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OPTIMIZATION OF WAX ADDITION LEVEL IN SUNFLOWER OIL OLEOGELS VIA RESPONSE SURFACE METHODOLOGY

Mustafa Öğütcü*[1], Elif Albayrak[2], Elif Sultan Karabayır[1]
[1] Department of Food Engineering, Faculty of Engineering, Çanakkale Onsekiz Mart University, Çanakkale, Türkiye
[2] Bayramiç Vocational College, Çanakkale Onsekiz Mart University, Bayramiç, Çanakkale, Türkiye

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ABSTRACT
In this study, oleogels were produced with a wax mixture (sunflower wax, SW; beeswax, BW) instead of single wax in order to reduce the total wax addition level and maintain the optimal properties of the oleogels. The textural and thermal properties were evaluated using the response surface methodology to determine the optimum wax addition level to form a gel with the similar properties to margarine. The firmness values and melting point were dominated by SW levels in the wax mixture. The XRD patterns showed that all of the samples had the β′ polymorphic form. The optimization results showed that oleogel prepared with 0.20%-SW and 2.80%-BW had lowest melting peak (46.42 °C). In conclusion, using a wax mixture instead of single wax type, a structurally stable gel with a lower melting point could be formed at a lower wax addition level.
Keywords: Oleogel, optimization, stability, thermal, wax mixture, response surface methodology

AYÇİÇEĞİ YAĞI OLEOJELLERİNDEKİ MUM İLAVE SEVİYESİNİN YANIT YÜZEY YÖNTEMİYLE OPTİMİZASYONU

ÖZ
Bu çalışmada, oleojellerin istenilen optimal özelliklerini korumak ve mum katkı miktarını azaltmak için tek mum yerine mum karışımı (ayçiçeği mumu, SW; balmumu, BW) kullanılarak oleojeller üretilmiştir. Tekstürel ve termal özellikler, margarine en yakın özelliklere sahip bir jel oluşturmak için optimum mum ilave seviyesini belirlemek üzere, yanıt yüzey yöntemi kullanılarak değerlendirilmiştir. Oleojel örneklerinin sertlik ve ergime noktası değerlerinin, daha çok mum karışımındaki SW oranına bağlı olduğu gözlemlenmiştir. XRD desenleri, tüm numunelerin β' polimorfik forma sahip olduğunu göstermiştir. Optimizasyon sonuçlarına göre, en düşük ergime noktasına (46,42 °C) sahip oleojelin %0,20SW ve %2,80BW ile hazırlanan oleojel olduğu belirlenmiştir. Sonuç olarak, tek mum yerine bir mum karışımı kullanılarak, daha düşük bir mum ekleme seviyesinde, daha düşük bir ergime noktasına sahip, yapısal olarak da kararlı bir jel oluşturulabileceği tespit edilmiştir.
Anahtar kelimeler: Oleojel, optimizasyon, stabilite, termal, mum karışımı, yanıt yüzey yöntemi

* *Correspondence author* / Yazışmalardan sorumlu yazar
✉: mustafaogutcu@gmail.com ☏: (+90) 286 218 0018 🖷: (+90) 286 218 0541

Mustafa Öğütcü; ORCID no: 0000-0001-8686-2768
Elif Albayrak; ORCID no: 0000-0002-1202-403X
Elif Sultan Karabayır; ORCID no: 0000-0003-1152-120X

INTRODUCTION

Organogel is a popular technique used for structuring liquid oils in food science, particularly in the last decade. When oils are structured via organogelation, the products are called oleogels. Oleogels range in appearance from opaque to transparent, and have different colors depending on the source of the oil and the gelators used. On the other hand, oleogels have been seen as spreadable fat alternatives due to some advantages such as low saturated fat, no *trans* fat and no change in fatty acid composition, and a wide melting and spreadability ranges (Co and Marangoni, 2012; Hwang et al., 2012; Hwang et al., 2013; Patel et al., 2014; Öğütcü et al., 2015). In order to structure oil, many gelators were reported, such as fatty alcohols, fatty acids, mono and diglycerides, γ-oryzanol and β-sitosterol mixture, and natural waxes. Particularly, wax-based oleogels are most popular and known oleogel structure. When compared to other organogelators, waxes have some advantages. They are inexpensive and relatively easy to obtain according to other gelators. Besides, they provide stable oleogel structure with low addition level, a wide range of melting points, and various firmness and stickiness values (Co and Marangoni, 2012; Hwang et al., 2013). For the reasons mentioned above, many researchers have reported oleogels prepared with different waxes (Dassanayake et al., 2009; Hwang et al., 2012; Patel et al., 2014; Yılmaz and Öğütcü, 2014). The waxes mostly used as gelators in the literature are sunflower, rice bran, candelilla, carnauba, berry, shellac wax, and beeswax (Dassanayake et al., 2009, Hwang et al., 2012; Patel et al., 2014; Yılmaz and Öğütcü, 2014).

Particularly, spreadable margarine or butter is required to have some physical, sensory, and structural features compared to other fats. The most important features that play a key role in consumer preferences are smooth texture, a bright appearance, and spreadability. Furthermore, the melting points of such products are very important for consumer health. Hence, margarine should melt at body temperature to minimize cardiovascular risk (Co and Marangoni, 2012; Hwang and Winkler-Moser, 2020). In the literature, many studies have reported oleogels as an alternative to butter and margarine prepared with different waxes and vegetable oils (Hwang et al., 2013; Öğütcü and Yılmaz, 2014; Yılmaz and Öğütcü, 2014; Patel et al., 2014; Öğütcü et al., 2015; Yılmaz and Öğütcü, 2015). As a result of these studies, it was determined that the firmness, stickiness, color, polymorphic form, acid and peroxide values of the developed oleogels were similar to those of the commercial ones. On the other hand, the melting point of the developed oleogels was reported to be higher than that of commercial margarine, which stands as the biggest problem in front of the commercialization of oleogels (Hwang et al., 2013; Patel et al., 2014; Yılmaz and Öğütcü, 2014; Öğütcü and Yılmaz, 2014; Öğütcü et al., 2015; Yılmaz and Öğütcü, 2015; Mandu et al., 2020). As is well known, the melting point, saturated fat content, spreadability and stability of oleogels depend on wax addition levels and wax types. In order to lower the melting point, the wax addition level must be reduced; however, this also negatively affects the oil-binding capacity and stability of oleogels (Hwang et al., 2012; Mandu et al., 2020). Therefore, in the literature, stable minimum gel formation concentrations for wax oleogels were determined and higher addition levels were investigated (Hwang et al., 2012; Yılmaz and Öğütcü, 2014). All of the studies mentioned above used various wax types, except wax mixture. Very recent studies by Winkler-Moser et al. (2019 and 2023), Hwang and Winkler-Moser (2020), and Ghazani et al. (2022) reported oleogels prepared with wax blends; however, these studies did not include optimization. Moreover, Wettlaufer et al., (2021) reported characteristics of oleogels prepared with mixture of waxes and their hydrolyzates. Recently, Thakur et al. (2022) reported optimization of processing parameters of soybean oil-carnauba wax oleogels. The aim of this study was to optimize the wax mixture level to be used in the production of sunflower oil oleogels structured with a mixture of beeswax and sunflower wax, exhibiting a spreadability and melting point similar to margarine.

MATERIAL AND METHODS

Materials

The commercial sunflower oil was purchased from the local market in Çanakkale, Türkiye. The beeswax (BW) and sunflower wax (SW) were purchased from KahlWax (Kahl GmbH & Co., Trittau, Germany). All other chemicals used in analysis were purchased from Merck (Darmstadt, Germany) and Sigma-Aldrich (St. Louis, USA), and all chemicals were of analytical grade.

Preparation of oleogels

For the preparation of the oleogels, a certain amount of the sunflower oil and waxes (w:w) (BW and SW) were weighted into the beher-glass, and the mixture was heated at 80 °C in water bath. The wax addition level and sample codes are given in Table 1. When the waxes were completely melted, the oil-wax mixture was stirred at 80 °C and 200 rpm for 3 min on the magnetic heating plate (IKA RCT Basic, China). The temperature of the oil-wax mixture was controlled with a digital thermometer during the production process. After this process, the mixture was filled into the falcon tubes and cooled at 25 °C in the refrigerated incubator for 24 hours. At the end of the cooling process, the oleogels were stored at the same temperature, and the planned analyses were performed.

Table 1. Sample codes, variables and CCD matrix for oleogels prepared with sunflower oil, beeswax and sunflower wax blend.

	Treatment		Concentrations		Response Variables			
Samples	BW	SW	BW (%)	SW (%)	T_O (°C)	Melting Point (°C)	Firmness (g force)	Stickiness (g force)
S1	0	0	3	3	31.70	56.02	682.25	824.31
S2	-1	1	1	5	34.07	61.11	743.64	830.71
S3	0	-1.414	3	0.17	31.76	45.67	52.76	68.91
S4	-1	-1	1	1	32.40	51.15	29.66	39.79
S5	1.414	0	5.83	3	31.99	56.56	1291.94	1506.95
S6	0	0	3	3	31.83	53.84	645.84	806.96
S7	1	1	5	5	32.49	59.04	2008.74	2282.14
S8	0	0	3	3	31.53	55.66	664.04	815.64
S9	0	0	3	3	31.06	56.56	651.15	701.10
S10	0	0	3	3	30.41	58.35	700.94	802.05
S11	1	-1	5	1	31.20	50.71	288.10	324.90
S12	0	1.414	3	5.83	32.16	60.86	1625.32	1932.87
S13	-1.414	0	0.17	3	36.29	58.59	247.78	289.62
*OFM	-	-	5.50	0.50	-	48.69	205.16	220.20
*OM	-	-	2.80	0.20	-	46.42	-	-

SW: sunflower wax, BW: beeswax, OFM: oleogels with 5.50%BW and 0.50%SW OM: Oleogels with 2.80%BW and 0.20%SW, T_O: *onset temperature.*
*The results are predicted values obtained from the multiple response optimization results.

Measurements

The measurements of physicochemical properties

The oil-binding capacity (OBC) was measured by the method described in detail in Yılmaz and Öğütcü (2014). The color values of the prepared oleogels were determined using Precision Colorimeter (NR20XE, Shenzhen 3nh Tech. Co., China), and the results were expressed as L, a* and b*.

The measurement of crystal morphology and textural properties

The firmness and stickiness values of the oleogels were measured with a Texture Analyzer (TA-HD Plus, Stable Microsystems, UK) equipped with spreadability rig (TA 425; TTC). The textural measurements were performed at 25 °C, and the test specifications were as follows: test speed at 3.0 mm/sec, post-test speed at 10 mm/sec and distance of 23.00 mm (Moskowitz, 1987). The data obtained from the texture analyzer were evaluated using an instrument software (Texture Exponent v.6.1.1.0, Stable Microsystems, UK). The X-ray diffraction (XRD) patterns of the oleogels were determined by XRD (Empyrean PANalytical, Netherlands), and the XRD data were evaluated by an instrument software (X'Pert Highscore Plus, Netherlands). For the XRD measurements, the angular scans in the 2.0° to 50° 2-theta range were performed with a scan rate of 2°/min and a Cu source X-ray tube (λ = 1.54056 Å, 45 kV and 40 mA).

The measurement of thermal properties

For the determination of the melting points of the prepared oleogels, differential scanning calorimeter (DSC) were used (Hitachi DSC7020, Japan). For the thermal properties of the oleogels, 5-7 mg gel samples were weighted into aluminum pan (also used as a reference), and the pan was then closed hermetically. Then, the samples were heated from 25 to 90 °C at a heating rate of 5 °C/min under N_2 gas (flow rate at 50 ml/min) for the determination of the onset (T_o) and peak (T_p) temperatures of the oleogel samples. The T_O and melting temperatures of the samples were calculated with the instrument software (TA7000 Measurement 10.5v, Hitachi High-Tech Science Corp.) from the obtained thermograms. The example of DSC thermograms is given in Figure 1.

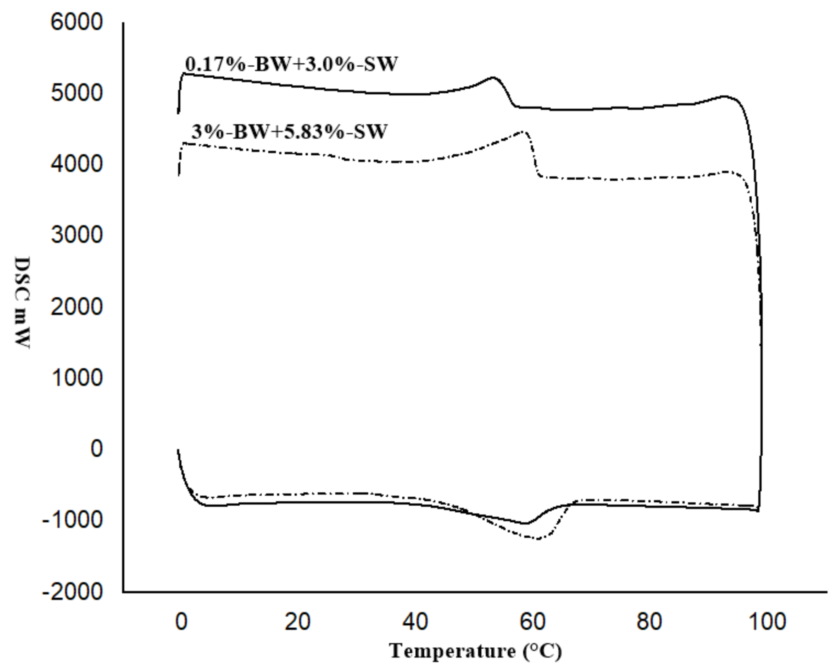

Figure 1. An example of the differential scanning calorimeter thermograms of the oleogels prepared with sunflower and beeswax mixtures.

Statistical Analysis

For the determination of the effects of the independent variables on the physicochemical, thermal and textural properties of the oleogels, the Response Surface Methodology (RSM) was used. A Central Composite Design (CCD) was applied, and the CCD matrix is given in Table 1. Each treatment code was repeated two times. The obtained data were analyzed with the Minitab 17 (17.1.0, 2013) software for surface plots and for the determination of the optimal concentrations. The second-order model, which was used for the description of the effects of the wax type and addition level on the response, is given as follows;

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_{ii}^2 + \sum_{i=1,j=2}^{k-1,k} \beta_{ij} X_i X_j + \varepsilon$$

where Xi and Xj are the independent variables affecting the response variable (Y), β_i and β_{ii}, β_{ij} are the coefficients of the linear effect and double interactions, β_0 is the intercept, and ε is the error.

RESULTS AND DISCUSSION

Physicochemical properties

Recent studies showed that oleogel structuring conditions such as gelator concentrations, types, shear and cooling rate etc. greatly affect the textural properties of oleogels (Blake and Marangoni, 2015a; Blake and Marangoni, 2015b; Thakur et al., 2022). Therefore, in order to determine and observe the effect of wax types and concentrations on oleogel properties, the oleogels used in the study were produced at both constant temperature and constant mixing and constant cooling rate. One of the most important problems with oil-based foods is that oil release negatively affects consumer preferences, which can reduce the reputation of manufacturers and cause economic losses (Öğütcü et al., 2017). Oil release and phase separation depend on the structural stability of the system, and as is well known, oil-binding capacity is one of the important indicators of the gel stability (Ghazani et al., 2022). High gel stability also means preventing oil release from gel structure or restricting of oil leakage (Ghazani et al., 2022). All of OBC values of the gels were higher than >98% (Table 2). There were no statistically significant differences between the oleogel samples in terms of OBC values ($p \geq 0.05$). Therefore, the effects of the waxes on the OBC values were not meaningful, according to these results. However, previous studies reported that SW-gels had lower crystal formation time and minimum gelling concentrations than BW-gels (Hwang et al., 2012; Yılmaz and Öğütcü, 2014). Waxes form structurally stable oleogels at minimum gel forming concentration and/or higher addition levels. Therefore, the SW-gels had higher OBC than the BW-gels at 3% addition level or less, as expected. In this regard, even the addition of SW at low concentrations increased the stability of the BW-based gels. Both the literature findings and our results showed that SW was more effective at restricting oil leakage. Recent research reported that oleogels with a mixture of SW and BW (1:1, 1:3 and 3:1) had higher oil binding capacity than pure ones (Ghazani et al., 2022). The most influential factor in consumer purchasing preferences is mostly appearance and color for food products (Öğütcü et al., 2015; Yılmaz and Öğütcü, 2015). The color values (L, a* and b*) of the oleogels were affected by wax type and addition level (Table 2). The color values of the oleogels showed that the L and b* values of the oleogels increased with an increase in the total wax addition level. Similar results were reported that olive oil oleogels prepared with BW and SW (Yılmaz and Öğütcü, 2014). Additionally, it is possible to produce oleogels of various colors, not only with different waxes, but also by varying the level of the wax added to wax mixtures.

Crystal morphology and textural properties

Polymorphism is defined as fats having different structural forms depending on their thermal history. Polymorphic forms affect the structural stability, texture and sensory properties of products as well as consumer perception. There are three main forms named α, β and β′. The stability of polymorphic forms decreases in the order $\beta > \beta' > \alpha$. In addition, especially from these polymorphic forms, the β′ polymorphic form is demanded in spreadable products or similar products, as it provides a desired, smooth and mouth-melting structure (Lawler and Dimick, 2008; Mandu et al., 2020). The α and β polymorphic forms are characterized by XRD pattern peaks around 4.10 and 4.50 Å, respectively, while the β′ form has a peak around 4.10 and 3.70 Å (Dassanayake et al., 2009; Le Reverend et al., 2010). The XRD patterns of oleogel samples are given in Table 3. According to the XRD patterns, all of the samples had peaks around 3.70 and 4.10 Å that showed the presence of the β′ polymorphic form. As a result, the oleogels were prepared with BW and SW mixtures had the desired smooth and stable structure like margarine and table spreads. On the other hand, the peak intensity increased with the increasing wax addition level. It was determined that the added SW level was more decisive on gel stability, especially when less than 5% BW was used as the gel agent. Previously studies reported similar results about the crystal structure of wax oleogels (Yılmaz and Öğütcü, 2014; Öğütcü et al., 2015; Mandu et al., 2020; Ghazani et al., 2022). The XRD patterns indicated that oleogels prepared with waxes have a margarine-like texture preferred and desired by consumer.

Table 2. Oil-binding capacity and colour values of the oleogels prepared with sunflower oil, beeswax and sunflower wax blends.

Samples	OBC (%)	L	a*	b*
S1	100±0.01	53.88±0.04[e*]	-1.98±0.01[b]	6.13±0.01[d]
S2	99.98±0.01	57.79±0.01[d]	-1.96±0.01[b]	6.02±0.01[e]
S3	99.97±0.01	32.74±0.01[h]	-1.21±0.01[ab]	4.13±0.01[i]
S4	99.41±0.51	24.28±0.04[i]	-0.55±0.01[a]	5.15±0.03[g]
S5	99.97±0.01	58.93±0.03[c]	-1.65±0.01[ab]	6.24±0.01[c]
S6	100±0.01	53.88±0.04[e]	-1.98±0.01[b]	6.13±0.01[d]
S7	99.98±0.01	63.58±0.01[b]	-1.79±0.01[b]	6.85±0.01[a]
S8	100±0.01	53.88±0.04[e]	-1.98±0.01[b]	6.13±0.01[d]
S9	100±0.01	53.88±0.04[e]	-1.98±0.01[b]	6.13±0.01[d]
S10	100±0.01	53.88±0.04[e]	-1.98±0.01[b]	6.13±0.01[d]
S11	99.99±0.01	48.43±0.03[f]	-1.90±0.01[b]	5.35±0.02[f]
S12	99.96±0.02	63.87±0.04[a]	-1.94±0.04[b]	6.52±0.01[b]
S13	99.78±0.27	43.17±0.01[g]	-1.61±0.01[ab]	4.27±0.01[h]

OBC: oil-binding capacity.
*The small letters show the differences among the samples in the same column ($p \leq 0.05$).

Table 3. The XRD patterns of the oleogels prepared with sunflower oil, beeswax and sunflower wax blends.

Samples	2-theta	d-sapcing (Å)
S1	2.75, 3.90, 19.43, 21.35, 21.47, 23.75, 40.21	32.10, 22.62, 4.56, 4.16, 4.13, 3.74, 2.24
S2	2.01, 4.00, 19.27, 21.50, 23.87, 46.85	43.77, 22.07, 4.60, 4.13, 3.72, 1.93
S3	4.77, 17.81, 18.62, 19.33, 21.36, 23.73	18.51, 4.98, 4.76, 4.58, 4.15, 3.74
S4	4.77, 18.74, 21.14, 23.54	18.50, 4.73, 4.20, 3.77
S5	3.90, 19.32, 21.42, 23.81, 35.94, 40.31	22.62, 4.58, 4.14, 3.73, 2.49, 2.23
S6	2.75, 3.90, 19.43, 21.35, 21.47, 23.75, 40.21	32.10, 22.62, 4.56, 4.16, 4.13, 3.74, 2.24
S7	3.88, 19.43, 21.48, 23.77, 40.22	22.75, 4.56, 4.13, 3.74, 2.24
S8	2.75, 3.90, 19.43, 21.35, 21.47, 23.75, 40.21	32.10, 22.62, 4.56, 4.16, 4.13, 3.74, 2.24
S9	2.75, 3.90, 19.43, 21.35, 21.47, 23.75, 40.21	32.10, 22.62, 4.56, 4.16, 4.13, 3.74, 2.24
S10	2.75, 3.90, 19.43, 21.35, 21.47, 23.75, 40.21	32.10, 22.62, 4.56, 4.16, 4.13, 3.74, 2.24
S11	19.32, 21.40, 23.79,	4.58, 4.15, 3.73,
S12	2.63, 3.84, 19.09, 19.70, 21.47, 23.79, 36.02	33.55, 22.96, 4.64, 4.50, 4.13, 3.73, 2.49
S13	3.94, 19.06, 19.72, 21.40, 23.77, 40.11	22.40, 4.65, 4.50, 4.15, 3.74, 2.24

As well as the color and crystal structures of foods, their textural properties are one of the most important parameters that affect consumer perception, preferences and purchasing decisions (Yılmaz and Öğütcü, 2015). Spreadability is one of the most important quality criteria among textural parameters in margarine and other table-spreads. Spreadability is defined as the degree to which the sample is homogeneously dispersed on a surface (Moskowitz, 1987; Yılmaz and Öğütcü, 2015). However, it is difficult to measure instrumentally; therefore, samples with medium firmness and stickiness are considered to be more spreadable (Yılmaz and Öğütcü, 2015). For the determination of the optimum concentration and effect of wax type on the thermal and textural properties (firmness and stickiness) of the sunflower oil oleogels, the RSM plots were created and are given in Figures 2a and 2b. Additionally, the Analysis of Variance (ANOVA) results for the textural properties are shown in Table 4. The results clearly indicated that the wax type and wax concentration significantly affected the firmness and stickiness values of the oleogels (Table 4). According to the ANOVA results, the quadratic coefficient, linear coefficient and interaction coefficient were significantly effective on both firmness and stickiness values. The R^2 and the adjusted-R^2 indicated the suitability and accuracy level of the model; hence, the R^2 and adjusted R^2 should not only have high values, but also be close to each other. The R^2 and adjusted-R^2 of the firmness were 99.65 and 98.40%, while the stickiness was 99.55 and 99.24%, respectively. The results demonstrated that more than 98% of the variable variance depended on wax concentrations. The firmness values of the oleogels ranged from 29.66– 2008.74 g force (0.30 – 19.70 N). The oleogels prepared with 1%BW and 1%SW had lower firmness values, while the oleogels prepared with 5%BW and 5%SW had higher firmness values, as expected. The textural measurements showed that both the total wax addition level and wax types were effective on the firmness and stickiness values of the oleogels. Similar to our findings, Ghazani et al. (2022) reported that hardness of gels increased with an increase in total wax addition level and depended on the wax types used in wax mixtures. There was a close positive correlation between the firmness and stickiness values of the gels. Hence, similar results were also observed for the stickiness values. The surface plots showed that the firmness and stickiness values of the oleogels increased with increasing wax concentration, and that, particularly, the SW level was more effective than the BW addition level on the oleogel textures (Figures 2a, 2b). For oleogels formed with plain BW, the minimum gel formation concentration should be 5% and above, not only for the stability of the gel but also for the high OBC value. However, the melting curve in DSC thermograms was reported to correlate with solid fat content (SFC) (Tengku-Rozaina and Birch, 2015). Accordingly, when the wax concentration increases, the SFC and melting temperature of the samples increase (Figure 1). Moreover, when a mixture of BW and SW is used as a gel agent, the SFC value decreases, and the production cost is reduced. At the same time, this means a healthier product. The above-mentioned results showed that using a mixture of BW and SW reduced the total wax concentration for stable oleogel formation, resulting in more economical, stable, healthier products than the oleogels formed using BW or SW alone.

Thermal properties

The melting point of such food products is an important parameter in terms of health as well as consumer preferences and perceptions. Generally, the melting point of the spreadable-breakfast margarine is 36 °C, the same as human body temperature as well as it has moderate firmness and stickiness values (Öğütcü and Yılmaz, 2014). The T_O temperature of the oleogels prepared with BW and SW blends ranged from 30.41 to 36.29 °C. In addition, the melting points of the prepared oleogels ranged from 45.67°C (0.17%SW:3%BW) to 61.11 °C (5%SW: 1%BW). Similar results were reported by Dassanayake et al. (2009), Hwang et al. (2012), Öğütcü and Yılmaz (2014) and Öğütcü and Yılmaz (2015). The thermal results indicated that the oleogels prepared with wax mixture allowed gel formation in the desired melting range (Table 1). The RSM plots of T_O temperature and melting point of the SW: BW mixture oleogels are given in Figures 2c and 2d, and the ANOVA results are shown in Table 4. The ANOVA results showed that the linear and quadratic coefficients were statistically significant, while the two-way interaction model coefficients were not. The R^2 and the adjusted R^2 were 94.92 and 91.29%, respectively. The results showed that the total wax addition level, and the type of the wax and its concentration in the mixture had an effect on the melting point of the oleogel, as well as the textural properties. As with the firmness and stickiness values of the oleogels, the surface plots of the melting point demonstrated that the SW was more effective than the BW on the melting point of the mixed oleogels. In other words, when the SW levels increased in the wax mixtures, the melting range of the oleogels increased. Similarly, Winkler-Moser et al. (2019) indicated that SW dominated oleogel formation when used with wax mixtures. A recent study indicated that the melting point of oleogels prepared with wax mixtures (sunflower wax, candelilla wax, and beeswax) not only depended on the total wax addition level but also on the wax types and concentrations in the mixture (Winkler-Moser et al., 2023). The results were optimized considering firmness and melting point of commercial margarines currently on the market. Figure 3 shows the optimum concentration of SW and BW to achieve the targeted result of firmness and stickiness to minimize the melting point. In terms of multiple optimization results, the firmness, stickiness and melting peak values of the oleogels (OFM) prepared with 0.50%SW and 5.50%BW were 205.16, 220.20 g-force and 48.69 °C, respectively (d=0.8829). The minimum melting point was 46.42 °C according to the optimization results (d=0.9512), independent of the firmness and stickiness values of the oleogels (OM) prepared with 0.20%SW and 2.80%BW (Table 1 and Figure 4). These results clearly demonstrated that the oleogels produced with SW and BW wax mixtures could be prepared with different firmness, stickiness and melting range values for their intended usage. Similar to our findings, Hwang and Winkler-Moser (2020) reported that by mixing candelilla (CW) and BW, the firmness of oleogel-based samples could be increased while their melting point could be tailored by the addition level of two waxes. Additionally, Wetlaufer et al. (2021) reported that molecular fractions of the waxes were effective on the thermal and textural properties of the oleogels.

Table 4. The ANOVA and regression results of textural features of the oleogels.

	Firmness	*p*-value	Stickiness	*p*-value	T_o (°C)	*p*-value	Melting Point	*p*-value
Intercept	75.50		57.40		35.44		47.32	
Model		0.001		0.001		0.008		0.001
Linear		0.001		0.001		0.009		0.001
BW	-70.70	0.001	-53.80	0.001	-2.42	0.004	-1.78	0.187
SW	-18.90	0.001	-10.30	0.001	-0.008	0.141	4.87	0.001
Square		0.004		0.017		0.008		0.032
BW*BW	11.57	0.031	8.53	0.173	0.32	0.003	0.19	0.165
SW*SW	20.21	0.002	21.34	0.007	0.05	0.504	-0.35	0.026
Interaction		0.001		0.001		0.807		0.551
BW*SW	32.92	0.001	72.89	0.001	-0.024	0.807	-0.10	0.551
Lack of fit		0.262		0.272		0.192		0.916
R^2	99.65		99.55		85.37		94.92	
Adj-R^2	98.40		99.24		74.93		91.29	

SW: sunflower wax, BW: beeswax, Adj; adjusted.

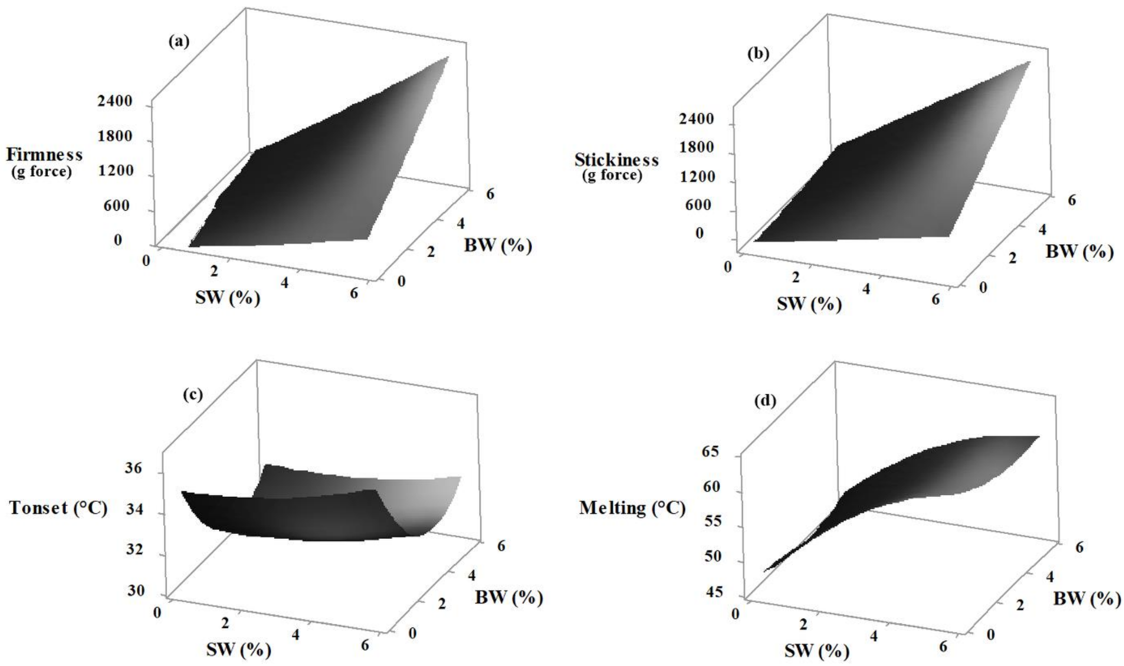

Figure 2. Effect of concentration of sunflower wax and beeswax on (a) firmness, (b) stickiness, and (c)tonset and (d) melting point values of the oleogels. SW: sunflower wax, BW: beeswax.

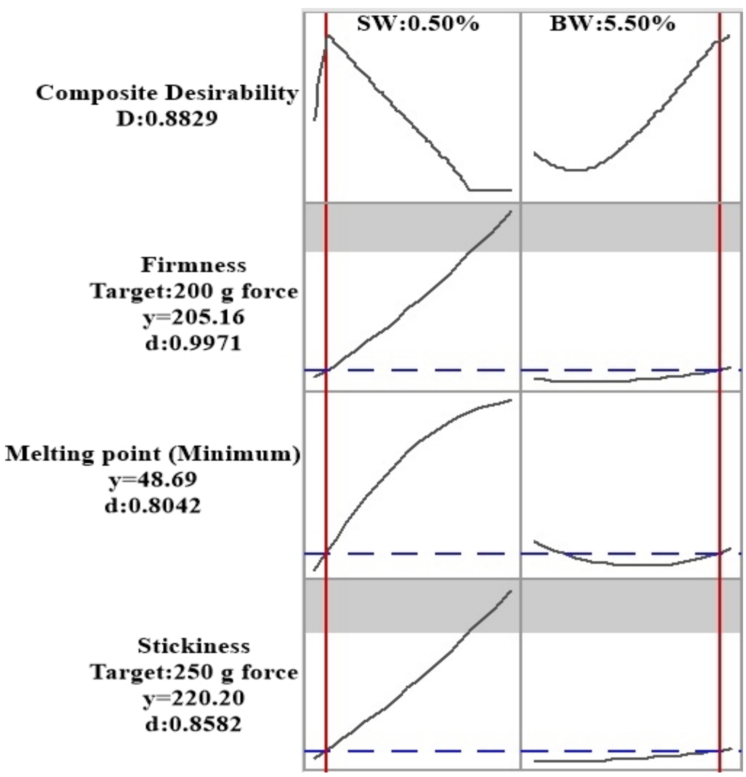

Figure 3. Multiple response optimization of oleogels prepared with sunflower oil, beeswax, and sunflower wax blends. d; desirability, y; predicted value. SW: sunflower wax, BW: beeswax.

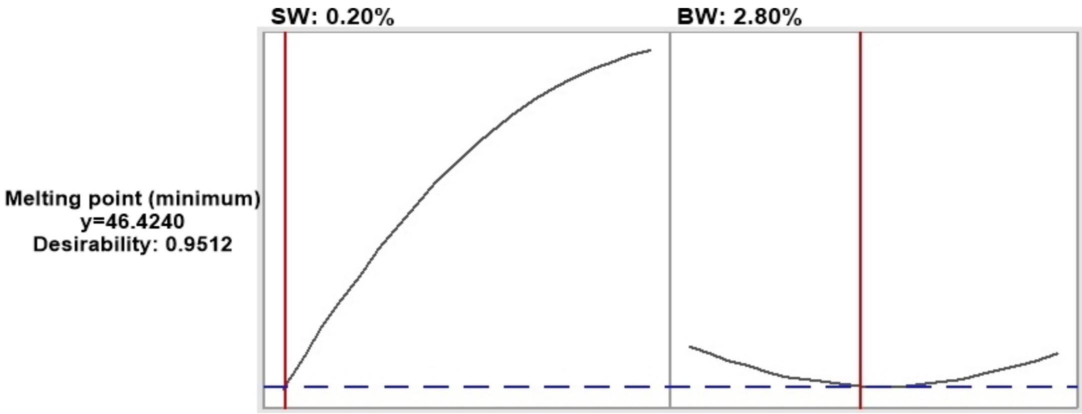

Figure 4. Response optimization of the minimized melting point of the oleogels prepared with sunflower oil, beeswax and sunflower wax blends. d; desirability, y; predicted value. SW: sunflower wax, BW: beeswax.

CONCLUSION

In this study, all oleogels, which were formed with beeswax and sunflower wax mixtures, had stable structures. According to the obtained XRD measurements, it was determined that all gel samples had the β' polymorphic structure. Again, it was observed that the melting ranges for all oleogels were 45.67–61.11 °C and that the firmness values changed between 0.30 and 19.70 N. In general, it was observed that the sunflower wax was more effective on the melting point and firmness values of the oleogels than the beeswax. As it is known, waxes have various melting points and purities depending on the sources obtained. Therefore, the addition levels of the waxes could be varied depending on the wax types and the structure and stability of the oleogels desired to be formed. When using low melting point waxes as gelators, it is necessary to increase the addition level of the wax for stable gel formation. In this case, the solid fat content and melting point of the oleogels increased. One of the remarkable results of this study is that by using a wax mixture instead of wax, a structurally stable gel with a lower melting point was formed at a lower wax addition level.

CONFLICT OF INTEREST

The author declares no conflicts of interest.

CONTRIBUTIONS

M. Öğütcü is responsible for the study and design of the work, data analysis and interpretation of results. E. Albayrak and E. S. Karabayır are responsible for carrying out the planned analysis and collecting data.

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