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Potential Application of Hydrocolloid-Based Oleogel and Beeswax Oleogel as Partial Substitutes of Solid Fat in Margarine

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Abstract: The purpose of this study was to produce margarine with reduced trans and saturated fatty acid contents using 10% beeswax oleogel and hydrocolloid-based oleogel containing 3.15% sodium caseinate, 0.5% guar gum, and 0.22% xanthan gum with a melting point, rheological and textural characteristics similar to palm oil. Oleogel samples were used as a substitute for palm oil and partially hydrogenated palm olein oil. Margarine (70% fat) formulated with these oleogels was investigated for solid fat content (SFC), melting point, and rheological and textural properties. The results showed that the replacement of 100% partially hydrogenated palm olein oil (PHPO) and 25% palm oil (PO) with beeswax oleogel and the replacement of 100% PHPO with hydrocolloid-based oleogel resulted in the production of margarine with rheological and textural properties similar to the commercial control margarine. In addition, these samples had a lower content of SFC and a higher melting point than the commercial control sample. The amounts of saturated and trans fatty acids also decreased. These were 28% and 80% in the sample containing beeswax and 15% and 73% in the sample with hydrocolloid-based oleogels for saturated and trans fatty acids, respectively. It was concluded that it is possible to manufacture margarine using the oleogel method while maintaining its physical characteristics and improving its nutritional properties.

Keywords: margarine; oleogel; beeswax; hydrocolloid; palm oil; rheological property

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1. Introduction

Fat is a significant component of many food products providing energy and fat-soluble vitamins (A, D, E, and K) and contributing to food's sensory and textural properties [1]. Solid fats are used in lipid-based food formulations to provide a suitable texture, high oxidative stability, and increased shelf life. Palm oil is produced from *Elaeis guineensis* belonging to the family of Arecaceae. This oil can provide sufficient solid oil in blends with unsaturated oils. It is a suitable source of fully natural hard fat for food products such as margarine, pastry, and shortening to provide solid fat functionality and increased stability [2]. Fat is chemically a mixture of triacylglycerol molecules. High melting point triacylglycerol molecules are crystalline at room temperature and form a three-dimensional

network that can physically trap lower melting point triacylglycerol molecules in this structural framework. These crystalline TAGs contain saturated and/or trans fatty acids [3]. To produce solid fatty acids, the process of partial hydrogenation was developed with the view that hydrogenated oils are derived from healthy vegetable oils and also have lower levels of saturated fatty acids, and gradually replaced the use of animal fats in food [4]. Partially Hydrogenated Oils (PHOs) are key ingredients in margarine, shortening, and bakery products. However, these oils are a source of trans fatty acids. Over the past twenty years, clinical and epidemiological studies have shown that consumption of trans fatty acids leads to an increase in low-density lipoprotein (LDL, bad cholesterol), a decrease in high-density lipoprotein (HDL, good cholesterol), and an increase in plasma TAG levels [5]. These effects increase the risk of cardiovascular disease and insulin resistance. It is also shown that some saturated fatty acids increase plasma LDL levels when substituted for carbohydrates in the diet, although the association between saturated fatty acid intake and cardiovascular disease is still debated [6]. Despite some disagreements in the scientific community, dietary guidelines recommend reducing the consumption of saturated fatty acids and eliminating trans fatty acids. In 2015, the US Food and Drug Administration announced that, with a three-year compliance period, hydrogenated oils should be removed from the GRAS list, meaning that after 2018, this primary source of trans fatty acids will not be allowed in foods [7]. Interestingly, even after much negative publicity and rejection of such foods, they are still seen in the market. This is because trans and saturated fatty acids are responsible for the formation of the network and play an important role in the texture and mouthfeel [3].

In recent years, using various methods, scientists have modified the physical properties of oil with low viscosity to be similar to the solid and elastoplastic properties of fat. Among these, a strategy called oleogel has been developed to reduce or replace fat [8]. In these systems, the oil combines with components that alter the physical properties of the oil by various molecular interactions such as hydrogen bonding and hydrophobic interactions so that its fluidity is reduced and its rheological properties are similar to those of fat. The continuous phase of these oil gels is lipid and shows the physical properties of the hydrogel [9,10]. Oleogels are produced by a direct method in the case of hydrophobic oleogelators such as wax, mono and diglycerides, and ethylcellulose, and by two indirect methods (using hydrophilic polymers), including emulsion and foam templated approaches [11].

Margarine or oleomargarine is food in the form of plastic or water in oil emulsion that should not contain less than 80% fat [12]. A high amount of margarine is fat (depending on the type of margarine), and the structural stability of margarine is affected by the available fat and the characteristics of the crystal network. Therefore, it is necessary to produce margarine with low saturation and desirable structural properties. Previous studies investigated the use of oleogels in margarine and spreads [12–15]. However, the number of studies investigating the use of oleogel as a solid fat substitute in margarine formulation is small. To our knowledge, the formulation of low content TFA and SFA margarine using hydrocolloid-based oleogel and beeswax oleogel has never been described. Therefore, the objective of this study was to produce margarine using hydrocolloid-based oleogel (HBO) and beeswax oleogel (BWO), whose attributes are similar to those of palm oil, and compare their properties with those of commercial margarine formula (control).

2. Materials and Methods

2.1. Materials

Palm oil (trans fatty acids = 0.7 %wt and saturated fatty acids = 52 %wt) and partially hydrogenated palm olein oil (trans fatty acids = 9.11 %wt and saturated fatty acids = 58 %wt) were kindly supplied by a margarine company. Non-fat milk powder, soy lecithin fluid, and sunflower oil (Behshahr Co., Behshahr, Iran, saturated fatty acids = 11 %wt) were purchased from a local grocery store. Xanthan gum, guar gum, sodium caseinate of bovine milk, and TBHQ were purchased from Sigma-Aldrich Chemical Co., Ltd. (St. Louis, MO, USA), and Palsgaard® DMG 0093 was purchased from Palsgaard A/S (Juelsminde,

Denmark). Beeswax also was purchased from KahlWax (Kahl GmbH & Co., Ltd., Trittau, Germany). Distilled water was used to prepare all samples.

2.2. Preparation of Beeswax Oleogel

The oleogels were prepared by heating a mixture of oil phase (sunflower oil (SFO) and beeswax (BW) with a melting point of 62–65 °C) at 80 °C for 30 min using a magnetic stirrer. For margarine formulation, an oleogel (OL) sample containing a concentration of 10% BW was selected as a substitute for palm oil (PO) and partially hydrogenated palm olein oil (PHPO), based on the preliminary study [16].

2.3. Preparation of Hydrocolloid-Based Oleogel

Considering that hydrocolloids are insoluble in oil, the preparation of this type of oleogels is performed indirectly, i.e., using the emulsion templated method. According to the methods of a previous study [17], first, concentrated oil in water emulsions (φoil = 0.6) were prepared using sodium caseinate (CN, 0–4 %wt), xanthan gum (XG, 0–1 %wt), and guar gum (GG, 0–1 %wt) with ultraturrax (Ultraturrax, IKA T25, Germany) at 13,500 rpm for 10 min. The emulsions were then dried using a heating oven at 70 °C to a constant weight. Afterward, to prepare the oleogels, the dried samples were homogenized using a household grinder for 20 s. All the samples were stored at 5 °C overnight before the experiments. The prepared sample with 3.15% CN, 0.5% GG, and 0.22% XG had the closest rheological properties, texture properties, and melting point to palm oil. Therefore, it was used for the margarine formulation.

2.4. Preparation of Margarine

In this study, margarine (70 %wt fat) with an oil phase consisting of a mixture of sunflower oil, palm oil, and partially hydrogenated palm olein oil and/or oleogel, soy lecithin, TBHQ, emulsifier, and an aqueous phase consisting of water, table salt, milk powder, and citric acid were produced. The aim was to replace partially hydrogenated palm olein oil (containing high trans fatty acids) and palm oil with oleogel. The oil phase of the margarine samples (70% fat) was prepared with the formulations shown in Table 1. The preparation process was as follows: the oil phase was heated at 60 °C for 10 min, then cooled to 45 °C, and the aqueous phase was added gradually to the oil phase. Both phases were homogenized at 10,000 rpm for 5 min. For mixing and crystallization, the resulting emulsion was then transferred to a Gastroback 42,909 ice cream maker at a temperature of 2–10 °C. Finally, the margarine samples were poured into plastic cups (250 gr) and stored at 4 °C.

Table 1. Formulation of margarine samples (70 %wt fat) prepared with beeswax oleogel (BWO) and hydrocolloid-based oleogel (HBO).

Sample	SFO%	OL%	PO%	PHPO%	Replacement Percentage
1	36	0	27	7	Control
2	36	3.50	27	3.50	50% (PHPO) with BWO or HBO
3	36	5.25	27	1.75	75% (PHPO) with BWO or HBO
4	36	7	27	0	100% (PHPO) with BWO or HBO
5	36	13.75	20.25	0	100% (PHPO) and 25% (PO) with BWO or HBO
6	36	20.50	13.50	0	100% (PHPO) and 50% (PO) with BWO or HBO

SFO: Sunflower Oil, OL: Oleogel, PO: Palm Oil, PHPO: Partially Hydrogenated Palm Olein Oil.

2.5. Measurement of Solid Fat Content

The solid fat content (SFC) was measured according to the direct method of AOCS (AOCS 16b-93) using nuclear magnetic resonance imaging (Bruker Minispec mq20 NMR spectrometer) at temperatures of 10, 20, 30, and 35 °C. Before analysis, the fat phase of the margarine was obtained by melting at 80 °C and after adding sodium sulfate (to ensure complete dehydration) and filtering with filter paper. One-third of the NMR tubes were

filled with filtered samples, and 4 tubes from each sample were placed in a chamber at 0 °C for 60 min. A tube of each sample was then placed at the mentioned temperatures for 30 min, and after this time, the solid fat content sample was read [15,17].

2.6. Differential Scanning Calorimetry

To investigate thermal behavior, 5–10 mg samples were placed in the differential scanning calorimeter (DSC, Mettler-Toledo, Greifensee, Switzerland) cell, and an empty pan was used as a reference. The oleogel samples were heated from 0 to 120 °C and margarine samples from 0 to 80 °C, with the temperature increasing at 5 °C/min. The melting temperature was calculated from the DSC thermogram [18,19].

2.7. Rheological Tests

All rheological tests were performed using a rheometer (Anton Paar Physica MCR 301, Anton Paar GmbH, Graz, Austria). To regulate the temperature, a Peltier plate system with a sensitivity of ±0.01 equipped with a water circulator was used, and to determine the rheological properties, parallel plate geometry (PP40) with a plate gap of 1 mm was used. In all tests, the samples were rested for about 1 min to achieve temperature equilibrium.

2.7.1. Strain Sweep Test

The values of elastic modulus (G′) and viscous modulus (G″) were determined in the strain range of 0.01–600% and a constant frequency of 1 Hz. The structure strength (G′ LVE), viscous modulus (G″ LVE) and loss tangent (tan δ, Equation (1)) in the linear viscoelastic region, and yield stress (τ_y) were reported.

$$tan\delta = \frac{G''}{G'} \quad (1)$$

2.7.2. Frequency Sweep Test

This test was to study the response of samples to frequency change. Considering the strain of the linear range obtained from the strain sweep test (0.1%), the frequency sweep was performed at a frequency of 0.1–100 Hz and at a constant strain of 0.1% to find the elastic modulus and viscous modulus as a function of angular frequency (ω, rad/s). The Power law or Ostwald model (Equation (2)) was fitted based on the data obtained from the frequency sweep diagrams. Model parameters were determined for all the samples, including structure strength (a, intercept) and gel structure type (b, slope).

$$G' = a\omega^b \quad (2)$$

2.7.3. Three Interval Thixotropy Test (3-ITT)

This test was performed according to the method of Tavernier et al. [20] with slight changes in three time intervals. The samples were subjected to linear range strain for 60 s in the first time interval. In the second time interval, the strain was applied above the linear range for 80 s (complete deformation), and in the third time interval, the samples were again subjected to the linear range for 60 s. The changes in the elastic modulus of the samples over time were obtained for these three time intervals. The following parameter was calculated:

$$\%\ R\ = \frac{G_{30}}{G_i} \times 100 \quad (3)$$

where % R is the structural recovery percentage, G_i is the initial storage modulus (in the first interval), and G_{30} is the storage modulus 30 s after deformation (third interval).

2.8. Spreadability Test

A stable micro system texture analyzer TA. XT Plus (Stable Micro Systems, Godalming, UK) equipped with a TTC Spreadability Rig (HDP/SR) and a 90° cone probe was used for testing the samples. The load was set to 5 kg (50 N). Each sample was then added to the top

and bottom cones. The top cone was placed at a distance of 25 mm from the bottom cone, moved at a rate of 3 mm/s, stopped up to a distance of 2 mm from the bottom cone and then decompressed. The force–time curve was then plotted. The maximum force represents the degree of hardness, and the spreadability was determined from the mean area under the curve (work of shear) [21].

2.9. Color Assessment

The color evaluation of the samples was performed based on the L*, b*, and a* system using HunterLab Colorflex EZ (HunterLab Inc., Reston, VA, USA). In this system, L*, a*, and b* indicate the degree of brightness, redness, and yellowness, respectively. It should be noted that before performing the test, the HunterLab was calibrated according to the manufacturer's instructions with white ceramics (L* = 92.24, a* = −1.28, and b* = 1.20). These indices were measured after production with three replications at ambient temperature [14,22].

2.10. Statistical Analysis

Data analysis was performed using SPSS Version 21. Quantitative data were presented using descriptive statistics as the mean and standard deviation of three replications of the experiment. A significant difference between samples means was determined using analysis of variance (ANOVA) at a significance level of 0.05.

3. Results and Discussion

3.1. SFC of Margarine

SFC is a factor that determines the percentage of solid fatty acids at a particular temperature and is usually measured by nuclear magnetic resonance [1]. Since SFC affects spreadability, firmness, mouthfeel, and stability, this value is considered a qualitative parameter of margarine texture [23]. The solid fat content in the refrigerator, at room temperature, and at body temperature is related to the spreadability, product stability, and mouthfeel, respectively [24]. Table 2 shows the solid fat content of margarine formulated with beeswax oleogel (BWO) and with hydrocolloid-based oleogel (HBO). As shown in the table, the highest amounts of solid fat at all temperatures were observed in the control sample, and SFC decreased with increasing replacement percentage. It has been reported that for spreadable margarine, the SFC at 10 °C should be not more than 32%, and to prevent any waxy sensation in the mouth, the SFC should be below 3.5% at 33.3 °C [24]. Therefore, given that the SFC of all samples was less than 21.59% at 10 °C and less than 2.13% at 35 °C, the margarine produced using these oleogels was spreadable and did not create a waxy taste in the mouth. Laia et al. [25] also stated that more than 10% SFC at 20 °C indicates instability and oil separation. The SFC of all the samples at this temperature was less than 10%, so none of the samples had a phase separation at room temperature. Similar results have been reported by Doan et al. [26], who reported that the solid fat content decreased with increasing replacement of palm oil with beeswax oleogel in confectionery filling. Additionally, in a study conducted by Demirkesen et al. [27], wax oleogels and palm-based shortening were used in gluten-free bakery products. The results showed that SFC decreased with increasing oleogel in the bread formulation. Several studies have shown that using wax oleogels in products such as cakes [28] and bakery products [29] has reduced the content of solid fat compared with control samples. In the present study, although the control sample (1) had a higher SFC at all measurement temperatures than the samples containing oleogel, all the samples had the desired margarine characteristics.

Table 2. Solid fat content (SFC) of margarines prepared with BWO and HBO.

Sample		10 °C	20 °C	30 °C	35 °C
BWO	1	21.59 ± 0.10 ^{a}	9.75 ± 0.30 ^{a}	4.31 ± 0.10 ^{a}	2.13 ± 0.02 ^{a}
	2	18.35 ± 0.00 ^{c}	7.36 ± 0.10 ^{c}	3.93 ± 0.08 ^{b}	2.09 ± 0.00 ^{b}
	3	16.14 ± 0.10 ^{e}	6.11 ± 0.10 ^{d}	3.69 ± 0.09 ^{c}	2.04 ± 0.01 ^{c}
	4	15.21 ± 0.10 ^{f}	5.51 ± 0.09 ^{e}	3.20 ± 0.10 ^{d}	2.01 ± 0.01 ^{d}
	5	11.31 ± 0.10 ^{h}	4.31 ± 0.10 ^{f}	2.90 ± 0.06 ^{e}	1.93 ± 0.02 ^{e}
	6	8.94 ± 0.30 ^{i}	4.01 ± 0.00 ^{g}	2.89 ± 0.04 ^{e}	1.92 ± 0.01 ^{e}
HBO	2	20.34 ± 0.10 ^{b}	9.28 ± 0.20 ^{a}	3.85 ± 0.09 ^{bc}	1.80 ± 0.06 ^{f}
	3	17.28 ± 0.30 ^{d}	8.31 ± 0.10 ^{b}	2.97 ± 0.07 ^{e}	1.20 ± 0.03 ^{g}
	4	13.40 ± 0.20 ^{g}	5.58 ± 0.09 ^{e}	2.84 ± 0.10 ^{e}	1.24 ± 0.03 ^{h}
	5	3.50 ± 0.20 ^{j}	0.80 ± 0.10 ^{h}	0.00 ± 0.00 ^{f}	0.00 ± 0.00 ^{i}
	6	0.90 ± 0.08 ^{k}	0.00 ± 0.00 ^{i}	0.00 ± 0.00 ^{f}	0.00 ± 0.00 ^{i}

Different small letters between rows show a significant difference between samples ($p \leq 0.05$). Data are presented as mean ± standard deviation.

3.2. Differential Scanning Calorimetry of Margarine

Figure 1A shows the melting profile of 10% beeswax oleogel (BWO), the control margarine, and sample 5 (replacement of 100% (PHPO) and 25% (PO) with BWO). The melting thermogram of hydrocolloid-based oleogel (HBO), the control margarine, and sample 4 (replacement of 100% (PHPO) with HBO) are shown in Figure 1B. In the industry, thermal profile determination is essential to control properties such as spreadability and thermal stability, especially at room temperature. The melting point of a fat or oil can be affected by the length of the FA chain (increasing the chain length leads to an increase in melting point), degree of unsaturation (increasing the degree of unsaturation leads to a decrease in melting point), the presence of TFA (TFA have higher melting point than the cis form) and polymorphisms (α—lowest melting point, β'—medium melting point and β—highest melting point) [30]. The control sample had two melting peaks in the melting range of 27 to 37 °C, revealing the fat content separated into two parts. The high melting point component consists of saturated triacylglycerols melted at higher temperatures and is the final peak. The soft component contains unsaturated fatty acids with a low melting point and is responsible for the first melting peak. This is in line with the composition of margarine, which contains palm oil, partially hydrogenated palm olein oil, and sunflower oil. Beeswax oleogel had multiple melting peaks and a melting range of 48 to 63 °C. As shown in the figure, with increasing replacement, the melting point, and melting range increased, so that sample 5 (100% (PHPO) and 25% (PO) with oleogel) was similar to the control in terms of rheological and textural characteristics and had a melting range of 26–50 °C. The wide melting range of this sample can be attributed to the heterogeneity of the compounds in margarine because the different compounds present (triacylglycerols of palm and partially hydrogenated palm olein oils, and fatty esters, *n*-alkanes, free fatty acids, and free fatty alcohols in beeswax) have different melting points. Other studies have reported similar results [14]. As shown in Figure 1B, the oleogel had a melting peak at 210 °C, and no peak of thermal degradation was observed. These results were similar to other studies that reported thermal stability and high melting points of hydrocolloid-based oleogels [31,32]. Furthermore, it was observed in Sample 4, which was similar to the control in terms of rheological and textural properties, that the melting point was transferred to high temperatures by increasing the amount of hydrocolloid-based oleogel in the margarine.

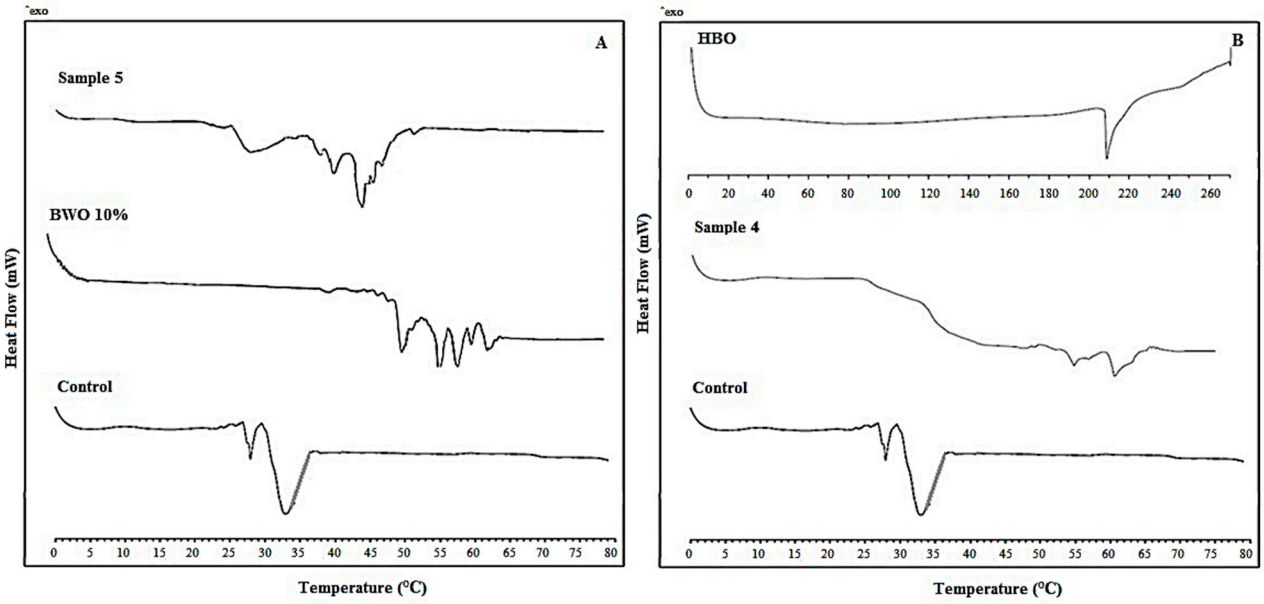

Figure 1. (**A**) melting profile of control margarine and beeswax oleogel 10% and sample 5. (**B**) Melting profile of control margarine and hydrocolloids-based oleogel (HBO), and sample 4.

3.3. Strain Sweep Test of Margarine

Table 3 lists the parameters of the strain sweep test, including the elastic modulus (G′ LVR), viscous modulus (G″ LVR), loss tangent, and yield stress (τy, G″ > G′) of the margarine samples. The results show that for all samples in the linear viscoelastic range, the storage or elastic modulus was more significant than the viscous modulus (G′ > G″), which indicates the predominant elastic characteristic. As shown in Table 3, the rheological behavior was different in samples containing hydrocolloid-based oleogel and beeswax oleogel. In samples containing beeswax oleogel, with increasing replacement percentage, the elastic modulus and viscous modulus decreased, and in most samples, there was no significant difference in terms of loss tangent or damping factor (tan δ = G″/G′). In addition, the yield stress (stress value of crossover point) increased moderately with the increasing replacement rate. In the study by Demirkesen et al. [27] investigating gluten-free baked products, it was stated that with an increasing percentage of shortening replacement with wax oleogel in bread dough, the elastic modulus decreased and the frequency dependence increased. Another study that investigated oleogels made with candelilla wax as a substitute for shortening in cookies found that the elastic modulus of cookies prepared with shortening was higher than that of cookies containing candelilla wax oleogel. This behavior was attributed to higher rheological properties of shortening compared with wax oleogel [33]. In the section on solid fat content, it was explained that, with increasing replacement, this parameter also decreased. The power law relationship between SFC and G′ ($G' = SFC^n$) for fat systems has been shown in many studies [34,35]. When the solid fat crystalline particles are less than a certain level, a strong network is not formed, and as a result, the elasticity is reduced. In the present study, the elastic modulus of beeswax-based margarine was closely related to the solid fat content. This relationship was not observed in the control samples or the margarine prepared with hydrocolloid-based oleogel; this may be because in margarine with beeswax oleogel, the building blocks of the gel are formed by fat crystals, whereas in margarine formed by hydrocolloid-based oleogel, the building blocks of the gel are hydrocolloids in addition to being formed by fat crystals. As for the control sample, G′ probably depends on other factors such as polymorphism, crystallization behavior, and margarine microstructure [36]. It is shown that fat plasticity is a function of two factors, SFC and crystal structure [37]. In a study by Liu et al. [36], the SFC and polymorphism of two types of commercial margarine with different textures were investigated. Their results showed that the SFC of the hard sample was less than the softer

sample, so they concluded that the SFC was not the only parameter affecting the strength of the crystal lattice. By analyzing the microstructure and polymorphism of the crystals, they found that the crystals of the harder sample were denser and of the β type, which is larger. They found that the softer sample had β′ crystals and a finer and softer texture.

Table 3. Parameters related to strain sweep test of margarines prepared with BWO and HBO.

Sample		τ_y (Pa)	Tan δ	G′ (Pa)	G″ (Pa)
BWO	1	989 ± 3^{d}	0.13 ± 0.003^{e}	$536{,}432 \pm 2354^{f}$	$69{,}736 \pm 2154^{f}$
	2	992 ± 4^{de}	0.13 ± 0.001^{e}	$1{,}082{,}967 \pm 3895^{a}$	$140{,}786 \pm 1573^{c}$
	3	990 ± 3^{d}	0.14 ± 0.001^{d}	$912{,}172 \pm 3500^{b}$	$127{,}704 \pm 1322^{d}$
	4	997 ± 1^{e}	0.13 ± 0.001^{e}	$909{,}367 \pm 2501^{b}$	$118{,}218 \pm 1215^{e}$
	5	1005 ± 3^{c}	0.13 ± 0.003^{e}	$539{,}775 \pm 2684^{f}$	$70{,}773 \pm 1387^{f}$
	6	1009 ± 4^{bc}	0.15 ± 0.002^{c}	$375{,}505 \pm 1900^{i}$	$56{,}326 \pm 1224^{i}$
HBO	2	1721 ± 7^{a}	0.11 ± 0.006^{f}	$427{,}257 \pm 1982^{h}$	$46{,}998 \pm 2700^{g}$
	3	1016 ± 5^{b}	0.13 ± 0.005^{e}	$477{,}719 \pm 3202^{g}$	$62{,}103 \pm 2845^{h}$
	4	996 ± 5^{ed}	0.13 ± 0.003^{e}	$547{,}702 \pm 3764^{e}$	$71{,}201 \pm 1935^{f}$
	5	610 ± 7^{f}	0.33 ± 0.005^{b}	$684{,}321 \pm 2495^{d}$	$225{,}826 \pm 4590^{b}$
	6	563 ± 6^{g}	0.45 ± 0.002^{a}	$708{,}569 \pm 3478^{c}$	$318{,}856 \pm 3348^{a}$

Different small letters between rows indicate a significant difference between samples ($p \leq 0.05$). Data are presented as mean ± standard deviation.

In the replacement with hydrocolloid-based oleogel, the samples' elastic and viscous modulus increased. It should be noted that this increase was greater for the viscous modulus, and these two became closer together in the linear viscoelastic range, the effect of which can be seen in the loss tangent. Loss tangent values greater than one indicate liquid behavior, and values less than one indicate solid behavior. For all the margarine samples, this value was less than one, indicating solid-like behavior. However, with increasing replacement percentage, the loss tangent increased, indicating a predominance of viscous behavior. Table 3 shows that sample 4 (replacement of 100% partially hydrogenated palm olein oil with HBO) had the same characteristics as the control sample. There was no significant difference between these two in terms of G″, tan δ, or yield stress. They also had the most similar levels of G′. Tanti et al. [4] reported that the elastic modulus increased with increasing shortening replacement with oleogel, as the full replacement of the shortening with the hydroxypropyl methylcellulose and methylcellulose oleogel resulted sandwich cookie creams which were too hard and stiff. The G′ of creams at 50% and 75% replacement was significantly higher than the control sample containing a higher percentage of shortening and the commercial sandwiched cookie creams. Therefore, the creams had suitable rheological properties at this replacement percentage. In another study, an increase in the replacement percentage of shortening with HPMC oleogel in muffins resulted in a decreased batter elastic modulus [38].

3.4. Frequency Sweep Test of Margarine

The frequency dependence of the storage modulus was investigated using the Ostwald model with a coefficient of $R^2 > 0.98$. The parameters related to this model are shown in Table 4. The factor value ranges were 188,044–461,400 for a and 0.10–0.47 for b. All the values were positive, demonstrating that storage modulus (G′) increased with angular frequency. With an increasing percentage of BWO replacement, factor a, which is a parameter indicating the strength of the structure, decreased similar to the elastic modulus, and factor b showed an increasing trend. This means that these margarine samples showed a behavior more dependent on frequency and that the frequency dependence of beeswax oleogel (0.5) was more than that of palm oil (0.15) and partially hydrogenated palm olein oil (0.1). Therefore, with increasing beeswax oleogel content in margarine, the frequency dependence (b) increased. It should be noted that stronger gels have high elastic modulus and are not frequency dependent, indicating that margarine obtained using BWO had a

weaker lattice structure [39]. Moreover, parameters a and b increased the percentage of hydrocolloid oleogel in margarine. Parameter a was in agreement with the result found for G′.

Table 4. Parameters of the power law (frequency sweep test) and structure recovery for margarines prepared with BWO and HBO.

Sample		a (Pa × s/rad)	b	Recovery%
BWO	1	232,620 ± 2051 ^{h}	0.10 ± 0.01 ^{f}	26.30 ± 1.50 ^{f}
	2	461,400 ± 3212 ^{a}	0.15 ± 0.02 ^{e}	27.40 ± 1.00 ^{f}
	3	422,155 ± 2982 ^{b}	0.18 ± 0.00 ^{c}	30.80 ± 1.90 ^{e}
	4	403,305 ± 2743 ^{c}	0.23 ± 0.02 ^{b}	31.90 ± 1.30 ^{e}
	5	239,777 ± 2037 ^{g}	0.40 ± 0.02 ^{a}	44.50 ± 2.80 ^{d}
	6	188,044 ± 2533 ^{i}	0.47 ± 0.01 ^{a}	51.60 ± 3.30 ^{c}
HBO	2	190,940 ± 2264 ^{i}	0.12 ± 0.01 ^{e}	32.60 ± 0.90 ^{e}
	3	235,150 ± 1835 ^{h}	0.13 ± 0.01 ^{e}	45.30 ± 1.70 ^{d}
	4	275,270 ± 2698 ^{f}	0.13 ± 0.00 ^{e}	56.00 ± 2.20 ^{c}
	5	377,883 ± 3263 ^{e}	0.17 ± 0.01 ^{ce}	66.00 ± 0.70 ^{b}
	6	391,442 ± 1988 ^{d}	0.20 ± 0.01 ^{b}	71.50 ± 2.30 ^{a}

Different small letters between rows show a significant difference between samples ($p \leq 0.05$). Data are presented as mean ± standard deviation.

3.5. Structure Recovery Test of Margarine

According to Table 4, an increasing trend was observed for structure recovery with an increasing replacement percentage, and the highest recovery was observed for sample 6 with the replacement of both oleogels (BWO and HBO). The recovery percentage of all margarine samples was between 26.3% and 71.5%. The lowest structure recovery was related to the control sample. Increasing the amount of beeswax oleogel in the formulation increased the structure recovery since the structure recovery of beeswax oleogel is much higher than the partially hydrogenated palm olein and palm oils. The increase in the recovery percentage of margarine containing hydrocolloid-based oleogel may also be due to the increase in oil phase uniformity in the formulation.

3.6. Spreadability Test of Margarine

From this test, two parameters of hardness and spreadability were obtained. These parameters are presented in Table 5. In previous studies, it has been reported that the maximum force required to penetrate the upper probe in the sample is considered hardness, and the spreadability is equivalent to the mean area under the positive curve (penetration curve) as smaller values of the area reflect more spreadability [40,41]. Hardness and spreadability are the most important features perceived by consumers. Kangchai et al. [42] reported that hardness is significantly associated with spreadability, the higher the hardness, the greater the resistance to spread (less spreadability). As shown in Table 5, with the increasing amount of beeswax oleogel in margarine, hardness significantly decreased, and spreadability increased.

Hwang et al. [43] observed that the hardness of margarine increased with an increasing amount of sunflower wax, and this parameter increased with increasing rice bran wax up to 5% wax but did not increase significantly from 5% wax to 7% wax. They also reported that the hardness of commercial spreads can be achieved with 3% wax oleogels in margarine; however, the hardness of stick margarine cannot be produced with 7% wax. The inconsistent results of this study with our study may be due to the concentration of wax used. There was no significant difference between sample 6 (replacement of 100% (PHPO) and 50% (PO) with BWO) and the control sample in terms of spreadability. It should be noted that according to the SFC results, all the margarine samples were spreadable at room temperature. Additionally, sample 5 (replacement of 100% (PHPO) and 25% (PO) with BWO) had the level of hardness closest to the control sample. Furthermore, in the present

study, the results showed that with increasing replacement of hydrocolloid-based oleogel, hardness increased and spreadability decreased. The hardness of sample 4 containing HBO was similar to that of the control sample, and according to the SFC results, all the samples also had good spreadability.

Table 5. Spreadability test parameters of margarine samples formulated with BWO and HBO.

Sample		Area under the Curve (g × sec)	Hardness (g)
BWO	1	2643 ± 53 ^{f}	2526 ± 43 ^{f}
	2	6190 ± 105 ^{a}	5108 ± 89 ^{a}
	3	6095 ± 123 ^{a}	4904 ± 27 ^{b}
	4	5508 ± 98 ^{b}	4325 ± 90 ^{c}
	5	3076 ± 91 ^{e}	2357 ± 32 ^{g}
	6	2633 ± 66 ^{f}	1651 ± 67 ^{l}
Sample		**Area under the Curve (g × sec)**	**Hardness (g)**
HBO	2	1710 ± 32 ^{i}	1609 ± 44 ^{i}
	3	2020 ± 64 ^{h}	2035 ± 58 ^{h}
	4	2430 ± 12 ^{g}	2314 ± 29 ^{g}
	5	3600 ± 33 ^{d}	3748 ± 27 ^{e}
	6	3855 ± 47 ^{c}	4012 ± 51 ^{d}

Different small letters between rows show a significant difference between samples ($p \leq 0.05$). Data are presented as mean ± standard deviation.

3.7. Margarine Color

Color is the first feature of appearance in the consumer's purchasing decision. Beta-carotene pigment was not added to the prepared samples to prevent color interference. The color indices are shown in Table 6. L* and b* values decreased and increased, respectively, with increasing replacement of beeswax oleogel. The control sample was the most transparent and had the least yellowish color. In addition, there was no significant difference between the samples in terms of a* values (redness–greenness), and all samples had little reddish tones. This may be due to the natural color of the beeswax; as the percentage of beeswax in the formulation increases, the lightness decreases, and the yellowness increases. Yılmaz et al. [44] analyzed the characteristics of cookies produced by shortening and beeswax; the results showed that the L* value was higher in cookies containing shortening, and there was no significant difference between the values of a*. The b* value of the cookie prepared with the shortening was lower than that of the cookie containing beeswax. In addition, the results related to the color of the samples containing hydrocolloid-based oleogel (Table 6) showed that the L* values varied from 83.61 to 88.14, and the highest value was for the control sample, which indicates a higher lightness of the sample. There was a significant difference between the samples in terms of a* values (0.28–1.3) and b* values (15.37–20.88). With increasing HBO replacement percentage, an increase was observed in these values. This is probably due to the color of the replaced oleogel.

Table 6. Color values in margarines prepared with BWO and HBO.

Sample		L*	a*	b*
BWO	1	88.14 ± 0.03 ^{a}	0.28 ± 0.006 ^{e}	15.37 ± 0.020 ^{k}
	2	87.17 ± 0.02 ^{d}	0.28 ± 0.000 ^{e}	16.56 ± 0.006 ^{h}
	3	87.04 ± 0.02 ^{e}	0.28 ± 0.000 ^{e}	17.40 ± 0.006 ^{f}
	4	86.29 ± 0.03 ^{f}	0.28 ± 0.006 ^{e}	17.42 ± 0.006 ^{e}
	5	84.01 ± 0.00 ^{h}	0.28 ± 0.006 ^{e}	20.82 ± 0.000 ^{c}
	6	83.61 ± 0.01 ^{j}	0.28 ± 0.006 ^{e}	21.37 ± 0.006 ^{a}

Table 6. *Cont.*

Sample		L*	a*	b*
HBO	2	88.04 ± 0.02 [b]	0.58 ± 0.006 [d]	15.47 ± 0.020 [j]
	3	87.69 ± 0.02 [c]	0.59 ± 0.006 [d]	15.85 ± 0.020 [i]
	4	87.19 ± 0.01 [d]	0.61 ± 0.006 [c]	17.24 ± 0.020 [g]
	5	84.68 ± 0.02 [g]	1.10 ± 0.006 [b]	20.45 ± 0.030 [d]
	6	83.96 ± 0.02 [i]	1.30 ± 0.006 [a]	20.88 ± 0.020 [b]

Different small letters between rows indicate a significant difference between samples ($p \leq 0.05$). Data are presented as mean ± standard deviation.

4. Conclusions

The process of producing low-saturated margarine using oleogel based on hydrocolloids and beeswax oleogel was performed to reduce the negative effects of solid fat consumption while maintaining appropriate texture and oxidative stability. This was performed by replacing the partially hydrogenated palm olein oil (PHPO) and palm oil (PO) with a hydrocolloid-based oleogel and a 10% beeswax oleogel in a margarine formulation. The results showed that the margarine samples with 100% PHPO and 25% PO replacement with BWO (sample 5) and 100% PHPO replacement with HBO (sample 4) had a lower solid fat content (SFC) and a higher melting point but similar rheological and textural properties to commercial control margarine. Due to the amount of saturated and trans fatty acids in oils used in the margarine formulation, and with this percentage of oleogels replacement (sample 4 and sample 5), the amount of saturated and trans fatty acids decreased. This reduction was 28% and 80% in the BWO replacement and 15% and 73% in the HBO replacement for saturated and trans fatty acids, respectively.

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