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Hazelnut oil-besswax and hazelnut oil-sunflower wax oleogels were performed very successfully in cookie preparation, and the cookies were liked by the consumers better than cookies made with the commercial bakery shortening.

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**A Introduction** 

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### ARTICLE

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**Texture, Sensory Properties and Stability of Cookies Prepared with Wax Oleogels** 

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Shortenings are the essential components of high quality baked foods. Their effects on dough structure formation and desired final product attributes depend mostly to their solid fat content and  $\beta'$  crystalline polymorphs. Saturated and *trans* fatty acids present in shortenings pose some important negative health considerations. Hence, alternative plastic fats with lower or zero quantity of saturated and trans fatty acids are in high demand. Oleogels are gel networks of liquid edible oils with no trans and very low saturated fatty acids. In this study, sunflower wax (SW) and beeswax (BW) oleogels of hazelnut oil were used in cookie preparation against commercial bakery shortening (CBS) as control, to compare the textural, sensory and stability properties of the cookies. The basic chemical composition, textural properties, some physical attributes of the cookies were compared. Sensory texture/flavor profile analysis (T/FPA) and consumer hedonic tests were also accomplished. Furthermore, the changes in cookie texture and stability were monitored during 30 days storage at room temperature. It was found out that in almost all quality properties, the oleogel cookies resembled CBS cookies. T/FPA results present detailed data for literature. Consumer hedonic scores indicated that oleogel cookies were better than CBS cookies and were also well accepted by consumers. Wax oleogels can be used as cookie shortenings successfully.

Shortenings are a class of fat products tailored to provide special functional and nutritional features in foods like bakery products, confectionary, fried foods, in icing and fillings, etc. The name originates from the expected results to 'shorten' or tenderize baked foods. Particularly in baked goods, shortenings impart tenderness, structure, mouthfeel, and flavor. Shortenings are considered as quasi-plastic materials, varying in viscosities from flowing liquids to non-flowing solids <sup>1,2</sup>. In baked foods including cookies, shortenings act as lubricants to contribute the plasticity of the final product, as well as to stabilize the air cells formed during mixing to yield tenderness, eating characters and preserving quality. Furthermore, during dough formation shortening covers gluten and starch granules to prevent their adherence to each other, and hence provides the development of well aerated, tender, lubricated and processable viscoelastic dough structure. In addition to flavor retention, mouthfeeling of the final products enhances to desirable levels by using shortenings 3-6.

To impart their functional roles in baked foods, shortenings must possess some properties in right proportion and amount. These include the ratio of liquid to solid fats (or solid fat content- SFC), level of inherent plasticity and oxidative stability of the shortenings used. If the SFC is very high, the dough will be stiff and aeration will be poor. Contrarily, if the SFC is too low, there would be no adequacy to entrap the air bubbles during mixing, hence very hard and oiling out structure

may appear. Moderate levels of SFC together with  $\beta'$  type crystal polymorphs have shown to promote optimum creaming and tenderness in baked foods including cookies <sup>1-4</sup>. Plasticity of a shortening is defined operationally to be smooth, not grainy, holding its shape on a surface, but readily deforming upon a force applied <sup>1</sup>. In a more common sense, plasticity is defined as the correct balance of hardness to stand as a solid to flowing viscous liquid under a shearing force to cause a permanent deformation <sup>7</sup>. Hence, shortening plasticity is a function of SFC and crystal structure. For better quality cookies, moderate levels of SFC at given temperatures and  $\beta'$ type polymorphs are required <sup>1-3</sup>. The oxidative stability of shortenings affects directly the shelf-life of the baked food. The SFC, presence of antioxidants, and processing and storage conditions determine the extent of the oxidation. Hence, shortening with balanced saturated and unsaturated fatty acids composition might be better for both stability as well as health benefits<sup>1</sup>.

Although many different types exist, cookies are usually high fat and high sugar products <sup>3,4</sup>. Shortenings, margarines or butter are the most preferred fats in cookie production, and all have substantial amounts of saturated or even *trans* fatty acids. Negative health effects of *trans* and saturated fats are well accepted phenomenon currently, and strategies to reduce them in processed foods are challenged by researches. Since plastic or structured fats are required for cookies to create desired quality, flavor and shelf-life, alternative shortenings with lower level of saturated and *trans* fatty acids are in demand

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commercially <sup>4-6,8,9</sup>. Biscuits were made with olive and sunflower oil against solid fat, and consumer perception of quality and healthiness was determined<sup>8</sup>. It was indicated that when consumers have not been informed about the content of the biscuits, their liking was lower with the liquid oils, but when the nutritional claim and label were provided, their sensory perception and liking increased. Alternative shortenings or fats with lower levels of *trans* and saturated fats are in search for healthy and better quality baked foods. Organogels or better called oleogels as edible fats are the new conceptual products to satisfy this need. Oleogels are selfstanding, thermo-reversible, anhydrous, physically entrapped three dimensional viscoelastic oil gel networks formed by the addition of small molecules called organogelators which form the self-assembled crystalline fibers via non-covalent interactions. Formation of the gels requires physical inter-chain interactions like hydrogen bonding, van der Waals forces and  $\pi$ stacking between organogelator molecules. Many types of organogelators including fatty alcohols, long chain fatty acids, hydroxy fatty acids, monoglycerides, lecithin, sorbitan tristearate, phytosterol and oryzanol mixtures, ceramides, waxes and wax esters, some polymers and others have been studied. For edible food applications, an organogelator must be food-grade, safe, effective in lower concentrations, easily available and cheap. Among others, plant and animal waxes have become preferable organogelators due to good organogelation functionality, better suitability for different products and relatively low price and commercial abundance <sup>10</sup>-. Edible oleogels, their productions and applications, as well as the properties of different organogelators were masterfully published 14.

There are limited numbers of studies reporting the usage of oleogels as cookie shortenings. Oleogel potential food applications including spreadable fat alternatives and margarines, restriction of oil migration in confectionary, controlled release of nutraceuticals and emulsions have been documented by Hughes et al.<sup>10</sup>. A monoglyceride gel emulsion against some all-purpose shortenings in cookie production was investigated. It was shown that the all-purpose shortening was superior than the gel in cookie quality <sup>4</sup>. In another similar study <sup>5</sup>, oil-water-cellulose ether emulsions against shortenings were used to prepare biscuits. It was indicated that the use of the emulsion yielded biscuits with 33% less fat with very similar texture characters, but consumer acceptance was a little lower than that of the shortening biscuits. In another study of the same research group 9, mechanical and acoustic properties of biscuits made with regular shortenings and liquid oilhydrocolloid mixtures were compared. It was shown that oil/xanthan gum mixture yields more elastic and resistant to breaking biscuits. Furthermore, biscuits with shortenings and oil/hydrocolloid mixtures have shown to confer similar mechanical and mouthfeeling properties. An emulsion of sunflower oil, water and cellulose either was used instead of shortening in biscuits <sup>15</sup>, and creep and oscillatory test were applied to dough. It was indicated that deformation was higher and considerable structural stabilization at lower frequencies were present in dough containing the emulsion. Shellac oleogels were used as shortening alternative for cake preparation. The texture and sensory properties of the cakes prepared with the oleogels were comparable to the standard cake <sup>16</sup>. Lately, a new approach was undertaken to prepare edible oleogels with water soluble polymers (methylcellulose and xanthan gum), and to use them in cake formulation to

compare with reference batches prepared with oil, commercial shortening or cake margarine. It was shown that the dough properties of oleogel batter were more similar to the oil batch, but the texture properties resembled more to the shortening and margarine batches <sup>17</sup>. These promising results implied the necessity for further studies. Not only food functional properties, but also clinical nutritional study of monoglyceride gels used in baked goods was investigated. Sugar free cakes and cookies were made with the gel or compositionally equivalent products, and were served to humans. It was shown that there were no postprandial lipid responses in the treatment groups, indicating no hazard of oleogel application in bakery products applications <sup>18</sup>.

Based on the above summarized studies, it was imperative to investigate in detail the application of different oleogels in baked foods. Hence, in this study we compared the finished products properties, sensory descriptions and consumer preferences of cookies prepared with hazelnut oil-beeswax and hazelnut oil-sunflower wax oleogels against commercial bakery shortening. Although investigation of the dough properties was not our objective, the physico-chemical, textural, flavor and texture profile analysis, consumer tests and storage stabilities of the prepared cookie samples were accomplished.

#### **B** Experimental

#### Materials

The refined hazelnut oil used to prepare the oleogels was purchased from Cotanak Oil Co. (Ordu, Turkey). The manufacturer provided the fatty acid composition of the oil as 0.03% myristic acid, 5.87% palmitic acid, 0.2% palmitoleic acid, 2.64% stearic acid, 82.7% oleic acid, 9.50% linoleic acid, 0.07% linolenic acid, 0.13% arachidic acid and 0.02% behenic acid. Beeswax 8108 was purchased from KahlWax (Kahl GmbH & Co., Trittau, Germany). It was defined as a whitish solid pellet with faint odor and 62-65 °C melting range, and classified as GRAS additive. Beeswax (BW), also recognized as E 901 approved food additive worldwide, is an organic wax produced by bees from the genus Apis mellifera L. It consists of 70-71% total esters, 1-1.5% free alcohols, 9-11% free acids and 12-15% hydrocarbons <sup>19,20</sup>. Sunflower wax 6607L were also obtained from KahlWax (Kahl GmbH & Co., Trittau, Germany). It is a yellowish solid pellet with soft/characteristic odor and melting range of 74-80 °C, and free from dangerous chemicals as indicated by the manufacturer. Sunflower wax (SW) is a hard, crystalline, high melting-point vegetable wax obtained during winterization of sunflower oil. It is known to contain long chain saturated C-42 to C-60 esters derived from fatty alcohols and fatty acids <sup>21</sup>. Commercial bakery shortening (CBS) containing 79% vegetable oils (44% saturated and trans fats <0.80%, palm, cottonseed, canola, safflower, sunflower and linseed oils), water, emulsifiers (sunflower lecithin, mono and diglycerides, polyglycerol esters and propan-1,2-diol esters) whey powder, salt (max. 0.30%), citric acid, potassium sorbate, Vitamins (A, D and E) and beta-carotene, was purchased from local store. Wheat flour, wheat starch, sugar, table salt, edible sodium bicarbonate and sodium pyrophosphate mixture and all other utensils were purchased from local stores.

#### **Oleogel Preparation**

Oleogels of both waxes at 5% (w/w) organogelator addition level were prepared according to our previous studies <sup>13,22</sup>. Briefly, hazelnut oil and each of the SW and BW were placed into separate beher-glasses and heated in a water bath at 90 °C. When the waxes melted completely, and all at isothermal conditions, each wax was added into the oil and stirred for 5 min. Then this mixture was poured into glass cups, and awaited at ambient temperature overnight without any stirring to form the oleogels. These stock oleogels were analyzed and used for the production of the cookies.

#### **Measurement of Oleogel Properties**

The solid fat content (SFC) of the oleogels and commercial bakery shortening (CBS) were measured with a Minispec Bruker NMR Analyzer mq20 (BrukerOptics, Inc.). The samples were first completely melted in water-bath at 90 °C. Secondly, 3.5 ml of each sample were taken into NMR tubes and conditioned in waterbath at 0 °C for 1 h. Then, the tubes conditioned at 20 °C for 30 min, before data recording. The calibration of the NMR was accomplished with standard solutions including 0, 31 and 73.5% solid fat. The melting temperature and enthalpy of the samples were measured with a Perkin-Elmer 4000 Series Differential Scanning Calorimeter (DSC) (Groningen, The Netherlands). Around 5-7 mg sample were weighed into aluminium pans and the pans were hermetically sealed. The samples were heated from room temperature to 140 °C by applying 10 °C /min heating rate. Then, the samples were cooled to -20 °C by 10 °C/min rate and kept for 3 min at that temperature for full crystal formation. Finally, the samples were heated to 100 °C by 5 °C/min heating rate. From the thermograms, the thermal parameters of the samples were calculated using the Pyris 1 Manager software of the instrument. Furthermore, the hardness and stickiness values of the samples were measured with a Texture Analyzer TA-XT2i (Stable Microsystems, Surrey, UK) by placing 150 g samples gelled in plastic cups in a custom-built block using a 45° conic acrylic probe. The method of penetration test was selected with 3.0 mm/s penetration speed into 23 mm depth, and then the probe was pulled out from the sample at 10 mm/s speed. The parameters were calculated by using the instrument software (Texture Exponent v.6.1.1.0, Stable Microsystems). The peroxide values (PV) were measured according to Cd8-53 method <sup>23</sup>. The X-ray diffraction (X-RD) patterns of the samples were taken with a Rigaku D-Max Rint 2200 model X-Ray Diffractometer (Rigaku Int. Corp, Tokyo, Japan). Angular scans ( $2\theta = 2.0$  -50° by 2 °/min) were performed using a Cu source X-ray tube ( $\lambda = 1.54056$  Å) at 40 kV and 40 mA. Data analysis was performed with MDI Jade 7 Materials Data Inc. (Livermore, USA) software program.

#### **Cookie Preparation**

Three different types of cookies were prepared with the recipe formulae given in Table 1. First the standard cookie mixture was prepared as dry mixture, and it consisted of 65% wheat flour, 19% sugar, 11% maltodextrin, 3.5% wheat starch,

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0.5% baking powder (2:3, sodium pyrophosphate: sodium bicarbonate) and 1% salt. Then, 24.07% of each oleogel or CBS and 7.15% liquid homogenized egg were mixed altogether in a bawl with a kitchen mixer (Kitchen Aid, Michigan, USA) for 5 min at 250 rpm, and 5 min at 25 rpm. The cookie dough was sheeted to a thickness of 4 mm by a noodle making machine (Imperia, Moncalier, Italy) and cut 8 cm diameter by using a circular cutting die. Twenty cookies were placed on a perforated tray and baked in an electric oven (Inoksan FPE 110, Bursa, Turkey) at 175 °C for 15 min. After cooling to room temperature, the cookie samples were packed in zipped polypropylene bags and stored at room temperature until analysis. The tray used, the height of tray inside the oven and all other cooking conditions were the same for all cooking experiments. Two separate batches of cookie production for each of the three types of the cookies were made. A picture of the prepared cookie samples is given in Fig. 1.

Table 1. The recipe formulations used to prepare the cookies.

Ingredients (%)	Cookie-I	Cookie-II	Cookie-III
Cookie Mixture <sup>1</sup>	68.77	68.77	68.77
Homogenized egg	7.15	7.15	7.15
CBS <sup>2</sup>	24.07	-	-
BWO <sup>3</sup>	-	24.07	-
SWO <sup>4</sup>	-	-	24.07
Total	100	100	100

<sup>1</sup> Cookie mixture: 65% wheat flour, 19% sugar, 11% maltodextrin, 3.50% wheat starch, %0.50 baking powder (0.20% sodium pyrophosphate and 0.30% sodium bicarbonate) and 1% salt; 2 CBS: commercial bakery shortening: 79% vegetable oils (44% saturated and *trans* fats <0.80%, palm, cottonseed, canola, safflower, sunflower and linseed oils), water, emulsifiers (sunflower lecithin, mono and diglycerides, polyglycerol esters and propan-1,2-diol esters) whey powder, salt (max. 0.30%), citric acid, potassium sorbate, Vitamins (A, D and E) and beta-carotene 3BWO: beeswax oleogel, 4SWO: sunflower wax oleogel.



**Figure 1.** The cookies produced with, (a) CBS, (b) BW Oleogel, (c) SW Oleogel.

#### **Physico-Chemical Properties of Cookies**

The physico-chemical properties were measured the next day after cookie preparation. The surface color of the cookie samples in at least 10 cookies were measured at different points with a Minolta CR-400 colorimeter (Konica Minolta Sensing, Osaka, Japan). Weight and dimensions (diameter and width) of at least 10 randomly selected cookies were recorded with a digital caliper (CD-15CP, Mitutoyo Ltd, Andover, UK). The moisture content of the samples was measured with OHAUS MB45 moisture analyzer (Switzerland), and water activity with AQUA Lab 4TE instrument (Decagon Devices, USA) at room temperature according to the instrument manuals. The ash of the cookies was measured by the AOCS Ba 5a-49 technique <sup>24</sup>. The pH values of the cookie samples were measured by immersing the electrode into the liquid suspension prepared by dissolving 25 g ground cookie in 100 mL neutral pure water. The fat content of the cookie samples was assessed by the Soxhlet technique according to AOAC 920.39 method <sup>25</sup>. The combustion energy values of the cookie samples were determined with a Leco AC-350 bomb calorimeter (St. Joseph, USA) according to its manual. Texture Analyzer TA-XT2i (Stable Microsystems, Surrey, UK) was used to determine the hardness and fracturability values of the cookies according to the technique modified by Piga et al.<sup>26</sup>. 2 mm cylindrical probe with 30 kg load cell was used for the puncture test. First, samples of cookie were placed in the center and fixed on the heavy duty platform. The puncture test was performed with gradients of 3.0 mm/s entrance, 0.5 mm/s inside sample, and 10 mm/s backing speed until 7 mm deepness with 20 g triggering force. Finally the Texture Exponent v.6.1.1.0 software (Stable Microsystems) was used to calculate the hardness and fracturability from the force-time curve. The maximum peak force was used to the calculate hardness value. Fracturability was determined at point where the plot has its first significant peak during the probes first compression into the cookie <sup>27</sup>.

#### **Texture/Flavor Profile Analysis of Cookies**

The Texture/Flavor Profile Analysis (T/FPA) of the cookies was accomplished according to Meilgaard et al.<sup>28</sup>. There were 12 panelists (7 female, 5 male, aged 23-45), voluntarily incorporated in this study, and they were assured about the safety and edibility of the samples being tested by a signed consent form. The panelists were staff and graduate students in our department. Around 10-15 hours of panel training were completed over a week within several sessions. First, the technique was taught to the panelists by the moderator. Then, through participation of the panelists, the texture/flavor descriptive terms were developed for cookie samples by using a broad range of different cookies available commercially. A line scale of 1 for minimum intensity to 5 for maximum intensity anchored on it was used. The panel determined and defined 3 cookie surface properties, 4 cross-sectional texture properties, 5 mouth properties sensed during mastication, and 2 flavor properties to profile the textural and flavor properties of the cookie samples. These descriptive terms together with their definitions are given in Table 2. With a broad range of different foodstuffs as reference materials, the panelists were trained with the scale about the intensity of each attribute defined to a satisfactory level of panel variability. During each panel, the three different cookies were placed on a plate coded with 3 digit random numbers. The panelists were provided samples together with water and expectoration cup under daylight in a sensory laboratory at room temperature. Duplicate samples were analyzed in a randomized setting in different sessions. T/FPA of cookies was also duplicated for each of the two production batches.

**Table 2.** The panel defined descriptive terms and their definitions for the texture/flavour profile analysis (T/FPA) of the cookie samples.

Descriptor	Definition					
Surface Properties						
-Glossiness	The level of cookie surface					
	brightness/opaqueness.					
-Colour	The level of desirable surface color of the					
	cookie.					
-Smoothness	The level of cookie surface homogeneity.					
<b>Cross-Sectional</b>						
Properties						
-Compactness	The amount of pores present interior of the					
	cookie.					
-Pore distribution	The diameter and distribution of the pores.					
-Crust thickness	The level of observable crust thickness.					
-Internal colour	The observed inner section color of the					
	cookie.					
-Colour Differences	The differences between of the crust and					
	interior color of the cookie.					
<b>Mouth Properties</b>						
-Bite Hardness	The resistance of cookie structure against					
	the biting.					
-Fracturability	The brittleness status during chewing.					
-Sandiness	The level of roughness or sandiness in the					
	mouth.					
-Dispersebility	The level of diffusibility in the mouth.					
-Meltdown	The level of diffusion or melting in the					
	mouth.					
Flavour Properties						
-Flavor	The total perception of flavour and aroma					
	when cookie swallowed.					
-Sweetness	The perceived sweetness level of cookie					
	during chewing.					

#### **Consumer Test of Cookies**

In order to assess the consumer hedonic scores of the cookies, a 5-point scale (1 for dislike extremely to 5 for like extremely) was used to measure the sensory attributes of appearance, texture, flavor, smell and acceptability. 200 different volunteer consumers tested each of the 3 different cookies coded with numbers.

#### **Storage Study of Cookies**

Two separate batches of cookies were produced under the same experimental conditions for the storage study. The cookies were placed into zippered polypropylene bags and stored at room temperature for 30 days. Samples were withdrawn every 10th. day and analyzed for texture (hardness and fracturability), moisture content, and peroxide value.

#### **Statistical Analysis**

All physico-chemical measurements data were given as means with standard deviation. The samples were compared with Anova and Tukey's test. The sensory analysis data were compared with non-parametric Kruskal-Wallis and Dunn's tests. The Minitab v.16.1 <sup>29</sup> and SPSS package programs <sup>30</sup> were used for the statistical analysis. The level of confidence was at least 95% in this study.

#### C Results and Discussion

#### Properties of the shortening and oleogels

Some important physico-chemical and textural properties of the BW and SW oleogels and CBS used in the cookie productions are presented in Table 3. The solid fat content (% SFC) values measured at 20 °C have indicated that CBS had significantly higher solid fats than that of the oleogels made with hazelnut oil and the waxes. This is quite an expected result since, it is a well-known phenomenon that organogelation of edible liquid oils does not interfere with their fatty acid composition or saturation level <sup>14</sup>. In fact, this situation has been conferred as one of the main nutritional advantage of the oleogels against hard fat products <sup>10,11,13,22</sup>. It was stated that the solid fat index of different types of commercial shortenings range from 1.5 to 52% at 21 °C<sup>1</sup>. It was also indicated that the presence of some solid fats in shortenings is essential for dough to shorten, or the air cell to develop to yield the preferred structure of baked goods <sup>3,4</sup>. Shortening, thus; must have a moderate level of solid fats for appropriate dough formation. Contrarily, oleogels do not need high levels of solid fat to behave as hard fat stocks. The gel network created by organogelation resembles truly a plastic fat as shown in many studies <sup>13,14,22</sup>. As can be observed from Table 3, the hardness and stickiness values of the two oleogels are lower than that of the CBS, but still they are high enough to behave the oleogel as plastic fat. Our previous <sup>13,22</sup> and many other literatures not to list all here <sup>11,14</sup> separately pointed out this fact. In a study <sup>3</sup>, bakery fat, margarine, hydrogenated fat and sunflower oil were compared for their effect on cookie quality. It was shown that liquid oil was not good enough to entrap the air cell to yield better texture, and it yielded harder cookies. Similarly, consumer hedonic expectations for biscuits made with saturated fat and liquid oils (sunflower and olive oils) have shown that, consumer acceptance was initially lower with liquid oils but

enhanced after informing the consumers with label and health effect informations <sup>8</sup>. In another study, shellac oleogels containing around 12% solid fat at 20 °C were used in cakes, and were found comparable in functionalities of texture and sensory properties to the cake margarine which contained around 25% solid fat at the same temperature <sup>16</sup>.

There was a significant difference between the oleogels samples and CBS for melting temperature and enthalpy. The melting temperatures of oleogels are directly related to the organogelator molecule used to create them, as previously shown <sup>13,22</sup>. Since the melting range of SW (74-80 °C) is higher than that of the BW (62-65 °C) as stated by the producers, the melting point of the corresponding oleogels change dependently. Whereas, the melting enthalpy of CBS is higher than those of the oleogel samples, since it contains much higher amounts of solid fats. These melting behaviours of the oleogel samples are very suitable for baked foods, since they indicate the respective plasticity.

Likewise, the moderate levels of hardness and stickiness compared to the CBS indicate the good enough level of plasticity. It was claimed that plasticity of fats are the function of two factors, SFC and crystal structure<sup>1</sup>. The X-RD patterns of the oleogels and CBS are presented in Fig. 2. The wide-angle region peaks at around 3.70-4.65 Å were observed in all three samples. Small-angle region peak at around 20-23 Å was present in all three samples, but peaks at around 13.84 and 39.75 Å were only found in the CBS sample. These wide-and small-angle region peaks were directly related to the  $\beta'$ polymorph type, which is usually observed in fine triglyceride crystals<sup>31</sup>. The crystalline nature of the wax oleogels also confirmed in previous studies  $^{11,13,22}$ , that was oleogels are in  $\beta'$ type polymorph. This polymorph is characterized with very homogeneous, smooth, creamy and fine texture <sup>11,13,14,22</sup>. The crystal sizes of the samples were also calculated and given in Table 3. In ordinary cookie production, the plastic shortenings were mixed with ingredients to incorporate air bubbles or to cream. For this purpose,  $\beta'$  polymorph and finer crystals were claimed to be better <sup>1</sup>. Hence, the oleogels with similar melting ranges and textural values would yield the very similar results which we have tested in the final products, the cookies.

Table 3. The physico-chemical properties of the oleogels and commercial bakery shortening (Mean±Sd).

Sample	PV	SFC	Tm	ΔHm	Hardness	Stickiness	<b>Crystal Size</b>
	(meqO <sub>2</sub> /kg)	(%, 20°C)	(°C)	(°C)	(N)	(N)	(Å)
BWO	0.36±0.01a	3.30±0.01b	47.67±0.33b	6.64±0.63c	2.73±0.12c	2.12±0.30b	43-64c
SWO	0.30±0.01a	3.64±0.00b	61.02±0.18a	10.45±0.83b	4.18±0.42b	1.68±0.11c	47-89b
CBS	0.33±0.01a	$29.73 \pm 0.39a$	$45.92\pm0.00b$	25.08±0.01a	14.21±0.97a	8.75±0.79a	247-279a

<sup>a-c</sup>Letters show the significant differences within each column (p < 0.05)

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Figure 2. The XRD patterns of the shortenings used in cookie preparation, (a) BW Oleogel, (b) SW Oleogel, and (c) commercial bakery shortening.

The oxidative stability of the shortening or oil used in cookie production is also important for the shelf life of the final products. The peroxide values (PV) of the oleogels were neither different from that of CBS, nor high (Table 3). It was stated that the fat or oil as delivered to bakery should have a PV less than  $1 \text{ meq/kg}^{-1}$ . There is no problem associated with oleogel oxidation observed in the cookies.

#### Physico-chemical properties of the cookies

Picture of the cookies prepared with the two oleogels and CBS is shown in Fig. 1. Samples of the cookies were analyzed for some common quality and composition parameters and the results are presented in Table 4. The instrumental luminosity (L\* values) of the cookies revealed that, the lighter surface color was on the cookie-I sample prepared with CBS, and the darker was cookie-II prepared with BW oleogel. This situation can be also observed from the picture presented in Fig. 1. Contrarily, the a\* values (redness-greenness) indicate no significant difference and a little greenish tones. The b\* value of cookie-I was lower than those of the other two, indicating lower level of yellowness. Although the stock oil was hazelnut oil in both oleogels, still some color differences occurred in the cookies surfaces, because of the different organogelators used to prepare the oleogels. Although color data of the oleogels are not shown in this study, it was shown in previous studies <sup>13,22</sup>, respectively. In one study <sup>4</sup>, the surface color of cookies made with monoglyceride gel and other hard fats were reported (L\* values of 59-63, a\* values of 7.6-9.2 and b\* values of 27-32). The difference between our samples and theirs can be attributed

to the different recipes used and cooking conditions applied. In our study, since the same recipe and cooking conditions were used in cookie preparation, the color difference can only be attributed to the color differences of the shortening or oleogels used.

The average weight and dimensions of the cookie samples are presented in Table 4. The weight and thickness of the cookies prepared with CBS were higher than those of the cookies made with the oleogels, whereas the diameters were not different. It was also evident that the moisture content of cookie-I made with CBS was higher than the other two cookies (Table 5), possibly explaining the higher weight of the cookie samples. The ratio of diameter: thickness was described as spread ratio to discuss the effect of fat type on cookie texture. It was indicated that the higher the spread ratio, the lower the aeration in cookie dough. Furthermore, sunflower oil was found to yield higher spread ratio than that of margarine and hydrogenated fats <sup>3</sup>. The spread ratio for cookie-I made with CBS was the lowest (5.62) compared to the oleogel cookies (7.18 and 6.85). Hence, it can be concluded that the aeration was better in cookie-I prepared with CBS than that of the cookies-II and III prepared with the oleogels. This result is consistent with Jacob and Leelavathi<sup>3</sup>. In another study, low saturated shortening and ultrasound treated shortening were compared for baked foods. It was shown that ultrasound treated shortening caused cookie height to increase and cookie spread to decrease 32.

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Table 4. Some physical properties of the cookies produced by bakery shortening and oleogel samples (Mean±Sd).

Sample	L	a*	b*	Weight	Diameter	Width	Hardness	Fracturability
				(g)	(mm)	(mm)	(N s)	(N s)
Cookie-I	84.13±2.03a	-0.41±0.09a	28.97±1.05b	12.41±0.57a	51.70 1.30a	9.20±0.54a	47.13±4.83a	24.57±3.01c
Cookie-II	79.34±1.76b	-0.35±0.72a	33.55±1.20a	10.68±0.80b	51.80 2.47a	7.21 ±0.4b	31.85±3.91c	33.10±2.60b
Cookie-III	81.77±0.85b	-0.04±0.70a	32.91±0.42a	10.14±0.76b	51.96±0.83a	7.58±0.69b	36.89±5.66b	35.97±2.32a
<sup>a-c</sup> Le	tters show	the signifi	cant differen	ces within	each colu	mn for e	each property	(p < 0.05).

Table 5. Some chemical properties of the cookies produced by bakery shortening and oleogel samples (Mean±Sd).

Sample	Fat	Ash	Moisture	Water	pН	PV	Energy
	(%)	(%)	(%)	Activity		(meqO <sub>2</sub> /kg)	(kJ/g)
Cookie-I	23.61±0.13a	0.79±0.07a	5.72±0.01a	0.46±0.014a	7.54±0.01a	0.45±0.02a	5360.75±5.02a
Cookie-II	28.12±0.25b	0.53±0.07a	2.63±0.48b	0.17±0.05b	7.35±0.01b	0.43±0.01a	5734.60±2.55b
Cookie-III	26.88±0.21c	0.61±0.14a	2.32±0.06b	0.17±0.01b	7.39±0.01b	0.40±0.01a	5794.30±2.83c
<sup>a-c</sup> Lett	ters show the	significant	differences with	in each c	column for	each property	(p < 0.05).

The hardness and fracturability values of the cookie samples were also measured and compared (Table 4). Cookie-I made with CBS was significantly harder than cookie-II and III made with the oleogels. Contrarily, the fracturability of cookie-I was the lowest among others. The effects of bakery fat, margarine, hydrogenated fat and liquid sunflower oil on cookie dough hardness and cookie breaking strength were compared <sup>3</sup>. It was shown that dough hardness was higher in hydrogenated fat samples, but cookie breaking strength was highest in sunflower oil containing samples. These findings concur with ours that less harder cookies made with the oleogels are more fracturable. Since oleogels contain more liquid oil than CBS, their effect on cookie hardness and fracturability are rather similar to sunflower oil reported by Jacob and Leelavathi<sup>3</sup>. This situation can also be attributed to the very fine crystals and  $\beta'$  polymorph of the oleogels, which seem to not support aeration in cookie dough <sup>1,3</sup>. In another study <sup>4</sup>, monoglyceride gel, dried and hydrated monoglycerides and Crisco oil were used in cookie preparation, and it was shown that monoglyceride gel containing cookies resulted in greater dough firmness, lower width and length of the cookies and the same cookie breaking strengths. The researchers suggested that all purpose shortening is better than the monoglycerides in overall cookie quality. Sunflower oil-water-cellulose ether emulsions replaced fat in biscuit production, and it was shown that penetration force and three-point break force for biscuits were not significantly different than the control sample. It was also suggested that higher hardness in biscuits is a quality defect and might be due to liquid oil or lower percentage of total solid fat <sup>5</sup>. In another study <sup>9</sup>, shortenings in biscuits were compared with liquid oil/hydrocolloid mixtures for textural properties. When shortening was replaced by the oil/xanthan gum system, the biscuit proved to be more elastic, resistant to breaking and noisy during penetration. The same research group <sup>15</sup> have also compared creep and oscillatory rheological test for biscuits made with cellulose emulsions as shortenings, and found out that dough with the emulsion had greater deformation as well as higher structural stabilization. Furthermore, the methoxyl and hydroxypropyl levels of the cellulose ether did not exert an

effect on the dough rheological properties. Edible grade shellac oleogels were used in spreads, chocolate paste and cake production. The texture profile results of the cakes made with the oleogel were comparable with the cake made with margarine <sup>16</sup>. The same research group <sup>17</sup> also prepared edible oleogels based on water soluble polymers and applied them to cake production. It was indicated that cake batter properties of the oleogel batch were more close to oil batch, but the textural properties of the cakes were similar to those of the cakes prepared with shortenings. Our findings concur with these results to indicate that oleogels composed of very fine  $\beta'$  type crystals are good enough to create less hard but more fracturable cookie texture compared with commercial bakery shortening (CBS) used. The results of spread ratio and texture measurements suggest acceptable cookie texture with the oleogels used.

Some chemical properties of the cookie samples are given in Table 5. Ash, moisture contents and water activity values of cookie-I were a little higher than the other two, whereas the fat content of the oleogel cookies (cookie-II and III) were higher than that of the CBS cookie. Likewise, combustion caloric value of the oleogel cookies was higher, respectively. As shown in Table 1, all three cookie types were prepared with the same added amounts of fat (24.07% each), but the CBS is itself an emulsion product composed of 79% vegetable oil and other ingredients. Contrarily, oleogels are full-fat products composed of 95% hazelnut oil and 5% organogelator wax. Hence, the difference in cookie fat content and caloric value must be attributed to this difference. Higher moisture content and hence water activity in cookie-I can also be attributed to the amount of water present in the CBS and/or to the higher water retention capacity of this type. Moisture levels of 1.3-2.7% were measured in cookies made with regular and sonicated allpurpose shortenings <sup>32</sup>. Monoglyceride gel, dried and hydrated monoglycerides and crisco oil were used in cookie formulae and it was shown that the moisture of the final products was around 4-8%. Goldstein and Seetharaman<sup>4</sup> pointed out that cookie height correlated with increasing moisture content. The same finding was detected for our samples, the highest

thickness (Table 4) of cookie-I concur with highest moisture content (Table 5). For cookies and similar products, water activity below 0.3 is usually preferred for improved microbial and enzymatic stability, as previously suggested <sup>5</sup>. Hence, oleogel cookies might have longer biological shelf life. There were no significant differences among the samples for the measured pH and peroxide (PV) values. It was indicated that the fat delivered to bakery foods must have PV below 1 meq/kg to extend the shelf life of the baked food <sup>1</sup>. The peroxide value of biscuits prepared with corn, sunflower and high oleic sunflower oils were monitored during storage, and maximum level of peroxide was between 1-1.5 mmol/kg oil determined after 166 days of storage <sup>33</sup>. Hence, the PV's measured in our cookie samples are quite low and acceptable.

#### Sensory properties of the cookies

The sensory texture/flavor profile analysis (T/FPA) results are presented in Table 6. Non-parametric statistical comparison of the scores was completed with the median values. There were 15 sensory definition terms determined by the panel to sensorially describe and compare the cookies. The scale was between 1 for minimum intensity and 5 for maximum intensity perceived for each attribute. Definitions of all sensory terms are given in Table 2. The surface appearance of the cookies was described by 'glossiness', 'color', and 'smoothness' terms. There were no statistically significant differences among the samples for these properties. Surface glossiness was a measure of cookie surface brightness and ranged around 3.25-3.90, indicating a moderately high level. The instrumental luminosity (L\* value) level shown in Table 4 indicated the most luminous sample cookie-I, but here the panel found its glossiness a little lower than the others. The panel determined the level of surface color as the desirability of the color, not the tones. The color scores of the oleogel cookies (cookie-II and III) were a little higher than that of the CBS cookie-I, but they were not significant statistically. The level of cookie surface homogeneity determined as smoothness was found to be 4.0 on the scale. This is a relatively high score indicating homogeneous surface, which can be also observed in Fig. 1.

As the cross-sectional properties, there were 5 sensory terms determined by the panel. The panelists broke half one cookie with their hands and then evaluated the cross-sectional properties. 'Compactness' determines the amount of pores present in the interior of the cookies, and higher compactness indicates lesser amounts of pores. Cookie-I had significantly lower scores than those of cookie-II and III, indicating a better air bubbling inside the cookie. Cookie dimensional measures given in Table 4 also confirm this situation. 'Pore distribution' defines the diameter and size distribution of the pores present in the interior of the cookies. There was no significant difference among the samples, and the scores were all over three. The 'crust thickness' was the level of observable crust thickness on the cookie cross-section. Although statistically not different, crust thickness values of the oleogel cookies were a little higher than that of cookie-I made with CBS. As shown in Table 4, these two cookies had also higher fracturability values, and instrumental fracturability might be related to sensory crust thickness. A similar conclusion was made for biscuits made with oil/hydrocolloid mixtures <sup>9</sup>, that the higher the breaking force (in our case fracturability), the higher the hard and crunchy perceptions. The inner section colors of the cookies were measured with 'internal color' term, and no difference was observed among the samples. The difference between the crust and inner section color was determined with the 'color difference' term. There were some perceived color differences of surface and interior section as evident with the measured scores around 3.70-3.80, but there was no significant difference among the cookies. These cross-sectional definition terms scores indicate quite similar or comparable measures with the oleogel cookies compared to CBS cookies.

**Table 6.** The texture/flavor profile analysis (T/FPA) results of the cookie samples (Mean±SE; Me).

<b>Description Terms*</b>	Cookie-I	Cookie-II	Cookie-III
Surface glossiness	3.27±0.35	3.81±0.12	3.90±0.21
	3.00	4.00	4.00
Surface Colour	$3.45 \pm 0.20$	4.00±0.19	$4.00 \pm 0.27$
	3.00	4.00	4.00
Surface smoothness	$3.81 \pm 0.18$	4.09±0.09	3.81±0.29
	4.00	4.00	4.00
Compactness	$3.00\pm0.19$	3.90±0.31	4.00±0.23
	3.00b	4.00a	4.00a
Pore distribution	$3.36 \pm 0.24$	$3.72 \pm 0.33$	4.09±0.16
	4.00	4.00	4.00
Crust thickness	$3.00{\pm}0.23$	$3.72 \pm 0.38$	3.81±0.22
	3.00	4.00	4.00
Internal colour	3.18±0.26	$3.72 \pm 0.23$	$4.00 \pm 0.27$
	3.00	4.00	4.00
<b>Colour Differences</b>	3.81±0.29	3.81±0.29	3.72±0.27
	4.00	4.00	4.00
Bite Hardness	$3.00 \pm 0.27$	4.27±0.30	4.18±0.22
	3.00b	5.00a	4.00a
Fracturability	$3.09 \pm 0.28$	4.27±0.27	4.18±0.29
	3.00b	5.00a	4.00a
Non-Sandiness	3.36±0.24	3.36±0.31	4.09±0.34
	3.00	4.00	4.00
Dispersibility	3.45±0.15	$3.72 \pm 0.42$	4.45±0.24
	3.00	4.00	5.00
Meltdown	$3.27 \pm 0.30$	3.90±0.31	4.36±0.24
	3.00b	4.00ab	5.00b
Flavour	3.36±0.31	4.00±0.33	4.18±0.35
	3.00	4.00	5.00
Sweetness	$4.98 \pm 0.10$	4.97±0.15	$4.98 \pm 0.10$
	5.00	5.00	5.00

\* The scale of 1 at the left end for minimum and 5 at the right end for maximum intensity was used.

<sup>a-c</sup>Letters show the significant differences within each line (p < 0.05).

Five sensory definition terms were used to define the mouth properties of the cookies (Table 6). 'Bite hardness' was the resistance of cookie structure at first bite by teeth. Cookie-I made with CBS had significantly lower scores of bite hardness than those of cookie-II and III made with the oleogels. The opposite measurements of textural hardness values were indicated in Table 4. Furthermore, the textural fracturability values (Table 4) seem to agree with this sensory measurement. Sensory 'fracturability' described as the brittleness level of the cookies during chewing were also significantly different among the samples, and cookie-I was significantly lower than the other two samples. If we believe that sensory fracturability is related to textural fracturability, the results concur this time. In one study <sup>6</sup>, biscuits with different shortenings and oil/gel system were formulated and sensory evaluation by free choice technique was carried out. The consumers developed sensory terms for first bite, during chewing and during bolus aggregation, and swallowing stages. Hardness and crunchy were described at first bite, while crisp and hard were measured during chewing. Although sensory evaluation techniques are totally different, some similar terms with our studies were used. It was indicated that as fat level decreased, the biscuits became more harder, drier and less flavored. Tarancón et al.<sup>9</sup> also indicated that for biscuits made with oil/hydrocolloid mixtures, high number of micro-fractures measured instrumentally correlate well with the perception of mealy, crunchy and brittle sensations. This result also agrees with our findings that sensory fracturability and instrumental fracturability values are in good accordance. The panel in this study used the 'nonsandiness' term to measure the cookie roughness level during chewing, and determined no significant difference among the cookies. The higher the non-sandiness score, the lower the roughness in the mouth were measured, as the panel reported. Tarancón et al.<sup>6</sup> used the 'easy to chew' and 'mealy' terms as similar definitions to 'non-sandiness' term in this study. 'Dispersibility' defined as the level of diffusivity in the mouth was also measured. The samples did not show any difference for this term. Similar to this, but defining the actual diffusion or solubilisation (melting) of the cookies in the mouth was measured with the 'meltdown' term. The meltdown of cookie-II made with SW oleogel was a little higher than the other two. This might be caused by the higher melting point of the SW oleogel (Table 3). This term may resemble the 'easy to swallow' or 'fat mouthfeel' term defined in the study of Tarancón et al.<sup>6</sup>.

In this study, the panel determined two sensory terms to measure the flavor properties of the cookie samples, namely 'flavor' and 'sweetness' terms. The 'flavor' term defines the total perception of flavor and aroma associated with regular cookies after it is swallowed. 'Sweetness' was the perceived regular level of sugar sweetness in the cookies. There were no significant differences among the cookies for both terms. 'Sweet', 'roasted flavor', 'fat flavor' and 'buttery flavor' were used in the panel of Tarancón et al. <sup>6</sup> as well.

Unfortunately we could not found sensory descriptive analysis studies made with cookies prepared with oleogels. There is only one study <sup>16</sup>, in which shellac oleogels were used in not cookie but cake formulations and sensory properties were measured. Sensory definition terms of 'volume', 'cell size', 'moistness', 'stickiness', 'sponginess', and 'crumbliness' were used to compare the samples. It was indicated that the cakes prepared with shellac oleogels had sensory scores comparable to the cakes prepared with margarine. In our study, we also defined the oleogels cookies against CBS cookies with the panel and found comparable or even better sensory scores. This data would be very beneficial for the literature.

Sensory success of a food product can only be proved by consumer tests. Hence the cookies were evaluated by 200 consumers with the aim of 5-point hedonic scale, and the results are presented in Fig. 3. Consumers scored the appearance, texture, flavor, smell and overall acceptability of the three cookie samples presented with number codes. Apparently there is no significant difference among the samples for the measured attributes. All measured scores were above 3.80 value indicating a good level of consumer preference. Although there was no significant difference among the samples, the scores measured for the oleogel cookies (cookie-II and III) were always a little higher than those measured in cookie-I made with CBS. This result indicated that oleogels yield well accepted and preferred cookies by the consumers. In one study <sup>5</sup>, biscuits with sunflower oil-water-cellulose ether emulsions were prepared and tested by consumers for appearance, color, texture, flavor, sweetness and overall liking. Similar to our results, they have indicated well accepted level of scores for biscuits prepared with the emulsions against regular shortenings. In another study<sup>8</sup>, it was shown that consumers like less biscuits with lower level of fats or liquid oils, but when informed about the health effects their liking for biscuit made with liquid oils enhances. Hence, oleogel properties of being free from trans fatty acids and very low in saturated fat can be advertised to consumer for higher acceptance.



Figure 3. Consumer hedonic scores for the cookies (1- dislike extremely to 5- like extremely; Mean $\pm$ SE; n = 200).

#### Storage stability of the cookies

A short-time (30 days) storage study at room temperature was carried out to observe any changes in the cookie samples prepared with the oleogels and CBS. At every 10th. days, the hardness, fracturability, moisture content and peroxide values were monitored, and the results are shown in Fig. 4. Cookie hardness exhibited (Fig. 4a) a different trend of changes during storage. For cookie-I made with CBS first an increase and then a decrease in hardness at 30th. day was detected. The hardness

of cookie-II made with the BW oleogel increased during storage, whereas hardness of cookie-III made with SW oleogel decreased significantly and gradually during storage. After 30 days of storage the hardness of cookie-I and II were not significantly different from each other but significantly higher than cookie-III. Hence, it can be concluded that BW oleogel was more similar to CBS in terms of cookie hardness during storage. Textural fracturability values of the cookies during 30 days storage were presented in Fig. 4b. Cookie-I and II had a primary increase followed by decrease at 30th. day. Contrarily, fracturability of cookie-III decreased gradually during storage, just like hardness. At the end of the storage period, the fracturability level of cookie-II was the highest, and that of cookie-III the lowest. For these two textural properties, it is clear that cookie-II produced with BW oleogel changed slightly, and would be better in terms of shelf life.

The changes in the cookie moisture level and peroxide value (PV) during the storage are presented in Fig. 4c and 4d. In all cookie samples, the moisture content increased gradually during storage. After 30 days, the moisture content of cookie-I was significantly higher than the other two, and moisture contents of cookie-II and III were not significantly different

from each other. Piga et al. <sup>26</sup> monitored the water activity of 'Amaretti' cookies, wrapped in polyvinylchloride (PVC) film or aluminium foil (ALL), for 35 days of storage. It was shown that cookies in PVC had a progressive and slow decrease in water activity, while the opposite was evident in ALL stored samples. The water activity of the cookies in that study was between 0.3 and 0.75, indicating around %4.5-7.5 moisture levels. Overally, there was not a large change in moisture content of the cookies under the stated storage conditions in this study to predict good level of freshness in the cookies. The peroxide values in the cookie samples increased very little during storage, thus the change was not significant statistically. After 30 days, all PV's were below 0.55 meqO<sub>2</sub>/kg oil. Clearly the oil used to prepare the oleogels (hazelnut oil) was fairly stable in the cookies during storage. Stauffer <sup>1</sup> indicated that shortenings used in bakery foods must have a PV below 1.0 meqO<sub>2</sub>/kg to be suitable. Furthermore, under better storage conditions with more suitable packaging materials, the oxidative stability as well as textural durability of the cookies might be much improved.



Figure 4. The changes of the hardness (a) and fracturability (b) values, and of the moisture (c) content and peroxide value (d) during storage in the cookie samples.

#### **D** Conclusions

In this study, the physico-chemical properties, sensory definitions, consumer hedonic scores and storage stabilities were accomplished for cookies prepared with two different oleogels against commercial bakery shortening. The textural and compositional properties of the oleogel cookies were quite comparable to regular cookie made with the shortening. Fifteen different definition terms were used by the panel to sensorially examine and compare the cookies. These data serve, by far the very important addition to this line of the literature. Most positive attributes scores of the oleogel cookies were equal or even higher than that of the regular cookies, indicating good quality. Likewise, consumer hedonic data indicated that oleogel cookies are more preferred and better accepted than the regular cookies. During the short term storage at room temperature, the cookie samples preserved their quality with minimum changes. In conclusion, this study points out that wax oleogels can be very successful in cookies as shortenings, and they provide very healthy fat without any trans fatty acids and with very low levels of saturated fatty acids.

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#### Notes and references

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