak} (°C)	38.75 ± 1.36c	41.22 ± 0.20 b	42.46 ± 0.52 b	47.07 ± 0.62a
	ΔH (J/g)	2.29 ± 0.74c	1.41 ± 0.19 d	6.28 ± 0.86a	3.52 ± 0.40 b
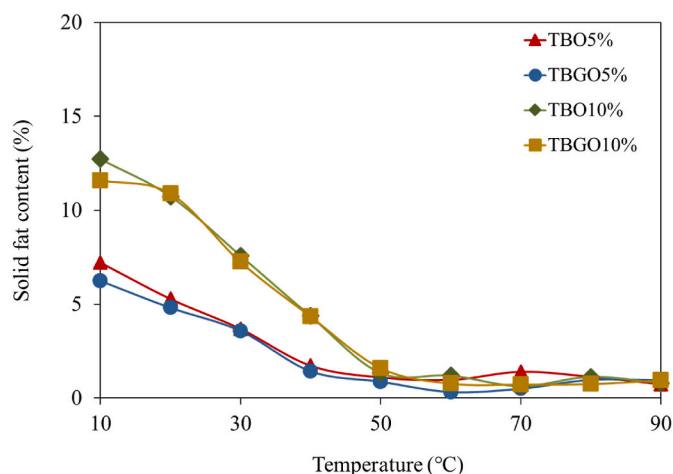
Fig. 4. Solid fat content of TM oil-oleogels.

Fig. 5. FTIR spectra of TM oil-oleogels and TM oil.
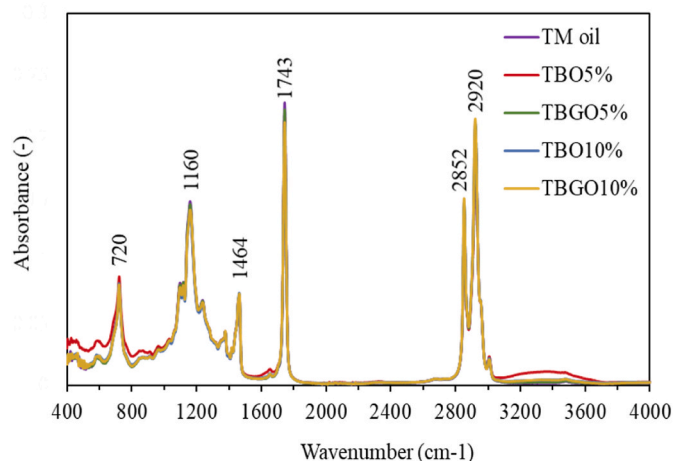

Fig. 6. XRD patterns of TM oil-oleogels.

2019b). Furthermore, the spectrum of oil and oleogel was consistent at specific peaks. Thakur et al. (2022) also observed that carnauba wax oleogels had a similar spectrum to that of soybean oil. Therefore, these results suggest that TM oil is physically solidified without changing chemical composition through oleogelation.

XRD can be utilized to analyze the internal structures of oleogels. As shown Fig. 6, the XRD patterns clearly showed that diffraction peaks of all oleogel samples were almost the same. All oleogel samples have diffraction peaks at 19.4°, 21.3°, and 23.7°, corresponding to spacings of 4.5, 4.1, and 3.7 Å, respectively, which indicate the presence of β, α, and β′ polymorphic forms in the structure (Fayaz et al., 2017). In particular, when the concentration of the gelator increased and GMS was added, a wider and sharper peak was shown. In according to Zhang et al. (2021), the strong diffraction peaks at 4.14 and 3.74 Å indicate that the crystal structure is mainly due to the beeswax gelator. Therefore, the oleogels of TM oil are formed by the crystallization of beeswax molecules.

Fig. 7 display microscope images of oleogels with single and binary gelators at the different concentration (5 and 10 %). TBO5 % and TBO10 % samples showed a large number of needle-like crystals that are not homogeneously dispersed phase. More denser crystal network was observed as BW concentration increased. TBGO 5 % and TBGO10 % exhibit fewer crystals and smoother surface. The difference in the crystal morphology and distribution of BW oleogels was more clearly observed with increasing concentration and adding GMS. As reported in the results of Hwang et al. (2015), a denser crystal network appeared to be associated with the increased hardness of oleogels.
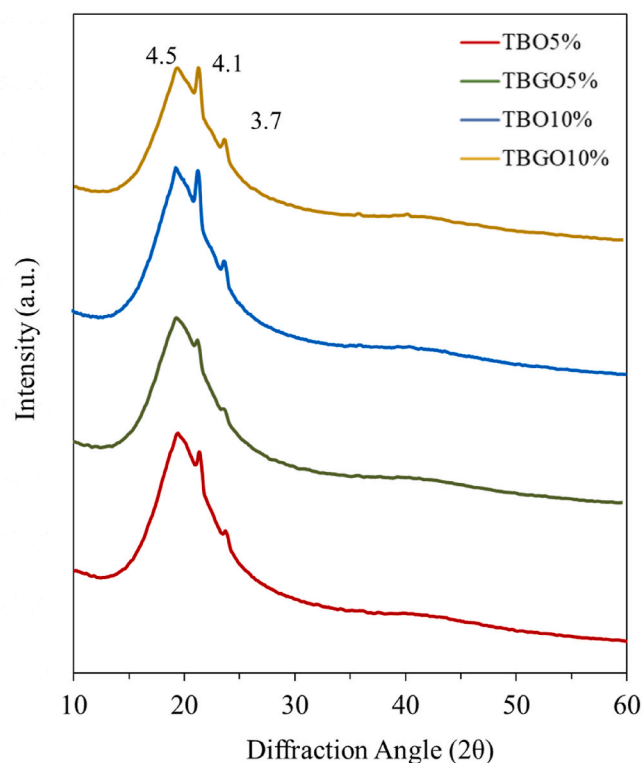
4. Conclusions

Although the TM oil are subject to food regulations managed by Generally Recognition of Safety (GRAS) and recognized as a nutritionally valuable oil containing unsaturated fatty acids, the utilization of TM oil has been very limited and it is mostly discarded during protein purification processes. Therefore, in this study, the expand TM oil in food applications, the oleogels were prepared with BW and GMS and their thermodynamic, rheological, and crystallization behaviors were investigated. The TM oil used in this study contains 73.6 % unsaturated fatty acids, composed of 46.1 % oleic acid (C18:1) and 25.1 % linoleic acid (C18:2). The applying the oleogelation technology using BW and GMS to TM oil, succeeded in solidification without changing the chemical composition. The use of GMS with BW increased gel strength, which was consistent with increased OBC, the rheological and thermal properties of oleogels. The interaction of BW and GMS on oleogel containing TM oil affects the structural, rheological, and thermal properties of oleogel, which is valuable as a solid fat replacer. Also, the addition of GMS increased the thermal stability and gel strength of the oleogel. Therefore, the use of oleogels prepared with a binary gelators (BW and GMS) is better than those made of single oleogelator. This study is anticipated to be advantageous in the future for manufacturing baked goods, as it can form a stronger gel when mixed with other food ingredient at high temperatures, potentially substituting for animal fat or solid fats. However, the rheological and thermal properties of TM oleogel were examined with a fixed ratio of 3:1. Further research is needed to explore the effects of the interaction between BW and GMS at various blending ratios in order to comprehend the gelation mechanism of TM oil oleogels. More structural research and sensory evaluation of TM oil-oleogel will be necessary for practical food applications as a healthy solid fat replacer.

CRediT authorship contribution statement

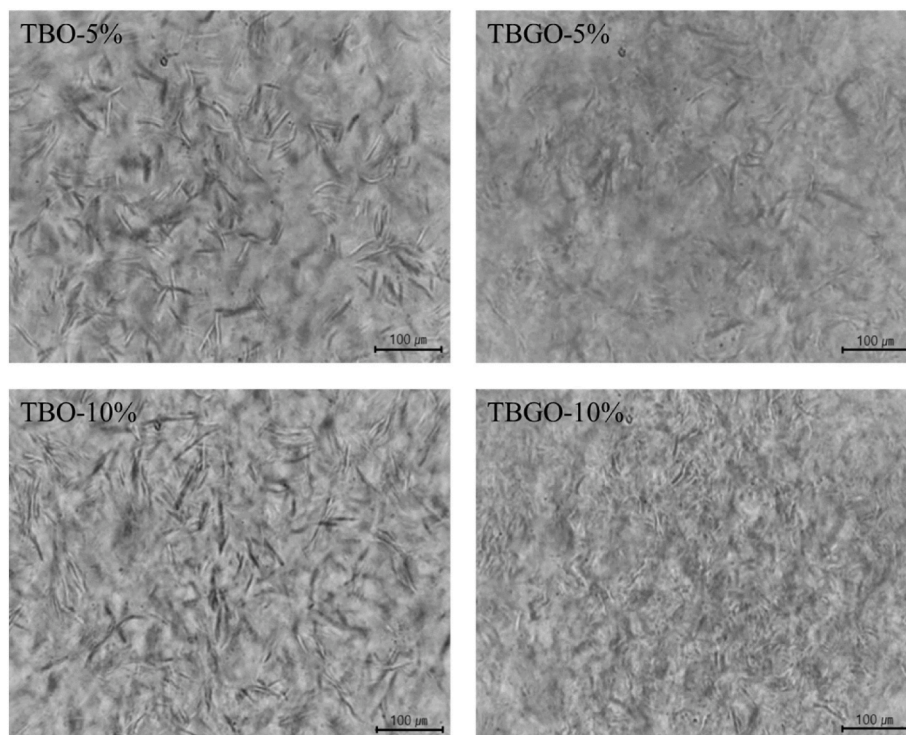**Sohui Jeong:** Conceptualization, Methodology, Formal analysis,

Fig. 7. Microscopic images of TM oil oleogels.

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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