Rapeseed Oil Based Oleogels for the Improvement of the Fatty Acid Profile Using Cookies as an Example

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In this study rapeseed oil based oleogels with sunflower wax, ethyl cellulose, monoacylglycerols, or mixtures of it as oleogelators are characterized in comparison to conventional bakery margarines or partially hydrogenated peanut fat with regard to their oil-binding capacity, hardness, and rheological behavior. Oleogels prepared with 5% and 10% sunflower wax and 5% monoacylglycerols with 5% ethyl cellulose, respectively, show comparable properties as the conventional fat phases with regard to the mentioned parameters. The rheological properties reveal that storage and loss moduli of oleogels prepared without sunflower wax are significantly lower in comparison to conventional margarines. In addition, cross-sectional views allow a very deep insight in the 3D structure of the oleogels via cryogenic scanning electron microscopy. For oleogels consisting of ethyl cellulose, entrapped oil droplets can be visualized and the pore size distribution is established by a semiautomatic approach. The use of oleogels results in a reduction of saturated fatty acids from 29%–47% to 7%–13% and in an increase of polyunsaturated fatty acids of at least 22% in oleogel-based cookies. Cookies prepared with oleogels are accepted by the consumer, even if the overall evaluation is slightly worse in comparison to the conventional products.
Practical Applications: The application of rapeseed oil based oleogels with sunflower wax, ethyl cellulose, monoacylglyerols, or mixtures is a very good possibility to replace fat phases in bakery products high in saturated fatty acids and to improve the fatty acid profile of the foods. This results in food with a higher content of monounsaturated oleic acid and polyunsaturated linolenic acid recommended for a healthier nutrition.

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1. Introduction

According to the German "Guidelines for Fine Bakery Products,"[1] fine bakery products contain at least 10% fat or oil resulting in a high-fat bakery product. The viscoelastic and mechanical properties of fats and oils applied during the process of production provide for desired characteristic features in bakery products.[2]

The fat phase limits the formation of a stable gluten network by evenly coating flour particles while kneading the dough, which prevents shrinking after the lamination and the cookies get a so-called short texture.[3] In addition, the interaction of the fat phase with other components of the dough directly affects the processability and characteristics of the cookies in regard to dough handling, lubricity, elasticity, shortness, tenderness, flakiness, and texture.[4,5] Moreover, the type and structure of the lipid phase influences the sensory properties of the product either by directly contributing to the taste and smell or by the formation of degradation products during processing or storage.[6,7] While the solid fraction of the fat phase tends to contribute to the structure of the dough or the finished baked product and traps air bubbles during dough preparation, the liquid fraction is responsible for a moist mouthfeel, lubricity, and a soft bite.[8]

In the past, the availability of cheap raw materials and the possibility to produce fat with specific physical properties caused a use of partially hardened fats like peanut fat or shortenings as fat phase in the production of bakery goods.[9] At the beginning of the 1990s hardened fats were replaced by fats of tropical origin such as palm oil due to the discussion about adverse health effects of *trans* fatty acids occurring in high amounts in partially hydrogenated fats.[10,11] Today the extensive use of palm oil is also under public discussion as palm plantations are still cultivated in sensitive areas that are important for the climate. In addition, palm oil is declined due to its high potential for the formation of 3-monochloro-1,2-propandiol and glycidyl fatty acid ester that pose a health risk.[12,13] Interesterification of liquid vegetable oil with completely hydrogenated fat results in semisolid fats that also could be used in bakery products, because they are free of *trans* fatty acids, but they contain a high amount of saturated fatty

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acids. Therefore, even if saturated fatty acids are seen less critically as *trans* fatty acids, solid fat based on interesterified liquid oil is not a recommendable alternative.[14]

A more promising alternative are oleogels on the basis of rapeseed oil. Edible oleogels are simply defined as gels of a structurant with an organic liquid oil as solvent that allows to formulate food products with a nutritionally favorable fatty acid composition.[15] While in solid or semisolid fats high-melting triacylglycerols create a network in which low-melting triacylglycerols are integrated, oleogels are formed by the use of an organooleogelator such as ethyl cellulose (EC), monoacylglycerols, fatty acids, alcohols, or waxes. After heating, melting, and cooling they form a 3D network in which the liquid phase is embedded in a gel-like structure.[16] These oleogels are self-standing, thermoreversible, and viscoelastic. The characteristic physical and rheological properties of oleogels are significantly influenced by type and concentration of the oleogelator,[17,18] whereby even a small amount of solid gelator is sufficient to produce oleogels with solid characteristics. Moreover, the properties of oleogels can further be adapted and optimized in a wide range by variation of the oleogelation temperature and the cooling rate.[19]

The type of the edible oil used for oleogelation is critical for the nutritional quality of the oleogel.[20,21] For example, rapeseed oil is characterized by a high level of monounsaturated oleic acid ($\approx$60%), a low level of saturated fatty acids ($\approx$7%), and a favorable ratio between omega-6 linoleic acid ($\approx$20%) and omega-3 alpha-linolenic acid ($\approx$10%) recommending the oil for human consumption.[22,23] Therefore, the addition of a structurant to rapeseed oil during oleogelation allows to bring the technological properties of the oil closer to those of solid fats, usually used in the production of fine bakery products, without changing the positive nutritional properties of the oil.

In literature a lot of different oleogels are described and characterized but only few papers have been published showing the application of oleogels in foods[24–27] and especially in bakery products.[17,28–31]

In a review on edible oil structuring Patel and Dewettinck[16] stated that the studies carried out to date found that the rheology of intermediate products (cake batter and cookie dough) is much less developed in oleogel-based bakery goods compared to those prepared using commercial bakery fats. The end products show comparable texture, stability, and sensorial properties. Goldstein and Seetharaman[32] showed that compared to all-purpose shortenings the use of monoacylglycerol based gel emulsions resulted in a higher firmness of the cookies. Patel et al.[33] used shellac oleogel based w/o emulsions (20 wt% water) as a shortening alternative for the preparation of cakes and found comparable functionalities (texture and sensory attributes) to the standard cake. In another study on cellulose emulsions as fat replacers in biscuits Tarancón et al.[34] demonstrated that biscuits formulated with either olive oil or sunflower oil and hydroxypropyl methyl cellulose (HPMC) with oil, water, and thickener in the proportions of 47%, 51%, and 2%, respectively, had the closest sensory properties to shortening biscuits.

The use of waxes as structuring oleogel components was also investigated in various bakery product studies. Doan et al.[35] found that carnauba wild wax, berry wax, candelilla wax, beeswax, and sunflower wax [SFW] were efficient oleogelators forming strong gels at concentration <2% w/w. The excellent crystal-

Figure 1. Photographs of oleogels based on rapeseed oil and different concentrations and/or combinations of sunflower wax (SFW), ethyl cellulose (EC), and monoacylglycerides (MG).

lization properties of waxes to liquid oil are explained with the low polarity, long chain length, and high melting point of the compounds resulting in well-formed networks with strong oil-binding properties even at low concentrations.[36] Jang et al.[28] described desirable spreadable properties and soft eating characteristics for cookies after replacement of the shortening by oleogels of canola oil and candelilla wax, although the firmness of the oleogels was lower than that of the shortening at room temperature. In addition, Mert et al.[10] demonstrated that cookies became harder when shortenings were totally replaced by oleogels on the basis of candelilla wax and canola oil. The usefulness of waxes for structuring oleogels was also shown by Yilmaz and Ogutcu,[37] who used sunflower wax and beeswax together with hazelnut oil to replace shortenings from cookies. Almost all properties of the oleogel cookies were comparable to the control cookies showing the good applicability of wax-based oleogels for the replacement of shortenings.

The aim of the present study was to compare the properties of oleogels prepared with different types of oleogelators, sunflower wax, ethyl cellulose, and monoacylglyerols as well as mixtures of these three oleogelators to conventional fat phases. In addition, the processability of these oleogels for the preparation of cookies and the properties of the cookies should be investigated.

2. Results and Discussion

In the present work, three different oleogelators (SFW, EC, and monoacylglycerides [MG]), individual or in mixtures, in different proportions to refined rapeseed oil have been used for the preparation of the oleogels (**Figure 1**). The properties of the oleogels were compared to partially hardened peanut fat and three different types of bakery margarine. Finally, cookies were prepared by use of the oleogels. Their properties were compared to features of cookies prepared with conventional fat phases.

2.1. Oil-Binding Capacity of Oleogels

For oleogels, the oil-binding capacity (OBC) describes the ability of the oleogelator to keep the liquid phase (rapeseed oil) in a

Table 1. Oil-binding capacity.

Sample	Oil-binding capacity of conventional solid fats [%]				Oil-binding capacity of oleogels [%]					
Temp.	PNF	Margarine A	Margarine B	Margarine C	SFW5	SFW10	SFW5 EC5	EC2.5 MG2.5	EC5 MG5	EC7.5 MG2.5
8 °C	99.9 (0.0)[a]	99.9 (0.0)[a]	99.9 (0.0)[a]	99.9 (0.0)[a]	99.6 (0.4)[a]	99.9 (0.1)[a]	96.3 (0.6)[a,b]	93.0 (5.1)[b]	98.9 (0.4)[a]	92.1 (0.7)[b]
20 °C	99.9 (0.0)[a]	99.6 (0.2)[a]	99.5 (0.2)[a]	99.4 (0.2)[a]	98.9 (0.3)[a]	99.7 (0.1)[a]	95.7 (2.0)[a,b]	92.0 (4.1)[b]	91.9 (1.5)[b]	86.2 (1.2)[c]
30 °C	85.2 (0.7)[b]	99.9 (0.0)[a]	99.9 (0.6)[a]	99.9 (0.0)[a]	97.6 (1.4)[a]	99.5 (0.0)[a]	89.0 (1.1)[b]	76.2 (3.4)[c]	87.0 (2.6)[b]	86.1 (1.1)[b]

Calculations of significant differences were performed by JMP via the application of a Tukey-Kramer test of all samples ($p < 0.05$). Significant differences were indicated by different letters (a-d).

stable structure, even when external forces affected the system. A high oil-binding capacity of the solid fat is critical for the final quality of the bakery product, because at low oil-binding capacity the liquid phase of the vegetable oil flows into the baked goods during storage.[38] **Table 1** shows the oil-binding capacity of partially hardened peanut fat and three margarines in comparison to different types of oleogels. The oil-binding capacity of the margarines is equal or higher than 99.4%, even at a temperature of 30 °C showing the high stability of the formed structure to bind the liquid phase. Partially hardened peanut fat also showed a high oil-binding capacity at temperature up to 20 °C, but at a higher temperature the solid structure of the hardened fat released some liquid material (14.8%) as a consequence of centrifugal treatment. This revealed the lower structural stability of peanut fat at a higher temperature in comparison to the margarines. A similar oil-binding capacity as margarines and a better OBC than partially hardened peanut fat at 30 °C has the oleogel with 10% SFW as oleogelator. The oil-binding capacity is equal to 99.5% or higher with no significant influence of the temperature. Also, SWF5 showed a high oil-binding capacity of 99.6% at 8 °C that decreased with increasing temperature, but the differences to SWF10 were not significant. The other oleogels showed significantly lower results for the oil-binding capacity with values ranging between 76.2% (SFW5EC5, 30 °C) and 98.9% (EC2.5MG2.5, 8 °C). The high oil-binding capacity of SFW5 and SFW10 even at higher temperature shows that the structure of the oleogels remained solid, ensuring a high gas retaining ability, which is necessary for the size and stability of gas bubbles during the heating in the baking process.[39,40] The longer the gas bubbles are retained in the dough, the larger is the volume of the final product.

The other oleogels (mixtures of EC and MG) showed significantly lower oil-binding capacities, which indicates a more limited applicability of these oleogels in the production of bakery products. Especially the limited ability of oleogels with lower OBC and lower stability at higher temperature to stabilize gas bubbles before and during the early stages of baking, will result in more compact products. These results correlate with the findings of Yilmaz and Ogutcu[37] that, in comparison to beeswax, MG cannot form stable (nonflowing) gels due to worse oil-binding capacity and higher concentrations of MG were needed.

2.2. Hardness of Oleogels

An important parameter for the characterization of oleogels is the hardness that describes the mechanical properties and gives an idea about the force that is necessary to deform the oleogel. This parameter directly influences the applicability of the oleogel for the preparation of the final food product.

The hardness of the conventional margarines ranged between 2.43 and 3.69 N at 20 °C and as expected showed a temperature dependency with decreasing values from 8 to 30 °C (**Table 2**). The hardness of conventional margarines was significantly lower at 8 and 20 °C, but higher at 30 °C compared to partially hardened peanut fat. Although it has been supposed that the rheological feature hardness depends on the solid fat content (SFC) of the fatty phase, some studies showed that not only SFC is responsible for the decrease of hardness with increasing temperature. Moziar et al.[41] also did not find a linear correlation between hardness and SFC. Narine and Marangoni[42] stated in a review that hardness depends on the nature of the microstructure and Rousseau and Marangoni[43] showed the correlation between hardness as rheological feature and as determined by sensory evaluation.

In comparison to PNF and margarines the hardness of different oleogels was much lower at 8 and 20 °C except for EC7.5MG2.5. The hardness of this oleogel at 8 °C was comparable to margarine A and B and an even higher hardness than the conventional solid fats at 20 °C was observed, which was only comparable with PNF.

It is also noticeable that the hardness of most of the oleogels was already very low at 20 °C and a further increase in temperature to 30 °C did not result in a significant change. On the other hand, the decrease in hardness was significant for the conventional fats, if the temperature was increased from 20 to 30 °C. Only the oleogel EC7.5MG2.5 showed a noticeable hardness at 30 °C, even higher than for the conventional solid fats. The hardness of the margarines was only comparable to oleogels consisting of SFW10 and EC7.5MG2.5 as oleogelator. The other oleogels were too soft which made a replacement of margarines by these types of oleogels in the processing difficult.

Table 2 also shows the effect of the concentration of the oleogelator on the hardness of the oleogel. With increasing amount of SFW in the oleogel from 5% to 10% the hardness increased significantly and even at 30 °C oleogel SFW10 showed a hardness comparable to conventional fats. In a mixture with SFW5 and EC5 the hardness of the resulting oleogel was lower (but not significant) in comparison to the pure application of SFW5. For the application of EC in combination with MG also an increased hardness was found for an increasing amount of EC as already stated by Zetzl et al.[44] At EC2.5 and MG2.5 the concentration of EC was far below the critical EC concentration of 6% that produce oil gelation as proposed by Zetzl et al.[44] for rapeseed oil. The authors stated that at lower concentration of EC in the oleogel

Table 2. Hardness of conventional fats and oleogels with different oleogelators.

Sample Temp.	Hardness of conventional solid fats [N]				Hardness of oleogels [N]					
	PNF	Margarine A	Margarine B	Margarine C	SFW5	SFW10	SFW5 EC5	EC2.5 MG2.5	EC5 MG5	EC7.5 MG2.5
8 °C	30.41 (3.41)[a]	6.15 (0.64)[c]	6.09 (0.48)[c]	10.62 (0.71)[b]	0.33 (0.04)[e]	1.93 (0.12)[d,e]	0.21 (0.02)[e]	0.10 (0.01)[e]	3.68 (0.20)[c,d]	5.53 (0.24)[c]
20 °C	5.27 (0.35)[a]	2.43 (0.16)[c]	2.58 (0.08)[c]	3.69 (0.33)[b]	0.28 (0.03)[e]	1.59 (0.07)[d]	0.15 (0.02)[e]	0.02 (0.00)[e]	1.02 (0.09)[d]	5.44 (0.60)[a]
30 °C	0.26 (0.01)[e]	1.34 (0.08)[c]	0.79 (0.04)[d]	2.29 (0.12)[b]	0.26 (0.03)[e]	1.34 (0.10)[c]	0.19 (0.01)[e]	0.03 (0.00)[e]	0.96 (0.02)[c,d]	4.63 (0.56)[a]

Calculations of significant differences were performed by JMP via the application of a Tukey-Kramer test of all samples ($p < 0.05$). Significant differences were indicated by different letters (a-d).
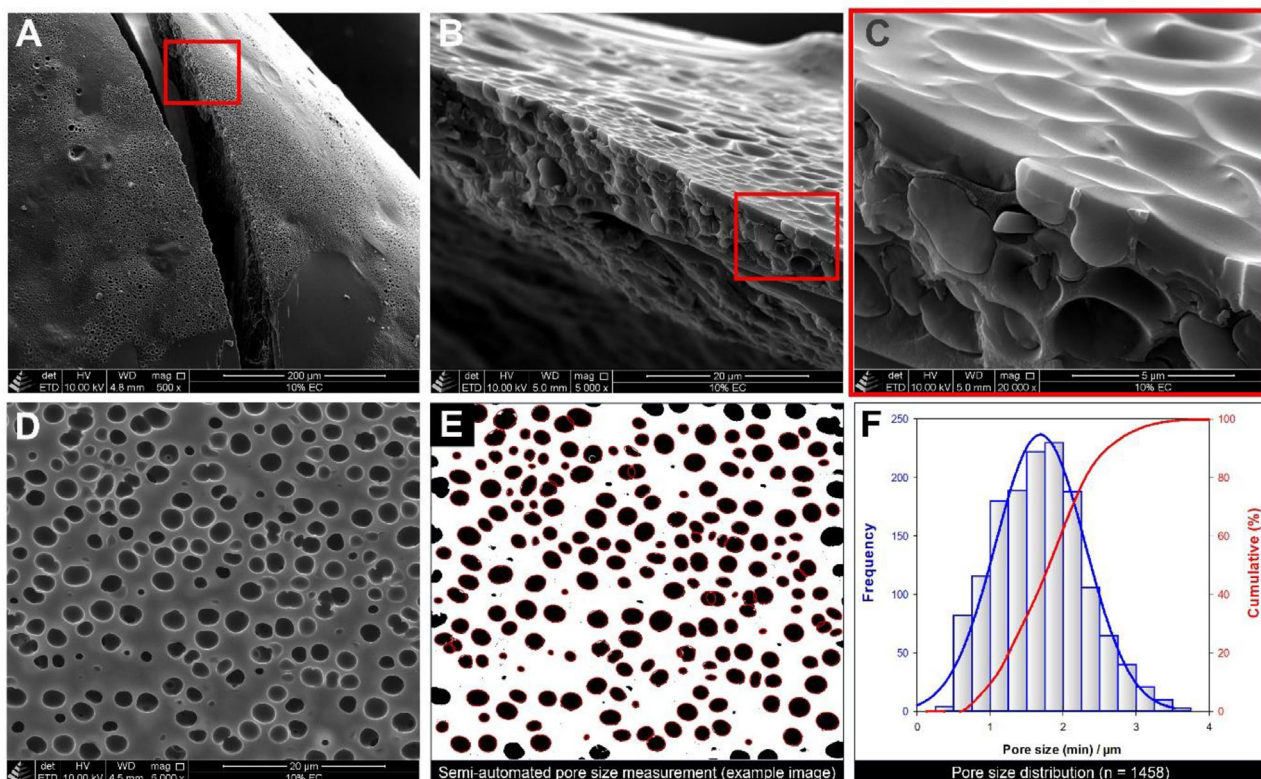
Figure 2. Cryo-SEM images of de-oiled oleogels based on rapeseed/canola oil and 10% ethyl cellulose (EC) at different magnification levels: A) 500×, B) 2500×, C) 20 000×, D) 5000× [the red rectangle in A/B highlights the zoom-in region shown in (C)]; E) evaluated pores (highlighted in red) from image (D) after processing with the Fiji ParticleSizer Plug-in, F) corresponding pore size distribution as result from the stack analysis of 13 images [similar to (E)] with 1458 pores.
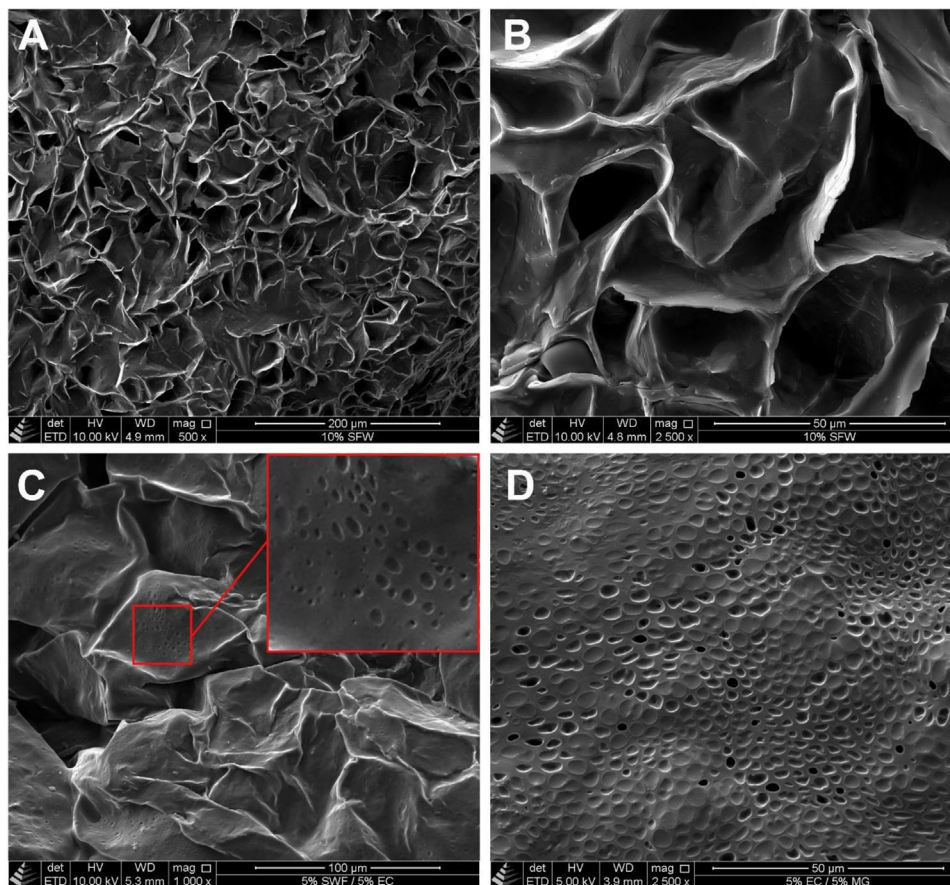
the mixture could be considered as a sol, in which particles of EC swollen with solvent thickened the mixture to some extent but are not able to form a gel (Figure 1). With higher concentration of EC and MG hardness increased and the oleogel got more and more of a gel-like character. In this mixture amphipathic molecules like MG act as plasticizer by interaction with gel junctions formed by weak van der Waal's interactions or hydrogen bonding between polymer strands.[45]

2.3. Structure of Oleogels

Cryogenic scanning electron microscopy (Cryo-SEM) imaging allowed to get a deep insight into the 3D structure of oleogels con-taining rapeseed oil stabilized by EC, SFW, MG, or mixtures of them. Cryo-SEM images of de-oiled oleogel structures have already been shown by other authors[44,46,47] but only viewed from above the sample surface and not in cross section.

The low-magnification image of an oleogel with 10% EC (**Figure 2**A) shows exemplarily that de-oiling of oleogels is only partial and some regions still exist with a smooth surface, where an oil film covers the pores completely. Nevertheless, there are many distinct regions showing the typical oleogel microstructure: The exposed polymer network of the 10% EC oleogel which is described as "coral-like" by Zetzl et al.,[47] exhibits a huge number of small regularly shaped round pores that are relatively evenly distributed all over the oleogel surface and would have been

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Figure 3. Cryo-SEM images of de-oiled oleogels based on rapeseed/canola oil and 10% sunflower wax (SFW) at two magnification levels: A) 500×, B) 2500×, on 5% SFW and 5% ethyl cellulose (EC): C) 1000×, and on 5% EC and 5% monoacylglycerides (MG): D) 2500×.

filled with rapeseed oil without de-oiling (Figure 2D). As also exemplarily shown in Figure 2A, occasional cracks occurred in the sample surface in some sample preparations during the plunge freezing step in nitrogen slush in the course of cryo-SEM analysis. This cross-section view allows an even deeper insight into the 3D structure of the gel network in the range of several micrometers in depth in which even the entrapped oil droplets in the discreetly compartmentalized pores can be visualized (Figure 2B,C). Thus, cross-sectional views clearly reveal that the exposed pore structure, which is visible on the partially de-oiled sample surface continues inside the oleogel.

Images with higher magnification (Figure 2D) also show, that some pores have holes or are fused together on the oleogel surface, which demonstrates that these pores seem to be partially interconnected via thin channels in the stabilizing polymer network as also suggested by Gravelle et al.[48] The scaffold-like polymer-containing walls between the (originally/normally oil-filled) pores have a thickness of ≈0.2–7 µm (average thickness: 1.5 µm) explaining the high stability of the oleogel containing EC.

A semiautomatic approach for the image evaluation was applied for determining the pore size distribution. The first thresholding step for distinguishing the pores from the background was carried out individually for each of the chosen images with

the standard thresholding tool in *ImageJ*. After that, the complete image stack was analyzed automatically with the Particle-Sizer plug-in. In contrast to manual measurements of a few hundred pores,[47] with this procedure more than thousand pores can be measured within minutes, descriptive statistical data and the number based pore size distribution included. Figure 2E,F exemplarily shows the analyzed pores (outlined in red) and the corresponding histogram of the pore size distribution (stack of 13 binary images similar to Figure 2F). Even coalesced pores can be recognized and evaluated individually by applying the integrated ellipse fitting mode. In the case of the analyzed 10% EC oleogel the number-based pore size distribution shows a size range from 0.4 to 4.3 µm. The median minimal Feret diameter (F_{min}) of the pores is 1.67 µm, the maximal Feret diameter (F_{max}) is 2.18 µm, respectively, which results in an aspect ratio (F_{max}/F_{min}) of 1.3, meaning that the round pores exposed on the oleogel surface are slightly elongated. This is in good agreement with punctual size measurements of pores inside the gel in the cross-section image from Figure 2B.

In comparison to the EC oleogel, the 10% SFW oleogel reveals a completely different structure (**Figure 3**A,B). The SFW oleogel presents a very irregular branched 3D network consisting of large thin-walled (curved) platelets with a thickness of less than 1 µm (≈0.2–0.8 µm, average thickness: 0.23 µm). The very porous net-

Table 3. Limit of linear viscoelastic region of conventional solid fats and oleogels.

Sample	Limit of LVR of conventional solid fats [%]				Limit of LVR of oleogels [%]					
Temp	PNF	Margarine A	Margarine B	Margarine C	SFW5	SFW10	SFW5 EC5	EC2.5 MG2.5	EC5 MG5	EC7.5 MG2.5
20 °C	0.009 (0.001)[d]	0.014 (0.001)[d]	0.020 (0.001)[d]	0.020 (0.001)[d]	0.009 (0.001)[d]	0.008 (0.001)[d]	0.028 (0.002)[d]	0.084 (0.006)[c]	0.535 (0.029)[a]	0.182 (0.011)[b]

Calculations of significant differences were performed by JMP via the application of a Tukey-Kramer test of all samples ($p < 0.05$). Significant differences were indicated by different letters (a-d).
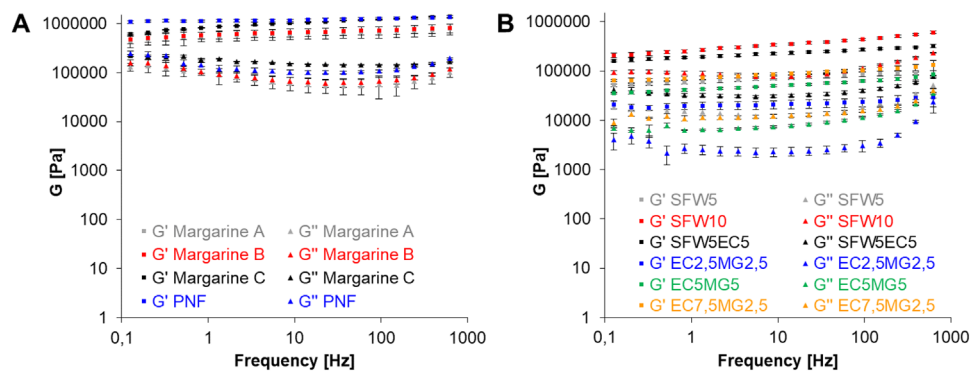
Figure 4. Rheological data from frequency sweeps of A) conventional solid fats and B) oleogels plotted as elastic and viscous moduli (G' and G'') versus frequency. Measurements were carried out at 20 °C. PNF: partially hardened peanut fat; SFW: sunflower wax; EC: ethyl cellulose; MG: monoacylglycerides.

work has large cavities with diameters in the range of 6–65 µm, which, without the de-oiling procedure, are normally filled with "free" oil. The platelet/sheet like structure of SFW-based oleogels was already shown by other authors.[49,50] When 5% SFW and 5% EC were used for the oleogelation, the predominant structure of the oleogel was very similar to the SWF oleogel, since the platelet like structure is still visible (Figure 3C). Only higher magnifications reveal that the coral-like structure with small round pores resulting from EC is present in the oleogel as well. This underlines that both structurants are still able to build their own 3D network if they are mixed together. In case of the oleogel containing 5% EC and 5% MG, the cryo-SEM images reveal a similar pore structure like oleogels based on EC only (Figure 3D). However, it was observed that significantly more pores were still filled with oil, so it appears that the de-oiling process gets hampered by the presence of monoglycerides. This observation can probably be explained by the fact, that EC limits the molecular mobility of MG in the oleogels since the amphipathic MG molecules act as plasticizers via the formation of hydrogen bondings with the unsubstituted hydroxyl groups of the EC polymers.[51,52] The obvious synergistic effects between MG and EC result in an incomplete de-oiling process which underlines their higher viscoelastic properties and a better oil-binding capacity compared to oleogels based on one oleogelator only.

2.4. Rheological Behavior of the Oleogels

In rheological investigations, amplitude tests are usually used in a first step to determine within which deformation range the substance structure of a sample remains intact by measuring stor-

age (G') and loss modulus (G'') as the deformation increases. For this, the limit of the linear-viscoelastic range was determined by means of amplitude tests within a range for the deformation of 0.001%–100% at a frequency of 1 Hz. The examined margarines as well as the 6 different oleogels showed a comparable behavior in the amplitude sweep, whereby the storage modulus was larger than the loss modulus (data not shown). That means for all investigated samples the elastic behavior dominated over the viscous behavior so that a gel character could be recognized for all samples.

Table 3 shows the limits of linear viscoelastic regions (LVR) of margarines and oleogels. For margarines, the limits of LVR varied between 0.007% and 0.029% deformation. For the oleogels, a comparable LVR could only be observed at concentrations of 5% and 10% SFW and 5% SFW + 5% EC. All oleogels with a combination of MG and EC showed a significantly higher limit of the LVR, which indicates that MG and EC containing oleogels display a higher stability and a lower spreadability compared to margarines. The synergistic effects of oleogels based on the combination of MG and EC were also observed within other studies.[52] **Figure 4** shows the rheological data from frequency sweeps of conventional solid bakery margarines, partially hardened peanut fat (Figure 4A) and different oleogels (Figure 4B). The subsequent frequency tests were performed with a constant deformation of 0.005% in the range of 0.02–100 Hz to compare the time-dependent deformation of margarines and oleogels. The linear curves with $G' > G''$ observed for oleogels and the margarines indicated good tolerance as a function of deformation, suggesting a good stability during long-term storage. In the direct comparison of the oleogels with the margarines it is
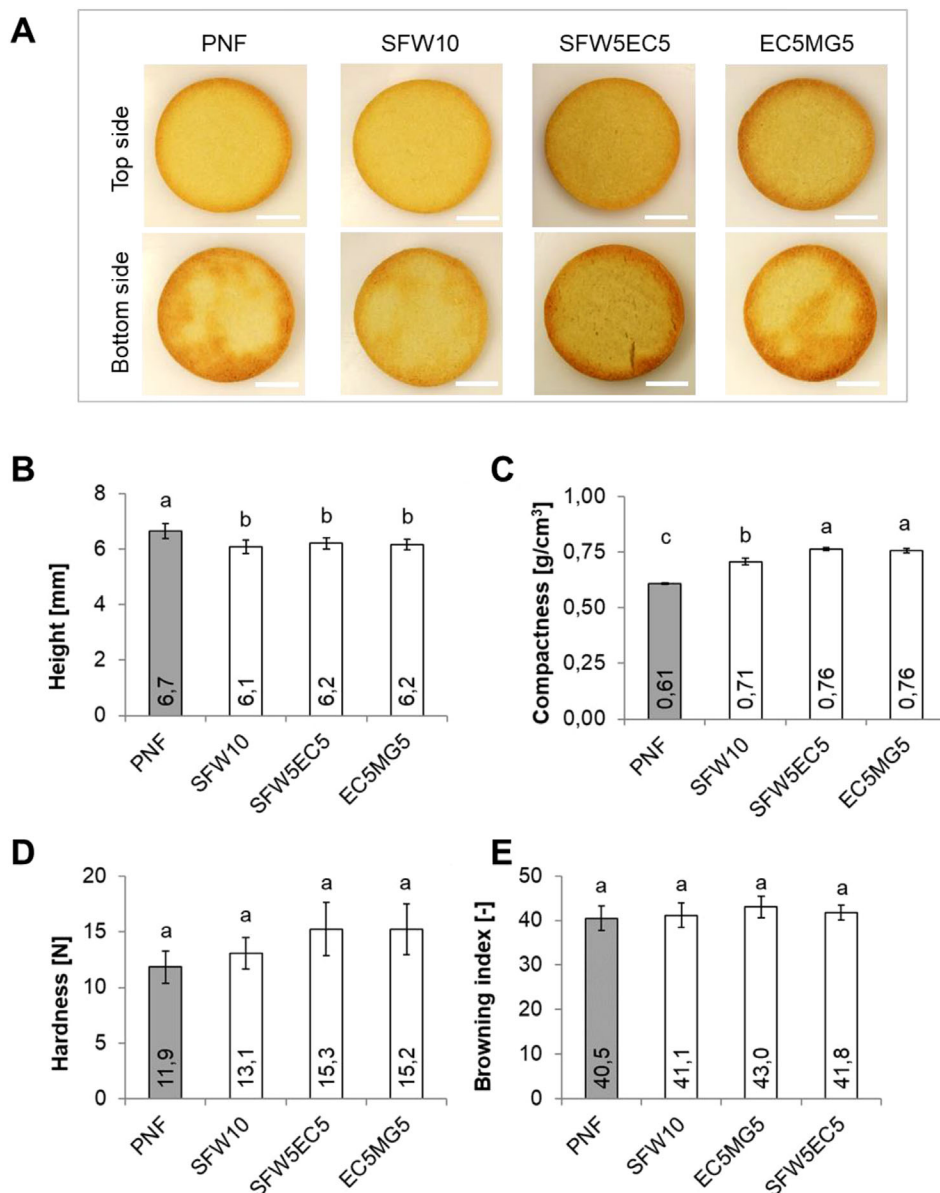
Figure 5. A) Photographs of baked cookies based on partially hardened peanut fat and oleogels, scale bars represent 2 cm (A); and B) height, C) compactness, D) hardness, and E) browning index of baked cookies. PNF: partially hardened peanut fat; SFW: sunflower wax; EC: ethyl cellulose; MG: monoacylglycerides. Different letters in the same line indicate statistically significant differences at $P < 0.05$ in a Tukey post-hoc test.

noticeable that the storage and loss moduli of the oleogels were significantly lower, especially for the oleogels without SFW. However, the oleogels with 10% SFW and 5% SFW + 5% EC showed a behavior that was well comparable with margarines and therefore they displayed a good temporal deformation behavior.

2.5. Preparation of Oleogel-Based Cookies

The three oleogels SFW10, SFW5EC5, and EC5MG5 were selected for further analyses and chosen for the preparation of cookies, because of their comparable characteristics to margarine and partly also to partially hardened peanut fat. In addition,

these three oleogels were identified as "most promising oleogels" based on the experience of the executing baker with regard to the criteria plasticity, malleability, suppleness, and dough forming characteristics (data not shown), wherefore the other oleogels were excluded within the following analyses. The cookies prepared with partially hardened peanut fat showed a better visual impression while the dough and the cookies prepared with SWF10, EC5MG5, and SFW5EC5 were a little brittle and fissured (**Figure 5**A). However, the mentioned oleogels were also suitable for the preparation of cookies.

Moreover, cookies processed with different kind of oleogels showed a significantly less height than the ones based on conventional solid fat phases even if the difference to the cookies
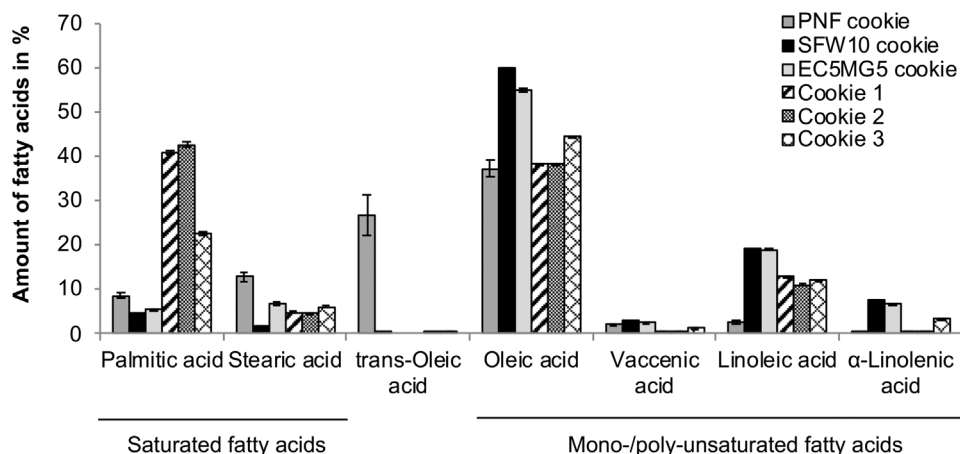
Figure 6. Fatty acid composition of conventional and oleogel-based cookies. PNF: partially hardened peanut fat; SFW: sunflower wax; EC: ethyl cellulose; MG: monoacylglycerides; Cookie 1–3: conventional cookies from the supermarket.

prepared with hydrogenated peanut fat was only about 10% or 0.5 mm (Figure 5B). The investigation also revealed significant differences in the compactness of the different cookies with more compact products prepared with oleogels. The cookies prepared with parts of EC (Figure 5C) turned out to be the most compact cookies, while the application of SFW10 resulted in products with middle compactness. Although the hardness of hydrogenated peanut fat (5.27 N at 20 °C) was much higher than the hardness of the different oleogels (1.59 N (SFW10), 0.15 N (SFW5EC5), and 1.02 (EC5MG5), respectively) the resulting cookies showed different results (Figure 5D). The lowest hardness was found for cookies prepared from hydrogenated peanut fat and the highest for cookies from oleogels with parts of EC. However, these results were not significant due to the great variation of the results. No significant differences were found in regard to the browning index of the different cookies showing that the replacement of the fatty phase by oleogels did not enhance or retard the browning reactions during baking (Figure 5E).

2.6. Fatty Acid Composition

One objective of the present work was the application of rapeseed oil based oleogels for the preparation of bakery products to improve their fatty acid composition in relation to more healthy unsaturated fatty acids and less saturated ones. **Figure 6** shows the fatty acid composition of different shortbread cookies produced industrially (cookie 1–3) and cookies produced with partially hydrogenated fat in comparison to cookies produced with rapeseed oil based oleogels (exemplarily for SFW10 and EC5MG5).

It can be seen that the use of rapeseed oil based oleogels significantly reduced the proportion of saturated fatty acids (29%–47% vs 7%–13%). On the other hand, the proportion of monounsaturated and polyunsaturated fatty acids (50%–62% vs 84%–90%) increased by at least 22%. These results address the advice of the FAO/WHO[53] and the European Society of Cardiology[54] who recommend a maximum of 10% of the total energy intake by saturated fatty acids in the human diet and even the recommendations of the American Heart Association, who gives an upper limit of 7%.[55] In addition, several studies demonstrated that re-

placing saturated and *trans* fatty acids with polyunsaturated fatty acids instead of reducing saturated fatty acids further reduces the risk of coronary heart diseases.[56–59] Therefore, the application of rapeseed oil based oleogels leads to a significant improvement of the fatty acid profile in cookies.

2.7. Sensory Evaluation (Ranking Test and Descriptive Test with Quality Evaluation) of the Cookies

One of the most important characteristic features of food is the sensory quality, which determines the buying decision of consumers.[60] Sensory tests are used when raw materials are exchanged during product development, to check if the modified products are comparable to conventional products.[61]

To investigate possible differences between cookies based on partially hardened peanut fat and oleogels as well as to figure out if there are differences between wax oleogel cookies and EC/MG oleogel cookies, sensory analysis was only performed with three different cookie samples: PNF, SFW10, and EC5MG5. Moreover, EC5SFW5 cookies were excluded from sensory analysis because consumer tests are very labor intensive and time consuming and a fourth sample would have complicated the evaluation for the testers.

The ranking test revealed significant differences between the three cookie samples prepared with different fat phases, as the critical Friedmann value (7.81 for $\alpha = 0.05$) was lower than the theoretical Friedmann test value (9.89, according to DIN ISO 8587:2010-08). Cookies with conventional fat were most popular, closely followed by cookies prepared with oleogel EC5MG5. Cookies prepared with oleogel SFW10 grouped at the lowest level of acceptance. **Figure 7** also shows that oleogel-based cookies were rated worse for all defined product attributes compared to cookies made with conventional fat. Cookies made with oleogel SFW10 differed slightly in smell and appearance from those with PNF. The use of oleogel EC5MG5 also shows a worse result in terms of appearance and smell but the differences for the other descriptors were less pronounced. It should be mentioned that partially hardened peanut fat does not contain any peanut flavor or aroma anymore since the refining process of the fat removes
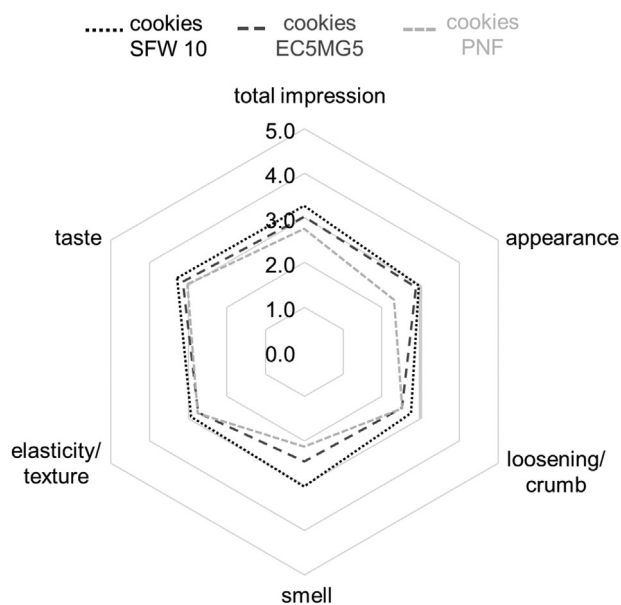
Figure 7. Sensory assessment of the product properties and overall impression of cookies prepared with two different oleogels in comparison to conventional prepared cookies. PNF: partially hardened peanut fat; SFW: sunflower wax; EC: ethyl cellulose; MG: monoacylglycerides.

these compounds.[9] Therefore, the differences between oleogel and peanut fat cookies regarding smell and taste result from the oleogel itself and are not based on a missing peanut aroma. This is also underlined by the comparison of EC5MG5 and SFW10 oleogel-based cookies, as the cookie with EC5MG5 is more similar to the control. It seems that the applied wax leads to a significantly different mouthfeel due to a change in melting behavior and besides that the waxy taste was considered as unpleasant. Probably, a slight reduction of SFW to 7.5% in combination with a small amount of EC 2.5% would be more comparable to the control.

3. Conclusions

The results of the project showed that it is possible to prepare oleogels on the basis of SFW, EC, MG or mixtures of them to replace conventional fat phases for the preparation of cookies. The sensory analysis showed that cookies prepared with oleogels were accepted by the consumer, but they are not comparable with the reference products, yet. Additional modifications need to be made to increase consumer acceptance and to make oleogels marketable.

In addition to the application of oleogels with one oleogelator the application of mixtures seems to be a promising way to prepare tailor-made oleogels. Also, the application of SFW resulted in oleogels with suitable properties for the preparation of cookies but since SFW is not approved the application in food is not yet allowed in Europe.

In the future it is also necessary to investigate and evaluate the application of oleogels based on the mentioned oleogelators for the preparation of other and more sophisticated bakery products like puff pastries or sand cake. An evaluation of the storage stability of the products is essential.

4. Experimental Section

Materials: Refined rapeseed oil used for the preparation of oleogels was kindly provided by Broekelmann + Co Oelmuehle GmbH + Co (Germany) and SFW by Karl GmbH & Co. KG (Germany). The rapeseed oil contained the following fatty acid composition: $C_{16:0}$: 4.5%, $C_{16:1}$: 0.3%, $C_{18:0}$: 1.2%, $C_{18:1\ D9}$: 61.7%, $C_{18:1\ D11}$: 3.0%, $C_{18:2}$: 18.2%, $C_{18:3}$: 7.1%, $C_{20:0}$: 0.5%, $C_{20:1}$: 1.2%, $C_{22:0}$: 0.3%, $C_{22:1}$: 0.3%, $C_{24:0}$: 0.1%, and $C_{24:1}$: 0.1%. MG (DIMODAN HR KOSHER, $C_{16:0}$: 92%, $C_{18:0}$: 5.5%, C_{20}: 1.5%, and $C_{22:0}$: 0.5%) and EC (Ethocel Standard 100 Premium) were donated by DDP Specialty Products Germany GmbH & Co. KG (Germany). Bakery margarines were obtained from Vortella Lebensmittelwerk, W. Vortmeyer GmbH (Germany). Partially hardened peanut fat (26.7 ± 4.5% *trans* oleic acid), wheat flour, baking powder, sugar, and three kinds of industrial shortbread cookies were purchased from local stores.

Preparation of Oleogels: Oleogels were produced by the addition of an oleogelator (EC, MG and/or SFW) in different proportions to rapeseed oil, whereby the total concentration of all oleogelators should not exceed a value of 10%. In the case of MG and SFW the mixture was stirred in glass bottles with a stirring speed of 300 rpm, heated at 80 °C in a polyethylene glycol bath and the temperature was maintained for 5 min. In the case of an oleogel based on EC, the mixture was heated at 180 °C and the temperature was kept for 30 min. Afterward, the mixture was poured into 60–120 mL plastic beakers or 500 mL rectangular aluminum containers and immediately stored at −20 °C for at least 24 h, to allow the oleogelator to form the 3D network. During this study six different oleogels which displayed suitable processability characteristics in pre-trials were prepared and analyzed: 5% SFW (SWF5), 10% SFW (SWF10), 2.5% EC + 2.5% MG (EC2.5MG2.5), 5% EC + 5% MG (EC5MG5), 7.5% EC + 2.5% MG (EC7.5MG2.5), and 5% SFW + 5% EC (SFW5EC5).

Cookie Preparation: For the preparation of cookies, a recipe of a standard baking test for cookies based on partially hardened peanut fat was used. In the case of cookies prepared with oleogels, partially hardened peanut fat was completely substituted by the oleogel. In the first step, 180 g partially hardened peanut fat or oleogel were mixed with 135 g sugar and 67.5 g water for 2 min using a planetary stirrer (model N-50, Hobart GmbH, Offenburg, Germany). In the second step, 450 g wheat flour and 1.35 g baking powder were added, and all ingredients were kneaded for 1 min. The dough was rolled out at a height of 6 mm and 48 cookies with a diameter of 6 cm were cut out. Cookies were baked for 13 min at 200 °C.

Oleogel Structure: Structure analysis was carried out by cryo-SEM. For exposing the internal structure of the oleogels small pieces of each sample ($\approx$1 cm^3, stored at 4 °C for at least 24 h after oleogel production) were cut out of the bulk sample with a scalpel and plunge-frozen in liquid nitrogen. The resulting freeze fracture fragments were de-oiled by immersion in isobutanol (Emsure, Merck, Darmstadt, Germany). Depending on the gel stability the immersion time varied between 30 and 60 s for the EC oleogels and up to 15 min for the SFW oleogels, respectively. After decanting the supernatant (isobutanol and dissolved rapeseed oil), the fragments were placed in a petri dish and dried under airflow, followed by cryo-SEM sample preparation consisting of the following steps: sample fixation on a plane cryo transfer shuttle with conductive mounting medium (1:1 mix of Tissue-Tek O.C.T compound and colloidal graphite, Agar Scientific Ltd., Stansted, UK), plunge freezing in nitrogen slush ($T = −210$ °C), transfer to precooled cryo chamber ($T = −135$ °C, PP2000 T, Quorum Technologies Ltd., Laughton, UK) for sublimation of residual ice contamination (15 min at −90 °C) and sputter-coating with a thin platinum layer in argon atmosphere (30 s coating at $\approx$5–10 mA current). After final transfer to the chamber of the field emission SEM (Quanta 250 FEG, FEI, Brno, Czech Republic), the analyses were carried out under high vacuum conditions at −135 °C with an accelerating voltage of 10 kV.

The number-based pore size distributions of the pores of the EC oleogel sample were determined based on the size and shape properties of their exposed cross section on the oleogel surface. For this purpose, several images ($\approx$5–15, magnification: 5000×) were randomly taken of different positions on the oleogel surfaces. Using the image analysis software ImageJ (v1.52p) in combination with the ParticleSizer-Plugin,[62] the pore outlines were measured automatically, whereas the thresholding step was carried

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out manually on each image for eliminating not fully exposed pores. Morphometric parameters of individual pores, which directly describe the external dimensions of each pore in the oleogel surface (e.g., median minimum Feret diameter, F_{min}), were extracted from the data output sheet and further processed using SigmaPlot (v14.0).

Oleogel Texture: Texture and hardness of solid fats and oleogels were analyzed using the Texture Analyser TA-XT2i (Stable Micro Systems Ltd., England) equipped with a stainless-steel cylindrical probe (0.25 in. diameter). The samples were prepared in cylindrical containers with a diameter of 6 cm and a height of 7 cm. To investigate their behavior at different temperatures (fridge temperature, room temperature and high temperature) the samples were pre-tempered at 8, 20, and 30 °C for 48 h before starting the tests. Four individual measurements were performed for each sample using a test speed of 2 mm s^{-1} and the same return speed for the cylindrical probe to go back to the initial position. The hardness was analyzed via measuring the maximum force, which is needed to penetrate the sample to a depth of 10 mm. Hardness was given as N.

Oil-Binding Capacity: The ability of a solid fat or oleogel to retain the oil was analyzed via centrifugation at 8, 20, and 30 °C. Each sample was analyzed with three replicates. In brief, 10 g of the sample were pre-tempered for 48 h in 15 mL centrifugation tubes at the corresponding temperature, followed by centrifugation at 4.000 g for 30 min using a temperature adjustable Sigma 3-18K centrifuge (Sigma Laborzentrifugen GmbH, Osterode am Harz, Germany). To quantify the oil leakage after centrifugation, liquid oil was removed from the centrifugation tubes and the weight of the remaining solid fat was determined. The OBC, which represents the mass of the remaining sample as percentage of the total sample mass, was calculated using following equation

$$OBC\ (\%) = \left(\frac{mass\ of\ remaining\ sample}{total\ mass\ of\ sample} \right) x\ 100 \qquad (1)$$

Rheological Behavior: The rheometer MCR 302 (Anton Paar, Austria), equipped with a Peltier system, a water bath (Julabo, Germany), and a plate/plate geometry (PP25) with a diameter of 25 mm, was used for the analysis of the viscoelastic behavior of solid fats and oleogels. The measuring gap was set at 1 mm. For rheological analyses the samples (stored at −20 °C) were cut out of the bulk sample using a scalpel and carefully transferred to the rheometer. For amplitude sweeps the samples were kept at 20 °C for 10 min to equilibrate and the storage (G') and the loss (G'') moduli were acquired as a function of strain (0.001%–100%) at a frequency of 1 Hz, to determine the limit of the linear viscoelastic region (LVR). A frequency of 1 Hz was chosen since this frequency should recapitulate the breakdown process of oleogel kneading during cookie-dough preparation. Frequency sweeps (0.02–100 Hz) were performed at 20 °C within the LVR at a strain of 0.005% to analyze the time-dependent deformation behavior of solid fats or oleogels. A number of three individual replicates was used for each experimental run.

Analysis of Cookie Properties: In total, three independent baking trials per fat phase were performed and 12 cookies per baking trial were analyzed regarding their height, weight, volume, compactness, texture, color, and sensory quality.

Height, Weight, Area, and Compactness: In brief, the height of the cookies was measured with a caliper, the weight was monitored by a precision balance and the area of the cookies was calculated automatically using the open source image processing and analysis program ImageJ. Then the volume of the cookies was calculated by the height and the area. The compactness of the cookies was calculated by dividing the weight of a cookie in gram by its corresponding volume in cubic centimeters.

Texture: The texture of the cookies was analyzed by using the Texture Analyser TA-XT2i (Stable Micro Systems Ltd., England) with a three-point bending setup and an elongated blade probe with which the cookies had been broken by using a test speed of 2 mm s^{-1} and the same return speed for the cylindrical probe to go back to the initial position. The measuring device approached the sample at a speed of 1 mm s^{-1}. The test speed was set to 3 mm s^{-1} and the back speed to 10 mm s^{-1}. After touching the surface of the cookie, the blade moved down 10 mm, breaking the

cookie in the process. The resulting maximum force corresponded to the hardness and brittleness of the cookies.

Color: The determination of the color of the biscuit surface was carried out with a Chroma-Meter (CR-310, Minolta Co., Ltd., known today as Konica Minolta, Japan). The parameters L (lightness/darkness), a (redness/greenness), and b (yellowness/blueness) were measured at ten different points on the surface of 12 different cookies for each baking trial of the different fat phases. Therefore, each result comprises 360 color measurements. The browning index[63] was calculated using the $L*$, $a*$, $b*$ values with the following formula

$$Browning\ index = \frac{100*(x - 0.31)}{0.17} \quad with\ x\ \frac{(a + 1.75*L)}{(5.64*L + a - 3.012*b)}$$

$$(2)$$

Cold Fat Extraction: In order to characterize the fat phase 20.0 g of cookies (crushed with a mortar) were weight into an Erlenmeyer flask. 100 mL petroleum ether were added, and the content of the Erlenmeyer flask homogenized for 15 s with an Ultra-Turrax T25 digital (IKA Werke GmbH & Co. KG, Germany) at 10.000 rpm. The flask was closed with a glass stopper and the mixture was shaken for 16 h on the horizontal shaker Certomat SII (B. Braun Biotech International GmbH, Germany) at 200 rpm. Afterward, the mixture was qualitatively filtered over a prefolded cellulose filter paper MN 875 ¼, 240 mm diameter (MachereyNagel GmbH & Co. KG, Germany) into a round bottom flask. Finally, the solvent was evaporated by a rotary evaporator R134 (Buechi Labortechnik GmbH, Germany) at 40 °C and 90 mbar, and last residues of petroleum ether were removed for 10 min with a gentle stream of nitrogen.

Fatty Acid Composition: The analysis of the fatty acid composition of the fat extracted from cookies by cold extraction was performed on the base of methods DGF-C-VI 10 (13) and C-VI 11 d (98),[64] with some modifications. In brief, one drop of extracted fat was put into a screw-top glass test tube, 1 mL of heptane (Merck KGaA, Germany) and 50 μL sodium methylate solution (30% solution in methanol for synthesis by Merck KGaA, Germany mixed with methanol by J.T. Baker, USA) were added and the glass treated with a vortex shaker (IKA Werke GmbH & Co. KG, Germany) for 1 min. Then 100 μL Millipore water were added to the solution and after treatment with the vortex shaker the solution was centrifuged at 3000 rpm for 5 min. The lower aqueous phase was removed, 50 μL hydrochloric acid with a concentration of 1 mol L^{-1} and methyl orange as indicator added and the solution mixed by shaking. The lower aqueous phase was discarded, two spatula tips of sodium sulfate were added. Afterward, the glass test tube was centrifuged at 3000 rpm for 5 min. The organic phase (1 μL) was injected in a HP5890 gas chromatograph (Agilent Technologies Sales & Services GmbH & Co. KG, Germany), with a capillary column, CP-Sil 88 (100 m long, 0.25 mm ID, film thickness 0.2 μm). The temperature program was as follows: From 155 °C; heated to 220 °C (1.5 °C min^{-1}), 10 min isotherm; injector 250 °C, detector 250 °C; carrier gas 36 cm s^{-1} hydrogen; split ratio 1:50; detector gas 30 mL min^{-1} hydrogen; 300 mL min^{-1} air and 30 mL min^{-1} nitrogen. The peak areas were computed by the integration software and percentages of fatty acid methyl esters (FAME) were obtained as weight percent by direct internal normalization.

Sensory Analysis: Hedonic sensory studies are often used to determine consumer preference of products[65] and to evaluate whether changes in the composition of food or the substitution of ingredients lead to changes in consumer preference. In hedonic tests, untrained consumers are used as testers. In the present sensory test, the number of testers was $n = 56$. Regarding the test procedure the testers only got the information which type of bakery product was being examined. The exact composition and ingredients were not specified in order to avoid possible influences, except for SFW, as this is not an approved food additive so far. Due to the consumption of SFW-containing cookies, testers had to provide informed consent. The samples were handed out in neutral packaging and the sample quantity and temperature was always the same.[66]

For sensory tests, cookies were produced with peanut fat as conventional fat phase as well as with oleogel EC5MG5 and with the oleogel

SFW10, respectively. A ranking test according to DIN ISO 8587:2010-08 was carried out to determine consumer preference. The test persons were instructed to sort the samples according to their overall impression in a descending order, from "very popular" to "not popular." The evaluation was carried out in accordance with method DIN ISO 8587:2010-08 using the Friedmann test.[67] The test persons were untrained examiners, which is why an error probability of $\alpha = 5\%$ was assumed in the evaluation.[68]

Finally, the product properties and the overall impression should be assessed. The rating scale was created by the German school grading system and ranged from 1 to 5, with 1 for "very good" and 5 for "very bad." Based on the identified descriptive terms, a comparison between bakery products with different types of oleogels and with conventional fat can be drawn. In this way the sensory strengths and weaknesses of oleogels for the production of cookies were highlighted.

Statistical Analysis: Mean and standard deviation (SD) were calculated from the results of the experiments described above. Calculations of significant differences were performed by JMP (SAS Institute GmbH, Cary, USA) via the application of a Tukey–Kramer test of all samples ($p < 0.05$). Significant differences were indicated by different letters.

Ethical Guidelines

Ethics approval was not required for this research.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

M.S.: conceptualization (equal), funding acquisition (supporting), project administration (equal), investigation (equal), validation (equal), writing – review, editing (equal); N.E.: investigation (equal); S.W.: sensory investigation (equal), writing – review, editing (equal); S.N.: project administration (equal), investigation (equal), validation (equal), writing – editing, review (equal); B.H.: investigation (equal), validation (equal), writing – review, editing (equal); B.M.: funding acquisition (equal), project administration (equal), supervision (equal), validation (equal), writing – original, review, editing (equal).

Data Availability Statement

Research data are not shared.

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