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Fat replacers based on oleogelation of beeswax/shellac wax and healthy vegetable oils

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ABSTRACT

Oleogels based on the combination of oleogelators and vegetable oils are a fat substitute alternative to obtain commercial fats (CF) with reduced saturated fatty acids (SFA), high polyunsaturated fatty acids (PUFA), and without trans fats (TFA). This study evaluated oleogelation to develop beeswax/shellac wax (BW/SW) with linseed (LO) and canola (CO) oil-based oleogels with a solid gel-like texture and high PUFA. The oleogelation conditions were optimized, and then oleogels were characterized. The optimal conditions were: oleogelator percentage of 10% (w/w), BW/SW ratio of 70/30 (%w/w), heating time of 30min, and heating temperature of 92 and 100 °C for BW/SW-LO and BW/SW-CO oleogels, respectively. The BW/SW-LO and BW/SW-CO oleogels presented firmness values of 423.6 ± 2.81 and 288 ± 2.99g, respectively, with a solid-like rheological behavior comparable to gel. Also, oleogels showed a lower SFA content (12.5 and 9.35%, respectively) than CF (35.9%), and no TFA was produced during oleogelation. The BW/SW-LO oleogel presented a higher PUFA content (68.4%) than the BW/SW-CO oleogel (29.1%). In addition, the oleogels showed peroxide values during storage below the maximum acceptable limit for edible fats and oils. Therefore, oleogels with a healthy fatty acid profile and low lipid oxidation are excellent alternatives to replace CF, offering promising food applications.

1. Introduction

The application of commercial vegetable fats is extensive in the food industry owing to their considerable influence on the texture of food products and their high oxidative stability (Saghafi et al., 2019). These commercial vegetable fats (CF) have been extensively utilized in the bakery and confectionery products industry, usually containing large amounts of saturated (SFA) and/or trans (TFA) fatty acids. However, high consumption of SFA and TFA increases the risk of obesity, diabetes, and coronary heart disease (Naeli et al., 2022). Therefore, partially hydrogenated vegetable fat (the primary source of TFA and a vital ingredient of CF has been excluded from the generally recognized as safe list by the Food and Drug Administration since June 2018 (Bhandari et al., 2020). Moreover, it has been pointed out that the replacement of industrially produced TFA with healthier oils and fats, mono and polyunsaturated fatty acids, reduces the risk of cardiovascular diseases (Martins et al., 2018). In this context, the formulation of CF with vegetable oils that have a higher proportion of polyunsaturated fatty acids (PUFA) than SFA is a significant challenge for food industry experts, as low levels of SFA and free of TFA in fats can lead to loss of their functionality (Naeli et al., 2020).

In recent years, structured vegetable oils by oleogelation systems have been suggested as the most promising alternatives to CF, allowing the reduction of the SFA and TFA content in food products (Naeli et al., 2022). Oleogelation systems are defined as the entrapment of the liquid oil in a three-dimensional network using the low concentration of one or more oleogelator agents (Lim et al., 2017). This technique changes the physical structure of vegetable oils based on the physicochemical properties of oleogelators to a solid gel-like network known as an oleogel, without any TFA production (Limpimwong et al., 2017). The

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common oleogelators for oleogelation include lipid-based oleogelators (e.g., plant waxes, fatty alcohols, phospholipids, mono- and diacylglycerols, sorbitan esters, or phytosterols) and polymeric oleogelators (e.g., ethylcellulose) (Giacintucci et al., 2018). There is an increasing interest in applying plant waxes as oleogelators due to their abundance and gelling ability (Patel 2018). In addition, wax-based oleogels have proven to be very efficient because, even at low concentrations, they can crystallize in a well-formed network with strong oil-binding properties (Doan et al., 2018). In this regard, beeswax (BW) has been widely used to create oleogels that provide solid fat-related features, such as thermoreversible processing, texture, and melt-in-mouth effect (Martins et al., 2018). BW is produced by bees from the genus *Apis mellifera* L and has been approved and certified as safe by the FDA. Clinical studies show it has plasma cholesterol-lowering and antimicrobial effects. BW has been used in foods as a coating agent, texturizer, additive carrier and as an oleogelator for oil structuring (Martins et al., 2018). Properties of BW oleogels have been studied with medium and long-chain triglycerides, sunflower, hazelnut oils and, with a mixture of fish-olive-linseed oil (Gómez-Estaca et al., 2019). On the other hand, shellac wax (SW) has attracted significant interest regarding the development of oleogels for the food industry. SW, the refined form of Lac, is a natural resin secreted by insects of the genus *Kerria* mainly in Southeast Asia. SW is one such small-molecular-weight component that has displayed excellent oil gelation behavior. In addition to structuring the liquid oil, SW also stabilizes oil continuous emulsions at different water content levels by adding an emulsifier (Patel 2018). Nowadays, SW can be applied in the fabrication of oleogels with different textures and thermal properties by altering their concentrations (Yuan et al., 2021). SW has been used as an oleogelator in rapeseed oil (Patel & Dewettinck, 2015) to structure a liquid oil-based thermoreversible soft matter system and to produce oleogel-based emulsions, spreads, chocolate paste, and cakes (Patel 2018).

To improve such oleogel properties as texture (e.g., firmness), rheological properties, and storage stability, the combination of different oleogelators in oleogel formulations has been proposed (Doan et al., 2018; Hwang, 2020). For example, to design oleogels with rheological and textural properties similar to a CF, the mixture of candelilla wax and glycerol monostearate with canola oil (Kim et al., 2022), rice bran wax, BW and candelilla wax with sunflower oil (Oh et al., 2017), BW, mono-diglycerides and sorbitan monostearate with avocado oil (Pérez-Monterroza et al., 2014), monoglycerides, BW, and propolis wax with pomegranate oil (Fayaz et al., 2017) have been studied. These systems of oleogelator combinations could have synergistic interactions and alter the structural framework and rheological properties of oleogels that are crucial to industrial applications (Doan et al., 2018). Winkler-Moser et al. (2019) demonstrated that both the melting property and the firmness of oleogels can be improved by mixing candelilla and bee waxes.

The final properties of oleogels depend not only on the structurant agents such as BW and SW but also on the solvent (oil) characteristics. Oils rich in unsaturated fatty acids produce more firmly structured oils or gels, leading to a high oil retention capacity (Trujillo-Ramírez et al., 2019). A greater degree of unsaturation eases the formation of a larger number of interpolymer (oleogelator agent) bonding points (Cabrera et al., 2020). Several studies have reported the formulation of oleogels with highly unsaturated vegetable oils such as canola and linseed oil (CO and LO) with BW and SW separately, dissolving quickly in these oils and exhibiting strong structures capable of successfully retaining the oil phase. Furthermore, BW and SW have low melting points (65 and 80 °C, respectively), favoring the stability of the oils against oxidative deterioration (Han et al., 2022; Patel, 2018). The CO which has only 7% SFA, but 62% MUFA, mainly oleic acid, and 32% PUFA, which include two essential fatty acids: linoleic (omega-6) and linolenic (omega-3) acid (Jang et al., 2015). In turn, LO contains 9% of SFA, 17% of MUFA, and 74% of PUFA, mainly omega-3 (50–60%), vitamins A and E (Stoltz et al., 2012). However, the rheological and textural properties of oleogels depend not only on the degree of oil unsaturation but also on the variables involved in the oleogels preparation process, such as the type and concentration of oleogelator, heating and cooling temperature, heating time, and agitation (Thakur et al., 2022). Thus, developing new oleogelation systems requires optimizing different parameters to prepare oleogels as fat substitutes. However, to our knowledge, oleogels based on the combination of BW/SW and vegetable oils (CO and LO) have not been tested as alternatives to CF with a healthy fatty acid profile. In this sense, combining proportions of two structuring agents, such as waxes, instead of one, with unsaturated vegetable oils can adjust the crystallization and gelling behavior of oleogels so that the desired properties (firmness, solids content, and melting temperatures) are achieved, thus reducing the need for large amounts of wax (Doan et al., 2018). Therefore, to achieve the process conditions to obtain a food product with specific characteristics, in this research, an experimental design was applied. According to the above considerations, the aim of this study was to evaluate the oleogelation process to develop BW/SW and vegetable oil-based oleogels with solid gel-like texture and high PUFA. First, the oleogelation process was optimized through the Taguchi method. Thus, the effect of four independent variables: oleogelator percentage, heating temperature, heating time, and BW/SW ratio on firmness was evaluated. Afterward, the optimized BW/SW-CO and BW/SW-LO oleogels were characterized according to firmness, oil binding capacity, fatty acid profile, rheological properties, and oxidative stability. It was hypothesized that the oleogel formulations can be considered alternative fat replacers.

2. Materials and methods

2.1. Materials

SW, SSB® Cera 2 was acquired from SSB Stroever GmbH & Co. KG. (Bremen, Germany). BW was purchased from Sigma Aldrich (St. Louis, MO, USA). The canola oil was purchased from Alimentos Soy Saludable SPA (Osorno, Chile), and linseed oil was purchased from Aceitería Dumont (Santiago, Chile). Commercial vegetable fats (CF: 100% fat, made from soybean oil, hydrogenated and interesterified soybean oil, hydrogenated and interesterified palm kernel butter, propylene glycol, T.B.H.Q., and citric acid) was purchased in a local market (Temuco, Chile). All other chemicals used were of analytical grade.

2.2. Preparation of oleogels

Oleogels were prepared using combinations of BW and SW with vegetable oils such as CO and LO. The oleogelation process was carried out by magnetic stirring at 300 rpm, using an oleogelator content of 4–10%, heating temperatures between 84 and 100 °C, and heating times of 10–30 min. The oleogels were cooled to room temperature for at least 2 h and later refrigerated at 4 °C (Supplementary material: Fig. S1). After 24 h, the samples were prepared for analysis (Pang et al., 2020; Patel & Dewettinck, 2015).

2.3. Experimental design

An experimental design using the Taguchi method was applied to prepare the BW/SW-CO and BW/SW-LO oleogels through an orthogonal matrix L_9 (3^4) with nine experiments, four variables, and three working levels applying the criterion “the bigger, the better” looking for a solid gel-like texture (firmness). In the preparation of oleogels, the effect of four independent variables, such as oleogelator percentage (4, 7, and 10%), heating temperature (84, 92, and 100 °C), heating time (10, 20, and 30 min), and BW/SW ratio (70/30, 50/50 and 30/70 %w/w) on firmness was evaluated. The optimized theoretical value of the Taguchi method was determined considering a triplicate for each design point using the Minitab 19 software (Minitab LLC, Pennsylvania, USA). The optimized theoretical equation (OTE) was determined by considering the average of the response with the greatest impact, identifying the most important variables and working level on oleogel firmness. Table 1 shows the orthogonal matrix used with the design variables. The OTE was calculated according to Eq. (1):

$$OTE = T + \left[WL_{Oleogelator} - T\right] + \left[WL_{Time} - T\right] + \left[WL_{Temperature} - T\right] + \left[WL_{BW/SW} - \mathrm{T}\right] \tag{1}$$

where T is the total average of responses of the experimental runs, and WL corresponds to the working level of each variable in the equation.

2.4. Characterization of oleogels

2.4.1. Textural analysis

The textural properties of optimized oleogels and CF (control) were evaluated according to Millao et al. (2023). The samples were evaluated by penetration test using the TA.XT PlusC texture analyzer (Stable Micro System, Surrey, UK) with a 5 kg load cell. Thirty grams of oleogel were placed in a glass and stored at 4 °C for 24h. The samples were removed from the storage site just prior to testing. A compression test was carried out with a 5 mm cylinder probe, penetrating the sample in its container at 1 mm/s to a distance of 10 mm. The probe was returned to its original position at a constant speed (1 mm/s). The maximum positive force, which indicates the sample's firmness, was recorded and expressed in grams.

2.4.2. Oil binding capacity (OBC)

The OBC was evaluated by performing the methodology described by Millao et al. (2023). The OBC test was performed by centrifuge method where the oleogel was subjected to centrifugation at 7000 rpm for 40 min. Then 1 g of sample was weighed into 15 mL tubes. After centrifugation, the released oil was removed by inverting the tubes, and the mass of the sample was recorded. The percentage of OBC was calculated using Eq. (2).

$$OBC(\%) = \left(1 - \left(m_i - m_f\right) / m_i\right) \times 100 \tag{2}$$

where m_i is the weight of the sample before centrifugation, and m_f is the weight after the oil drainage.

2.4.3. Confocal Laser Scanning Microscopy (CLSM)

The internal phase distribution in oleogels was analyzed using an Olympus FV 1000 spectral confocal microscope system (Olympus Corporation, Tokyo, Japan). Nile Red (30 μM solution in acetone) was used to stain the oil phase of the samples. The samples were exposed to an argon laser (488 nm) with a 20X objective to image the microstructure of the samples.

Table 1
Firmness of oleogels using the orthogonal matrix L_9 (3^4).

Design point	Oleogelador (%)	Time (min)	Temperature (°C)	BW/SW ratio (w/w)	Firmness (g)	
					BW/SW-CO	BW/SW-LO
1	4	20	85	70/30	56.8 ± 3.33	100.8 ± 5.22
2	4	30	92	50/50	32.6 ± 1.80	62.4 ± 1.22
3	4	40	100	30/70	15.8 ± 1.73	35.5 ± 2.17
4	7	20	92	30/70	69.2 ± 2.26	164.7 ± 3.24
5	7	30	100	70/30	197.8 ± 4.38	250.7 ± 5.24
6	7	40	85	50/50	98.0 ± 1.04	188.3 ± 7.79
7	10	20	100	50/50	214.6 ± 4.58	350.6 ± 4.42
8	10	30	85	30/70	116.9 ± 2.33	253.9 ± 5.33
9	10	40	92	70/30	287.9 ± 2.97	442.7 ± 9.75

2.4.4. Fatty acid profile

The fatty acid profile of oleogels, vegetable oils and CF were determined. The fatty acid composition was determined by the AOCS Official Method Ce 1f-96, using gas chromatography (7820A, Agilent Technologies, USA) equipped with a flame ionization detector. This analysis was done in duplicate.

2.4.5. Rheological analysis

Rheological measurements of oleogels and CF were made using a rheometer (Discovery HR2, TA Instruments, New Castle, DE, USA), equipped with a flat parallel plate geometry (stainless steel, 50 mm diameter, 1000 μm gap). TRIOS software package (TA Instruments, New Castle, DE, USA) was used to control the equipment and acquire rheological parameters. The steady-shear flow measurements were made at 25 °C in a shear rate range of 1–100 s^{-1}. The range of linear viscosity values in the samples was obtained from the plot of elastic modulus (G′) versus oscillatory strain (%). Thus, the viscoelastic behavior of the samples was measured under oscillations at 1 Hz and from 0.001 to 20%. In the frequency sweep test, the temperature was held at 25 °C, and the response of moduli (G′ and G″) to increase frequency (0.1–100 Hz) at a strain of 0.01% within the linear viscosity region was measured. Finally, temperature sweep tests were performed to evaluate the thermal stability of the samples from 5 °C to 90 °C at a linear heating rate of 5 °C/min, a shear strain of 0.01%, and 1 Hz. Samples were performed in triplicate.

2.4.6. Differential scanning calorimetry (DSC) analysis

DSC measurements were carried out in a DSC 1 STAR System (Mettler-Toledo, Greinfensee, Switzerland) to determine the thermal stability of oleogels. Subsequently, 23–26 mg of oleogel were analyzed. An empty pan was used as a reference, and N_2 gas was used as a purge gas. The thermal profile of oleogel samples was determined in the 25–150 °C (heating) and 150-25 °C (cooling) range at a scan rate of 5 °C/min. The onset and peak temperature of melting or crystallization were calculated using the STARe Software (DB V12.10, Mettler-Toledo, Greifensee, Switzerland). Samples were performed in triplicate.

2.5. Lipid oxidation of oleogels

2.5.1. Peroxide value

The peroxide value (PV) of oleogels, vegetable oils, and CF were evaluated according to the AOCS method cd 8–23 (AOCS 1996). The samples (5 g) were treated with a 30 mL acetic acid: chloroform (3:2 v/v) solution and then with a saturated solution of potassium iodide (0.5 mL), and the liberated iodine was titrated with 0.01 N solution of sodium thiosulfate. PV was reported as milliequivalents of peroxide per kilogram oil. The PV was calculated in triplicate according to Eq. (3):

$$PV = ((S - B) \times M \times 1000) / W \tag{3}$$

where S is the titer of sample in mL, B is the titer of blank in mL, M is the molarity of the thiosulfate solution, and W is the weight of sample in g.

2.5.2. Rancimat test

The Rancimat test, an accelerated technique used to assess the oxidative state of edible fats and oils, was used. For the measurements, a 5 g sample of oleogel, vegetable oil, and CF was exposed to a high air temperature of 110 °C and an airflow of 20 mL/h using the Metrohm Rancimat 743 (Herisau, Switzerland; AOCS, 1996). The results are given in a conductivity curve as a function of time; thus, the induction period

(IP) was determined from the inflection point of the conductivity curve (Naeli et al., 2022). All samples were analyzed in triplicate.

2.5.3. Oxidative stability

The oxidative stability of optimal oleogels, vegetable oils, and CF was evaluated by performing the methodology described by Millao et al. (2023) with some modifications. The oxidative stability of the samples was determined by accelerated oxidation using PV. The PV of the samples was evaluated at 25 °C and under accelerated storage conditions at 50 °C for 35 days. All samples were analyzed in triplicate.

2.6. Statistical analysis

The results presented are the average and standard deviation calculated from these replicate measurements. A two-way analysis of variance (ANOVA) was carried out with a significance level set at 0.05. Duncan's test was performed in the case of significant differences detected with the ANOVA. Statistical analyses were performed with the InfoStat software 2014.

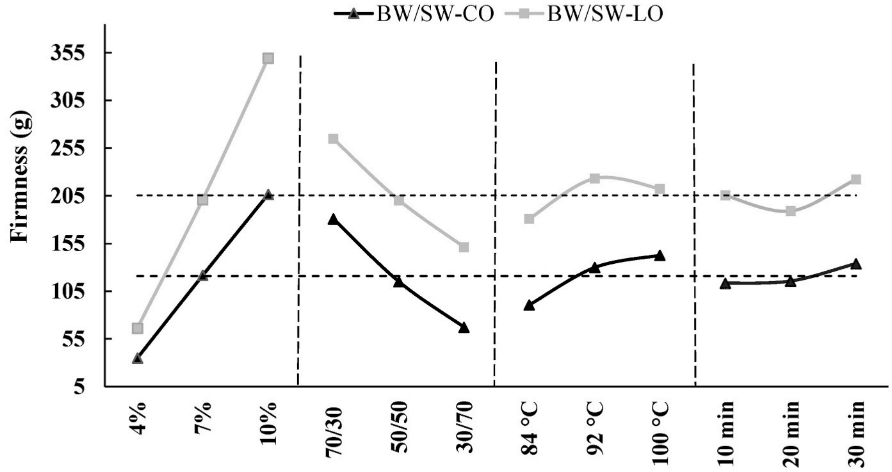
3. Results and discussion

3.1. Optimization of the BW/SW and vegetable oil-based oleogels

The oleogelation process was optimized through the Taguchi method using an orthogonal array L_9 (3^4). Thus, the effect of four independent variables: oleogelator percentage (4, 7, and 10%), heating temperature (84, 92, and 100 °C), heating time (10, 20, and 30 min), and BW/SW ratio (70/30, 50/50 and 30/70 %w/w) on firmness was evaluated. After evaluating each variable, the firmness values of the BW/SW-CO and BW/SW-LO oleogels ranged from 15.8 to 287.9 g and 35.5–442.7 g, respectively (Table 1). Firmness is a critically relevant textural property that affects consumer products' processing, shelf life, and acceptability (Blake et al., 2018).

Fig. 1 shows the inclination degree of the slope in terms of firmness in the oleogel formulation, indicating that the greater the difference between levels for a variable, the greater the magnitude of change in firmness. The results showed that the oleogelator content for the BW/SW-CO and BW/SW-LO oleogels was the most influential variable in terms of firmness, with differences of 171.4 and 278.5 units, at a 10% working level. Several studies have reported that as the oleogelator content increases, the firmness of oleogels based on vegetable waxes and oils increases due to the increasing solid volume fraction of dense networks, which are highly efficient at entrapping the surrounding liquid oil (Blake et al., 2018; Thakur et al., 2022).

The other most influential variable regarding firmness was a BW/SW ratio with differences of 113.5 and 113.3 units between working levels (1–3) for the BW/SW-CO and BW/SW-LO oleogels, respectively. The most relevant working level was 70/30 (%w/w) of the BW/SW ratio, with a higher content of BW than SW. In this sense, preliminary results revealed that the firmness of oleogels prepared with BW was significantly higher ($p < 0.05$) than oleogels prepared with SW (Supplementary material: Fig. S2). In our case, oleogels designed with BW and SW achieved different firmness values in their structures; however, the two oleogel formulations were significantly different ($p < 0.05$) in the firmness values compared to the commercial vegetable fat (CF) used as a control. For this reason, BW/SW combinations with highly unsaturated vegetable oils were evaluated to design new oleogel systems, which may become a promising strategy as a fat substitute to produce healthier and higher quality food products, reducing the limitations and constraints of using a single oleogelator in the formulation of oleogels. According to Doan et al. (2018), the interaction between two wax-type oleogelators can alter the crystallization behavior and the resulting microstructural network. The heating temperature had less influence on firmness than the variables mentioned above, with differences of 52.1 and 42.2 units, in which the most significant levels were 100 and 92 °C, for the BW/SW-CO and BW/SW-LO oleogels, respectively. All heating temperatures used during the oleogelation process were sufficient to induce gelation as they were above the melting temperature of the oleogelants: 63 °C for BW (Blake et al., 2018) and 85 °C for SH (Patel, 2018). Finally, the heating time was the variable that had the least influence on firmness, with differences of 20.3 and 33.1 units between the working levels



Working Levels	Oleogelator (%)		BW/SW ratio (%w/w)		Temperature (°C)		Time (min)	
	CO	LO	CO	LO	CO	LO	CO	LO
1	35.1	66.2	180.8	264.7	90.6	181.0	113.5	201.0
2	121.7	201.2	115.1	196.1	129.9	223.3	115.7	189.0
3	206.5	344.7	67.3	151.4	142.7	207.9	133.9	222.1
Δ (1,2,3)	171.4	278.5	113.5	113.4	52.1	42.2	20.3	33.1

Fig. 1. Effect of working levels of each variable in the formulation of BW/SW-CO and BW/SW-LO oleogels on the firmness. The greater the difference between the work levels, the greater the magnitude of the change in firmness.

for the BW/SW-CO and BW/SW-LO oleogels, respectively. The low contribution of the time variable is the result of the selected working levels, which were adequate for forming a solid network known as an oleogel but not necessarily for increasing firmness.

On the other hand, the firmness results for the BW/SW-CO and BW/SW-LO oleogels were subjected to statistical analysis (ANOVA) to determine how the control variables influenced the response. The coefficients of determination (R^2) for the independent variables of oleogelator percentage, BW/SW ratio, heating temperature, and heating time were 64.1 and 82.9%, 28.4 and 13.9%, 6.4 and 2.0%, 1.1 and 1.2%, for the BW/SW-CO and BW/SW-LO oleogels, respectively. Also, the R^2 for these four variables was significant ($p < 0.05$), with 99.9% indicating a high affinity or association of the independent variables with the firmness. Therefore, the optimal process conditions to formulate oleogels with solid gel-like texture were: oleogelator percentage of 10% (w/w), BW/SW ratio of 70/30 (%w/w), heating temperature of 92 °C for BW/SW-LO and 100 °C for BW/SW-CO, and heating time of 30 min. Under these process conditions, oleogels showed optimal experimental firmness values for BW/SW-CO (288.0 ± 2.99 g) and BW/SW-LO (423.6 ± 2.81 g), showing a solid structure after the oleogelation process. In addition, the optimized theoretical equations (OTE) of BW/SW-CO and BW/SW-LO oleogels contributed 267.5 and 421.5 g on the firmness, respectively, agreeing with the optimal experimental values. Finally, the Taguchi experimental design made it possible to determine which process optimization variables had the most significant impact on product quality with the minimum number of assays (Morales et al., 2017).

Table 2 shows the firmness of the optimized oleogels and CF sample. The firmness of the BW/SW-LO oleogel was significantly higher ($p < 0.05$) than the BW/SW-CO oleogels. This is due to LO having a more important proportion of PUFA than CO. Firmer oleogels can be obtained using oils with a higher degree of unsaturation, which could lead to a more efficient chain-polymer interaction and thus increase the structural and physical stability of the oleogels (Pang et al., 2020). Moreover, the van der Waals forces present in wax-based oleogels are more potent in oleogels with highly unsaturated vegetable oils such as LO, producing crystals that promote a more stable network structure (Han et al., 2022). Similar behavior was reported by Millao et al. (2023), who produced ethylcellulose oleogels with linseed and canola oil with firmness values of 443.03 and 32.57 g, respectively.

On the other hand, the firmness of the BW/SW-LO oleogel showed no significant difference ($p > 0.05$) compared to CF (419 ± 3.78 g), while the firmness of BW/SW-CO was significantly ($p < 0.05$) lower than CF. However, depending on the texture (solid or semi-solid) of oleogels made with waxes and vegetable oils, they can be used as a substitute for fats in different food matrices such as spreads, chocolate paste, and cakes (Patel, 2018), beef burgers (Khiabani et al., 2020) and baked goods (Jang et al., 2015).

3.2. Oil binding capacity and microstructure of oleogels

Oil binding capacity (OBC) is another crucial indicator that expresses the strength and nature of oleogelator-oil interaction, especially when the gel is mechanically stressed (Doan et al., 2018). A high oil entrapment capacity in oleogels reflects the degree of entrapment of liquid oil in the 3D network by the oleogelator (Pandolsook & Kupongsak, 2017). Therefore, the BW/SW-LO and BW/SW-CO oleogels had a high oil binding capacity (OBC>97%) with no significant differences ($p > 0.05$) between them, leading to less oil leakage in the proposed systems (Table 2). Oleogels must possess high OBC because they contain a high liquid volume fraction, in our case 90% w/w. Failure to structure the liquid fraction (having low OBC) will result in oil leakage, and as a consequence of this, deterioration of the surrounding food matrix (Blake et al., 2018). Similar OBC values of 96.3% and 97.3% were reported in oleogels based on BW (5% w/w) with pomegranate oil (Fayaz et al., 2017) and carnauba wax (8% w/w) with soybean oil (Thakur et al., 2022), respectively.

Table 2
Characterization of oleogels optimized, vegetable oils and commercial fat.

Parameters	BX/SW-CO	BX/SW-LO	CF	CO	LO
Firmness (g)	288.0 ± 2.99 ^{a}	423.6 ± 2.81 ^{b}	419.0 ± 3.78 ^{b}	–	–
OBC (%)	97.7 ± 0.42 ^{a}	98.0 ± 0.73 ^{a}	–	–	–
PV (meq O_2/Kg oil)	2.24 ± 0.15 ^{a}	1.98 ± 0.00 ^{b}	0.91 ± 0.11 ^{c}	1.45 ± 0.10 ^{c}	0.58 ± 0.00 ^{d}
$IP_{110^\circ C}$ (h)	4.26 ± 0.28 ^{c}	1.26 ± 0.09 ^{e}	22.9 ± 0.45 ^{a}	4.94 ± 0.17 ^{b}	1.45 ± 0.04 ^{d}
Fatty acids (%)					
C8:0	nd	nd	0.08 ± 0.00	nd	nd
C10:0	nd	nd	0.35 ± 0.00	nd	nd
C12:0	nd	nd	7.64 ± 0.01	0.02 ± 0.00	0.02 ± 0.00
C14:0	0.17 ± 0.00	0.16 ± 0.00	2.94 ± 0.01	0.08 ± 0.00	0.06 ± 0.00
C15:0	nd	nd	nd	0.02 ± 0.01	0.02 ± 0.00
C16:0	6.13 ± 0.01	6.83 ± 0.00	11.11 ± 0.00	4.70 ± 0.00	5.24 ± 0.01
C17:0	0.12 ± 0.00	0.06 ± 0.00	0.08 ± 0.00	0.13 ± 0.00	0.06 ± 0.00
C18:0	1.73 ± 0.01	4.80 ± 0.00	12.94 ± 0.00	1.59 ± 0.00	3.34 ± 0.00
C20:0	0.53 ± 0.01	0.17 ± 0.00	0.35 ± 0.00	0.52 ± 0.00	nd
C21:0	nd	nd	nd	0.03 ± 0.00	nd
C22:0	0.33 ± 0.01	0.19 ± 0.00	0.33 ± 0.00	0.33 ± 0.00	0.14 ± 0.00
C24:0	0.34 ± 0.01	0.34 ± 0.00	0.12 ± 0.00	0.08 ± 0.00	0.08 ± 0.00
Σ SFA	9.35 ^{b}	12.5 ^{c}	35.9 ^{a}	7.50 ^{e}	8.96 ^{d}
C14:1	0.03 ± 0.00	nd	nd	nd	nd
C15:1	nd	nd	nd	0.01 ± 0.00	nd
C16:1	0.23 ± 0.00	0.10 ± 0.00	0.09 ± 0.01	0.23 ± 0.00	0.08 ± 0.00
C18:1 (ω-9)	59.8 ± 0.04	18.7 ± 0.01	17.6 ± 0.01	60.5 ± 0.01	19.4 ± 0.00
C20:1n9	1.19 ± 0.03	0.18 ± 0.00	0.32 ± 0.00	1.19 ± 0.00	nd
C22:1 n9	0.21 ± 0.00	nd	nd	0.22 ± 0.00	0.02 ± 0.00
C24:1	0.13 ± 0.00	nd	nd	0.13 ± 0.00	nd
Σ MUFA	61.5 ^{b}	19.0 ^{d}	18.0 ^{e}	62.3 ^{a}	19.5 ^{c}
C18:2n6 (ω-6)	19.0 ± 0.01	15.9 ± 0.01	40.8 ± 0.02	19.6 ± 0.00	15.6 ± 0.00
C18:3n3 (ω-3)	10.1 ± 0.04	52.5 ± 0.01	5.28 ± 0.01	10.5 ± 0.01	55.8 ± 0.00
C20:2n6	nd	nd	nd	0.08 ± 0.00	0.04 ± 0.00
C20:3n6	nd	nd	nd	nd	0.03 ± 0.00
C20:3n3	nd	0.05 ± 0.00	nd	nd	0.12 ± 0.00
Σ PUFA	29.1 ^{c}	68.4 ^{b}	46.1 ^{c}	30.2 ^{d}	71.5 ^{a}
TFA	nd	nd	nd	nd	nd
ω-6/ω-3	1.88	0.30	7.73	1.87	0.28

Different letters on the same row indicate statistically significant differences ($p < 0.05$); Oleogels optimized (BW/SW-CO and BW/SW-LO), CO (canola oil), LO (linseed oil), and CF (commercial fat); OBC (Oil binding capacity); PV (peroxide value); $IP_{110^\circ C}$, (Induction period of oxidation at 110 °C); nd: not detected; Σ SFA: Sum of saturated fatty acids; Σ MUFA: Sum of monounsaturated fatty acids; Σ PUFA: Sum of polyunsaturated fatty acids; TFA: Trans fatty acid; ω-6: Linoleic acid; ω-3: Linolenic acid.

In addition, the microstructure of the BW/SW oleogels was examined using confocal laser scanning microscopy. Both the BW/SW-CO (Fig. 2A) and BW/SW-LO (Fig. 2B) oleogels exhibited platelet-like crystals homogeneously distributed throughout the oleogel structure, which is in line with previous studies reporting that wax-based oleogels show platelet-like crystals in discrete and aggregate forms (Doan et al., 2018; Khiabani et al., 2020). The microstructure of wax-based oleogels forms platelet-like crystals due to the van der Waals interactions that cause the liquid oils to immobilize in a three-dimensional structure (Fayaz et al., 2017). On the other hand, no differences were observed in crystal distribution and size between the optimal oleogels BW/SW-CO and BW/SW-CO. These oleogels have the same combination (BW/SW) and percentage of oleogelator (10% w/w), differing only by the type of vegetable oil. According to Doan et al. (2018), one of the oleogel preparation parameters that mainly affects the gelling behavior is the nature and chain length of the chemical components present in the waxes, resulting in a firm gel during the formation of wax crystals in the form of platelets.

3.3. Fatty acid profile of oleogels

Table 2 shows the fatty acid profile of oleogels compared to oils and CF. As expected, CO and LO contained mainly long-chain fatty acids with different degrees of unsaturation. As a result, these oils presented SFA content of 7.50 and 8.96%, MUFA of 62.3 and 19.5%, and PUFA of 30.2 and 71.5%, for CO and LO respectively, which is consistent with previous reports by other authors (Jang et al., 2015, Franco et al., 2020). However, a significant decrease ($p < 0.05$) was observed in the fatty acid profile of oleogels compared to oils, attributed to the oleogelation process. The SFA composition of the vegetable oils, oleogels, and CF sample was represented mainly by palmitic (C16:0) and stearic (C18:0) fatty acids. At the same time, a considerable percentage of dodecanoic acids (C12:0) was observed in the CF sample. Also, the MUFA and PUFA profile of all samples was represented by oleic (C18:1, ω-9), linoleic (C18:2n6, ω-6), and alpha-linolenic (C18:3n3, ω-3) acids. The CF sample presented the highest SFA content of 35.9%, compared to BW/SW-LO and BW/SW-CO oleogels with 9.35 and 12.5%, respectively. Similar behavior was reported by Naeli et al. (2022), who compared oleogels based on ethyl cellulose and hydroxypropyl methyl cellulose biopolymers with commercial shortenings with a high SFA content (47.8%). The BW/SW-CO oleogel displayed a high MUFA content with 61.5% compared to the BW/SW-LO oleogel (19.0%) and CF (18.0%), where oleic acid was in the majority of total oil. BW/SW-LO oleogel showed a high PUFA content of 68.4%, represented mainly through linolenic acid (52.5 ± 0.01%) followed by linoleic acid (15.9 ± 0.01%), compared to BW/SW-CO oleogel and CF with PUFA content of 29.1% and 46.1%, respectively. Moreover, linoleic acid accounted 19.0 ± 0.01% and 40.8 ± 0.02% for BW/SW-CO oleogel and CF, respectively. Finally, it was shown that no TFA was produced during the oleogelation process using heating temperatures of 92 °C (BW/SW-CO) and 100 °C (BW/SW-LO). According to the study reported by Bhat et al. (2022), edible oils subjected to cooking temperatures below 200 °C have a minimal effect on TFA generation, while temperatures above 200 °C may increase the level of TFA.

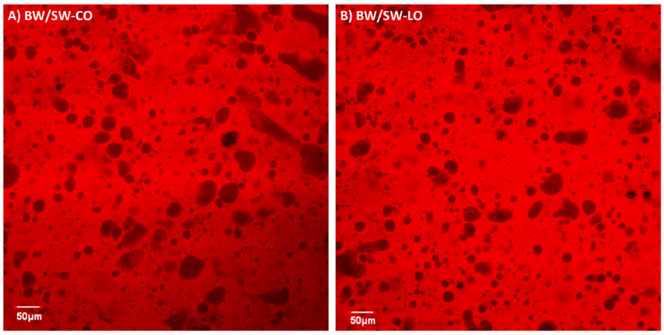


Fig. 2. Confocal microscopy images of (A) BW/SW-CO oleogel and (B) BW/SW-LO oleogel. The lipid component is shown in red and crystal formation is displayed in black. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Another essential aspect for analysis of fatty acid profile is the omega-6/omega-3 ratio in the oleogels compared to oils and CF. An excessive amount of omega-6 relative to omega-3 (omega-6/omega-3 ratio) facilitates the pathogenesis of many diseases such as cardiovascular, cancer, inflammatory and autoimmune diseases, while an increase in omega-3 in relation to omega-6 (omega-6/omega-3 ratio less than 1) exert suppressive effects, reducing the risk of chronic diseases (Simopoulos, 2002). In this regard, the BW/SW-LO (0.33) and BW/SW-CO (1.88) oleogels showed a lower omega-6/omega-3 ratio than the CF sample (7.73). Furthermore, it is essential to emphasize that the BW/SW-LO oleogel had a healthier fatty acid profile.

3.4. Rheology of oleogels

Parameters derived from rheological studies of the oleogels compared to CF included a sweep of viscosity, elastic modulus G′, and

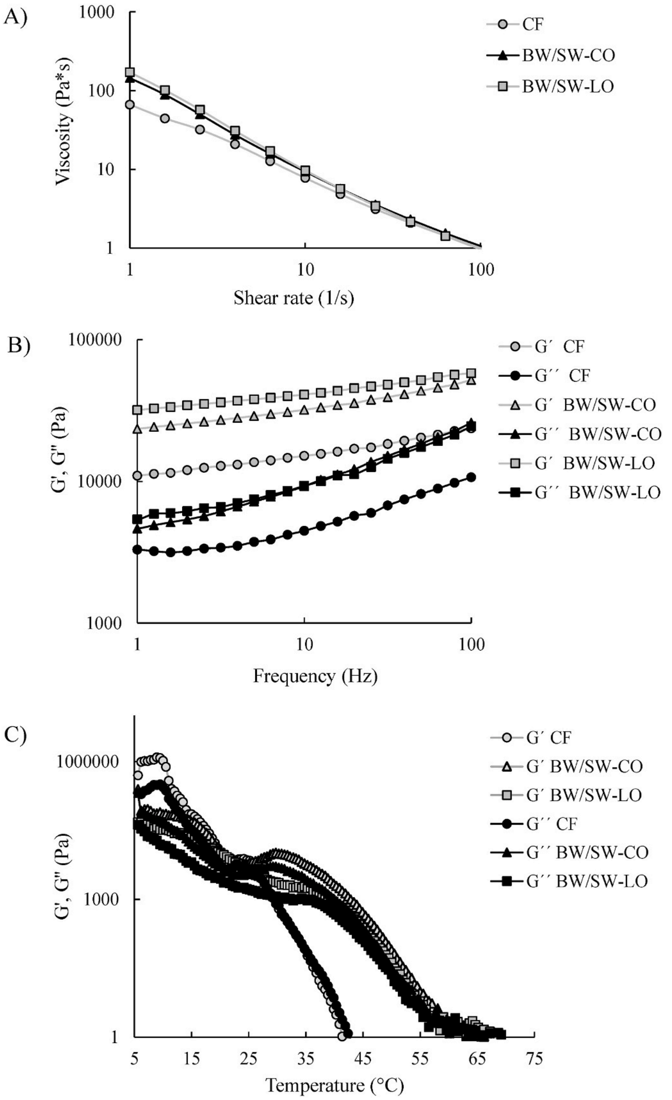


Fig. 3. (A) Viscosity sweep, (B) frequency sweep, and (C) temperature sweep curves for optimal oleogels and control (CF).

viscous modulus G″ as a function of frequency and temperature, respectively (Fig. 3). The results showed that the viscosity of BW/SW-CO and BW/SW-LO oleogels decreases similarly as the shear rate increases, indicating a shear-thinning behavior of oleogels (Fig. 3A). Other studies have also reported similar shear-thinning behavior of oleogels based on waxes and vegetable oils (Thakur et al., 2022; Trujillo-Ramírez et al., 2019). Both oleogels presented higher viscosity than CF, especially at lower shear rates. The ability of the oleogelator to entrap liquid oil provides a non-flowing gel-like property (Meng et al., 2018). Likewise, BW and SW-based oleogels exhibit the formation of multiple crystal networks, promoting more shear stability than CF (Han et al., 2022). However, when the shear rate was higher than 10 (1/s), the oleogels and CF exhibited similar behavior, indicating that the rearrangement of the structured and crystallized network of oleogels in a certain direction under shearing offered less resistance to flow.

Frequency sweep rheograms were performed in the linear viscoelastic region of the oleogels compared to CF, as shown in Fig. 3B. Both oleogels and CF showed a similar behavior with storage modulus (G′) values higher than loss modulus (G") values over the entire frequency range (1–100 Hz), demonstrating that the samples exhibit solid-like behavior comparable to a gel. It is worth noting that the G′ of a sample indicates the elastic portion and represents the solid properties, while G″ indicates the viscous portion and represents the liquid properties (Naeli et al., 2022). Moreover, both the G′ and G″ parameters showed slightly higher values with the increase in frequency, suggesting the existence of slight relaxation processes occurring even at short time scales and indicating a weak dependence of G′ and G″ over the frequency applied. This behavior has been reported before for wax-based oleogels (forming solid gels in the frequency range of the linear viscoelastic region) (Guneser et al., 2021; Thakur et al., 2022). The BW/SW-LO oleogel showed higher G′ values than the BW/SW-CO oleogel and both oleogels showed substantially higher G' than CF, indicating higher resistance to shearing deformation due to the formation of more robust structural gel networks by wax crystal fractions (Naeli et al., 2020). These results are not consistent with the results of the texture analysis, where the BW/SW-LO oleogel showed differences with BW/SW-CO and similar firmness to CF, which could be explained by differences in crystal network properties. In oleogels structured using oil with a higher polyunsaturated fatty acid content, such as LO, the network formation with a greater number of small crystals has been observed (Han et al., 2022). Thus, it could be favoring a higher resistance to compressional deformation of BW/SW-LO oleogel during textural analysis. In addition, the crossover point between G′ and G″ was not observed where $G' = G''$, which indicates that the oleogels were relatively stable in the frequency range, showing that the oleogels could have sufficient stability during storage (Guneser et al., 2021).

Fig. 3C shows the thermodynamic properties of the oleogels, evaluated by the temperature sweep test from 5 to 90 °C. In all samples, G′ and G″ decreased with the increase in temperature, indicating a decrease in the elastic properties but an increase in viscosity. This behavior versus temperature is typical in lipid systems (Naeli et al., 2022). Oleogel samples exhibited higher values of G′ relative to G″ from 5 °C to 55 °C, showing dominant elastic properties (solid gel-like state). In turn, decreases in G′ and G″ values were observed when the temperature increased, producing a phase transition in the oleogels superior to the CF sample. Then, from temperatures above 65 °C (close to the melting point), the crossover of G′ and G″ indicated a liquid behavior. By contrast, the CF sample presented higher values of G′ than G″ up to a temperature of 30 °C. After 30 °C, G″ was higher than G′, suggesting that the system had converted into viscous or liquid matrices. Similar behavior was reported by Lim et al. (2017) in oleogels prepared from natural waxes (candelilla, carnauba, and BW) with vegetable oils, where the two viscoelastic parameters decreased with the increase in temperature until they reached the crossing of G′ and G″ curves at temperatures between 46 and 74 °C, respectively.

3.5. Differential scanning calorimetry of oleogels

Two melting peaks (1 and 2) were detected in BW/SW-CO and BW/SW-LO oleogels due to the heterogeneous chemical composition of the systems given by the two types of waxes; these melting peaks were at 50.1 ± 1.09 and 74.2 ± 0.09 °C for BW/SW-CO and at 51.3 ± 0.21 and 76.3 ± 0.43 for BW/SW-LO (Table 3). Both BW and SW are heterogeneous materials composed of long-chain esters, n-alkanes, and fatty acids in the case of BW (Blake et al., 2018), and of fatty alcohols and esters in the case of SW (Patel et al., 2013), which explains the melting peaks found (Sivakanthan et al., 2023). The melting temperature of BW oleogels reported by other authors is around 50 °C, although it depends on the BW concentration (Martins et al., 2016). For SW oleogels, more than one melting peak has been detected at temperatures near 55, 66, and 79 °C (Patel et al., 2013). Similarly, two crystallization peaks (1 and 2) have been found in BW/SW-CO and BW/SW-LO oleogels (Table 3), meaning that crystals were formed from two different crystallization stages. This may be the result of inhibited crystal growth due to the low compatibility between different components of the oleogel (Martins et al., 2016). Other authors have reported two crystallization peaks for BW oleogels (43 and 29 °C) and three crystallization peaks for SW oleogels (69, 60, and 50 °C) (Martins et al., 2016; Patel et al., 2013).

Overall, the results revealed significant differences ($p < 0.05$) in thermal behavior between the two oleogels, where the melting and crystallization peaks of the BW/SW-CO oleogel showed a slight decrease in temperature (about 2 °C) compared to the BW/SW-LO oleogel. Differences in the formation of a solid solution (or gel) are caused by solute-solute interactions among the different components of the oleogels (Qiu et al., 2023). In our case, the oleogelator composition in both oleogels is the same. However, in terms of the percentage of total saturated and unsaturated fatty acids, the canola and linseed oil show minimal differences, explaining the similarities in their thermal behavior of the oleogels and reflecting the similar stability of crystal networks.

3.6. Lipid oxidation and oxidative stability of oleogels

The peroxide value (PV) and induction period of 110 °C ($IP_{110^\circ C}$) were used to measure the oxidation state of the BW/SW-CO and BW/SW-LO oleogels, fresh oils (CO and LO), and CF sample (Table 2). In the first stage of the oxidation process, fatty acids react with oxygen to form odorless compounds such as peroxides (Khiabani et al., 2020). In addition, a higher $IP_{110^\circ C}$ implies that the samples take longer to generate volatile oxidation products and, therefore, higher oleogel and oil stability (Naeli et al., 2022). The results revealed that oleogels significantly increased the ($p > 0.05$) PV compared to fresh oils. At the same time, oleogels significantly decreased ($p > 0.05$) the $IP_{110^\circ C}$ compared to oils before the oleogelation process. As mentioned, LO and CO comprise an important proportion of unsaturated fatty acids susceptible to oxidation; therefore, exposure to high temperatures during the oleogelation process explains this increase in lipid oxidation.

On the other hand, by comparing the BW/SW-LO and BW/SW-CO oleogels with CF, both oleogels showed a significantly higher ($p < 0.05$) PV and a significant decrease ($p > 0.05$) in the $IP_{110^\circ C}$ than the CF sample. This behavior can be attributed to the fact that the CF sample has a high percentage of SFA and contains commercial antioxidants (TBHQ and citric acid) that enhance its stability against lipid oxidation. Although oil oxidation may lead to gel quality deterioration, in this case the PV values of both oleogels were far below the maximum value allowed by international standards, up to 15 meq O_2/kg oil (CODEX-STAN 210, 1990) for cold-pressed oils.

The oxidative stability of oleogels was evaluated through PV determination at 25 °C and under accelerated storage conditions at 50 °C for 35 days (Fig. 4). During storage at 25 °C (Fig. 4A), the results showed that the BW/SW-LO and BW/SW-CO oleogels had no significant differences ($p > 0.05$) between them in the PV. However, the BW/SW-LO and BW/SW-CO oleogels' PV was significantly increased ($p < 0.05$) through

Table 3

Thermal properties of optimized oleogels.

Oleogels	$T_{om\ 1}$ (°C)	$T_{pm\ 1}$ (°C)	$T_{om\ 2}$ (°C)	$T_{pm\ 2}$ (°C)	$T_{oc\ 1}$ (°C)	$T_{pc\ 1}$ (°C)	$T_{oc\ 2}$ (°C)	$T_{pc\ 2}$ (°C)
BW/SW-CO	41.5 ± 2.50^a	50.1 ± 1.09^a	66.3 ± 1.76^a	74.2 ± 0.09^a	49.1 ± 0.27^a	47.5 ± 0.30^a	41.5 ± 0.06^a	39.7 ± 0.14^a
BW/SW-LO	38.7 ± 4.94^a	51.3 ± 0.21^a	68.7 ± 2.83^a	76.3 ± 0.43^b	50.7 ± 0.32^b	48.9 ± 0.46^b	42.5 ± 0.41^b	40.3 ± 0.25^b

Different letters on the same column indicate statistically significant differences ($p < 0.05$); $T_{om\ (1\text{-}2)}$: onset melting; $T_{pm\ (1\text{-}2)}$: peak melting; $T_{oc\ (1\text{-}2)}$: onset crystallization: $T_{pc\ (1\text{-}2)}$: peak crystallization.

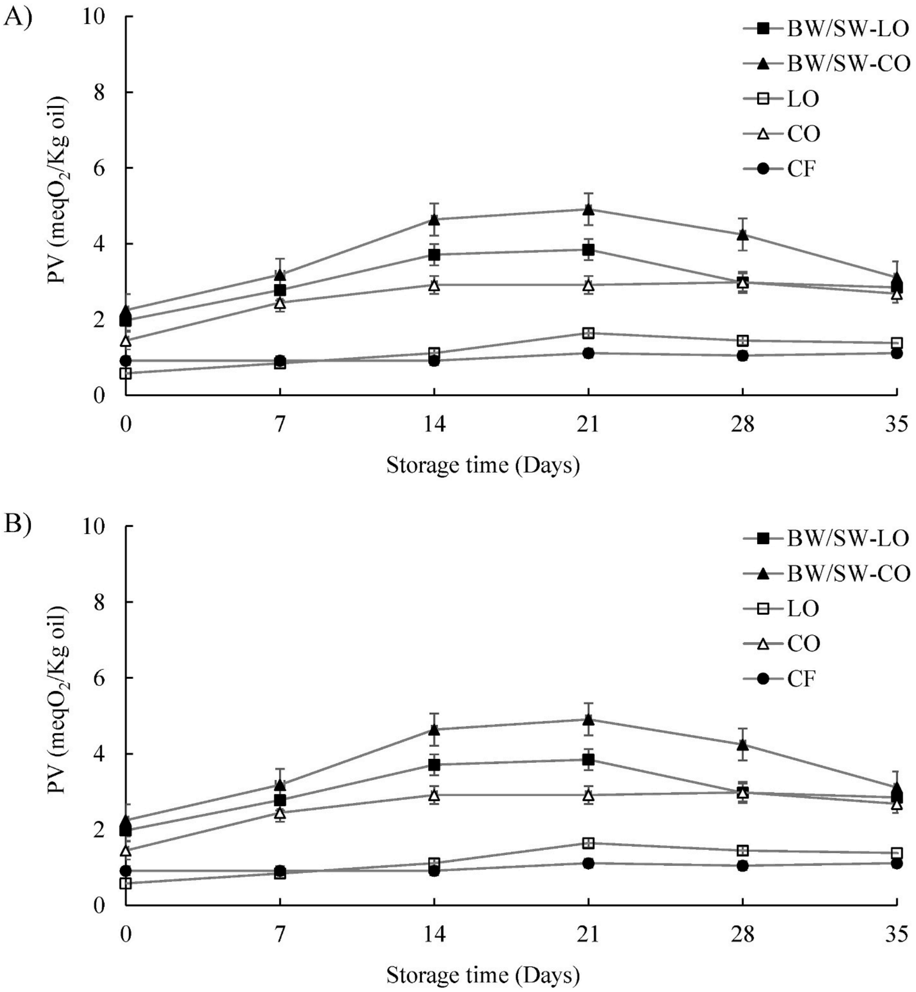


Fig. 4. Effect of storage temperature at 25 °C (A) and 50 °C (B) on the PV of the BW/SW-CO and BW/SW-LO oleogels, fresh oils (CO and LO), and CF sample on the storage period for 35 days.

storage time, reaching a maximum PV at 21 days. After that, both oleogels decreased, likely due to the degradation of primary oxidation compounds (hydroperoxides) in other compounds, such as carbonyls and aldehydes (Fu et al., 2020). It was also observed that the BW/SW-CO and BW/SW-LO oleogels maintained higher PVs for the 35 days of storage than fresh oils, as mentioned above, attributable to the oleogelation conditions (temperature, time, and oleogelator content) of both vegetable oils. Similar behavior was reported by Sobolev et al. (2022), who demonstrated that oleogels prepared with waxes and vegetable oils had shown a slight increase in PV compared to fresh oils due to the oleogelation process.

As in the previous case, during storage at 50 °C (Fig. 4B), the BW/SW-LO oleogel, compared to the BW/SW-CO oleogel, showed no significant differences ($p < 0.05$) in PV. However, both oleogels presented a maximum at 7 days of storage, then a significant decrease ($p < 0.05$) in PV was observed as storage days progressed. These results can be explained by the composition of oleogels possessing high unsaturated fatty acid contents; therefore, they are more susceptible to lipid oxidation caused by increased storage temperature. In contrast to Fig. 4A, no significant differences ($p > 0.05$) in PV were observed between the BW/SW-CO and BW/SW-LO oleogels and their fresh oils. Some authors have reported a protective effect of oleogel structuring agents on oil oxidation (Fu et al., 2020; Hwang, 2020). Yet when the storage temperature was increased, there was no protective effect of the BW/SW mixture on the structure of the oils.

4. Conclusions

This study showed that under optimal oleogelation conditions, oleogels could be obtained using BW/SW blend and vegetable oils (CO and LO) with optimal experimental firmness values similar to the optimized theoretical values and high oil binding capacity. In addition, oleogels showed a lower SFA content than CF; no TFA was produced during oleogelation. Also, the BW/SW-LO oleogel presented a high

PUFA content compared to the BW/SW-CO oleogel, with a slight reduction compared to their oils.

Rheological studies showed that the viscosity of oleogels decreases similarly as the shear rate increases, indicating a shear-thinning behavior. The oleogels presented higher viscosity than CF, especially at lower shear rates, due to the formation of crystalline networks in their structure, which provided higher shear stability. Both oleogels showed similar behavior with storage modulus (G') values higher than loss modulus (G") values over the entire frequency range, demonstrating that the samples exhibit solid-like behavior comparable to a gel.

The peroxide value of the BW/SW-CO and BW/SW-LO oleogels was far below the maximum value allowed by international standards, indicating that the oleogels did not present excessive damage by lipid oxidation during the oleogelation process.

Consequently, the formulations of the BW/SW-LO and BW/SW-CO oleogels with a healthy fatty acid profile (high polyunsaturated fats acids, low saturated fats and without trans fats) and low lipid oxidation are excellent alternatives to replace fats, offering promising food product applications. Furthermore, these findings provide a better understanding of oleogels based on the BW/SW combination with highly unsaturated vegetable oils, and it would be interesting to extend their application as a fat substitute with the same functionalities as commercial vegetable fats, such as organoleptic characteristics, cost effectiveness, and food quality.

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CRediT authorship contribution statement

Eduardo Morales: Investigation, Methodology, Writing – original draft. **Nicole Iturra:** Methodology, Writing – review & editing. **Ingrid Contardo:** Methodology, Writing – review & editing. **Marcela Quilaqueo:** Writing – review & editing. **Daniel Franco:** Writing – review & editing. **Mónica Rubilar:** Conceptualization, Funding acquisition, Project administration, 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

The data that has been used is confidential.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.lwt.2023.115144.

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