ORIGINAL ARTICLE

Oleogels of Some Plant Waxes: Characterization and Comparison with Sunflower Wax Oleogel

Emin Yilmaz[1] (iD) · Eda Keskin Uslu[1] · Ceren Öz[1]

Abstract This study aimed to develop the oleogels of tea wax (TWO), rapeseed wax (RWO), orange peel wax (OWO), rose wax (RsWO), and beery wax (BWO), and to compare their properties with sunflower wax oleogel (SWO). Since the literature lacks most of these new oleogels, the gained information would be valuable. The wax oleogels were analyzed for their main physicochemical, thermal, structural, rheological, and sensory properties. The minimum gelation concentrations (C*) ranged from 1.0% (SWO) to 25% (RWO), with gelation times of 0.3–205 min, respectively. While polarized light microscopy images showed large crystal aggregates, X-ray diffraction patterns proved the presence of β′ polymorph crystals and some amorphous solids. Rheological analyses indicated that the samples had a weak-gel structure, and were stable until around 30–70 °C temperatures. Further, the oleogel samples showed a thixotropic rheological behavior with force-related recovery at 10 °C. Quantitative sensory descriptive analysis (QDA) proved that there were some differences among the samples for sensory hardness, spreadability, liquefaction, rancid, waxy, and cooling attributes. Also, tea aroma in TWO, bitterness in OWO, and rose aroma in RsWO were quite dominant. In conclusion, RWO and OWO seemed unsuitable for food applications due to very high C* and bitterness, while RsWO and BWO could be used in food formulations.

✉ Emin Yilmaz
  eyilmaz@comu.edu.tr

[1] Faculty of Engineering, Department of Food Engineering, Çanakkale Onsekiz Mart University, Çanakkale, 17020, Turkey

J Am Oil Chem Soc (2021) **98:** 643–655.

Introduction

There has been a demand for solid fats in the food industry, and to fulfill this need, oil chemists developed oil structuring strategies like hydrogenation, interesterification, and fractionation to produce plastic consistency fat from liquid vegetable oils. A relatively new technology called oleogelation is developed in the last decade to provide alternative structured fats without enhancements of saturated and *trans* fatty acids, contrary to the existing technologies. These gels are formed by adding various oil gelling agents, called organogelators, to initiate lattices of lipid aggregates organized as liquid crystals, fibrillar lattices, or micellar packages. Indeed, these structures are soft, elastic materials without any compositional or isomeric change in the fatty acids of the starting liquid oil. Hence, they are nutritionally preferable, but structural and sensory properties need to be improved (Co and Marangoni, 2012; Dassanayake et al., 2009; Sagiri et al., 2018).

The current literature (Co and Marangoni, 2012; Sagiri et al., 2018) indicates that most of the thermal and rheological properties of the oleogels are governed by the organogelators used to create them together with the techniques used to form the gels. There are around 50 organogelators, not to list them all, searched for edible applications and classified as low molecular weight gelators (LMWG) and polymeric gelators (PG). Regardless of their chemical nature, all of them must be food grade, safe, technologically feasible, abundant, and cheap to be commercialized (Co and Marangoni, 2012; Mattice and Marangoni, 2018; Sagiri et al., 2018).

Natural plant waxes were among the most studied organogelators. They are mainly composed of hydrocarbons, wax esters, long-chain fatty acids and alcohols, triterpenoids, and others. The kinds and proportions of the chemical components in the waxes determine the properties of the oleogels created (Mattice and Marangoni, 2018; Toro-Vazquez et al., 2007; Yılmaz et al., 2015). Some plant waxes like rice bran wax (Dassanayake et al., 2009), sunflower wax (Yılmaz et al., 2015), candelilla wax (Toro-Vazquez et al., 2007), carnauba wax (Blake and Marangoni, 2015), beeswax (Yılmaz et al., 2015), sugarcane wax (Rocha et al., 2013), berry wax (Doan et al., 2017), and fruit wax (Okuro et al., 2018) have been the most widely researched waxes for oleogel development. Most of these waxes are deemed as GRAS or approved by the FDA as food additives (Mattice and Marangoni, 2018).

Generally, plant waxes were classified within LMWG. Waxes act as crystal network providers to entrap liquid oil to form the gel structure. A typical minimum concentration (critical concentration, C*) was determined for each wax at which gelling could occur (Co and Marangoni, 2012; Mattice and Marangoni, 2018). After complete melting of the wax in liquid oil, and during the cooling period, new wax crystals are formed through some junction zones, and within these networks, the liquid oil is entrapped similar to the regular lipid crystals created with saturated triglycerides. Most wax oleogels resemble β′ polymorph crystals with different lamellar packings. Since different waxes contain diverse components that can improve or decline crystallization state due to alteration of the saturation equilibrium between crystallizable wax components and liquid oil, oleogels with different rheologies could be generated (Blake and Marangoni, 2015; Mattice and Marangoni, 2018). It is stated that wax oleogels were viscoelastic in nature, and deformation can occur by force as ductile or brittle types. They generally display shear thinning behavior with low thixotropic recovery rates. Further, the cooling rate and presence of shear affect the rheology of wax oleogels quite diversely depending on the wax type (Blake and Marangoni, 2015; Co and Marangoni, 2012; Mattice and Marangoni, 2018).

The aim of this study was to provide information about oleogel forming abilities and of properties of tea wax, rapeseed wax, orange peel wax, rose wax, and berry wax in comparison with sunflower wax. Since the literature lacks information about the plant wax oleogels listed above, it could be important to share these new data. Further, sunflower wax oleogel is accepted as the gold standard (mainly because it is the best performing plant wax in oleogels according to the literature) for wax type oleogels to compare with the others listed above. Descriptive sensory analysis was also included to evaluate the possibility of food applications.

Materials and Methods

Materials

Refined-winterized sunflower oil (Trakya Birlik Oil Co., Tekirdağ, Turkey) was purchased from the local market and used to prepare the oleogels. The sunflower seed wax (6607 L), tea leaf wax (6614), rapeseed wax (6237), rose wax (6692), and berry wax (6290) were provided by Kahlwax Co. (Kalh GmbH &Co., Trittau, Germany), and orange peel wax (E00068) was provided by Koster Keunen Holland B.V. (Raambrug, Holland). The producers provided only general descriptions and product code numbers (given in the parentheses above) of the waxes. We used these waxes as received without any further purification or modification. All used chemicals and solvents were of analytical grade quality and were purchased from Sigma Chem. Co. (St. Louis, MO, USA) and/or Merck (Darmstadt, Germany).

Preparation of the Oleogels

Initially, to determine the minimum gelation concentrations (C*) of the waxes used in this study, serial addition levels of the waxes were prepared (oil: wax = 70:30 to 99:1, w/w). To form the oleogel, weighed proportions were mixed and heated in a water bath at 90 °C until complete melting (30 min) and mixed vigorously for homogeneity. Then, the tubes were taken out to the ambient temperature and stayed overnight. The C* was determined visually by turning the tubes to check upon flowing. The tubes without flow and solid appearance were selected to determine the C* values (Hwang et al., 2012). The RSO and OWO were prepared at their C* (25% and 15% by weight), while the rest were prepared at 10% wax addition level. The prepared oleogels could be observed in Fig. 1.

Physicochemical Properties of the Oleogels

The oleogels were heated in a water bath for 30 min at 90 °C, and then removed and cooled at room temperature (23 ± 2°C) until crystallization. The gelation time (GT) was considered to be the time in minutes until the oleogels solidified. They were considered gels when, after tilting tube 90°, the flow was not initiated (Yılmaz et al., 2015).

The oil binding capacity of the samples was determined by the modified method of Yılmaz et al. (2015). First, the oleogels were preheated (90 °C, 30 min) in a water bath to complete melting. Then, 1 mL of melted oleogels was placed into tared Eppendorf tubes and kept at 4°C for 1 hour. After gel formation, the tubes were weighed again and then centrifuged at 10.000 rpm for 15 min at room
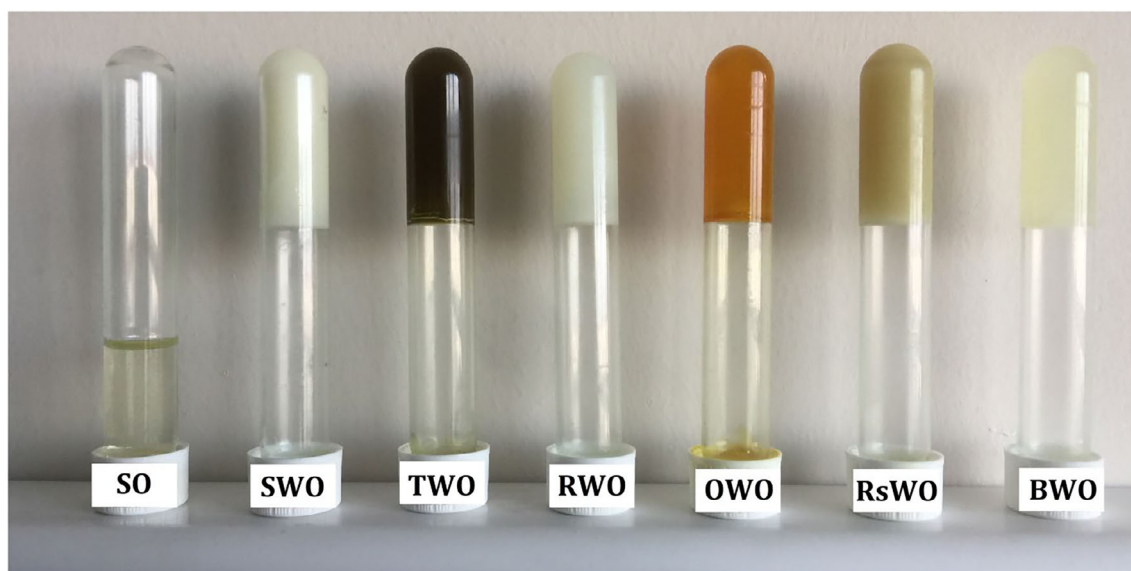
Fig 1 Sunflower oil (SO) and the oleogels prepared (BWO, berry wax oleogel; OWO, orange peel wax oleogel; RsWO, rose wax oleogel; RWO, rapeseed wax oleogel; SWO, sunflower wax oleogel; TWO, tea wax oleogel)

temperature. The tubes were turned over, and liquid oil was drained. Finally, the tubes were weighed again, and oil binding capacity values were obtained gravimetrically.

The solid fat contents (SFC) of the oleogel samples were assessed with a Minispec Bruker NMR Analyzer mq20 (Bruker Optics, Inc.). The oleogels were melted in a water bath (90 °C, 30 min) and then transferred to NMR tubes (3.5 mL). The tubes were conditioned in another water bath at 0°C for 1 hour, and then at 20 °C for 1 hour. The instrument was calibrated with standards including 0, 31, and 73.5% of solid fat. The measurements were carried out according to method 16b-93 (AOCS, 1998).

Colors of the oleogel samples were measured with Minolta CR-400 (Konica Minolta Sensing, Osaka, Japan) colorimeter according to CIE standards, and the L, a*, and b* values were recorded (Yılmaz et al., 2015).

The samples' peroxide values were determined according to method Cd 8–53 (AOCS, 1998).

Thermal Properties of the Oleogels

The samples of each oleogel (5–8 mg) were weighed into aluminum pans and sealed hermetically. The onset, peak temperatures, and enthalpies of both crystallization and melting were determined using a Perkin-Elmer 4000 Series Differential Scanning Calorimeter (DSC) (Groningen, The Netherlands). The temperature program was as follows: heating the samples from 20 to 100 °C at 10°C min^{-1} rate; cooling the samples to −30 °C at 10°C min^{-1} rate, and keeping the samples for 3 min at that temperature for full

crystal formation; finally, heating the samples again to 100 °C at 5°C min^{-1} heating rate. The parameters of the analysis were calculated using Pyris 1 Manager Software on the instrument (Dassanayake et al., 2009; Yılmaz et al., 2015).

Structural Properties of the Oleogels

Morphologies of the oleogel samples were visualized with an Olympus CX31 polarized light microscope (PLM, Olympus Optical Co., Japan) equipped with a CCD Canon color camera. First, the oleogel samples were melted in a water bath at 90 °C for 30 min, and then one drop of melted sample was put on a glass microscope slide and covered by a slip. The sample was kept for one night in a refrigerator and then 2 hours at room temperature before viewing under the microscope at room temperature (23 ± 2°C), controlled by an air conditioner. The best images among the replicates were selected (Yılmaz et al., 2015).

The X-ray diffraction patterns of the oleogel crystals were analyzed with a PANalytical Empyrean model (The Netherlands) X-ray diffractometer according to method Cj 2–95 (AOCS, 2012). The samples at ambient temperature (23 ± 2°C) were loaded to samples holder, and then angular scans (2θ) were performed from 2.0° to 50° by 2°/min scan rate with a Cu source X-ray tube (λ = 1.54056 Å, 40 kV and 40 mA). Data analysis was completed with X'Pert HighScore Plus software (Malvern Panalytical Ltd., Royston, UK) (Yılmaz et al., 2015).

Rheological Analyses of the Oleogels

A DHR 2 rheometer (TA Instruments, USA) was used with cross-hatched parallel plate geometry ($\phi = 40$ mm, gap 0.9 ± 0.1 mm), and a Peltier system ($\pm 0.1°C$) under the lower plate to assess the rheological properties of the oleogel samples. For each oleogel sample, first amplitude sweep (strain = 0.01–100%) tests were performed at 10°C with 1 Hz frequency to determine the linear viscoelastic region (LVR). The LVR was observed from the stress sweeps as a plateau for the storage (G´) and loss (G″) modulus. Within the LVR, the samples could be measured in the nondestructive range, and for valid rheological data, measurements must be completed within the LVR (Mezger, 2014). After determining the strain within the LVR, frequency sweep tests were completed for each sample at 10 °C with the LVR strains determined (0.02–0.26%) and frequencies from 0.1 to 100 Hz. For each sample, the loss factor (tan δ) was followed against the applied frequency values.

To determine the structural recovery abilities of the oleogels, a time sweep test was applied. In this test, time-dependent viscoelastic behavior was determined with shearing under constant dynamic conditions. Three strain gradient regions were selected and applied to each sample at 10°C with 1 Hz frequency. To simulate the resting condition, strains at LVR for each sample were applied at the first time region for 180 s. The LVR strains determined previously with the amplitude sweep tests were 0.023, 0.038, 0.039, 0.267, 0.061, and 0.064% for the SWO, TWO,

RWO, OWO, RsWO, and BWO samples, respectively. Then, to make a structural breakdown in the samples, a strain value of 1.0% constant for each sample was applied for another 180 s to simulate the force-related breakdown (higher force region ensuring the $LVR_{strain} \leq$ Strain condition) region. Finally, a strain of 0.001% constant for each sample was applied for 900 s to simulate the structural-recovery (lower force region, ensuring $LVR_{strain} \geq$ Strain condition) region. This test provided graphics about the samples' structural recovery abilities exposed to stress and released from the stress.

Lastly, a temperature ramp test was carried out from 0°C to 80 °C by 1°C min^{-1} rate at 1 Hz frequency within the linear viscoelastic region. The soak time was 120 s for all. All samples were tested three times, and the results were presented as the average values.

Descriptive Sensory Analysis of the Oleogels

Quantitative Descriptive Analysis (QDA) was used to describe the oleogel samples sensorially (Meilgaard et al., 1991; Yılmaz et al., 2015). There were seven female and four male panelists aged between 22 and 47 incorporating voluntarily into the study. A consent form was signed, indicating that the samples were edible and safe. The panel was trained under the moderation of a panel leader for at least 10 hours to select, define, and score the sensory descriptive terms. The panel was developed in seven different sensory description terms to assess the samples. The sensory terms, their definitions, and references are listed in Table 1. Different hard fats, margarines, wax oleogels, and some foodstuffs were used to train the panelists. A 10 cm line scale anchored from 0 at the left end for minimum intensity to 10 at the right end for maximum intensity was used to quantify the sensory attributes for each sample. In each session, three randomly selected samples coded with 3-digit numbers were served to the panel. All tests were carried out at room temperature under daylight, and the panelists were provided with water, bread slices, apple slice, and an expectoration cup. Duplicate samples were analyzed in different sessions held in different days for each of the replicates of oleogel production.

Statistical Analysis

In this study, the oleogel production was replicated two times. All analyses were completed for both replicates, and all samples were analyzed at least in duplicate. All data were given as mean values with SD. The ANOVA with comparisons of means by Tukey's test was completed for data analysis. Statistical analysis was performed with Minitab v.16.1 software (Minitab, 2010). The level of confidence was at least 95%.

Table 1 The panel defined sensory descriptive terms, their definitions, and references

	Definition	References
Hardness	Force required to push a knife into the sample	Min: Yoghurt Max: Tallow
Spreadability	Easiness to spread sample over a bread loaf	Min: Chewing gum Max: Cream cheese
Liquefaction	Amount of liquid oil once the sample was spread on a bread loaf	Min: Tallow Max: Sunflower oil
Rancid	Aromas associated with oxidized oil	Min: Fresh sunflower oil Max: Sunflower oil stored 4 days at 60 °C
Waxy	Aromas associated with waxes	Min: Absent Max: Shred sunflower wax
Cooling	Cold feeling inside mouth	Min: None Max: Menthol candy
Other	Tea aroma/Bitterness/Rose aroma	Min: Absent Max: Green tea/caffeine solution (0.1%)/rose water

Results and Discussion

Physicochemical Properties

The six different plant wax-based oleogels developed in this study are shown in Fig. 1. Some physicochemical properties of the oleogel samples are presented in Table 2. The minimum gelation concentrations (C*, %) of sunflower wax oleogel (SWO), tea wax oleogel (TWO), rapeseed wax oleogel (RWO), orange peel wax oleogel (OWO), rose wax oleogel (RsWO), and berry wax oleogel (BWO) were determined as 1.0%, 7.0%, 25.0%, 15.0%, 8.0%, and 5.0%, respectively. The addition levels of the waxes in those oleogels were at the minimum gelation concentrations (C*) for RWO and OWO (25% and 15%, respectively) and at 10% (w/w) for the rest. The C* (%) of the waxes indicated that sunflower wax (SW) could form stable oleogel at a 1% addition level, while it was 25% for the rapeseed wax (RW). The C* values of the SW and berry wax (BW) were found quite closer to the values reported previously (Mattice and Marangoni, 2018; Patel, 2016), while tea wax (TW), rapeseed wax (RW), orange peel wax (OW), and rose wax (RsW) were given in this study. Mattice and Marangoni (2018) reported the C* values of SW and BW as 0.5% and 6%. These values are lower than those found in this study, but it must be known that the source and purity of the waxes could be different. Clearly, C* values of RW and OW are quite high to be practically usable as organogelators. This might be due to the nature and composition of the waxes or due to their level of purity. Actually, plant waxes with high molecular weight fatty acid and alcohol esters and n-alkanes were shown to have relatively lower C* values (Mattice and Marangoni, 2018). In this study, the molecular species compositions of the studied waxes were not analyzed, nor did the producers provided the data. Hence, research need is identified. At this stage, RW and OW do

not seem appropriate candidates for oleogel preparation. Blake and Marangoni (2015) indicated that if wax has C* around 1–2% and its oleogels are prepared at a 5% addition level, the gel could have too much solid character rather than gel. This situation was observed truly for SWO, but to a lesser extent for TWO, BWO, and RsWO. Since the C* of RSO and OWO were high, we selected a 10% addition level for the rest to make a more meaningful comparison. It was previously (Mattice and Marangoni, 2018; Yılmaz et al., 2015) suggested that wax addition levels should not exceed 10 wt% in wax oleogels. Higher organogelator addition levels would create some sensory defects in products and some possible health consequences during long-term consumption. Further, the sustainability and cost of the wax sources must be considered.

The gelation times (GT) at the ambient temperature indicated (Table 2) that SWO was formed at 0.3 min, while RWO formed after 205 min. Therefore, this prolonged gel formation time would also restrict the usability of RW as an organogelator. GT would depend on the organogelator type and addition level, melting temperature and duration, cooling rate (if any), formation with cooling or ambient temperature setting, and oil type used (Co and Marangoni, 2012). A very long GT would not be preferable, especially during actual commercial productions, but moderate GT with low C* values and high gel stability would be preferable for applying a candidate gelator.

The oil binding capacities (OBC) of the waxes were above 99% (Table 2), and in agreement with previous literature for the plant waxes (Mattice and Marangoni, 2018; Patel, 2016; Toro-Vazquez et al., 2007). It was stated that OBC generally depends on the microstructure (wax crystal surface area and pore size) and intermolecular interactions leading to adsorption of liquid oil on available surfaces (Mattice and Marangoni, 2018). These findings agree with previous reports (Mattice and Marangoni, 2018; Patel, 2016)

Table 2 Some physicochemical properties of the plant wax oleogels

	Minimum gelation concentration (C*, %)	Gelation time (min)	Oil binding capacity (%)	Solid fat content (%, 20 °C)	L	a*	b*	Peroxide value (meqO$_2$/kg)
SWO	1.0 ± 0.0e	0.3 ± 0.02f	100.0 ± 0.0a	6.5 ± 0.5a	73.5 ± 0.0a	−2.8 ± 0.0d	5.9 ± 0.5c	5.7 ± 0.1c
TWO	7.0 ± 0.5c	13.7 ± 3.7e	99.8 ± 0.0a	5.3 ± 0.4b	18.1 ± 3.3f	1.5 ± 0.1b	6.5 ± 0.3c	21.8 ± 1.0a
RWO	25.0 ± 1.5a	205.2 ± 0.5a	99.4 ± 0.0a	7.2 ± 1.6a	58.1 ± 0.5b	−4.4 ± 0.0e	−2.1 ± 0.0d	5.2 ± 0.0c
OWO	15.0 ± 0.5b	30.2 ± 0.0c	100.0 ± 0.0a	3.4 ± 0.2c	43.7 ± 1.0d	7.7 ± 2.2a	20.4 ± 2.1a	11.1 ± 0.2b
RsWO	8.0 ± 0.0c	27.3 ± 0.3d	99.9 ± 0.0a	3.3 ± 0.5c	51.4 ± 3.5c	−4.8 ± 0.4e	15.8 ± 3.0b	10.3 ± 0.6b
BWO	5.0 ± 0.5d	53.0 ± 4.5b	99.9 ± 0.0a	2.9 ± 1.6c	36.2 ± 3.5e	−1.7 ± 0.4c	4.6 ± 0.9c	10.7 ± 2.2b

Values represent mean of replicates ± SD and letters represent significant differences among the samples ($P \leq 0.05$). For each replicate, 100 g of samples were produced.

BWO, berry wax oleogel (10%); OWO, orange peel wax oleogel (15%); RsWO, rose wax oleogel (10%); RWO, rapeseed wax oleogel (25%); SWO, sunflower wax oleogel (10%); TWO, tea wax oleogel (10%).

WILEY AOCS

that wax crystals are quite good at immobilization of almost all free liquid oil in an oleogel structure. The new waxes were found quite useful enough for their oil binding abilities.

The CIE color values (L, a*, b*) of the oleogels were also measured (Table 2, Fig. 1). Clearly, there is a significant variation among the samples. Since the same liquid oil (sunflower oil) was used to create the oleogels, this color difference is due to the waxes used. Since TWO has a dark color, it might create some problems for food applications. Likewise, OWO and RsWO present observable color tones, and care must be taken during their use. Sometimes, a particular color could also be preferable in some food products, like cakes or meat products.

Peroxide values of the oleogels are presented in Table 2. The lowest values (5.2 and 5.7 meqO$_2$ kg^{-1}) were in the RWO and SWO, and the highest value was in TWO (21.8 meqO$_2$ kg^{-1}). Since the same liquid oil was used and the same conditions were applied during oleogel preparation, this variation must be due to the organogelators added. Since the waxes were used as provided by the manufacturers, their oxidative stability or presence of oxidable components in them depended on the wax type, purity, and storage conditions. Further, different waxes might have different components susceptible to oxidation during the oleogelation process, in which the oil: wax blend heated to 90 °C and mixed for 30 min, and stored overnight at room temperature. It could also be possible that some waxes might have pro-oxidant components that promote unsaturated fatty acids oxidation in the liquid oil used. In our previous study (Yılmaz et al., 2015), sunflower and beeswax oleogels were shown to be quite stable against oxidation during storage. In this study, PV were measured only in the fresh samples. Overall, the TWO sample seems not acceptable due to its very high PV. Further, the nitrogen gas atmosphere and/or vacuum could be utilized during oleogel preparation to control oxidation.

Thermal Properties

Crystallization and melting onset and peak temperatures and enthalpies were measured both in the oleogel samples and in the waxes used as the organogelators (Table 3). The order of crystallization peak temperatures for the waxes was SW > TW > RsW > OW > BW > RW, while the peak melting temperature order was SW > TW > RsW > BW > RW > OW. Further, TW, OW, and BW had two different fractions having two different crystallization and melting peak temperatures. The higher values were selected for sorting. The corresponding oleogels had lower peak melting and crystallization temperatures than those of the waxes, respectively. Among the oleogels, the crystallization and melting peak temperatures were ordered as SWO > TWO > RsWO > OWO > BWO > RWO, and SWO > TWO > RsWO > BWO > RWO > OWO, respectively (Table 3). Obviously, higher melting point waxes yielded higher melting point oleogels. The melting range and peak temperature of an oleogel could be designed by selecting the wax type and addition level according to the purpose of the application. Nevertheless, other factors like legal issues, sensory appropriateness, availability, and price must also be considered.

Thermal properties of structured fats, including oleogels, would be of great importance for technological process designs related to heat exchange operations, products storage and distribution facilities, and others. Since different plant waxes used in this study might most probably contain different components at different proportions, their own thermal behavior were quite divergent, and hence the oleogels produced from them hereof. Generally, a wide melting range (onset to peak temperatures) was evident for almost all waxes, indicating that they had complex chemical nature. The melting range of the oleogels was rather less wide than the waxes, possibly due to the decreased proportion of the wax in the total oleogel composition. When the peak temperature difference between the melting and crystallization temperatures of the oleogel samples was considered, the largest difference was observed in RWO samples (13.99 °C). Likewise, the lowest difference was in RsWO sample (2.86 °C). As the melting and crystallization peak temperature difference extends, a hysteresis problem might occur in the prepared oleogel, indicating a loose gel structure with some liquid oil leakages on the surface. The phenomenon was explained as the suppression of the visualization of endothermic melting due to crystal network disintegration and melting of solid particles during the heating process (Co and Marangoni, 2012; Mattice and Marangoni, 2018). Especially during oleogel storage, the development of hysteresis is not desired, and waxes with lower melting ranges could be preferred in oleogel production.

Structural Properties

The microstructure of the plant wax oleogels is shown in Fig. 2. In these polarized light microscopy (PLM) images, wax crystals generally appear as white, bright scenes, while liquid oil is observed as dark surroundings. As shown in Fig. 2, there was a crowd of wax crystals, since the waxes' addition level was at least 10% by weight. It was quite hard to identify the wax crystal morphology due to the large, dense aggregates of the crystals. These pictures were taken under static conditions and only on the fresh samples with the given addition levels. Dassanayake et al. (2009) presented spherulitic wax crystals for carnauba wax, while needle-like crystals for sunflower wax were observed in olive oil oleogels by Hwang et al. (2012). Further, it was claimed that fiber-like, smaller needle crystals form

Table 3 Thermal properties of the plant waxes and the oleogels developed from the plant waxes

		Crystallization			Melting		
		Onset$_c$ (°C)	Peak (T$_c$) (°C)	ΔH$_c$ (J g^{-1})	Onset$_m$ (°C)	Peak (T$_m$) (°C)	ΔH$_m$ (J g^{-1})
Waxes	SW	73.80 ± 0.01	67.95 ± 0.24	−199.79 ± 4.38	69.39 ± 0.03	78.26 ± 0.12	193.13 ± 5.11
	TW-Fr.1	45.30 ± 0.06	42.54 ± 0.09	−3.04 ± 0.30	35.31 ± 0.16	44.13 ± 0.35	4.83 ± 0.23
	TW-Fr.2	57.71 ± 0.97	53.63 ± 0.01	−11.22 ± 0.66	48.93 ± 0.18	61.62 ± 0.82	14.49 ± 0.58
	RW	17.65 ± 0.12	16.07 ± 0.62	−6.03 ± 0.79	14.45 ± 0.61	29.73 ± 0.13	80.47 ± 3.00
	OW-Fr.1	11.42 ± 0.17	9.19 ± 0.11	−6.31 ± 0.01	6.57 ± 2.77	13.92 ± 0.23	4.42 ± 0.10
	OW-Fr.2	40.25 ± 0.09	37.33 ± 0.12	−4.94 ± 0.27	18.22 ± 0.15	21.42 ± 0.23	1.57 ± 0.11
	RsW	52.54 ± 0.39	46.71 ± 0.21	−148.91 ± 5.25	30.23 ± 0.30	50.12 ± 0.23	151.99 ± 1.54
	BW-Fr.1	14.88 ± 0.86	12.60 ± 1.39	−10.31 ± 0.33	8.63 ± 1.22	14.47 ± 0.32	9.78 ± 2.41
	BW-Fr.2	36.81 ± 0.13	34.18 ± 0.04	−61.95 ± 3.86	39.94 ± 0.01	44.15 ± 0.19	48.94 ± 4.56
Oleogels	SWO	63.23 ± 0.16	61.70 ± 0.12	−17.34 ± 0.09	53.85 ± 3.36	65.07 ± 0.01	18.63 ± 0.18
	TWO-Fr.1	41.62 ± 0.52	39.26 ± 0.47	−1.44 ± 0.18	37.22 ± 2.72	42.33 ± 0.11	2.11 ± 2.33
	RWO	6.88 ± 0.21	3.24 ± 0.12	−7.35 ± 1.10	11.26 ± 0.12	17.23 ± 0.23	11.11 ± 0.55
	OWO-Fr.2	19.73 ± 0.03	19.30 ± 0.21	−0.05 ± 0.01	6.76 ± 0.38	8.27 ± 0.01	0.06 ± 0.05
	RsWO	36.42 ± 0.11	33.91 ± 0.25	−5.39 ± 1.23	23.42 ± 2.81	36.77 ± 0.02	4.58 ± 2.50
	BWO-Fr.2	18.99 ± 0.01	17.02 ± 0.11	−5.17 ± 0.34	13.91 ± 1.53	22.58 ± 0.01	4.96 ± 0.74

BW, berry fruit wax; BWO, berry wax oleogel (10%); Fr.1, fraction one; Fr.2, fraction two; OW, orange peel wax; OWO, orange peel wax oleogel (15%); RsW, rapeseed wax; RsWO, rose wax oleogel (10%); RWO, rapeseed wax oleogel (25%); SW, sunflower wax; SWO, sunflower wax oleogel (10%); TW, tea wax; TWO, tea wax oleogel (10%).
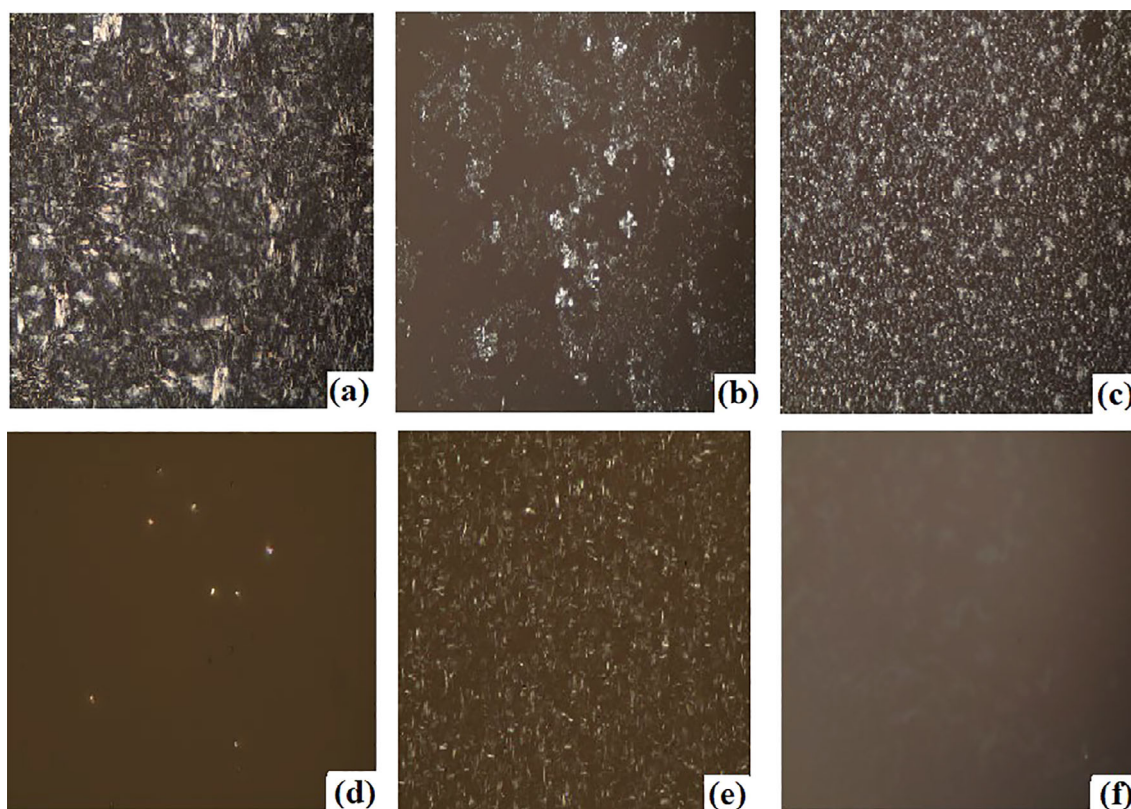

Fig 2 Polarized light microscopy (PLM) images of the oleogels (a. SWO, b. TWO, c. RWO, d. OWO, e. RsWO, f. BWO, with 40× magnification)

stronger, hard gels with higher oil binding capacities (Dassanayake et al., 2009). In our previous study (Yılmaz et al., 2015), different crystal morphologies with sunflower wax and beeswax oleogels were observed. Although, in this study, the PLM pictures could not permit us to observe the differences due to higher concentration, different morphologies with different waxes were possible. Also, cooling rate and shearing were shown to affect gel crystal morphology (Mattice and Marangoni, 2018). Further, it was indicated that smaller crystal sizes in large numbers produce smoother texture in solid fats, which were preferred during better palate perception (Sato, 2001). Blake and Marangoni (2015) used cryo-scan electron microscopy to image the microstructure of de-oiled wax oleogels. Contrary to previous findings, they found that rice bran, sunflower, and candelilla wax crystals were platelet-like in morphology as observed with cryo-electron microscopy, and PLM images in previous studies, including theirs, were misperception due to limited magnification. Further, they suggested that surface area and surface roughness might be the real factors contributing to the oil binding capacity. Although we could not manage to make cryo-electron microscopy, the oleogels in this study were observed quite smooth and uniform by eye observation and rheological analyses and included densely packed small crystal aggregates.

The X-ray diffraction patterns of the oleogel samples are presented in Fig. 3. The short spacing corresponding to different wide-angle peaks could be observed in Fig. 3. The SWO presented 3.69, 4.05, and 4.42 Å peaks; the TWO showed 3.65, 4.05, and 4.60 Å peaks; the RWO sample had 3.69 and 4.47 Å peaks; the OWO sample included 4.08 and 4.49 Å peaks; RsWO sample showed 3.72, 4.12, and 4.43 Å peaks; and finally BWO had 4.51 Å peak with different intensities, respectively. It was acknowledged that the diffraction pattern obtained from a sample is the combination of all molecules found in the sample (AOCS, 2012). Hence, in these samples, the patterns result from all crystals formed by the added waxes, including different chemical components and sunflower oil triglycerides. According to the AOCS method (AOCS, 2012), if a sample shows a peak at 4.2 Å, its polymorphic form is α, if a sample contains 3.8 and 4.2 Å peaks, it must be β′ polymorph, and if the peak appears at 4.6 Å position, then it should be of β polymorphic form. Stahl et al. (2017) stated only one different short spacing value as 4.15 Å for α form than those given in the AOCS method. Although there were no exact matches, all oleogel samples seem to fit the β′ polymorph type with their observed peaks, except the BWO sample. BWO sample had a single peak at 4.51 Å, and should have β polymorph crystals. In fact, in the study of Doan et al. (2017), a single peak at 0.415 nm was observed for the berry wax oleogels (BWO) with rice bran oil. It was

also indicated (Mattice and Marangoni, 2018) that berry wax includes over 95% of free fatty acids and forms hexagonal crystals. The X-ray data presented in this study agree with previous studies (Doan et al., 2017; Mattice and Marangoni, 2018). Differences for peak angles and intensities might be due to component differences like carbon chain length and proportions of different components of the added waxes. The peaks around 3.70 and 4.10 Å were observed for candelilla, carnauba, and rice bran wax oleogels with olive oil (Dassanayake et al., 2009). Similar peaks located near the β′ polymorph position were observed in our previous studies with sunflower and beeswax oleogels (Yılmaz et al., 2015). Further, the X-ray pattern graphs (Fig. 3) might also indicate the presence of amorphous triglycerides adhered to the surface of the crystals due to the broadened pattern observed in the peaks. Generally, β′polymorphic form of fat crystals are preferred for dispersibility, smooth texture, optimum melting, and good mouthfeel, among the other polymorphs for margarine and other spreadable fat products, while β polymorph is the preferred one for chocolate, confectionery, and shortening fats (Chrysam, 1996). Most of the wax oleogels prepared in this study could be used in margarine and spreadable fat products with respect to their crystalline form.

Rheological Properties

The amplitude sweeps were completed to describe the deformation behavior in the nondestructive range. This range is known as the linear viscoelastic region (LVR) and identified as the constant or plateau strain values on the storage modulus (G′) graphs before deformation occurs. This linearity limit strain values determined for each sample must be used for all other rheological measurements for correct analyses (Mezger, 2014). The determined LVR strain values for the SWO, TWO, RWO, OWO, RsWO, and BWO samples were 0.023, 0.038, 0.039, 0.267, 0.061, and 0.064%, respectively. Further, the sinusoidal response signals (phase-shift angle) were between 0° and 45° during the oleogel samples' measurements, indicating a true gel-like state. It is well known that the sine curves of the preset parameters and the measuring result show a time lag for the response signal, and it always takes values between 0° and 90°. A phase shift value of 0° indicates ideally elastic, a 90° value indicates ideally viscous, and values between 0° and 45° indicate gel-like deformation behavior (Mezger, 2014).

The oscillatory frequency sweep tests describe a sample's time-dependent behavior in the nondestructive range (LVR) to simulate fast motion on short timescales with high frequencies or slow motion on long timescales (or at rest) with low frequencies. In practice, frequency sweep
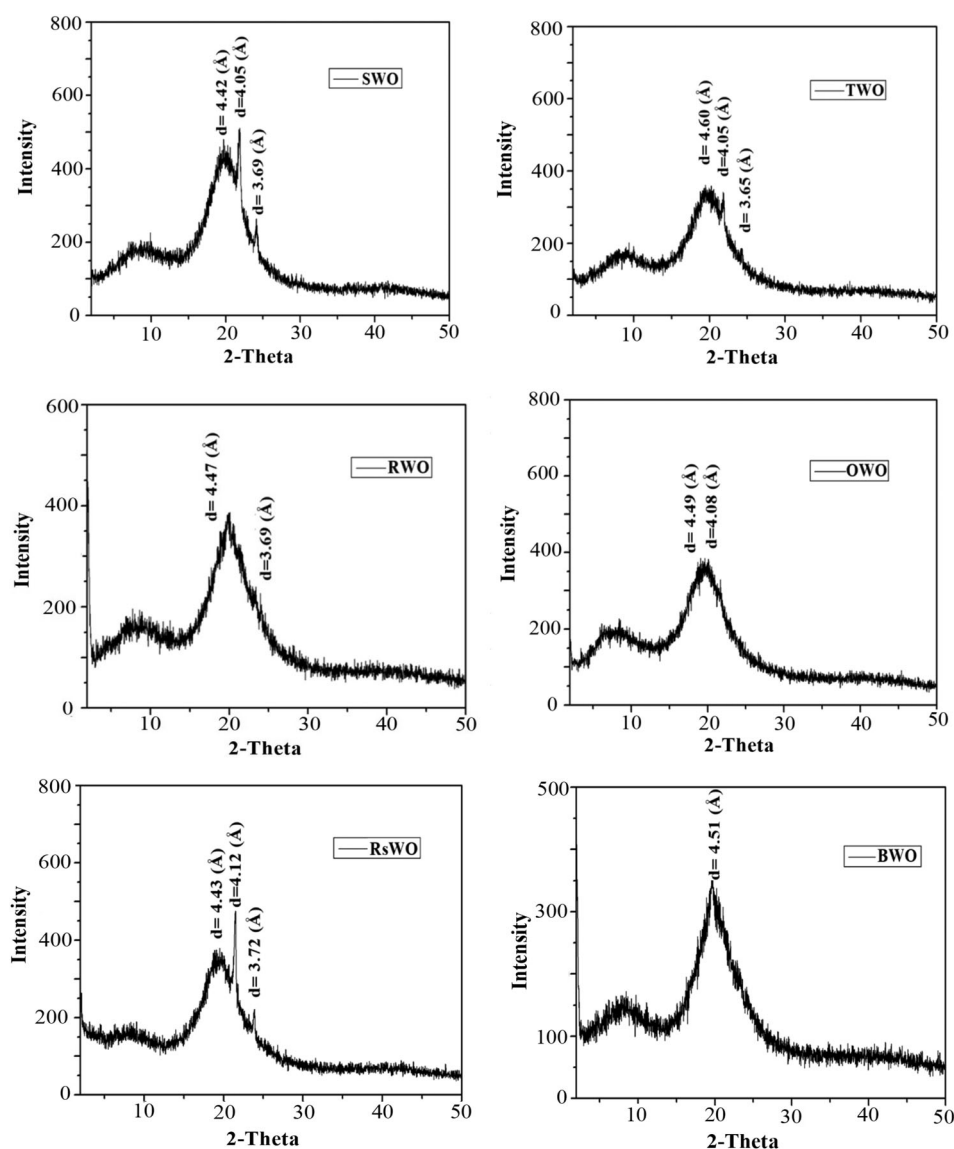
Fig 3 The X-ray diffraction patterns of the oleogel samples (BWO, berry wax oleogel; OWO, orange peel wax oleogel; RsWO, rose wax oleogel; RWO, rapeseed wax oleogel; SWO, sunflower wax oleogel; TWO, tea wax oleogel)

tests provide information about inner structure and behavior, and long-term stability for the gels (Mezger, 2014). The angular frequency range of 1–1000 rad s^{-1} were applied to the samples, and the loss factor or damping factor (tan δ) values were measured (Fig. 4). The loss (damping) factor describes the ratio of the two portions of the viscoelastic behavior (tan δ = G″ / G′). G′ (storage modulus) represents the elastic portion of the viscoelastic behavior and describes the sample's solid-like properties, while G″ (loss modulus) characterizes the viscous portion, which describes the liquid-like properties of the sample. In fact, these two parameters (G′ and G″) are the two components of the complex viscosity, which describes the entire viscoelastic behavior. These two parameters were

determined by measuring the stored deformation energy (for G′) and the deformation energy dissipated through the internal flow of the sample (for G″). It was also well recognized that for the gelled state, it should always be G′ > G″ (Mezger, 2014). As could be observed from Fig. 4, the samples' loss factor values were lower than zero, and between −1 and −7, which indicates the gelled state. In fact, throughout the applied frequency range, the tan δ values were always below zero, or always the storage modulus values (G′) were higher than the loss modulus (G″) values. This means that the gels' solid-like properties remain against the enhanced deformation force, indicating possible storage stabilities of the samples. But, the samples were found as not strong gels since their loss factor (tan δ)
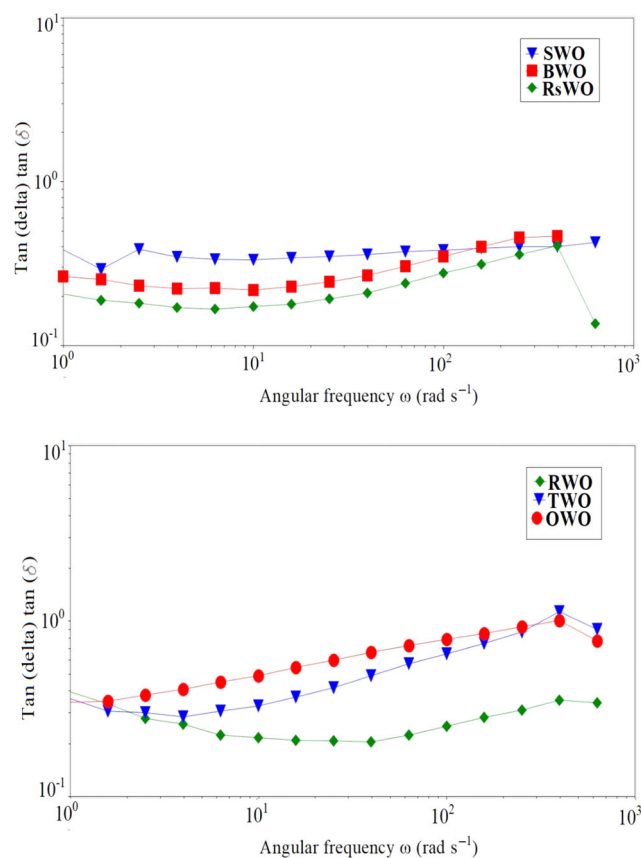
WILEY AOCS

Fig 4 Frequency sweep test results of the oleogel samples (BWO, berry wax oleogel; OWO, orange peel wax oleogel; RsWO, rose wax oleogel, RWO, rapeseed wax oleogel; SWO, sunflower wax oleogel; TWO, tea wax oleogel)

Fig 5 Time sweep test results of the oleogel samples (BWO, berry wax oleogel; OWO, orange peel wax oleogel; RsWO, rose wax oleogel; RWO, rapeseed wax oleogel; SWO, sunflower wax oleogel; TWO, tea wax oleogel)

values were not between 0.2 and 0.3, which indicates resistance to syneresis. Further, the order of measured G′ values from the highest to the lowest was SWO > RWO > RsWO > BWO > OWO > TWO, respectively, indicating that SWO had the highest stiffness, and TWO had the lowest stiffness. Overall, data from Fig. 4 indicate that these oleogel samples are genuinely in a gelled state, and have certain storage stability levels, but on more extended storage periods, syneresis may also develop. These rheological properties of the studied wax gels seem similar to the reported values obtained for the SW and fruit wax (FW) oleogels in the literature (Mattice and Marangoni, 2018).

Time-dependent oscillatory tests (time sweep tests) were also conducted (Fig. 5). In all oleogel samples, in the first region (simulating resting behavior at LVR), the complex viscosity (η*) values were between 200 and 40.000 Pa, and the storage modulus (G′) values were higher than those of the loss modulus (G″) values, indicating the presence of proper gel structure. In the second region, strong shear (1.0% strain) was applied to simulate structural breakdown,
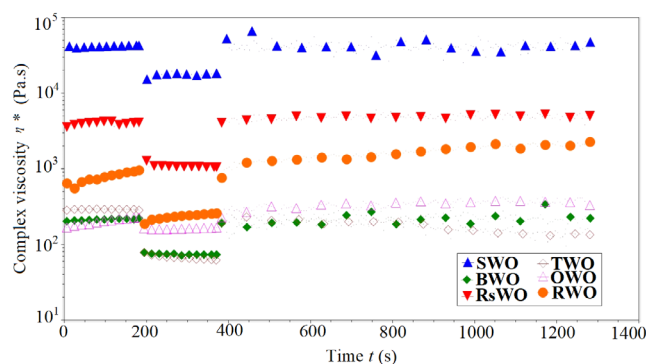
and complex viscosity in all samples were decreased significantly, indicating structural breakdown and loss of gelled consistency. Clearly, enough deformations have occurred due to the applied force. Finally, in the third region, after removing the strong force (strain was reduced to 0.001%), the structural recovery was observed with the increasing complex viscosity values, clearly observed in all samples (Fig. 5). This means that a structural reformation or regeneration was developed in the samples. Since complex viscosity values in the third region increased again to the levels observed in the first (resting) region, it could be easily said that full structural recoveries were present for these oleogel samples. This type of complete time-dependent recovery of the initial state upon reduction of the load is called thixotropic behavior. This behavior was observed for sunflower, candelilla, carnauba, rice bran, and fruit wax oleogels in previous studies (Mattice and Marangoni, 2018; Patel, 2016). The thixotropic behavior observed in the wax oleogels could be important and desired property in product applications in formulated food products where mixing, whipping, or other mechanical breakdown operations are unavoidable. After the unit operation, regeneration of the gelled state in the fat phase might provide the intended solid fat functionalities (Patel, 2016).

To observe the flow behavior of the oleogels at different temperatures, temperature ramp tests were also conducted under constant amplitude and frequency (Fig. 6). This test shows the temperature point where the gelled structure no longer continues at the G′ = G″ crossover point. Fig. 6 shows that at around 70 °C, the SWO melted completely, and the gel state was lost. Similar phenomena were observed for TWO at around 60 °C, for RWO at around 30 °C, for OWO at around 44 °C, for RsWO at around 50 °C, and for BWO at around 40 °C. The peak melting temperatures (Tm) measured by DSC (Table 3) were usually lower than those observed at the temperature
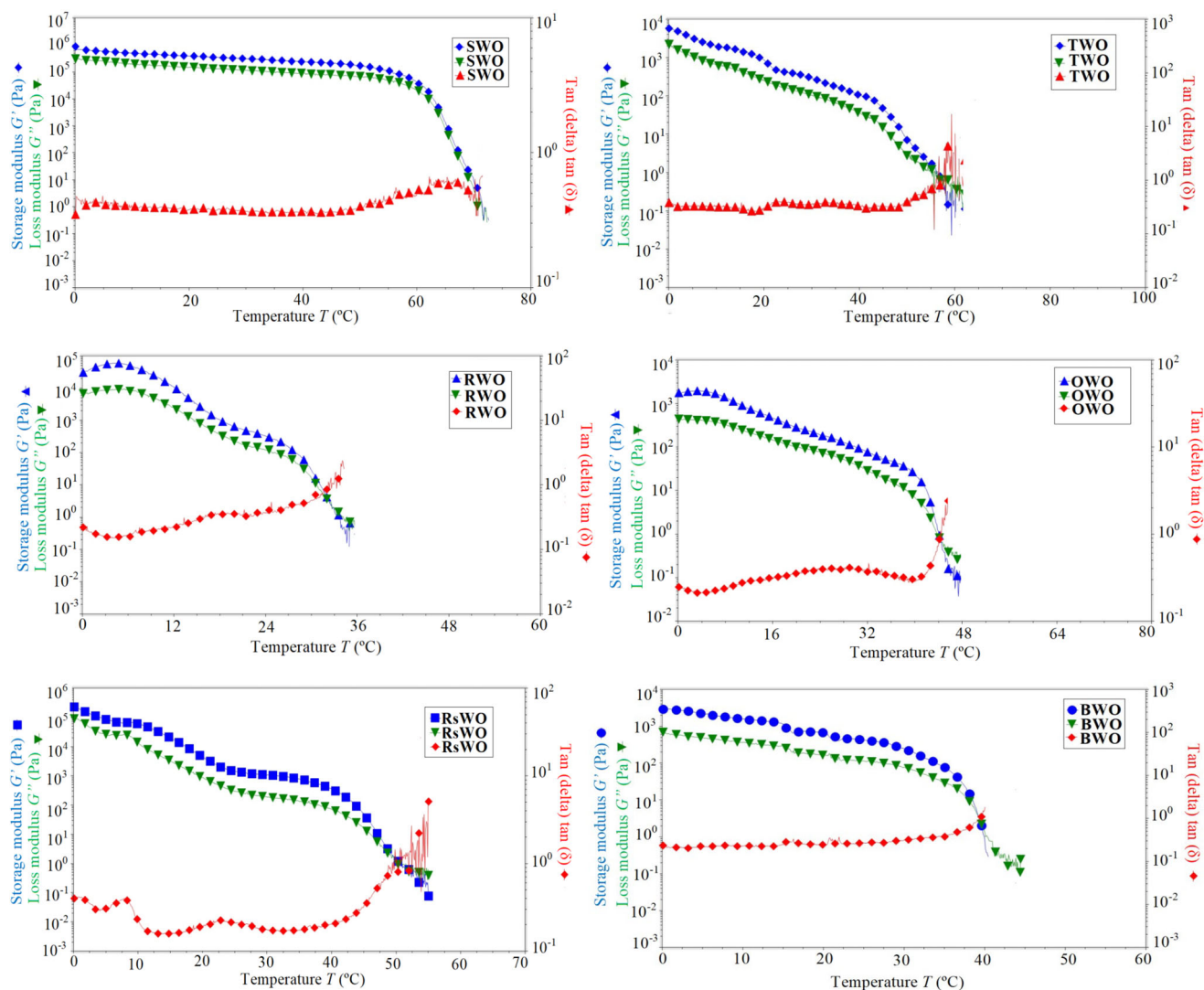
Fig 6 Temperature ramp test results of the oleogel samples (BWO, berry wax oleogel; OWO, orange peel wax oleogel; RsWO, rose wax oleogel; RWO, rapeseed wax oleogel; SWO, sunflower wax oleogel; TWO, tea wax oleogel)

ramp tests' crossover point. It could be probable that during these temperature tests, some junction zones remained until higher temperatures, and the instrument recognized the still present storage modulus values. Further, the damping temperature (tanδ line) of all samples crosses the storage modulus just before the crossover point ($G' = G''$), indicating that the glass transition temperatures of the oleogels were a few degrees below the melting temperature. These data could be important during the decision-making of oleogel applications in the heat processing of foods. Clearly, SWO was quite heat-stable, followed by TWO and RsWO samples.

Sensory Properties

The quantitative descriptive analysis (QDA) was applied to the oleogel samples, and the findings are summarized in

Table 4. The panel determined, defined, and used seven sensory descriptors to define the oleogel samples. The "hardness" was defined as the total force required to push a knife inside the sample by hand. Yoghurt was given for the lowest score (0) standards, while tallow was given for the highest score (10) standard. Significant differences were determined among the samples, and SWO had the hardest score (8.8), while TWO and OWO samples showed the lowest hardness (1.2 and 1.2) values. These sensory data fit very well with the rheological data, which indicated the highest G' value for SWO, and the lowest G' values for TWO and OWO samples. It could also be recalled that OWO was produced by adding 15% (w/w) wax, while the addition level was 10% in the SWO sample. Eventually, the behaviors of different waxes determined by rheology and sensory analysis were congruent. "Spreadability" was

WILEY AOCS

Table 4 Sensory quantitative descriptive analysis (QDA) test results of the oleogels

	SWO	TWO	RWO	OWO	RsWO	BWO
Hardness	8.8 ± 1.3a	1.2 ± 1.0c	2.6 ± 2.0b	1.2 ± 1.0c	3.2 ± 2.0b	2.2 ± 1.4b
Spreadability	6.5 ± 2.3b	8.8 ± 1.3a	8.5 ± 2.1a	8.8 ± 1.5a	8.0 ± 2.3a	8.7 ± 1.3a
Liquefaction	1.2 ± 1.7c	3.8 ± 1.3a	4.1 ± 2.1a	4.8 ± 2.4a	2.7 ± 2.5b	2.8 ± 1.5b
Rancid	1.2 ± 0.3c	3.2 ± 3.1b	0.7 ± 0.8d	5.2 ± 1.3a	2.3 ± 2.6b	2.6 ± 2.2b
Waxy	5.1 ± 2.9a	4.3 ± 3.3a	2.8 ± 2.5b	3.3 ± 3.0ab	3.5 ± 2.6ab	4.0 ± 3.1a
Cooling	2.5 ± 2.0b	3.6 ± 1.9b	3.0 ± 1.9b	3.3 ± 2.6b	5.0 ± 2.9a	4.1 ± 2.7a
Other (tea aroma, Bitterness, rose aroma)	0.0 ± 0.0	7.2 ± 2.6	0.0 ± 0.0	9.2 ± 1.1	8.1 ± 1.8	0.0 ± 0.0

Values represent mean of the replicates ± SD and letter represents significant differences among the samples ($P ≤ 0.05$).BWO, berry wax oleogel (10%); OWO, orange peel wax oleogel (15%); RsWO, rose wax oleogel (10%); RWO, rapeseed wax oleogel (25%); SWO, sunflower wax oleogel (10%); TWO, tea wax oleogel (10%).

defined as the easiness of a sample to be spread on the surface of a bread loaf. Except for the SWO sample, the other samples' spreadabilities were not significantly different, and all were higher than that of the SWO (6.5) sample. Clearly, there is an adverse relationship between spreadability and hardness, respectively. Apparently, the oleogel samples were usually quite well spreadable, like cream cheese used as the standard for maximum spreadability value. The sensory attribute "liquefaction" was defined as the amount of melted fat when it was spread onto a surface. It might provide an insight into the inner kinetic stability of the sample. It measured the amount of total melted fat when the sample was spread on a bread loaf. The melting could be due to both kinetic and thermal energy input. The panelists observed the amount of melted fat after the sample was spread on bread loaf; hence, as the value enhances, the amount of melting increases. Clearly, the melting of the OWO and RWO samples was higher. But, there was no complete melting since none of the samples had a score of 10. In fact, once the energy input was ceased, the melted samples recovered again to a solid state, as observed in the time sweep rheological measurements (Fig. 5). This recovery behavior was observed in previously studied wax oleogels, as well (Mattice and Marangoni, 2018; Patel, 2016). To quantify the amount of oxidation perceived in the samples, the panel measured the "rancid" attribute (Table 4). The highest rancid score (5.2) was in the OWO sample, followed by the TWO samples (3.2), while the lowest value (0.7) was in the RWO sample. Since the same oil and the same preparation technique were used, the differences must be due to the added waxes. Further, the peroxide value results (Table 2) showed that the higher values were in the TWO and OWO samples. The panel successfully identified samples with higher and lower scores of rancid attribute closely related to their PV. All oleogel samples had some "waxy" attribute, which was defined as the aromas associated with paraffin or other waxes. This attribute was significantly higher in the SWO, TWO, and BWO

samples. This attribute must quite be dependent on the kind and addition level of the wax used as the organogelator. RWO was only possibly prepared at its C* value of 25% by total oleogel weight. Although at this highest addition level, the waxy aroma was the lowest in that sample. It was quite possible that the rapeseed wax (RW) used in this study may contain components that were not wax esters but some other higher melting molecules. "Cooling" was defined as the cool sensation of a sample once it melted in the mouth space. It could be related to the melting rate in the mouth temperature. When solid fats melt in mouth space, some heat is absorbed for melting, and a sensation of cooling is occurred due to the lost heat of the body (Chrysam, 1996). Except for the highest scores (5.0 and 4.1) measured in the RsWO and BWO samples, the rest was not significantly different. Since some different aromas sensed in some of the samples, an "other" term was selected and defined for the individual samples. The TWO samples had some tea aromas, and panel quantified it with a 7.2 score on 10 maximum value of the green tea reference. This clearly shows that the TWO samples have quite a significant tea aroma. This situation could limit its food applications, but at the same time could provide an advantage in specific products in which tea aroma is desired. Similarly, the OWO sample had quite a significant amount of "bitterness" as measured with a 9.2 score. In fact, it was well known that orange seed oil and orange peel (Aydeniz Güneşer and Yılmaz, 2019) contain very bitter citrus flavonoids. The orange wax produced from orange peel most probably contains these very bitter compounds. Consequently, food applications of OWO could be limited, but some other applications might arise in the future due to the citrus flavonoids, which are acquainted as functional compounds (Aydeniz Güneşer and Yılmaz, 2019). Lastly, RsWO included significant rose aroma (8.1 mean score) in comparison with pure rose water (10) as the reference. Depending on product type and consumer expectations, this aroma might also be preferable. Sensory studies with

oleogels are limited in the literature, and findings in this study might contribute significantly. The final success of any food products mainly depends on its sensory quality and consumer acceptance.

Conclusions

In this study, the properties of the oleogels of tea wax, rapeseed wax, orange peel wax, rose wax, and berry wax were determined and compared with sunflower wax oleogels. Since the literature lacks about the data of most of these plant wax oleogels, the knowledge provided in this study could be very important. The results showed that due to higher C* values of RW and OW, their oleogels could not be appropriate for food applications. RsWO, BWO, and, to some extent, TWO could have practical applications since their thermal and rheological behaviors were good enough, as determined and compared with SWO. The high PV measured in the TWO samples must also be resolved. The main conclusion of this study is that these new plant waxes show quite diverse quality profiles for their oleogels. If further purification could be done for these new waxes to get their good gelling, sensorially suitable versions by the wax producers, they might find commercial usage for oleogel production. Further, this study indicated that wax oleogels were good in rheological and thermal properties in general, and new, cheaper, safe, sensorially suitable plant wax sources must be researched and developed for the approaching industrial applications of the oleogels for commercialization.

Conflict of Interest The authors declare that they have no conflict of interest.

References

AOCS. (1998) *Official methods and recommended practices* (Vol. **I and II**, 4h ed.). Champaign, IL: American Oil Chemists' Society.

AOCS. (2012) *AOCS Official Method Cj 2-95. X-ray diffraction analysis of fats. Official methods and recommended practices of the AOCS, 6th Ed. 2011–2012 methods and additions and revisions.* Champaign, IL: American Oil Chemists' Society.

Aydeniz Güneşer, B., & Yılmaz, E. (2019) Comparing the effects of conventional and microwave roasting methods for bioactive composition and the sensory quality of coldpressed orange seed oil. *Journal of Food Science and Technology*, **56**:634–642.

Blake, A. I., & Marangoni, A. G. (2015) Plant wax crystals display platelet-like morphology. *Food Structure*, **3**:30–34.

Chrysam, M. M. (1996) Margarines and spreads. In Y. H. Hui (Ed.), *Bailey's industrial oil & fat products* (Vol. **3**). New York, NY: Wiley-Interscience.

Co, E. D., & Marangoni, A. G. (2012) Organogels: An alternative edible oil-structuring method. *Journal of the American Oil Chemists' Society*, **89**:749–780.

Dassanayake, L. S. K., Kodali, D. R., Ueno, S., & Sato, K. (2009) Physical properties of rice bran wax in bulk and organogels. *Journal of the American Oil Chemists' Society*, **86**:1163–1173.

Doan, C. D., Tavernier, I., Sintang, M. D. B., Danthine, S., Van de Walle, D., Rimaux, T., & Dewettinck, K. (2017) Crystallization and gelation behavior of low- and high melting waxes in rice bran oil: A case-study on berry wax and sunflower wax. *Food Biophysics*, **12**:97–108.

Hwang, H.-S., Kim, S., Singh, M., Winkler-Moser, J., & Liu, S. (2012) Organogel formation of soybean oil with waxes. *Journal of the American Oil Chemists' Society*, **89**:639–647.

Mattice, K. D., & Marangoni, A. G. (2018) New insights into wax crystal networks in oleogels. In A. R. Patel (Ed.), *Edible oil structuring: Concept, methods and applications* (pp. 71–94). Cambridge, England: Royal Society of Chemistry.

Meilgaard, M., Civille, G. V., & Carr, B. T. (1991) *Sensory evaluation techniques*. Boca Raton, FL: CRC Press.

Mezger, T. G. (2014) *Applied rheology*. Graz, Austria: Anton Paar GmbH.

Minitab. (2010) *Minitab statistical software (version 16.1)*. State College, PA: Author.

Okuro, P. A., Tavernier, I., Bin Sintag, M. D., Skirtach, A. G., Vicente, A. A., Dewettinck, K., & Cunha, R. L. (2018) Synergistic interactions between lecithin and fruit wax in oleogel formation. *Food & Function*, **9**:1755–1767.

Patel, A. R. (2016) Understanding the oil-gelling properties of natural waxes. *Inform*, **27**:17–20.

Rocha, J. C. B., Lopes, J. D., Mascarenhas, M. C. N., Arellano, D. B., Guerreiro, L. M. R., & daCunha, R. L. (2013) Thermal and rheological properties of organogels formed by sugarcaneor candelilla wax in soybean oil. *Foodservice Research International*, **50**: 318–323.

Sagiri, S. S., Samateh, M., & John, G. (2018) Biobased molecular structuring agents. In A. R. Patel (Ed.), *Edible oil structuring: Concept, methods and applications* (pp. 25–52). Cambridge, England: Royal Society of Chemistry.

Sato, K. (2001) Crystallization behaviour of fats and lipids: A review. *Chemical Engineering Science*, **56**:2255–2265.

Stahl, M. A., Buscato, M. H., & Grimaldi, R. (2017) Low sat-structured fats enriched in α-linolenic acid: Physicochemical properties and crystallization characteristics. *Journal of Food Science and Technology*, **54**:3391–3403.

Toro-Vazquez, J. F., Morales-Rueda, J., Dibildox-Alvarado, E., Charo-Alonso, M., Alonzo-Macias, M., & Gonzalez-Chavez, M. M. (2007) Thermal and textural properties of organogels developed by candelilla wax in safflower oil. *Journal of the American Oil Chemists' Society*, **84**:989–1000.

Yılmaz, E., Öğütcü, M., & Karagül Yüceer, Y. (2015) Physical properties, volatiles compositions and sensory descriptions of the aromatized hazelnut oil-wax organogels. *Journal of Food Science*, **80**: S2035–S2044.