Food Research International 184 (2024) 114213

Contents lists available at ScienceDirect

Food Research International

journal homepage: www.elsevier.com/locate/foodres

Investigation of the influence of minor components and fatty acid profile of oil on properties of beeswax and stearic acid-based oleogels

Subajiny Sivakanthan [a,b,c], Sabrina Fawzia [d], Sagadevan Mundree [e], Terrence Madhujith [f], Azharul Karim [a,*]

[a] School of Mechanical, Medical and Process Engineering, Faculty of Engineering, Queensland University of Technology, Brisbane City, QLD 4000, Australia
[b] Department of Agricultural Chemistry, Faculty of Agriculture, University of Jaffna, Kilinochchi 44000, Sri Lanka[1]
[c] Postgraduate Institute of Agriculture, University of Peradeniya, Peradeniya 20400, Sri Lanka
[d] School of Civil and Environmental Engineering, Faculty of Engineering, Queensland University of Technology, Brisbane City, QLD 4000, Australia
[e] School of Biology and Environmental Science, Faculty of Science, Queensland University of Technology, 2 George St, Brisbane City, QLD 4000, Australia
[f] Department of Food Science and Technology, Faculty of Agriculture, University of Peradeniya, Peradeniya 20400, Sri Lanka

ARTICLE INFO

Keywords:
Minor components
Beeswax
Stearic acid
Fatty acid composition

ABSTRACT

Understanding the impact of minor components and the fatty acid profile of oil on oleogel properties is essential for optimizing their characteristics. Considering the scarcity of literature addressing this aspect, this study aimed to explore the correlation between these factors and the properties of beeswax and stearic acid-based oleogels derived from rice bran oil and sesame oil. Minor oil components were modified by stripping the oil, heating the oil with water, and adding β-sitosterol. Oleogels were then prepared using a mixture of beeswax and stearic acid (3:1, w/w) at a concentration of 11.74 % (w/w). The properties of oils and oleogels were evaluated. The findings indicated that minor components and fatty acid composition of the oils substantially influence the oleogel properties. Removing minor components by stripping resulted in smaller and less uniformly distributed crystals and less oil binding capacity compared to the oleogels prepared from untreated oils. A moderate amount of minor components exhibited a significant influence on oleogel properties. The addition of β-sitosterol did not show any influence on oleogel properties except for the oleogel made from untreated oil blend added with β-sitosterol which had more uniform crystals in the microstructure and demonstrated better rheological stability when stored at 5 °C for two months. The oil composition did not show any influence on the thermal and molecular properties of oleogels. Consequently, the oleogel formulation derived from the untreated oil blend enriched with β-sitosterol was identified as the optimal formula for subsequent development. The findings of this study suggest that the physical and mechanical properties as well as the oxidative stability of beeswax and stearic acid-based oleogels are significantly affected by the minor constituents and fatty acid composition of the oil. Moreover, it demonstrates that the properties of oleogels can be tailored by modifying oil composition by blending different oils.

1. Introduction

In recent years, there has been a surge in academic attention towards oleogels as a potential substitute for traditional solid fats high in *trans* fats and saturated fats. This shift is driven by mounting health concerns associated with the consumption of *trans* fats and saturated fats. Oleogels with gel-like characteristics are produced through oleogelation; an oil structuring technique that uses structurants/ oleogelators to entrap the liquid oil into a 3D gel network formed by the oleogelator (Pinto et al., 2021). Developing oleogels that can meet the requirements of industrial applications is challenging because the properties of oleogels are a function of the interaction effect of many factors such as the composition of oil (triglyceride and minor components) and oleogelators, their interactions, and processing conditions (Doan et al., 2017; Martins et al., 2018; Sun et al., 2022). Edible oils usually consisted of about 95 % triacylglycerols and about 5 % non-triacylglycerols such as partial (mono and di) acylglycerols, free fatty acids, phospholipids, pigments (chlorophylls, and carotenoids), phytosterols, tocopherols,

* Corresponding author.
E-mail addresses: ssubajiny@univ.jfn.ac.lk (S. Sivakanthan), sabrina.fawzia@qut.edu.au (S. Fawzia), sagadevan.mundree@qut.edu.au (S. Mundree), tmadhujith@agri.pdn.ac.lk (T. Madhujith), azharul.karim@qut.edu.au (A. Karim).
[1] Permanent address.

https://doi.org/10.1016/j.foodres.2024.114213
Received 9 November 2023; Received in revised form 20 February 2024; Accepted 10 March 2024
Available online 13 March 2024

tocotrienols, and other phenolic compounds, which are collectively known as minor components (Abad & Shahidi, 2020). The physicochemical properties of the oils are determined by the level of saturation of the oils, positional distribution of the fatty acids in the triacylglycerols, and minor components (Devi & Khatkar, 2016). Thus, while the influence of oil composition and gelator type plays a significant role in determining oleogel properties, it may not be consistent across all oleogel systems.

While there has been substantial scholarly interest in the influence of oleogelator, and processing conditions on oleogel properties, there has been a limited number of studies examining the impact of minor components (non-triglycerides) and fatty acid composition of the oil on these properties (Giacintucci et al., 2018; Scharfe et al., 2019; Scharfe, Niksch, et al., 2022; Scharfe, Prange, et al., 2022a, 2022b; Sun et al., 2022; Zhao et al., 2020). Even though extensive studies have been reported on using beeswax as an oleogelator owing to its excellent gelling abilities (Doan et al., 2018), comprehensive knowledge of the effect of oil composition including the degree of saturation and minor components on the beeswax and stearic acid-based oleogel is limited. Scharfe, Niksch, et al. (2022) studied the influence of minor oil components on the mechanism of gel formation of sunflower and canola oil oleogels using different types of waxes, however, the study did not consider the influence of fatty acid composition. Since the gel network and of different oleogel systems differ due to the interactions, it is important to investigate the influence of oil composition on oleogel properties when a new oleogel system is developed. In this context, the current study examines the impact of minor components and the fatty acid profile of sesame oil and rice bran oil on oleogels based on beeswax and stearic acid, a topic not previously addressed in existing literature.

Moreover, since the oleogelation process involves heating, it is crucial to examine the influence of oleogelation on the natural minor components present in the oil. Minor components present in the oils such as phenolic compounds, tocopherols, and β-carotene possess several health benefits (Uncu & Ozen, 2020). To the best of the authors' knowledge, no previous studies focused on the effect of oleogelation on oil minor components. Therefore, this study aimed to evaluate the changes in natural minor components of oils during oleogelation by comparing the total phenolic content, α-tocopherol content, β-carotene content, and 2,2′-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging activity of the oleogels and respective oils.

In this study, beeswax and stearic acid have been used as a combination at the ratio of 3:1 (11.74 %, w/w), which has been reported to exhibit synergistic effects in tailoring the properties of oleogel prepared from sesame oil and rice bran oil in our previous study (Sivakanthan et al., 2023). Both these oleogelators have been approved by the Food and Drug Administration as food additives. Further, the influence of the addition of β-sitosterol (5 % w/w of the total weight of oleogel) was also evaluated. Several *in-vitro* and *in-vivo* studies evidenced that β-sitosterol possesses a plethora of biological actions and health benefits including antioxidant, lipid-lowering, antimicrobial, antidiabetic, immunomodulatory, anticancer, and anti-inflammatory activities (Babu & Jayaraman, 2020). According to Schedule 25 of the Food Standards Code in Australia and New Zealand, the inclusion of plant sterols in margarine is allowed, provided that the combined amount of saturated and trans fatty acids does not exceed 28 % of the total fatty acid content in the margarine (Food Standards: Australia and New Zealand, 2016).

Hence, the primary objective of this research was to gain a comprehensive understanding of how oil composition and properties affect the characteristics of beeswax and stearic acid-based oleogels, as well as to explore the impact of oleogelation on the natural minor components present in the oils. To achieve this goal, sesame oil and rice bran oil were utilized individually and in various blends to produce oleogels, employing beeswax and stearic acid as the primary oleogelators, with the inclusion of β-sitosterol as an additive. In order to thoroughly investigate the role of minor components and fatty acid composition of oils in determining oleogel properties, several analyses were conducted on the oils, including assessments of viscosity, polarity, acid value, and peroxide value. Furthermore, to investigate further the influence of minor components present in the oils on oleogel properties, modifications were made to the composition of minor components through the addition of β-sitosterol, as well as through processes such as stripping (to remove minor components) and heating the oil with water (to increase the concentration of total polar compounds). Subsequently, the oleogels were subjected for the analysis for oil binding capacity, rheological, thermal, molecular, and microstructural properties, and oxidation behavior. The relationship between the oil composition and properties and properties of oleogels was evaluated through Pearson correlation. Since the properties of oleogels are an interaction effect of several parameters of oils and oleogelators, exploring the correlation between oil composition and oleogel properties is crucial for tailoring oleogel properties to meet specific industrial requirements. The insights gained from this study regarding the influence of oil minor components and fatty acid composition on novel oleogel systems could be highly beneficial in the selection of appropriate oils and additives to achieve the desired physical and mechanical properties of oleogels.

2. Materials and methods

2.1. Materials

Samples of sesame oil (Changs, Thailand) and rice bran oil (Alfa one, USA) were procured from a local grocery store in Brisbane, Australia. Additionally, the following chemicals and reagents were obtained from Sigma Aldrich, Australia: beeswax (refined), stearic acid, β-sitosterol ≥ 70 %, β-carotene pharmaceutical secondary standard, silicic acid for column chromatography (100–200 mesh, 75–150 μm), activated charcoal for column chromatography (~100 mesh), standards for GC–MS (Supelco 37 component FAME mix, linoleic acid methyl ester mix, heneicosanoic acid, methyl nonadecanoate), standard for LC-MS ((±)-α-Tocopherol) and other chemicals, and reagents.

2.2. Methods

This study has been conducted in four steps: 1. preparation of treated oils, 2. characterization of treated and untreated oils, 3. preparation of oleogel from treated and untreated oils, and 4. characterization of oleogels. Details of these steps are provided below. All experiments were performed in triplicates.

2.2.1. Preparation of treated oils

Oleogels were prepared using three kinds of oils such as regular oil (without any treatments), oils heated with water to increase the polar compounds, and oils devoid of non-triglyceride compounds prepared by stripping using column chromatography.

2.2.1.1. Heating oil with water. The oil samples were heated in the presence of water and then water was subsequently removed. The method used in this study was based on the methods reported by Scharfe, Niksch, et al. (2022) and with modifications. The heated oil was prepared in two sets; the first set of oil added with 2 % (w/w) of water (named as T1) was heated for 30 min at 60 °C with shaking at 250 rpm on a magnet stirrer and the second set of oil with 2 % (w/w) of water (named as T2) was heated for 1 h under the same conditions. The heated oil and water mixture was centrifuged at 3900 rpm for 20 min (Eppendorf 5810), and the top oil layer was collected. The traces of water in the oil were removed by drying under vacuum, and the oils were purged with nitrogen and stored at −70 °C until used for the experiments.

2.2.1.2. Stripping. The oils were stripped to remove the minor components according to the method explained by Abuzaytoun and Shahidi

(2006) with minor modifications. A glass chromatographic column (3.4 cm i.d. × 40 cm height) packed sequentially with two adsorbents suspended in n-hexane was used. The first layer comprised 50 g of activated silicic acid, followed by 50 g of activated charcoal in the second layer, and an additional 50 g of activated silicic acid at the top. Fifty grams of oil were mixed with an equal volume of n-hexane and then passed through the chromatographic column. The solvent in the eluent, referred to as the was evaporated under vacuum at 30 °C. Any remaining traces of the solvent were removed by flushing with nitrogen. The stripped oils were collected in 25 mL bottles, which were then purged with nitrogen and stored at −70 °C for future experiments (Abuzaytoun & Shahidi, 2006).

2.2.2. Preparation of oleogel

Oleogels were prepared by direct dispersion of gelators in the oil. Oils and oleogelators (beeswax: stearic acid, 3:1 at the concentration of 11.74 %, w/w) (Sivakanthan et al., 2023) were weighed into the tubes and heated at 80 °C for 10 min in a magnetic stirrer. After heating, the mixture was cooled at room temperature and stored at 20 °C for 48 h before analysis. The composition of oleogels is shown in Table 1.

2.2.3. Characterization of oils/oleogels

The treated and untreated oils were characterized by employing viscosity, polarity, acid value, β-carotene content, and total phenolic content.

2.2.3.1. Viscosity measurement. The viscosity of the oil samples was assessed using a Rheometer (MCR 302, Anton Paar, Austria) equipped with a Peltier system and concentric cylinder. The measurements were conducted with specific dimensions: bob diameter of 26.666 mm, bob length of 40.005 mm, cup diameter of 28.916 mm, active length of 120.2 mm, and positioning length of 72.5 mm. The dynamic viscosity was measured with a gap of 0 mm at a temperature of 25 °C, applying a shear rate ranging from 1 to 100 s^{-1}, and maintaining a constant sample volume of 18 mL.

Table 1
Composition of oleogels (per 100 g).

Oleogel name	Oil type	Oil (g)	Beeswax (g)	Stearic acid (g)	β-Sitosterol (g)
B-O	Blend oil (B)	88	9	3	–
SO-O	Sesame oil (SO)	88	9	3	–
RBO-O	Rice bran oil (RBO)	88	9	3	–
B + BS-O	Blend oil (B)	88	9	3	5
SO + BS-O	Sesame oil (SO)	88	9	3	5
RBO + BS-O	Rice bran oil (RBO)	88	9	3	5
B-S-O	Stripped blend oil (B-S)	88	9	3	–
SO-S-O	Stripped sesame oil (SO-S)	88	9	3	–
RBO-S-O	Stripped rice bran oil (RBO-S)	88	9	3	–
B-S + BS-O	Stripped blend oil (B-S)	88	9	3	5
SO-S + BS-O	Stripped sesame oil (SO-S)	88	9	3	5
RBO-S + BS-O	Stripped rice bran oil (RBO-S)	88	9	3	5
SO-T1–O	Sesame oil heated for 30 min (SO-T1)	88	9	3	–
SO-T2-O	Sesame oil heated for 60 min (SO-T2)	88	9	3	–
RBO-T1–O	Rice bran oil heated for 30 min (RBO-T1)	88	9	3	–
RBO-T2-O	Rice bran oil heated for 60 min (RBO-T2)	88	9	3	–

BS - β-Sitosterol, Blend oil composition - sesame oil: rice bran oil at the ratio of 4:5 (w/w) (Sivakanthan et al., 2023).

2.2.3.2. Polarity determination. The polarity of the oil samples was measured using Testo 270- a deep-frying oil tester (Testo Inc., Germany). The oil was preheated, and the amount of total polar compounds was measured at 45 °C (±4 °C). The values were reported in percentages

2.2.3.3. Fatty acid composition analysis. The fatty acid profile of oil and oleogel samples was determined using gas chromatography. Fatty acid methyl esters (FAMEs) were prepared according to the method by WHO (WHO, 2020). Briefly, the sample (50 mg) and recovery standard (heneicosanoic acid) were taken in a Teflon-lined screw-capped glass test tube and dissolved in toluene (1 mL). Then, 2 mL of BF_3 in methanol (7 % v/v) was added. Tubes were heated at 95 °C for 45 min in a water bath. The tubes were removed from the water bath after 45 min and allowed to cool to room temperature. Then, distilled water (5 mL), hexane (1 mL), and sodium sulfate (1 g) were added and vortexed. FAMEs in the hexane layer were collected and filtered through a 0.22 m PTFE syringe filter. Internal standard (methyl nonadecanoate) was added to the FAME and one microliter of FAME was used to inject into the GC–MS system (Shimadzu GCMS TQ-8040) equipped with a capillary column (Rtx-2330, 60 m × 0.25 mm, 0.20 μm) to identify and quantify the fatty acids. The reference standards such as Supelco 37 component FAME mix, and *cis* and *trans* linoleic acid methyl ester mix were used to identify and quantify the fatty acids using calibration curves. Helium was used as the carrier gas at the flow rate of 1 mL min^{-1}. The inlet temperature was set at 240 °C, with an injection volume of 1.0 μL and a split ratio of 22:1. Initially, the column oven temperature was held at 100 °C for 1 min, followed by a ramp of 10 °C per minute to reach 140 °C, a ramp of 6 °C per minute to reach 175 °C, a ramp of 10 °C per minute to reach 200 °C, and a final ramp of 5 °C per minute to reach 250 °C, where it was held for 4 min. The interface and ion-source temperatures for the GCMS-TQ were maintained at 260 °C and 230 °C, respectively. Mass spectrometric data was collected in the range of 45–600 *m/z* with a detector voltage set at 1150 V. The identification and quantification of detected components were performed using GCMS Solution and LabSolutions Insight software.

2.2.3.4. Acid value determination. The acid value of oils was determined according to AOCS Official Method Cd 3d-63 (AOCS, 2017).

2.2.3.5. Peroxide value determination. The peroxide value of oils was determined according to AOCS Official Method Cd 8–53 (AOCS, 2003) with some modifications. Briefly, 5 ± 0.05 g of sample was weighed into a 250 mL Erlenmeyer flask, and 30 mL of acetic acid: chloroform (3:2, v/v) was added and mixed well to dissolve the sample. Then, 0.5 mL of saturated KI solution was added, stoppered, and left to stand for 1 min in the dark with occasional shaking. Then, 30 mL of distilled water was added and titrated with 0.01 N sodium thiosulfate using the starch indicator. A blank determination also was conducted in parallel. The results were expressed as meq O_2/kg of oil or oleogel.

2.2.3.6. β-Carotene content analysis. β-carotene content of oil/ oleogel samples was assessed as explained by Abad and Shahidi (2020) with some modifications. A solution of hexane and acetone (70:30, v/v) was added to the oil/ oleogel (1:1, w/v) and vortexed for 1 min to dissolve the sample completely. Then the mixture was filtered through a 0.22 μm PTFE syringe filter and 200 μL of the solution was transferred to the 96-well plate and the absorbance was read at 430 nm in a plate reader (Synergy HTX multi-mode reader). The content of β-carotene was quantified from a standard curve of β-carotene (β-carotene pharmaceutical secondary standard) prepared under the same condition.

2.2.3.7. Phenolic compounds analysis

2.2.3.7.1. Extraction. The phenolic compounds in oils and oleogels were extracted by liquid–liquid extraction as explained by Antonini et al. (2015) and Abuzaytoun and Shahidi (2006) with some modifications. Five grams of oil was diluted with hexane (1:1, w/v) and methanol (8:2, v/v, methanol/water), vortexed, and then, kept in an ultrasonicator bath (Elma Elmasonic P, Germany) at 37 Hz for 15 min. Then the contents were centrifuged at 3900 rpm for 10 min (Eppendorf 5810) to collect the supernatant. The operations were repeated twice, and all supernatants were pooled. The collected supernatant was filtered using a 0.22 μm PTFE syringe filter. The supernatants were used for the total phenolic content analysis by Folin and Ciocalteu's method and radical scavenging activity by the 2,2′-diphenyl-1-picrylhydrazyl (DPPH) method as explained below.

2.2.3.7.2. Determination of total phenolic content. Folin and Ciocalteu's method: The quantification of total phenolics was conducted following the procedure outlined by Singleton and Rossi (1965) with some modifications. Ten microliters of extract were added to the Eppendorf tube containing 790 μL of Milli-Q water. Then 100 μL of Folin and Ciocalteu's reagent was added to the tube. The contents were mixed and the tube was left to stand for 5 mins at room temperature followed by the addition of 150 μL of 20 % sodium carbonate. The contents were thoroughly mixed, and the tube was left to incubate in the dark at room temperature (20 °C) for 2 h. Then, 200 μL of aliquots were plated triplicates onto a 96-well plate and the absorbance was read at 765 nm in a plate reader (Synergy HTX multi-mode reader). The phenolic content was determined using a gallic acid standard curve and reported in milligrams of gallic acid equivalents (GAE) per gram of extract. Each measurement was conducted in triplicate.

2.2.3.7.3. Determination of DPPH radical scavenging activity. A hundred microliter of the methanolic extract was taken in a 96-well plate and 100 μL of DPPH working solution (0.1 mM in methanol) was added. After a 30 min of reaction at room temperature (20 °C) under dark conditions, the absorbance of the mixture was measured at 517 nm in a plate reader (Synergy HTX multi-mode reader) using methanol instead of the sample as the blank (Blois, 1958). Inhibition of free radical DPPH as a percentage was calculated as follows.

$$\text{Inhibition } (\%) = (Abs_{blank} - Abs_{sample}) / A_{blank} \times 100$$

Where Abs_{blank} is the absorbance of the blank and Abs_{sample} is the absorbance of the test compound.

2.2.3.8. α-Tocopherol content analysis by LC-MS. α-Tocopherol content of oils and oleogels was analyzed using the UHPLC system as explained by Zaunschirm et al. (2018) with some modifications. Fifty milligrams of oil/ oleogel sample was dissolved thoroughly in 2 mL isopropyl alcohol and subsequently, it was filtered using a 0.22 μm PTFE syringe filter. Analysis was conducted by injecting 5 μL of sample into a UHPLC system (Dionex Ultimate 3000, Thermo Fisher Scientific, Austria) equipped with a C18 column (Kinetex EVO, 100 × 2.1 mm, 2.6 μm, Phenomenex). The mobile phase consisted of a mixture of methanol and water with a flow rate of 0.3 mL/min. A gradient elution was employed, initiating with 75 % methanol and 25 % LCMS grade water (LiChrosolv® water), gradually transitioning to 100 % methanol over 5 min, and maintaining this composition for 1.5 min. The initial conditions were reached again at the 7 min. α-Tocopherol was detected at a wavelength of 295 nm. α-Tocopherol reference standard was used to detect and confirm the peak. Data acquisition and processing were conducted using Thermo Xcalibur 3.0.63.3 software (Thermo Fisher Scientific Inc.). To quantify α-tocopherol, an external calibration curve of α-tocopherol ranging from 0.3 to 300 ppm was employed.

2.2.3.9. Oxidation induction temperature analysis. The oxidation induction temperature of the oil/ oleogels was assessed using thermogravimetric analysis (TGA) curves obtained from a dynamic (non-isothermal) heating experiment using a Simultaneous Thermal Analyzer (Jupiter STA 449 F3, Netzsch, Germany) as explained by Sivakanthan et al. (2023).

2.2.3.10. Microscopic analysis. The microscopic images of the oleogels were acquired using Nikon Eclipse LV100ND polarized light microscope equipped with a digital camera (Nikon DS-Fi2). A drop of the molten sample was placed onto a preheated glass slide and a cover slip was put on. It was then stored for 48 h at 20 °C prior to imaging. Bright-field images were captured at a magnification of 200× at 20 °C. The images were processed using ImageJ software (ImageJ 1.53e; Java 1.8.0_172, National Institutes of Health, USA). The images were converted into 8-bit grayscale images to measure the distance between crystals and the length of the crystals. Fractal dimension was obtained using the tool "fractal box counting" after converting the images to 8-bit binary images and default box sizes 2,3,4,6,8,12,16,32, and 64 with black background. Since the crystals were needle and platelet-shaped, the distance between the crystals was measured by drawing the lines between two crystals from the middle of the crystals. Further, since the crystals were arranged in both parallel and perpendicular directions to the cover glass, only parallel crystals were considered for measurements to get more accurate data.

2.2.3.11. Oil binding capacity determination. The oil binding capacity of the oleogels was determined as explained by Sivakanthan et al. (2023).

2.2.3.12. Rheological measurements. The rheological analysis (amplitude sweep, frequency sweep, and thixotropy) of the oleogels was performed according to Sivakanthan et al. (2023) using Anton Paar MCR302 Rheometer (Austria) with the TruStrain™ option, analysis software: RheoCompass™ (version 1.30.999.) and a Peltier temperature control unit. Sand-blasted parallel plate geometry (diameter of 50 mm, PP50-S) was used.

2.2.3.13. Thermal analysis. The melting and crystallization parameters were determined as explained by Sivakanthan et al. (2023) using Differential Scanning Calorimetry (DSC) (DSC 204 F1 Phoenix, Netzsch, Germany). Briefly, the samples (10 ± 1 mg) in sealed aluminium crucibles were subjected to heating and cooling cycles under a nitrogen atmosphere. Briefly, cooling was performed from 85 °C to 0 °C at the rate of 2 °C/min, keeping isothermally at 0 °C, and heating was performed from 0 °C to 85 °C at the rate of 5 °C/min.

2.2.3.14. Fourier transform infrared spectroscopy (FTIR) analysis. The molecular interactions between beeswax and stearic acid were analyzed using FTIR spectra of the samples recorded over the range of 4000 to 400 cm^{-1} using a Fourier transform infrared spectrometer (Nicolet iS50 FT-IR, Thermo Scientific, USA) at a resolution of 4 cm^{-1}. A total of 64 scans were collected at room temperature (20 °C).

2.2.4. Storage stability study

Storage stability of the oleogels at 5 °C and 20 °C was evaluated in terms of rheological properties. For this purpose, the samples were stored at 5 °C and 20 °C for two months and analyzed by amplitude sweep experiments as already explained under the section – rheological properties.

2.2.5. Statistical analysis

The statistical analyses were conducted using Minitab 21.1 (Minitab, LLC, USA). All values were presented as mean ± standard deviation. To assess the statistical significance, a one-way ANOVA and Tukey's test were employed at a significance level of 95 % ($p < 0.05$). Additionally, the correlation between different parameters was examined using the Pearson correlation method at a 95 % confidence interval.

3. Results and discussion

To acquire a clear insight into the oil composition on oleogel properties, oleogels were prepared from untreated and treated rice bran oil and sesame oil as a single oil and as a blend of both oils as determined in our previous study (Sivakanthan et al., 2023). Further, the influence of the addition of β-sitosterol (5 % w/w of the total weight of oleogel) was also evaluated.

3.1. Composition and properties of oils

Recent studies have reported that the composition of oil influences the properties of oleogels (Giacintucci et al., 2018; Scharfe et al., 2019; Scharfe, Niksch, et al., 2022; Scharfe, Prange, et al., 2022a, 2022b; Sun et al., 2022; Zhao et al., 2020). Since the composition of oil influences the physical and chemical properties of the oils, it is crucial to evaluate their physical and chemical properties. Table 2 shows the physicochemical characteristics and fatty acid composition of untreated and treated oils. Stripping and heating of oils were performed to change the composition of minor oil components by removal and addition of minor components, respectively to examine the influence of minor components on the properties of oleogels.

Stripping of oils by chromatography removes non-triglyceride components (minor components) such as free fatty acids, monoacylglycerols, diacylglycerols, pigments (carotenoids), tocopherols, and phospholipids (Abad & Shahidi, 2020). Heating the oil in the presence of water leads to the degradation of oil via different processes such as hydrolysis, oxidation, and polymerization (Khor et al., 2019; Tsai et al., 2023). As oil degrades, the acid value increases due to the formation of free fatty acids through the hydrolysis of triglycerides, the oxidative degradation of oil leads to the generation of peroxides, increasing peroxide value, and as the degradation progresses further, various polar compounds are formed, contributing to the increase in total polar compounds. Polar compounds such as free fatty acids, monoglycerides, diglycerides, and polymeric triglycerides are generated in the oils during hydrolysis, oxidation, and polymerization (Flores et al., 2021).

The results shown in Table 2 indicate that both stripping and heating with water significantly altered the properties of the oils as determined in terms of acid value, peroxide value, polarity, and viscosity. Similar observations have been reported by Scharfe, Prange, et al. (2022a) the canola oil, sunflower oil, and flaxseed oil. The stripped oils had the lowest peroxide value, acid value, and total polar compounds, which indicates that stripping has removed the free fatty acids, peroxides, and polar compounds effectively. An increase in peroxide value, acid value, and total polar compounds was observed in all heated oil samples indicating degradation of oils during heating the oil with water. Prolonged heating of the oil and the presence of water could have accelerated hydrolysis, oxidation, and polymerization (Sun et al., 2022). The acid value of rice bran oil was higher than that of sesame oil after heating, which shows that the sesame oil has undergone less degradation than rice bran oil. However, there were no significant differences in the peroxide values of these two oils heated for the same time. The total polar compounds of treated and untreated rice bran oil were significantly higher than that of corresponding sesame oil. The higher total polar compounds in rice bran oil could be attributed to the higher amount of initial polar components and a higher degree of hydrolysis and formation of free fatty acids compared to sesame oil. Furthermore, the total polar compounds showed a consistent rise with extended heating duration leading to pronounced degree of hydrolysis, and the formation of free fatty acids, when compared to sesame oil.

Untreated rice bran oil showed significantly higher viscosity compared to untreated sesame oil. Viscosity is related to saturated and unsaturated fatty acid content (Valantina et al., 2016). A higher degree of unsaturation is related to the lower viscosity of the oil (Scharfe, Prange, et al., 2022a). Since the degree of unsaturation of sesame oil is higher than that of rice bran oil, sesame oil had lower viscosity than rice bran oil. Stripping has significantly reduced the viscosity of both oils, whereas heating did not influence the viscosity of the oils. Scharfe, Niksch, et al. (2022) also reported a reduction in the viscosity of canola oil and sunflower oil after stripping. Untreated sesame oil had higher

Table 2
The physicochemical properties and fatty acid composition of the oils.

Oil	AV (mg KOH/ g of oil)	PV (meq O_2/kg)	Polarity (%)	Viscosity (mPa.s)	BC (mg/ kg)	Tocopherol (mg/kg)	TPC (mg/ kg)	DPPH inhibition (%)	OIT (°C)	Fatty acid group		
										SFA (%)	MUFA (%)	PUFA (%)
B	1.12 ±0.08^{de}	6.66 ±0.15^{c}	11.50 ±0.14^{e}	60.25 ±0.032^{b}	6.72 ±0.56^{b}	1217.23 ±63.95^{a}	35.20 ±3.42^{b}	66.42 ±1.06^{c}	336.90 ±0.14^{bc}	21.90 ±0.35^{a}	38.75 ±0.03^{b}	39.40 ±0.43^{bc}
SO	0.91 ±0.11^{e}	6.03 ±0.87^{c}	8.30 ±0.14^{f}	52.26 ±0.106^{c}	11.10 ±0.39^{a}	1115.15 ±37.45^{a}	65.62 ±3.08^{a}	75.56 ±0.26^{a}	339.90 ±0.42^{a}	16.67 ±0.56^{b}	40.74 ±0.95^{ab}	42.64 ±0.34^{a}
RBO	1.22 ±0.05^{de}	5.98 ±0.38^{c}	12.35 ±0.07^{d}	69.29 ±0.035^{a}	2.78 ±0.07^{c}	1168.50 ±91.03^{a}	20.17 ±3.46^{c}	31.90 ±0.23^{e}	331.10 ±0.42^{ab}	22.40 ±1.05^{a}	42.05 ±0.95^{ab}	35.15 ±1.34^{de}
B-S	<0.001 ±0.00^{f}	0.03 ±0.01^{d}	<1^{g}	26.18 ±0.014^{d}	0.70 ±0.01^{d}	70.96 ±6.47^{c}	6.88 ±0.11^{d}	0.56 ±0.21 ^{g}	324.75 ±0.78 ^{cd}	23.30 ±0.85^{a}	38.68 ±0.29^{b}	38.02 ±0.54 ^{cd}
SO-S	<0.001 ±0.00^{f}	0.02 ±0.01^{d}	<1^{g}	26.54 ±0.092^{d}	0.74 ±0.03^{d}	65.63 ±5.01^{c}	9.84 ±1.55^{d}	1.31 ±0.25 ^{g}	326.85 ±0.49 ^{cd}	16.63 ±1.48^{b}	39.89 ±1.01^{ab}	43.48 ±0.51^{a}
RBO-S	<0.001 ±0.00^{f}	0.02 ±0.00^{d}	<1^{g}	22.67 ±0.014^{e}	0.86 ±0.01^{d}	69.55 ±11.64^{c}	5.41 ±0.01^{d}	0.56 ±0.12 ^{g}	325.70 ±0.28 ^{cd}	21.34 ±0.74^{a}	41.55 ±0.25^{ab}	36.96 ±0.23 ^{cd}
SO-T1	1.40 ±0.08^{d}	9.86 ±0.10^{b}	11.10 ±0.14^{e}	52.26 ±0.025^{c}	10.88 ±0.18^{a}	567.09±8.10^{b}	62.13 ±0.01^{a}	71.64 ±0.09^{b}	329.40 ±1.13^{c}	14.98 ±0.68^{b}	43.50 ±2.12^{a}	41.55 ±1.48^{ab}
SO-T2	2.64 ±0.02^{c}	13.53 ±0.20^{a}	42.05 ±0.35^{b}	51.97 ±0.02^{c}	11.07 ±0.45^{a}	511.02 ±4.71^{b}	62.08 ±0.36^{a}	60.26 ±1.85^{d}	321.60 ±2.69^{de}	14.05 ±0.35^{b}	41.65 ±0.49^{ab}	43.90 ±0.42^{a}
RBO-T1	3.79 ±0.23^{b}	10.86 ±0.03^{b}	15.15 ±0.07^{c}	69.17 ±0.014^{a}	2.68 ±0.14^{c}	637.50 ±13.02^{b}	18.52 ±0.01^{c}	31.53 ±0.79^{e}	317.45 ±4.03^{e}	22.82 ±0.45^{a}	41.96 ±1.07^{ab}	33.85 ±0.49^{e}
RBO-T2	4.79 ±0.08^{a}	13.33 ±0.27^{a}	48.40 ±0.28^{a}	69.68 ±0.02^{a}	2.75 ±0.05^{c}	517.35 ±22.71^{b}	17.09 ±1.54^{c}	20.90 ±1.06^{f}	316.95 ±0.35^{e}	23.31 ±0.44^{a}	42.70 ±0.42^{a}	34.00 ±0.03^{e}

Different superscript letters (a-g) in the same column show a significant difference ($p < 0.05$). AV – Acid value; BC – β- Carotene; OIT – oxidation induction temperature; PV – peroxide value; TPC – total phenolic content, SFA – saturated fatty acids; MUFA – monounsaturated fatty acids; PUFA – polyunsaturated fatty acids

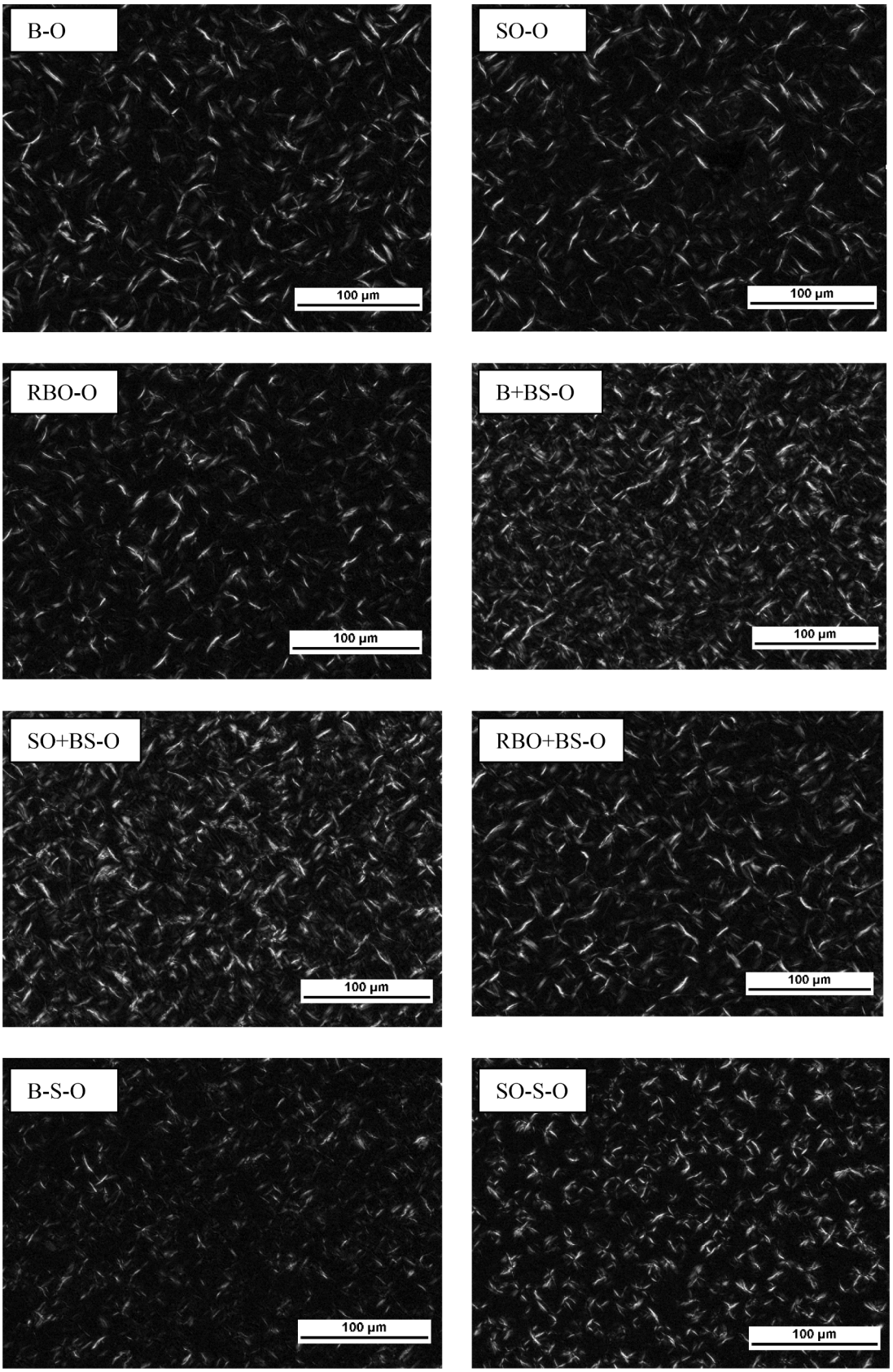

Fig. 1. Bright field polarized microscopy images of oleogels. Images were acquired at magnification 200× at 20 °C. Scale bar: 100 μm.

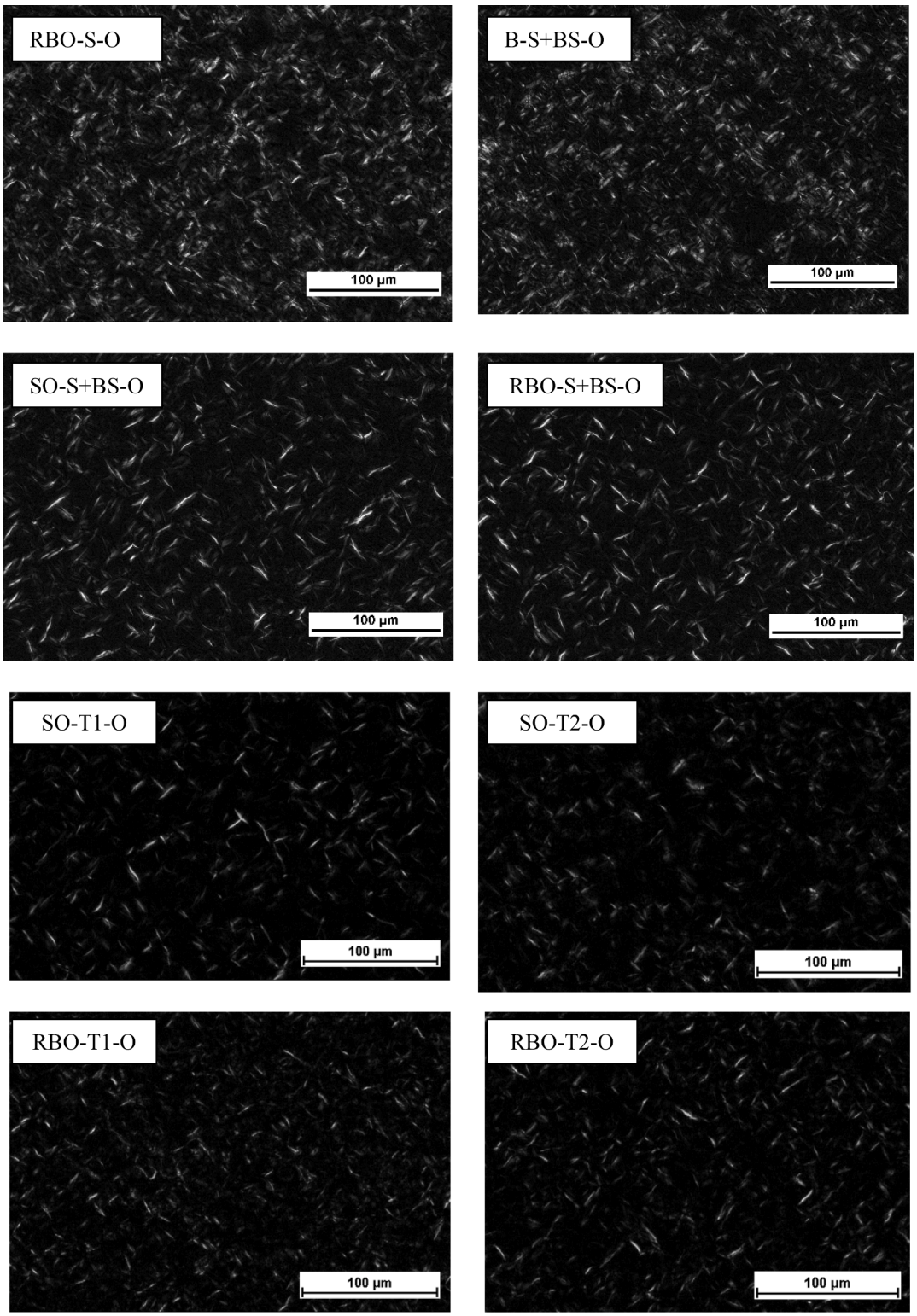

Fig. 1. (*continued*).

β-carotene content, total phenolic content, and DPPH scavenging activity than those of untreated rice bran oil whereas α-tocopherol content (ions were detected at *m/z* 429.372, $C_{29}H4_9O_2^+$ $[M + H\text{-}2H]^+$, *m/z* 430.378, $C_{29}H_{49}O_2^{+\cdot}$ $M^{+\cdot}$) (Supplemental file - Fig. 1) (Fu et al., 2021) of both untreated rice bran oil and sesame oil were not significantly different. Heating did not significantly change the concentration of both β-carotene and total phenolic content of both oils; however, the α-tocopherol content and DPPH activity showed a significant reduction after heating as indicated by the correlation analysis which showed a significant positive correlation between α-tocopherol content and DPPH activity (Table 5). The reduction in the DPPH activity could be due to the degradation of tocopherols or upon prolonged heating (Kmiecik et al., 2019). Stripping removed more than 75 % of the total phenolics and more than 90 % of the β-carotene from the oils. The saturated, monounsaturated, and polyunsaturated fatty acid compositions of sesame oil and rice bran oil were significantly different (Table 2). Sesame oil

contained significantly higher unsaturated fatty acid content than rice bran oil, whereas rice bran oil contained significantly higher saturated fatty acid content than sesame oil. Oil treatments did not have any significant influence on the fatty acid composition of the oils. The following sections explain how the oil composition influenced the properties of oleogels developed from sesame oil and rice bran oil using a synergistic mixture of beeswax and stearic acid as oleogelators.

3.2. Microstructure

Microscopic images of the oleogels were acquired using polarized light microscopy to assess the influence of fatty acid composition and minor components on the microstructure of oleogels. Polarized light microscopy images of the oleogels and the fractal dimension, length of crystals, and the distance between crystals are shown in Fig. 1 and Table 3, respectively.

As confirmed by Blake and Marangoni (2015) through scanning electron microscopy, all samples exhibited needle-like crystals, which are noted to be the 2D representation of platelet-like crystals. The uniformity of crystal mass distribution in the oleogels can be quantitatively assessed by calculating the fractal dimension (D) using the box-counting method (Scharfe, Niksch, et al., 2022). The higher the fractal dimension, the more uniform distribution of the mass with less cavities in the network (Frolova et al., 2022). Significant differences in the fractal dimension, average length, and distance between crystals were observed. Significantly higher fractal dimension values were reported for all oleogels made from untreated oils without β-sitosterol (B-O, SO-O, and RBO-O), and the oleogel made from untreated oil blend added with β-sitosterol (B + BS-O), which suggests more uniform arrangement of the crystals and less cavities compared to the other oleogels. More uniform distribution of the crystals in the oleogel made from untreated oil blend added with β-sitosterol (B + BS-O) compared to the oleogels made from either untreated sesame oil or untreated rice bran oil added with β-sitosterol could be due to the synergistic interactions between the composition of the oil blend, oleogelators, and β-sitosterol. However, the exact mechanism behind this effect could not be interpreted based on the results of this study. Further analysis of this oleogel using X-ray diffraction analysis for the type and arrangement of crystals in the gel structure would unravel the mechanism behind this interaction. Oleogels made from stripped oil regardless of the addition of β-sitosterol (B-S-O, SO-S-O, RBO-S-O, B-S + BS-O, SO-S + BS-O, and RBO-S + BS-O) and untreated sesame oil and rice bran oil added with β-sitosterol (SO + BS-O and RBO + BS-O) contained more irregular shaped crystals with less uniform arrangement. Even though the oleogels made using stripped oils contained smaller-sized crystals compared to other oleogels, the removal of oil minor components resulted in an irregular arrangement of the crystals in the gel network. The fractal dimensions reported in this study are in line with the values reported by Scharfe, Niksch, et al. (2022). Moreover, Scharfe, Niksch, et al. (2022) also reported that oleogels produced from stripped canola oil showed smaller crystals than the oleogels produced from untreated oils and the fractal dimension was highest for the samples produced from untreated oil. The oleogels produced from heated sesame oil and heated rice bran oil had significantly lower fractal dimensions than the oleogels made from respective untreated oils. This could be attributed to the negative influence of the high level of polar components on crystal arrangement. Therefore, the more uniform arrangements of the crystals in the oleogels produced from untreated oils could be attributed to the role of a moderate level of minor components in producing a more uniform microstructure. The observations of the present study clearly indicate that the natural minor components present in the oils played a crucial role in the microstructure of the oleogels, which in turn influenced the macrostructural properties of the oleogels. As per the results of the correlation analysis (Table 5), the fractal dimension of the oleogels had a significant correlation with the total polar compounds and viscosity of the oil, which indicates that the spatial arrangement of the crystals in the gel network is significantly influenced by the minor components of the oils. Further, the fractal dimension showed a significant positive correlation with other parameters of the oleogels such as structure recovery and oil binding capacity. The fatty acid composition of the oils did not have a notable influence on the microstructure of the oleogels. The length of the crystals and the distance between the crystals showed deviations between the measurements of the same sample since the crystals were oriented in different directions which could have led to manual errors in drawing the lines between the crystals. Therefore, the role of oil composition on the length of the crystals and the distance between crystals was not clear from the analysis of the results of this study. Scharfe, Niksch, et al. (2022) also noted that the impact of minor components on crystal sizes was inconclusive, possibly due to the presence of very small crystals that were difficult to differentiate from one another or due to poor contrast between the crystals and the background.

Table 3
Fractal dimension (D), length of the crystals, and distance between crystals as analyzed by ImageJ.

Sample	Fractal dimension (D)	Length of the crystals (μm)	Distance between crystals (μm)
B-O	1.968 ± 0.002^{a}	15.19 ± 2.50^{ab}	9.92 ± 4.48^{abcd}
SO-O	1.967 ± 0.001^{a}	15.79 ± 2.34^{a}	9.48 ± 3.80^{abcd}
RBO-O	1.965 ± 0.004^{a}	13.69 ± 2.64^{bc}	9.08 ± 3.14^{abcd}
B + BS-O	1.964 ± 0.004^{a}	12.61 ± 2.59^{cde}	7.69 ± 2.30^{cd}
SO + BS-O	1.229 ± 0.001^{h}	12.43 ± 2.42^{cde}	8.75 ± 3.30^{bcd}
RBO + BS-O	1.248 ± 0.011^{g}	12.89 ± 2.22^{cd}	10.37 ± 3.70^{abcd}
B-S-O	1.191 ± 0.010^{i}	8.40 ± 1.96^{h}	8.51 ± 2.90^{bcd}
SO-S-O	1.273 ± 0.002^{f}	9.64 ± 2.06^{gh}	11.28 ± 4.99^{a}
RBO-S-O	1.370 ± 0.019^{c}	9.57 ± 1.78^{gh}	9.14 ± 3.32^{abcd}
B-S + BS-O	1.413 ± 0.002^{b}	8.54 ± 2.27^{h}	7.80 ± 2.83^{d}
SO-S + BS-O	1.173 ± 0.015^{j}	11.76 ± 2.85^{def}	9.12 ± 3.41^{abcd}
RBO-S + BS-O	1.137 ± 0.002^{k}	11.03 ± 3.12^{efg}	10.87 ± 4.43^{ab}
SO-T1–O	1.090 ± 0.000^{l}	10.76 ± 1.54^{defg}	10.29 ± 3.37^{abcd}
SO-T2-O	1.326 ± 0.003^{d}	9.69 ± 1.94^{gh}	9.69 ± 1.94^{abcd}
RBO-T1–O	1.291 ± 0.002^{e}	10.35 ± 2.53^{fgh}	10.94 ± 3.07^{abcd}
RBO-T2-O	1.185 ± 0.005^{i}	11.55 ± 2.89^{defg}	10.71 ± 2.66^{abcd}

Different superscript letters (a-l) in the same column show a significant difference ($p < 0.05$).

3.3. Oil binding capacity

The oil binding capacity of the oleogel is the ability of the gel structure to retain the oil after being subjected to an external force (Flöter et al., 2021). The oil binding capacity of oleogels prepared using different oils is shown in Table 4. The oleogels prepared from stripped oils either with β-sitosterol or without β-sitosterol had lower oil binding capacity than the oleogel prepared using untreated oils. Oil binding capacity depends primarily on the microstructure of the gel network such as spatial distribution of mass and crystal shape and size (Ghazani et al., 2022; Manzoor et al., 2022).

A well-connected and homogenous gel network can better retain oil. The results of oil binding capacity are in line with the analysis of the microstructure of the oleogels. As already explained, the oleogels made from stripped oils showed irregular arrangements of the crystals with empty cavities, which led to the lower oil-holding capacity of the gels (Blake et al., 2014). A similar observation of less ordered crystal arrangement and poor oil binding capacity of the oleogels has been reported (Frolova et al., 2022). Significantly lower oil binding capacities of oleogels prepared using stripped oils indicate that the stripping has a negative influence on gel structure. This effect has been further confirmed by the Pearson correlation analysis (Table 5). The oil binding capacity of the oleogels showed a strong positive correlation with total phenolic content, DPPH scavenging activity, and β-carotene content of

Table 4
OBC, LVR, **G′** at LVR, loss factor, and structure recovery of oleogels.

Oleogel sample label	OBC (%)	LVR (%)	G′ at LVR (Pa)	Loss factor	Structure recovery (%)
B-O	99.83 ± 0.13^{a}	0.052 ± 0.0025^{a}	110,125 ± 784^{abcd}	0.141 ± 0.001^{bc}	33.26 ± 1.83^{a}
SO-O	99.82 ± 0.11^{a}	0.050 ± 0.0014^{a}	129,785 ± 2,326^{ab}	0.129 ± 0.001^{de}	26.67 ± 0.55^{c}
RBO-O	99.80 ± 0.05^{a}	0.050 ± 0.0020 ^{a}	93,945 ± 6,738 ^{cd}	0.149 ± 0.001^{ab}	32.13 ± 2.27^{ab}
B + BS-O	99.76 ± 0.27^{a}	0.050 ± 0.0057^{a}	109,541 ± 4,014^{abcd}	0.142 ± 0.003^{bc}	32.55 ± 1.88^{a}
SO + BS-O	99.97 ± 0.03^{a}	0.050 ± 0.0039^{a}	123,665 ± 7,502^{ab}	0.128 ± 0.005^{e}	25.63 ± 0.78^{c}
RBO + BS-O	99.92 ± 0.10^{a}	0.050 ± 0.0016^{a}	89,067 ± 1,21^{d}	0.156 ± 0.001^{a}	26.70 ± 0.47^{c}
B-S-O	99.23 ± 0.21^{a}	0.047 ± 0.0014^{a}	110,060 ± 6,236^{abcd}	0.143 ± 0.001^{bc}	31.24 ± 0.68^{ab}
SO-S-O	99.15 ± 0.11^{b}	0.047 ± 0.0057^{a}	119,515 ± 1,421^{abc}	0.123 ± 0.002^{e}	26.92 ± 0.33^{c}
RBO-S-O	98.68 ± 0.20^{b}	0.044 ± 0.0019^{a}	91,707 ± 9,549^{d}	0.132 ± 0.000^{cde}	27.95 ± 0.04^{bc}
B-S + BS-O	98.95 ± 0.16^{b}	0.048 ± 0.0053^{a}	104,715 ± 9,284^{bcd}	0.141 ± 0.005^{bcd}	29.14 ± 0.91^{abc}
SO-S + BS -O	98.96 ± 0.15^{b}	0.047 ± 0.0058^{a}	126,755 ± 912^{ab}	0.126 ± 0.001^{e}	29.03 ± 0.62^{abc}
RBO-S + BS-O	99.11 ± 0.45^{b}	0.047 ± 0.0017^{a}	85,635 ± 7,947^{d}	0.147 ± 0.002^{ab}	29.39 ± 0.36^{abc}
SO-T1–O	99.32 ± 0.36^{a}	0.050 ± 0.0011^{a}	134,700 ± 6,731^{a}	0.122 ± 0.005^{e}	25.72 ± 0.83^{c}
SO-T2-O	99.25 ± 0.15^{a}	0.050 ± 0.0024^{a}	130,830 ± 2,94^{ab}	0.125 ± 0.007^{e}	26.42 ± 1.42^{c}
RBO-T1–O	99.63 ± 0.27^{a}	0.049 ± 0.0004^{a}	123,400 ± 10,154^{ab}	0.148 ± 0.001^{ab}	26.82 ± 0.24^{c}
RBO-T2-O	99.43 ± 0.04^{a}	0.051 ± 0.0007^{a}	120,485 ± 13,116^{abc}	0.148 ± 0.001^{ab}	26.68 ± 0.84^{c}

Different superscript letters (a-e) in the same column show a significant difference ($p < 0.05$). OBC – Oil binding capacity.

the oil. Moreover, the oil binding capacity exhibited strong positive correlations with the viscosity of the oils which is significantly reduced after stripping and fractal dimension. Even though the viscosity of untreated sesame oil and rice bran oil differed significantly, the oil binding capacities of oleogels made using untreated sesame oil, rice bran oil, and their blends did not differ significantly. The higher fractal dimension of the oleogels resulted in a higher oil binding capacity indicating that more evenly distributed crystals can hold more oil than less-uniformly arranged crystals (Manzoor et al., 2022). Therefore, it could be concluded that the major reason for the differences in the oil binding capacities of oleogels made from untreated and treated oils is the minor components. The incorporation of β-sitosterol did not show a significant influence on the oil binding capacity of the oleogels.

3.4. Rheological properties

Rheological analysis of oleogels was performed to gain insight into the performance of oleogels under different mechanical conditions. Rheological properties were analyzed by amplitude sweep, frequency sweep, and thixotropy. Amplitude sweep experiments are used to examine the behavior of oleogels with increasing shear stress. Storage modulus (G′) and loss modulus (G″) as a function of shear stress are used to determine the linear viscoelastic range (LVR). The amplitude sweeps provide detailed information about network properties such as breakdown characteristics of the gel. In a typical amplitude sweep of an oleogel, there is an initial phase where both the storage modulus (G′) and the loss modulus (G″) remain relatively constant at low stress, indicating a stable gel structure. However, as stress increases, the ability of the gel to store deformation energy diminishes, leading to a decrease in G′. At a certain point, the viscous behavior of the gel becomes predominant, causing the sample to flow when G′ equals G″ (known as the flow point). The maximum value of G′ within the linear viscoelastic region (LVR) serves as an indicator of the strength of the gel strength (Scharfe, Prange, et al., 2022a).

According to the data provided in Table 4, and Fig. 2, there were significant differences in rheological properties between the samples. According to the information obtained from amplitude sweeps, there were no significant differences between the samples for LVR. This indicates that all oleogels have similar sensitivity to the applied stress. However, other parameters such as G′ at LVR, loss factor, and thixotropy were significantly different. A similar observation of non-significant differences in LVR and significant differences in the gel strength has been reported by Sun et al. (2022) for the monoglyceride oleogels developed from refined and unrefined walnut oils. G′ at LVR represents the elastic property of oleogels. A higher G′ value indicates a stronger gel structure and high deformation resistance (Doan et al., 2015). In the present study, oleogels made from both untreated and stripped blend oil, and sesame oil as well as the oleogels made from both rice bran oil and sesame oil heated with water had significantly higher G′ at LVR compared to the oleogels made from untreated and stripped rice bran oil. These results indicate that the oil type and very high polar compounds had a significant influence on the gel strength, whereas stripping did not have a significant effect.

The influence of oil type on gel strength could be correlated to the degree of saturation and the viscosity of the oils. The correlation analysis exhibited a significant strong positive correlation of G′ at LVR with peroxide value, total phenolic content, β-carotene content, and fatty acid composition of the oils. Sesame oil has a higher degree of unsaturation and lower viscosity compared to rice bran oil. The lower viscosity of the sesame oil enables a higher diffusion rate of the oleogelators (Scharfe, Prange, et al., 2022a) compared to rice bran oil. Scharfe, Niksch, et al. (2022) also reported that more unsaturated oil produced harder gels compared to less unsaturated oils for rice bran wax-based oleogels. Similarly, Yu et al. (2020), Yang et al. (2018), and Calligaris et al. (2014) also reported that the firmness of the oleogel is dependent on the fatty acid composition and viscosity of the oil and oils characterized by a higher degree of unsaturation and lower viscosity tended to yield firmer oleogels.

Significantly higher G′ at LVR of oleogels made from rice bran oil heated with water (RBO-T1–O and RBO-T2-O) compared to the oleogel made from the untreated rice bran oil (RBO-O and RBO + BS-O) and stripped rice bran oil (RBO-S-O and RBO-S + BS-O) could be attributed to the high concentration of polar compounds. Scharfe, Niksch, et al. (2022) also found out that the gels become harder with increasing polar compounds for the beeswax oleogels. The polar compounds could influence the gelling ability of wax and the resulting strength of oleogel due to the interactions with wax components, modifying the interactions between the crystals and increasing the solubility of wax (Scharfe, Niksch, et al., 2022). Further, these results did not show any significant influence on oleogel properties due to the addition of β-sitosterol.

The loss factor (also known as the loss tangent or tan δ) is a dimensionless quantity defined as the ratio of the G″ to the G′ of a material and it is a measure of energy dissipation during deformation. For oleogels, a low loss factor indicates that the material is resistant to flow and possesses a stable gel network. On the other hand, a high loss factor suggests that the gel structure is less stable, and the oleogel may experience flow or deformation more easily. The loss factor of oleogels can influence the sensory attributes of products. For instance, in the food industry, the texture and mouthfeel of products can be tuned by controlling the loss factor of oleogels. Lower loss factors may result in a creamier, more solid texture, while higher loss factors may lead to a more spreadable and softer texture. All oleogels made using rice bran oil

Table 5
Pearson correlations (r) between the parameters.

Parameter	Structure recovery	OBC	G′ at LVR	Loss factor	LVR	OIT of oleogel	AV of oil	PV of oil	Viscosity of oil	Polarity of oil
OBC	0.21									
G′ at LVR	−0.40*	−0.03								
Loss factor	0.35	0.40*	−0.64*							
LVR	0.01	0.24	0.20	0.08						
OIT of oleogel	0.01	0.68*	0.15	0.10	0.30					
The acid value of oil	−0.30	0.18	0.35	0.27	0.32	0.33				
PV of oil	−0.30	0.30	0.45*	0.08	0.39*	0.61*	0.89			
Viscosity oil	−0.04	0.62*	0.15	0.40*	0.34	0.77*	0.74	0.82*		
Polarity of oil	−0.26	0.15	0.33	0.12	0.34	0.31	0.88*	0.88*	0.63*	
TPC of oil	−0.319	0.36*	0.59*	−0.47*	0.29	0.76	0.23	0.59*	0.43*	0.34
DPPH activity of oil	−0.11	0.55*	0.45*	0.01	0.31	0.91*	0.30	0.64*	0.63*	0.34
β-Carotene content of oil	−0.281	0.36*	0.56*	−0.44*	0.28	0.78	0.24	0.61*	0.44*	0.36*
OIT of oil	0.21	0.54*	−0.03	−0.18	0.03	0.63	−0.44*	−0.15	0.14	−0.36*
PUFA content of oil	−0.16	−0.12	0.59*	−0.75*	0.00	0.00	−0.38*	−0.12	−0.35*	−0.15
Fractal Dimension	0.58	0.50	−0.11	0.34	0.10	0.55	−0.06	0.09	0.37*	−0.02
TPC of oleogel	−0.24	0.48	0.50	−0.19	0.26	0.83	0.17	0.52	0.47	0.27
DPPH of oleogel	−0.09	0.56	0.45	−0.02	0.32	0.89	0.25	0.59	0.59	0.30
BC content of oleogel	−0.27	0.46	0.51	−0.04	0.30	0.86	0.41	0.75	0.65	0.48
OIT of oleogel	0.00	0.68	0.15	0.29	0.30	1.00	0.33	0.61	0.77	0.31
Tocopherol of oil	0.16	0.79*	0.03	0.48*	0.24	0.93*	0.29	0.52*	0.82*	0.27
Tocopherol of oleogel	0.20	0.80*	−0.02	0.44*	0.21	0.91*	0.16	0.40*	0.74*	0.16

* p-Values of the corresponding correlation values are significant ($p < 0.05$). AV – acid value; BC – β- carotene; OIT – oxidation induction temperature; PUFA – polyunsaturated fatty acids; PV – peroxide value; TPC – total phenolic content

Parameter	TPC oil	DPPH activity of oil	BC content of oil	OIT of oil	PUFA content of oil	Fractal dimension	TPC of oleogel	DPPH of oleogel	BC content of oleogel	Tocopherol of oil
DPPH activity of oil	0.93*									
BC content of oil	0.99*	0.95*								
OIT of oil	0.52*	0.61*	0.52*							
PUFA content of oil	0.53*	0.33	0.51*	0.40*						
Fractal dimension	0.26	0.45*	0.27	0.59*	0.05					
TPC of oleogel	0.97	0.95	0.97*	0.66*	0.51*	0.37*				
DPPH of oleogel	0.92	0.99	0.94*	0.65*	0.38*	0.46*	0.96*			
BC of oleogel	0.95	0.97	0.96*	0.46*	0.33	0.30	0.94*	0.95*		
OIT of oleogel	0.76	0.91	0.78*	0.63*	0.00	0.55*	0.83*	0.89*	0.86*	
Tocopherol of oil	0.58*	0.80*	0.60*	0.65*	−0.08	0.65*	0.70*	0.79*	0.72*	
Tocopherol of oleogel	0.56*	0.77*	0.57*	0.74*	−0.02	0.70*	0.70*	0.77*	0.67*	0.99*

* p-Values of the corresponding correlation values are significant ($p<0.05$). AV – acid value; BC – β- carotene; OIT – oxidation induction temperature; PUFA – polyunsaturated fatty acids; PV – peroxide value; TPC – total phenolic content

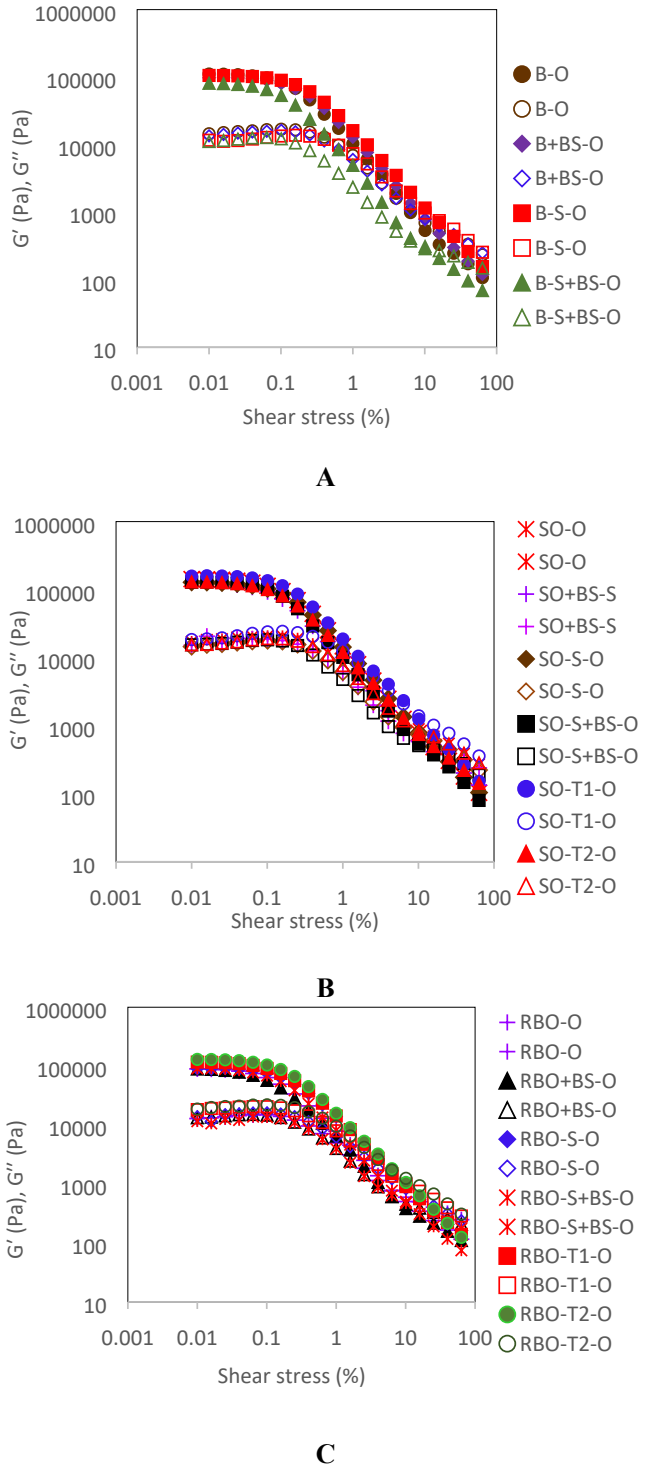

Fig. 2. Amplitude sweeps of oleogels. Closed series markers refer to the G′ and open series markers refer to the G″.

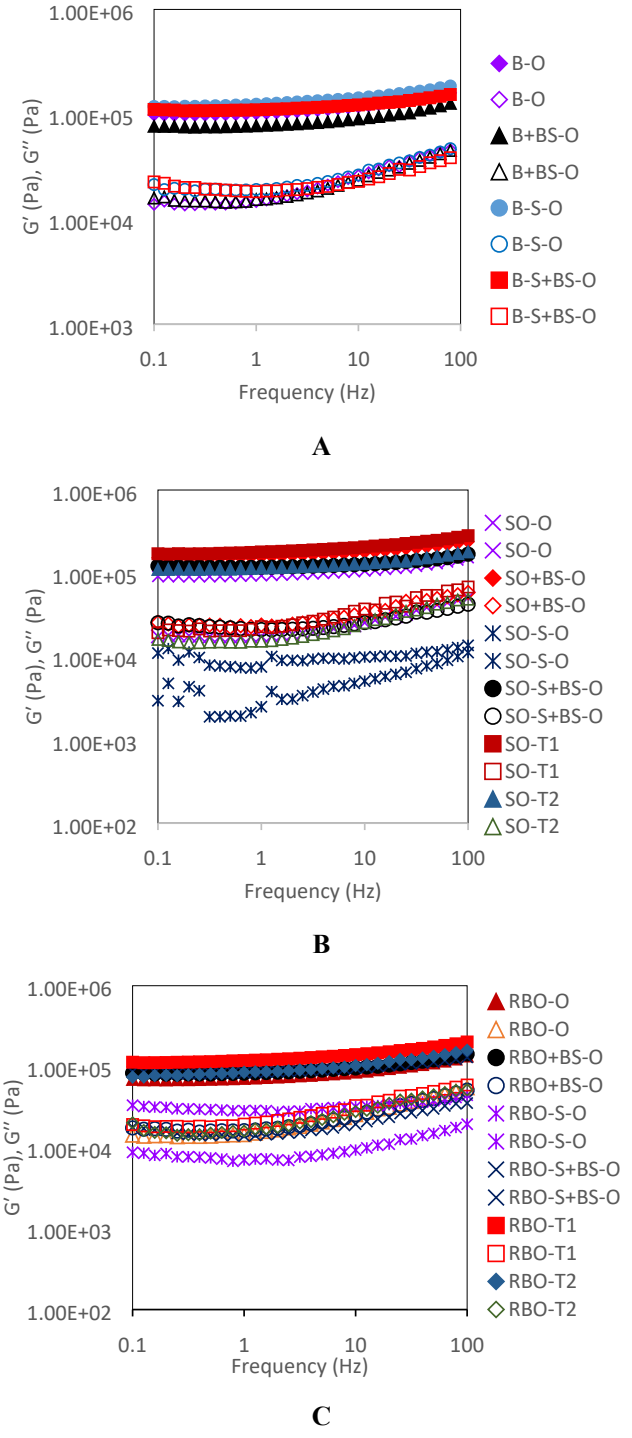

Fig. 3. Frequency sweeps of oleogels. Closed series markers refer to the G′ and open series markers refer to the G″.

alone had a significantly higher loss factor, whereas all oleogels made using sesame oil had significantly lowest loss factor indicating that sesame oil yielded a more stable network than rice bran oil.

Further, it can be concluded that the oil treatments such as stripping or heating with water did not have any significant influence on gel strength in terms of loss factor, whereas the oil type exerted a significant effect. These results are in line with the results of G′ at LVR as explained above, that is, sesame oil produced stronger gel compared to rice bran oil. These observations could be further explained by the results of the correlation analysis shown in Table 5. Loss factor showed a significant negative correlation with the degree of unsaturation, total phenolic content, β-carotene content, and G′ at LVR. That is, highly unsaturated oils (less viscous oil) with a high amount of minor components produce a more stable gel structure (low loss factor).

The frequency sweep experiment measures the response of a material in terms of G′ and G″ to a range of frequencies under controlled conditions. As shown in Fig. 3 (A, B, C), all oleogels had a higher G′ than G″ indicating gel-like behaviors of all samples. All oleogels prepared using stripped oils showed more frequency dependency compared to the

oleogels made from untreated oils and oils heated with water (Fig. 3 (A, B, C)). This could be attributed to the very low viscosities of the stripped oils. Even though untreated and heated sesame oils and rice bran oils had significantly different viscosities, the effect of the small differences in the viscosities of oils on the frequency dependency of oleogels was less prominent. Similar to the other rheological properties, the incorporation of β-carotene did not show any noticeable effect on frequency sweeps of oleogels.

Thixotropy is a time-dependent rheological manifestation of the gels where the material becomes less viscous over time under constant stress or strain (shear thinning behavior) and is a reversible process (Chen et al., 2019). In this study, a three-interval thixotropic experiment was conducted to evaluate the structure recovery ability in terms of recovery of the viscosity of the gel. The structure recovery ability as determined by thixotropy experiments (Table 4), all oleogels made using blend oils regardless of whether untreated or stripped and either added with β-sitosterol or not showed significantly higher structure recovery ability than others prepared using a single oil.

All oleogels made using stripped oils showed significantly less initial viscosity compared to the oleogels made from other oils (Fig. 4), however, initial viscosities of oleogels made from untreated and the oils heated with water were not significantly different. This could be due to the very low viscosity of stripped oils compared to other oils. Even though sesame oil resulted in higher gel strength than blend oil as determined by G′ at LVR and loss factor, the blend oil resulted in higher thixotropic behavior compared to the single oil. These results cannot be explained based on the minor oil components or viscosity of the oils. Therefore, it could be interpreted that there may be a synergistic interaction among both oils related to the thixotropic behavior of the oleogels. The synergistic effect among oils could be a result of new interactions in the blend oil among the fatty acids via hydrogen bonding, π-π stacking, electrostatic interactions, and van der Waals interactions (Manzoor et al., 2022). Several studies reported synergistic interactions among different combinations of oleogelators (Sivakanthan et al., 2022). However, there is no study on the use of a blend of different oils to tailor the properties of oleogels. In our previous study, we optimized the combination of oleogelators (beeswax and stearic acid) and oil blend (rice bran oil and sesame oil) (Sivakanthan et al., 2023) and the present study used the optimized formula to make oleogels. Further research focus should be directed on using oil blends to explore the synergistic effects of different oil types to enhance the oleogel properties. Further, the incorporation of β-sitosterol did not show any significant influence on the thixotropy of the oleogels. As per the correlation analysis shown in Table 5, structure recovery showed a significant strong positive correlation with the fractal dimension indicating that a more uniform arrangement of the crystals in the gel network leads to a higher structure recovery ability of the oleogels. Further, the structure recovery exhibited a significant weak negative correlation with the G′ at LVR. From this observation of this study, it can be concluded that even though G′ at LVR is related to the strength of the gel network, a higher G′ at LVR is not always related to a high structure recovery ability. Moreover, Ghazani et al. (2022) reported that the fractal dimension and elastic nature of the oleogels have a negative correlation, that is, an increase in fractal dimension results in a lower elastic constant. In our study, the fractal dimension and G′at LVR showed a negative correlation, however, the correlation was not significant.

Complex viscosity is a measure of a sample's resistance to shearing forces and it can be used to interpret the spreadability of the oleogel. Gels with high viscosity are more difficult to spread than gels with low viscosity (Palla et al., 2017; Thomas et al., 2023). As illustrated in Fig. 5, the complex viscosity of all oleogels was decreased with an increasing frequency indicating the shear thinning behavior of all oleogels. This behavior is in line with the observation reported in the literature (Kwon & Chang, 2022; Thomas et al., 2023). The oleogels based on stripped oils had significantly less initial complex viscosity compared to other oleogels, which could be attributed to the significantly less initial viscosity of the stripped oils compared to other oils.

Few studies have reported the significant influence of minor oil components on the rheological properties of oleogels. For instance, in the case of monoglyceride oleogels based on walnut oil, refined oil resulted in harder gels compared to crude oil and the authors have interpreted the reason as the naturally present phospholipids, pigments, tocopherols, and phenolics in the crude oil have negatively interacted with the gel formation (Sun et al., 2022). Scharfe, Niksch, et al. (2022) reported the impact of minor oil components on different wax-based oleogels. Gravelle et al. (2016) demonstrated the differences in the mechanical properties of ethylcellulose oleogels prepared using refined, bleached, and deodorized canola or soybean oils, and those made with cold-pressed flaxseed oil due to the presence of minor components and oil polarity. The differences between the results reported in these studies and the results reported in our study could be due to the concentration of minor components as well as the concentration of oleogelators used. The present study did not find any significant influence of minor components present naturally in the oils on the mechanical properties of beeswax and stearic acid-based oleogel made from rice bran oil, sesame oil, and their blend. However, polar compounds at higher levels showed a

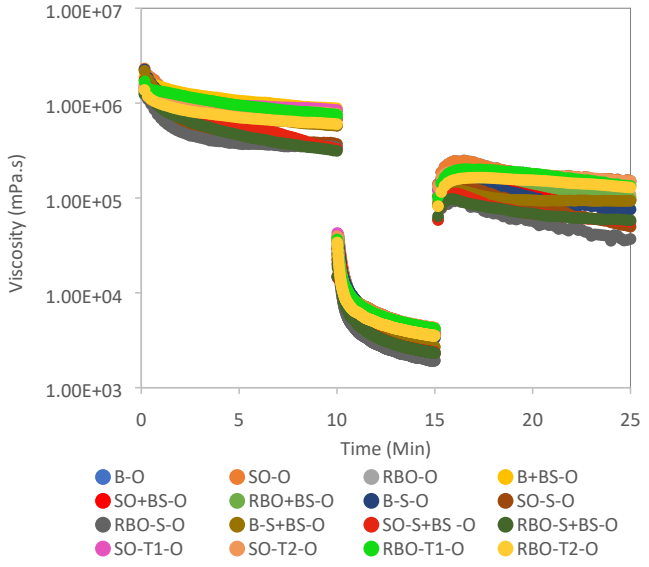

Fig. 4. Thixotropic behavior of oleogels.

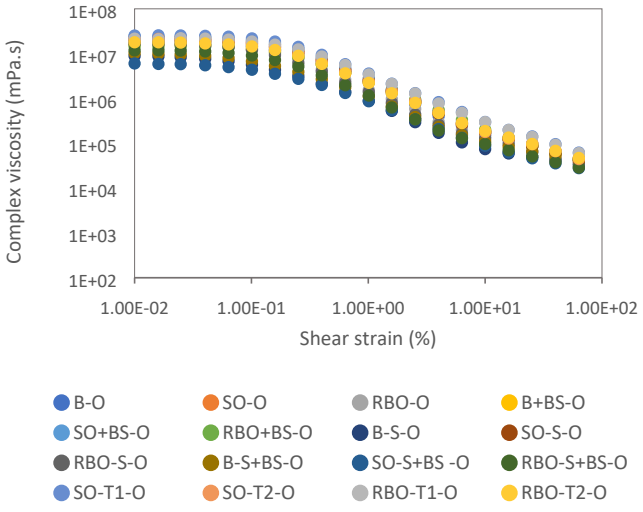

Fig. 5. Complex viscosity of oleogels.

significant effect. From these observations, it can be concluded that the effect of minor components at lower concentrations on the mechanical properties of beeswax and stearic acid-based oleogels is less prominent. Further, beeswax is reported as an effective gelator and the present study has used beeswax at the concentration of 9 % (w/w) as a mixture with stearic acid (3 %). The effect of minor components might be observable at lower concentrations of oleogelators. This could be further confirmed by the results of correlation analysis (Table 5). The polarity of the oil did not show any significant correlation with the rheological properties, whereas the total phenolic content and β-carotene content showed significant correlations with the rheological characteristics indicating that a moderate amount of minor components have a significant influence on oleogel characteristics, whereas a very high level of minor components did not have any significant influence on rheological characteristics of oleogels.

3.5. Thermal properties

The DSC heating thermograms and the parameters obtained from the thermograms of melting and crystallization are provided in Fig. 6 and Supplemental file (Table S1), respectively. The thermal properties such as onset crystallization, peak crystallization, onset melting, and peak melting temperatures of oleogels ranged between 46.50 ± 0.57 °C to 48.30 ± 0.42 °C, 45.80 ± 0.57 °C to 47.55 ± 0.49 °C, 24.65 ± 0.64 °C to 27.70 ± 1.27 °C, and 49.75 ± 0.07 °C to 51.30 ± 0.14 °C, respectively, and there were no significant differences among the oleogels for the thermal properties. The results indicate that the oil composition did not have any influence on the thermal properties of the oleogels. This observation was in line with the observation reported by Frolova et al. (2022). The thermal properties of oleogels primarily depend on the type and concentration of oleogelators regardless of the oil type and composition (Frolova et al., 2022; Pang et al., 2020). Regarding the sensory aspect, high melting point of the oleogel would result in a waxy mouthfeel, which could be a possible drawback of using beeswax as an oleogelator. Therefore, the utilization of beeswax oleogel with a high melting point can be optimized by exploring alternative options to suit various applications such as use in confectionaries or can be used to partially substitute the solid fat in margarine production or can be blended with low melting oils or oleogels.

3.6. FTIR analysis

Molecular interactions among the constituents of the oleogel were analyzed by FTIR. The FTIR spectra of the oils, neat oleogelators, β-sitosterol, and oleogels are shown in Fig. 7. In the functional group region of the spectra of oils and oleogels, three prominent peaks in the regions of 1743–1744 cm^{-1} corresponding to the ester carbonyl group, 2847–2853 cm^{-1} corresponding to asymmetric CH2 stretching, and 2914–2923 cm^{-1} corresponding to symmetric CH2 stretching and two less prominent peaks in the regions of 2953–2955 cm^{-1} corresponding to C—H stretching of an alkyl group (C—C) and 3007–3009 cm^{-1} corresponding to C—H stretching of the alkenyl group (C═C) (Li et al., 2022) were identified. Untreated and treated oil had similar peaks indicating that either stripping or heating with water did not cause any changes in the molecular interactions among the functional groups of the oils.

The absorption bands of all oleogels did not show any shifts in the symmetric CH_2 stretching band (2922 cm^{-1}) except for three oleogels such as RBO-O, RBO + BS-O, and SO-T2-O which showed small shifts (2921 cm^{-1}, 2917 cm^{-1}, and 2917 cm^{-1}, respectively) compared to the oils. However, the bands corresponding to asymmetric CH_2 stretching were observed between the wave numbers 2849——2853 cm^{-1}. When comparing the corresponding peaks of the oils, most of the samples are characterized by more shifts compared to the symmetric stretching peaks in the asymmetric stretching peaks. This observation is in line with the observation reported by Frolova et al. (2022), Martins et al. (2016), and Li et al. (2022) for the beeswax-based oleogels. The reason for the shifts in the peaks could be due to the decrease in the fluidity of the alkyl chains caused by van der Waals interactions (Li et al., 2022). Although β-sitosterol exhibited a peak indicative of hydrogen bonding, none of the oleogels displayed peaks in the spectral range of 3400–3550 cm^{-1}. This suggests the absence of hydrogen bonding in all of the samples. The reason for the absence of peak corresponding to hydrogen bonds in the oleogels incorporated with β-sitosterol could be the dilution effect by the oil. Therefore, it can be concluded that the oleogels structures were formed primarily through van der Waals interactions. Li et al. (2022) also reported similar findings for the beeswax-based oleogels. Further, from these results, it can be concluded that the minor components and fatty acid composition and resulting physicochemical

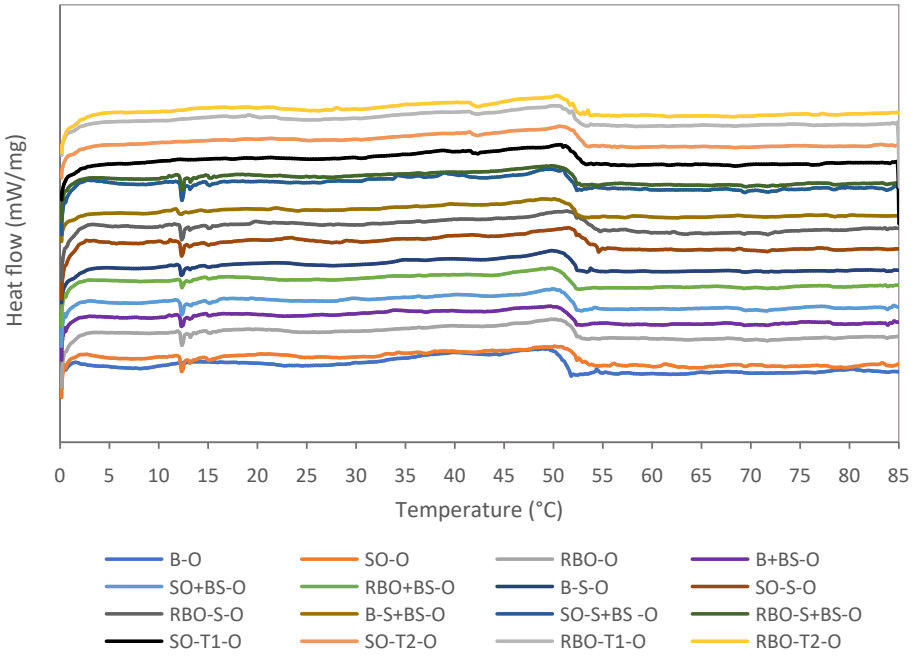

Fig. 6. DSC heating thermograms of oleogels.

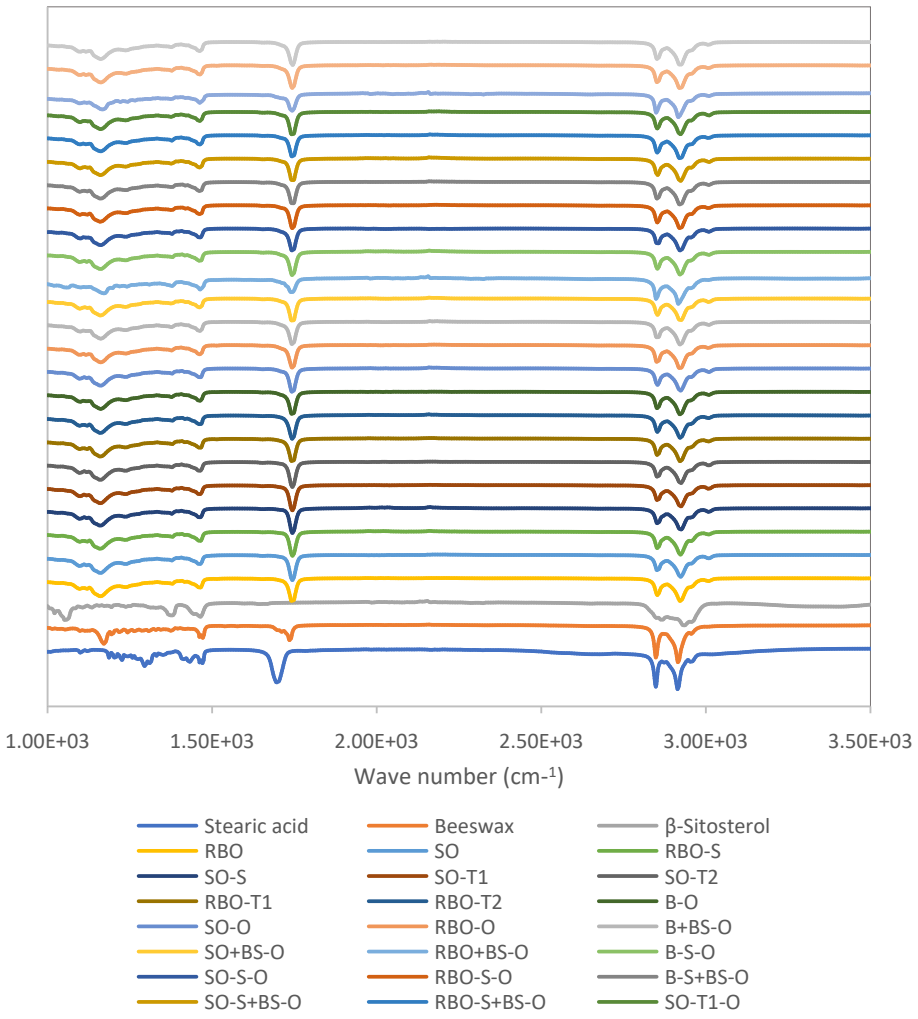

Fig. 7. FTIR spectra of oils, neat oleogelators and oleogels.

properties of the oils did not have any significant influence on the molecular interactions of the oleogels. Similar observations have been reported in the literature. For instance, Martins et al. (2016) demonstrated that the physicochemical properties of walnut oil do not exert any influence on the intermolecular forces within oleogels.

3.7. Antioxidant properties and oxidation behavior of oleogels

Table 6 shows the β-carotene content, α-tocopherol content, total phenolic content, DPPH radical scavenging activity, and oxidation induction temperature of oleogels. Total phenolic content, α-tocopherol content, and DPPH radical scavenging activity of all oleogels showed a similar trend. Total phenolic content and DPPH radical scavenging activity of untreated sesame oil-based oleogels were highest among all oleogels, whereas the oleogels produced from the stripped oils had the lowest values of these parameters indicating the removal of the minor components during stripping. α-Tocopherol content of all oleogels produced from untreated oils did not show any significant differences among them, however, the values were significantly less than corresponding oils, and lowest α-tocopherol contents were shown by the oleogels produced from stripped oils. The oleogels produced from the sesame oil and rice bran oil heated with water did not show much difference in the total phenolic content, α-tocopherol content, DPPH radical scavenging activity, or β-carotene content. All these parameters are in line with the respective parameters of the oils with significantly higher values for the total phenolic content, DPPH scavenging activity, and β-carotene content compared to the corresponding oils, however with significantly less α-tocopherol content. The higher values could be attributed to the phenolic compounds and β-carotene content in the beeswax (Martinello & Mutinelli, 2021). Significantly less α-tocopherol content in oleogels than in respective oils and in the oleogels produced from the heated oils compared to the oleogels produced from untreated oils could be attributed to the sensitivity of the α-tocopherol to heat (Kmiecik et al., 2019). The oxidation induction temperature of oleogels produced from untreated oils was significantly higher than the oxidation induction temperature of oleogels made from corresponding stripped oils. In line with the oxidation induction temperature of oils, sesame oil-based oleogels showed higher oxidation induction temperatures than that of rice bran oil-based oleogels except for the oleogels made from the oils heated for 1 h. Compared to the respective oils, the oleogels made from stripped oils had significantly lower values for oxidation induction temperatures. This could be attributed to the removal of antioxidants from the oils such as phenolic compounds, α-tocopherol, and β-carotene during stripping. The addition of β-sitosterol did not show any significant influence on the oxidation behavior of the oleogels. From these results, it could be concluded that the oleogelation process did not cause any significant changes in the antioxidant properties of the oleogels except for the reductions in α-tocopherol content. The Pearson correlation analysis (Table 5) showed a significant ($p < 0.01$) strong positive correlation of all these parameters of the oleogels with those of corresponding oils.

Table 6
β-Carotene content, α-tocopherol content, total phenolic content, DPPH radical scavenging activity, and oxidation induction temperature of oleogels.

Sample	β-Carotene content (mg/kg)	α-Tocopherol (mg/kg)	TPC (mg/kg)	DPPH radical scavenging activity (inhibition %)	OIT (°C)
B-O	7.85 ± 0.23^{cd}	463.98 ± 12.86^{a}	47.73 ± 1.68^{c}	68.84 ± 0.26^{c}	335.00 ± 1.41^{abc}
SO-O	10.40 ± 0.76^{ab}	464.07 ± 19.04^{a}	82.11 ± 1.46^{a}	78.54 ± 0.79^{a}	340.15 ± 0.21^{a}
RBO-O	5.42 ± 0.22^{e}	454.15 ± 9.72^{a}	28.80 ± 1.74^{d}	36.38 ± 0.79^{e}	332.30 ± 2.40^{bc}
B + BS-O	8.52 ± 1.80^{bc}	469.15 ± 8.98^{a}	49.33 ± 0.59^{c}	69.96 ± 0.26^{bc}	333.80 ± 0.57^{abc}
SO + BS-O	10.87 ± 0.18^{a}	440.08 ± 11.15^{a}	80.26 ± 2.56^{a}	78.73 ± 1.06^{a}	337.35 ± 1.20^{ab}
RBO + BS-O	5.87 ± 0.12^{de}	448.60 ± 14.49^{a}	29.50 ± 2.00^{d}	36.01 ± 0.24^{e}	330.00 ± 0.28^{c}
B-S-O	1.25 ± 0.08^{f}	18.97 ± 1.80^{e}	6.64 ± 0.68^{h}	3.92 ± 0.23^{j}	307.50 ± 1.41^{efg}
SO-S-O	1.31 ± 0.16^{f}	18.34 ± 1.71^{e}	12.76 ± 0.82^{fg}	6.90 ± 0.19^{i}	301.35 ± 1.77^{gh}
RBO-S-O	1.00 ± 0.21^{f}	20.26 ± 1.95^{e}	7.06 ± 1.21^{gh}	5.97 ± 0.53^{ij}	308.95 ± 1.48^{ef}
B-S + BS-O	1.51 ± 0.43^{f}	15.36 ± 1.10^{e}	6.59 ± 0.50^{h}	13.99 ± 0.26^{h}	302.35 ± 2.62^{fgh}
SO-S + BS-O	1.15 ± 0.15^{f}	17.36 ± 1.43^{e}	12.87 ± 0.67^{fg}	18.28 ± 0.53^{g}	299.25 ± 1.34^{h}
RBO-S + BS-O	1.02 ± 0.11^{f}	19.73 ± 1.93^{e}	7.04 ± 1.12^{gh}	15.49 ± 1.32^{h}	309.45 ± 1.63^{e}
SO-T1–O	11.69 ± 0.13^{a}	168.92 ± 6.85^{bc}	60.75 ± 1.06^{b}	71.64 ± 0.76^{b}	332.00 ± 2.12^{bc}
SO-T2-O	11.17 ± 0.49^{a}	138.18 ± 6.05^{cd}	60.93 ± 1.49^{b}	61.75 ± 0.52^{d}	322.50 ± 2.83^{d}
RBO-T1–O	5.30 ± 0.08^{e}	175.41 ± 9.71^{b}	19.71 ± 1.62^{e}	34.33 ± 0.51^{e}	322.00 ± 2.40^{d}
RBO-T2-O	5.27 ± 0.56^{e}	124.01 ± 4.49^{d}	16.55 ± 2.75^{ef}	24.07 ± 0.75^{f}	318.10 ± 0.42^{d}

Different superscript letters (a-j) in the same column show a significant difference ($p < 0.05$). TPC – total phenolic content; OIT – oxidation induction temperature.

3.8. Storage stability

The rheological analysis provides valuable information about the long-term stability and shelf-life of oleogels. Changes in rheological properties over time can indicate structural degradation, phase separation, or other undesirable changes that may impact the product's shelf-life (Almeida & Bahia, 2006). By monitoring the rheological behavior, manufacturers can assess the product's stability and make necessary adjustments to extend its shelf-life. Storage stability of the oleogels at 5 °C and 20 °C was evaluated in terms of rheological properties. For this purpose, the samples were stored at 5 °C and 20 °C for two months and analyzed by amplitude sweep experiments (Supplemental file - Fig. S2.). All oleogels stored at 20 °C for two months showed about 20–70 % reduction in the G′ at LVR regardless of the type of oil used indicating the changes in the gel structure at 20 °C with time due to post-crystallization events. However, the changes in the G′ at LVR of oleogels stored at 5 °C were significantly less (0 – 35 %) compared to the change that occurred in the oleogels stored at 20 °C for two months. The oleogel made from an untreated oil blend added with β-sitosterol (B + BS-O) stored at 5 °C did not show any changes in the G′ at LVR. Saw et al. (2023) also reported that at 25 °C of storage, superolein oleogels were theologically unstable due to insufficient supercooling effect. Therefore, storage at 5 °C would help maintain the structure of oleogel during long-term storage.

4. Conclusions

This study aimed to uncover the fundamental understanding of how minor components and fatty acid composition of the oils influence the properties of beeswax and stearic acid-based oleogels and the effect of oleogelation on the minor components of the oils. To gain reliable data on the influence of oil composition on oleogel properties and the influence of oleogelation on minor oil components, a comprehensive characterization of the composition and quality of oils and properties of oleogels was studied. Results show that both fatty acid composition and minor components of oil played profound roles in the properties of beeswax-stearic acid based oleogels. A moderate amount of minor components have a significant positive influence on oleogel properties. Further, the addition of β-sitosterol (5 %) to the untreated oil blend resulted in improved microstructure and rheological stability during long-term storage. This could be considered an advantage as the β-sitosterol could be incorporated into the oleogel formulation to enhance the rheological as well as the functional properties of the oleogel. Analysis of the oleogel prepared using the untreated oil blend incorporated with β-sitosterol by X-ray diffraction would help acquire comprehensive knowledge of the mechanism behind the improved microstructural properties. Further, the results showed that oleogelation did not cause any significant changes in the minor components of the oils except for α-tocopherol content. The results of this study would be useful for the successful development of oleogels as a superior alternative in terms of nutritional value for solid fats that contain a high content of saturated and *trans* fatty acids without compromising the texture of the food. Further studies on the influence of the composition of oleogelators such as beeswax and stearic acid on oleogels would be valuable to further advance the knowledge on the topic of this study.

CRediT authorship contribution statement

Subajiny Sivakanthan: Conceptualization, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Sabrina Fawzia:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing. **Sagadevan Mundree:** Conceptualization, Supervision, Writing – review & editing. **Terrence Madhujith:** Conceptualization, Supervision, Writing – review & editing. **Azharul Karim:** Conceptualization, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing – review & editing.

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

Data will be made available on request.

Acknowledgements

This work was financially supported by Queensland University of Technology, Brisbane Australia and Accelerating Higher Education

Expansion and Development (AHEAD): a World Bank-funded Sri Lankan government operation. The authors would like to acknowledge the laboratory facilities from the Central Analytical Research Facility (CARF) and the Centre for Agriculture and the Bioeconomy (CAB) of the Queensland University of Technology, Australia.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodres.2024.114213.

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