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Structural and sensory studies on chocolate spreads with hydrocolloid-based oleogels as a fat alternative

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ABSTRACT

In this study, chocolate spreads were designed using oleogels with two oils (olive and sunflower), and hydroxypropylmethylcellulose (HPMC) and xanthan gum (XG), as structuring agents. Thus, the lipid profile of the spreads can be improved by totally or partially replacing the coconut butter used in their preparation. Structural behaviour was analysed using confocal laser scanning microscopy (CLSM), small amplitude oscillatory rheology, and a spreadability test. A Free Choice Profile analysis was performed by consumers to determine the sensorial attributes that described the chocolate spreads. The results showed that the oleogels conferred consistency to the spreads because of the network formed by HPMC and XG. However, while coconut butter replacement at 50% gave a similar structure to the control spread, 100% replacement resulted in less homogeneous spreads. This trend might be attributed to the chemical compatibility between the oleogel-coconut butter, which led to stronger systems. Sensory evaluation showed that chocolate spread replaced by sunflower oleogel at 50% presented sensory attributes like the control spread with “creamy appearance”, “creamy texture”, and “cocoa flavour”. Therefore, using oleogels can be a viable and healthy alternative to replace the saturated fat present in chocolate spreads.

1. Introduction

Consumption of certain foods have a profound impact on health. To prevent a multitude of pathologies, such as obesity, cardiovascular disease, and cancer, it is necessary to follow healthy dietary guidelines. Currently, many foods contain solid fats, which have a high content of saturated and/or trans fatty acids; these fats are widely used in the food industry for their palatability, functionality, and texture (Co & Marangoni, 2012; Liu, Xu, & Guo, 2007; Pehlivanoglu et al., 2018). However, because of its influence on the sensory and functional properties of food (Pehlivanoglu et al., 2018), achieving the optimal substitution of solid fat is still a challenge (Lim, Inglett, & Lee, 2010). Previously, studies have partially or totally replaced solid fats in different foods with sources of lipids (Jacob & Leelavathi, 2007), carbohydrates (Gibis, Schuh, & Weiss, 2015; Rodríguez-García, Laguna, Puig, Salvador, & Hernando, 2013; Zahn, Pepke, & Rohm, 2010), and proteins (Paglarini, Martini, & Pollonio, 2019; Youssef & Barbut, 2009).

Recently, the replacement of saturated and/or trans fats in food

products has been studied making oleogels with high nutritional quality oils (Fayaz et al., 2017a; Pehlivanoglu et al., 2018). Edible oleogels are defined as a solid-like material with oil immobilised in a three-dimensional network with gelling capacity (Huang, Hallinan, & Maleky, 2018). Oleogelification is an emerging technology that allows semi-solid properties to be conferred to oils, without modifying their chemical characteristics (Lim, Jeong, Oh, & Lee, 2017; Luo et al., 2019; Patel et al., 2014a). For this, gelling agents are required to obtain an oleogel with textural properties like solid fat (Co & Marangoni, 2012; O'Sullivan, Barbut, & Marangoni, 2016). Hydroxypropylmethylcellulose (HPMC) is a biopolymer used as a stabilising additive in the food industry, because of its commercial availability and low cost (Abdolmaleki, Alizadeh, Nayebyzadeh, Hosseini, & Shahin, 2019). Several studies confirm HPMC as an oleogelator (Bascuas, Hernando, Moraga, & Quiles, 2020; Meng, Qi, Guo, Wang, & Liu, 2018a; 2018b; Oh & Lee, 2018; Patel & Dewettinck, 2015; Tanti, Barbut, & Marangoni, 2016a; 2016b) because it provides viscosity-enhancing properties (Zhao et al., 2009), giving oleogels with viscoelastic

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properties and high oil retention (Meng, Qi, Guo, Wang, & Liu, 2018c; Patel & Dewettinck, 2015). Besides, the addition of thickeners, such as xanthan gum (XG), increases the viscosity of the aqueous continuous phase of the emulsions, favouring the stability of the HPMC oleogels (Meng et al., 2018a; Patel et al., 2014a).

One industry that could benefit from the substitution of fat for oleogels is the spreads industry, where products are used in pastries and confectionery, and are mostly made of fat and sugar (Demirkesen & Mert, 2019; Manzocco, Calligaris, Camerin, Pizzale, & Nicoli, 2014). The fat content of these spreads can reach up to 60% and largely determines the sensory, rheological, and textural properties, because of the network of fatty crystals (Espert, Salvador, Sanz, & Hernández, 2020; Miele, Di Monaco, Masi, & Cavella, 2015). Consequently, the substitution of solid fat for liquid oil can greatly affect the chocolate spread's performance and, therefore, the quality of the product (Fayaz et al., 2017b). Our group has recently characterised the microstructure, rheological behaviour, and oxidative stability of oleogels prepared using HPMC and XG, indicating that sunflower or olive oleogels could have potential food applications (Bascuas et al., 2020). To date, very few studies exist regarding using oleogels in spreads. Fayaz et al. (2020b) and Patel, Rajarethinam, et al. (2014) studied the incorporation of wax- and shellac-based oleogels, respectively, in chocolate spreads but yet, no studies investigate using biopolymer-based oleogels in chocolate spreads.

The objective of this study is to evaluate the feasibility of using olive and sunflower oil oleogels, made with biopolymer oleogelators, to replace saturated fat in chocolate spreads by studying their structural properties (microstructure, rheology, and texture) and sensory attributes.

2. Materials and methods

2.1. Ingredients

Hydroxypropylmethylcellulose (HPMC; 4000 cP) was provided by Dow Chemical Company (Midland, MI, United States) and xanthan gum (XG; Satiatine CX 931) by Cargill R & D (Vilvoorde, Belgium). Water (Bezoya, Segovia, Spain, with a calcium content 6.32 mg/l), olive oil (O) (Hacendado, Mercadona, Spain), refined sunflower oil (S) (Consum, Spain), sugar (Disem, Spain), skimmed powder milk (1 g/100 g fat) (Central Lechera Asturiana, Spain), and cocoa powder (Chocolates Valor S.A., Alicante, Spain) were purchased in supermarkets. Coconut butter was supplied by Gracomsa (Catarroja, Valencia, Spain).

2.2. Oleogels preparation

The oleogels were prepared as described by Bascuas et al. (2020), and the emulsions were vacuum dried (Vaciotem-T, J.P. SELECTA, Spain) at 60 °C and -0.85 bar for 14 h. The dried products were ground in a grinder (Moulinex A320R1, Paris, France) for 5 s to produce the oleogels. Two oleogels were produced using olive oil; olive oleogels and sunflower oil; sunflower oleogels.

2.3. Spreads preparation

Chocolate spreads were prepared using the proportions shown in Table 1. One control spread with coconut fat (C) and four spreads with partial or total fat coconut replacement, by the oleogel, were made. Thus, a spread with 50% replacement of coconut fat by olive oil (CO), a spread with 50% replacement of coconut fat by sunflower oil (CS), and two 100% replacements of coconut fat by olive oil (O) or sunflower (S) oleogel were obtained. The rest of the ingredients used were common in all the formulations.

A food processor (TM31 Thermomix, Vorwerk, Wuppertal, Germany) was used to mix the ingredients. First, sugar, skimmed powder milk, cocoa powder, and mineral water were mixed at 70 °C for 6 min at

Table 1
Composition of the studied chocolate spreads.

Ingredients	g/100 g				
	C	CO	CS	O	S
Sugar	32.5	32.5	32.5	32.5	32.5
Oleogel	–	15	15	30	30
Coconut butter	30	15	15	–	–
Water	20	20	20	20	20
Skimmed powder milk	12.5	12.5	12.5	12.5	12.5
Cocoa powder	5	5	5	5	5

Control spread made with coconut fat (C); spread made with 50% coconut fat and 50% olive oleogel (CO); spread made with 50% coconut fat and 50% sunflower oleogel (CS); spread made with olive oleogel (O); spread made with sunflower oleogel (S).

speed 2 (200 rpm). After cooling at room temperature, fat (coconut butter and/or oleogel) was added and mixed in the processor for 3 min at speed 2. To achieve a suitable spread texture, the speed of the Thermomix was increased to 4 (400 rpm) and mixed for 2 min, and to 5 (500 rpm) for 1 min. After the spread was made, it was refrigerated at 5 °C. All analyses were performed 24 h after the spreads were made.

2.4. Structural properties

2.4.1. Microstructure analysis

Confocal scanning laser microscopy (CLSM) was conducted using a ZEISS 780 microscope coupled to an Axio Observer Z1 inverted microscope (Carl Zeiss, Germany). To visualise the samples, the C-Apochromat 40X/1.2 W water immersion objective was used. The images were obtained and stored with a resolution of 1024 × 1024 pixels using the microscope software (ZEN). The stains used were Nile Red, Fluorescein (FITC), and Calcofluor White (Fluka, Sigma-Aldrich, Missouri, USA). The Nile Red was used to detect fat, it was excited with the 561 laser line and was detected between 576 and 620 nm, the FITC stained protein and was excited with the 488 laser line and was detected between 499 and 525 nm, Calcofluor White stained polysaccharides and was excited with the diode line 405 and detected between 410 and 477 nm.

To observe and study the spread, a small amount of sample was placed on a slide, 20 µL of Nile Red solution was added and it was left to rest for 10 min. The same procedure was performed with FITC and Calcofluor White, and samples were covered with a glass coverslip.

2.4.2. Rheological measurements

To perform the rheology measurements, an AR-G2 controlled stress rheometer (TA Instruments®, New Castle, USA), coupled to a computer system (TA Instruments Universal Analysis 2000 Software) was used. A serrated plate-plate (40 mm) was used in all the experiments, with a gap of 1 mm. The tests were performed at 20 °C in duplicate. Small amplitude oscillation sweeps (SAOS) were performed to analyse the viscoelastic properties. To determine the extent of the linear viscoelastic region (LVR) stress sweeps (from 0.1 to 200 Pa with a logarithmic distribution, 10 points per decade) were conducted at 1 Hz. Frequency sweeps were performed between 0.01 and 10 Hz within the linear region (at 5 Pa) with values of storage modulus (G'), loss modulus (G''), and $\tan \delta$ recorded.

2.4.3. Texture measurements

A TA-XT plus Texture Analyzer equipped with the Texture Exponent software (Stable Microsystems, Godalming, UK) was used to determine the texture properties of the samples.

Spreadability of samples was measured using a TTC Spreadability Rig (HDP/SR) attachment. Samples were filled into a female cone (90° angle), with special attention made to avoid bubbles formation, and were penetrated 22.5 mm using the corresponding male cone (90° angle) at a speed of 1 mm/s. Force expressed in N, as a measurement of firmness, and area under the curve (AUC; N s), as a measurement of

spreadability, were recorded. Experiments were performed in duplicate.

2.5. Sensory analysis

The sensory analysis was conducted in a sensory room equipped with individual booths designed in accordance with ISO 8589:2007 (ISO, 2007), under artificial daylight and controlled temperature (22 °C).

Twenty untrained consumers (60% women, 40% men), with ages between 25 and 50 years old, took part in a Free Choice Profile analysis. In the first session, the terms used by each consumer describing the differences among spreads were generated by a Repertory Grid Method (RGM). Three samples were presented and each consumer described the similarities and differences among samples in their own terms. Consumers evaluated the appearance, taste, aroma and texture of the different spreads. In the second session, each consumer used their own list of terms by rating the intensity for each sample using a 10 cm unstructured line scale with the anchors "Not perceived" and "Intense". The samples were served in white plastic cups labelled with random three-digit codes and served at room temperature in random order following a Williams design. They were asked to evaluate the appearance by observing the sample, the aroma and after eating a spoonful of the sample with a plastic spoon evaluate the taste and texture. Water was provided to clean the palate between samples.

2.6. Statistical analysis

One-way analysis of variance (ANOVA) was applied to study the effects of fat on the rheological and texture parameters studied. The least significant differences (LSD) were calculated using the Fisher's test, and the significance was determined at $p < 0.05$. A Generalized Procrustes Analysis (GPA) was applied to the Free Choice Profile data. XLSTAT statistical software (2010.5.02 (Addinsoft, Barcelona, Spain)) was used.

3. Results and discussion

3.1. Structural properties

3.1.1. Microstructure analysis

Fig. 1 shows the distribution of ingredients in the spreads studied using CLSM together with the pictures of the different spreads. Using staining agents Nile Red, FITC, and Calcofluor White, the components of the spreads observed were fat in red, protein in green, and polysaccharide in blue. In the images stained with Nile Red and FITC (left column), the control spread (C) (Fig. 1a) comprises a continuous phase of green colour, consisting mostly of protein, in which a second continuous phase of fat stained with Nile Red is distributed and diffused homogeneously. There is a part of the spread's fat that forms small globules; however, another part is homogeneously distributed and interacts with the components of the matrix. Distributed in the matrix, grey particles can be seen, possibly corresponding to undissolved sugar (Fig. 1a). In images stained with Nile Red and Calcofluor (Fig. 1f), there is a homogeneous distribution of fat, stained pink, merged with the remaining components making up the continuous phase. These images show also isolated blue particles, which probably correspond to undissolved cellulosic material of cocoa.

When the spreads are made with a mixture of oleogel and coconut fat (CO and CS), their appearance is similar, regardless of the oleogel used (Fig. 1b and g; 1c and h). Furthermore, their structure is more like the control spread than those made only with oleogels (O and S) (Fig. 1d and i; 1e and j). In CO and CS spreads, the fat remains structured by the hydrocolloids used to make the oleogel: HPMC and XG. The presence of these polysaccharides is believed to partially prevent coalescence, because of the formation of a hard and thick interface layer limiting the mobility of the globules inside the matrix (Borreani et al., 2017; Meng et al., 2018a; Patel et al., 2014a). In addition, like the control spread, cocoa particles are distributed throughout the spread, along with

possible HPMC segments, not adsorbed at the droplet surface. As observed previously by Meng et al. (2018c), in soybean oil oleogels structured by cellulose ethers, only few segments of the HPMC chain are adsorbed at the droplet surface because of its rigid backbone, with the hydrophilic segments stretching to the aqueous phase.

In spreads with 100% replaced coconut fat using olive and sunflower oleogels (O and S) (Fig. 1d and e), part of the fat is bound with the protein, constituting a continuous phase which contains sugar and cocoa particles distributed in the spread. However, large fat globules (black arrows in Fig. 1d and e) can also be observed. Therefore, a different fat distribution can be seen if compared to C, CO, and CS spreads, which show a more homogeneous fat distribution. These larger fat globules are observed in pink, trapped in the three-dimensional hydrocolloids network stained in blue by Calcofluor (Fig. 1i and j). This structure has been previously described by Espert et al. (2017) and Meng, Qi, Guo, Wang, and Liu (2018b), in emulsions and in oleogels prepared with polysaccharides, respectively. In Fig. 1i and j, incipient coalescence is observed, especially in the S spread (black arrows). This destabilisation phenomena can determine many of the important properties of food such as appearance, shelf life, flavour profile, texture, and release characteristics (McClements, 2015). O and S spreads also show several undissolved particles stained in grey (red arrows), greater than in CO and CS spreads, likely corresponding to crystallised sugar and non-cellulose cocoa fibre.

It can be speculated that the process of making the spreads had a different effect on their stability. In spreads O and S, the shear force used resulted in a less structured and homogeneous spread with several undissolved particles, while the spreads with a mixture of saturated fat and oleogel (CO and CS) gave a structure like the C spread, with a higher proportion of fused components. Other studies have shown that incorporating a little solid fat in oleogels improves their structure. Patel (2015) observed that incorporating palm stearin in oleogels made with HPMC allowed a full recovery of the structure after the shear force applied, stopped.

3.1.2. Rheological measurements

The mechanical spectra of the chocolate spreads (viscoelastic modules as a function of frequency) at 20 °C are represented in Fig. 2. In all the spreads (Fig. 2a), the storage modulus (G') is greater than the loss modulus (G'') and all samples have a similar frequency dependency. This gel-like behaviour was also found by Espert et al. (2020) and Espert, Wiking, Salvador, and Sanz (2020) in spreads formulated with sunflower oil, and in spreads made with emulsions structured with hydrocolloids, such as methyl cellulose, HPMC, or XG. Furthermore, other filler spreads, formulated with oleogels using monoglycerides or waxes as structuring agents, also exhibited gel-like behaviour (Fayaz et al., 2017b; Patel et al., 2014b). Furthermore, Fig. 2b shows the variation of $\tan \delta$ (G''/G') with frequency. This parameter shows the presence or absence of changes in the internal structure of the sample. As seen in Fig. 2b, spreads elaborated with total fat replacement by oleogels (spreads O and S) show higher $\tan \delta$ values and a greater frequency dependence than the other spreads, thus having a lesser structured system. In contrast, spreads made with partial fat replacement (CS and CO), presented a $\tan \delta$ profile like spread C, with lower values than O and S spreads, corresponding to systems with a more solid internal structure.

Table 2 shows the values of G' , G'' , and $\tan \delta$ at the frequency 1 Hz for each of the chocolate spreads. Spreads CO and CS presented significantly higher G' values than spreads C, O and S. $\tan \delta$ values for spreads O and S were the highest, indicating a weaker internal structure, although, the difference in internal structure is much better appreciated in Fig. 2 along the evolution of frequency.

These results agree with microstructural observations, where the spreads made with partial fat replacement presented a more homogeneous structure and were more similar to the control spread. Thus, a more organised structure may favour improved rheological properties

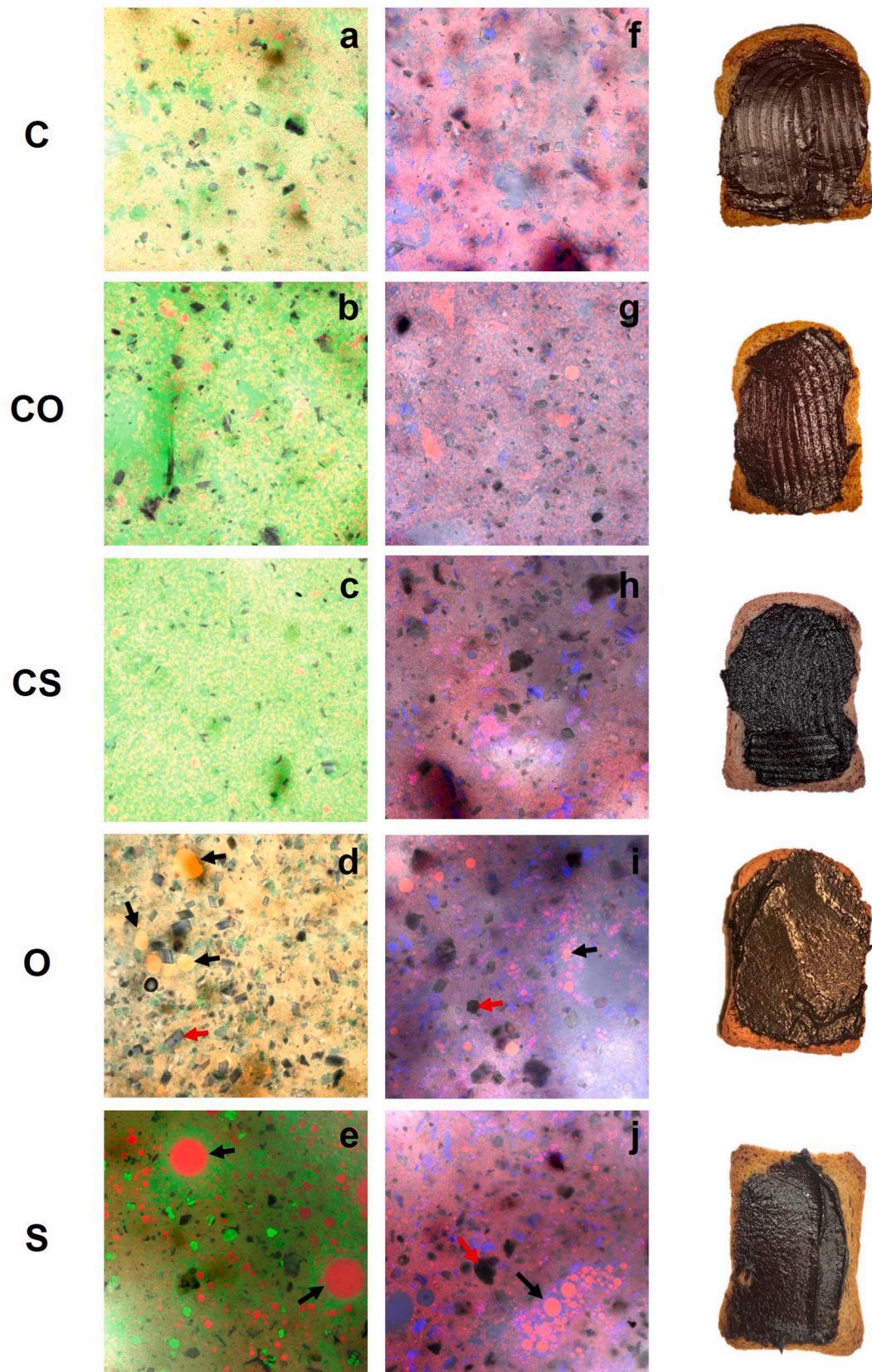


Fig. 1. Images taken with a confocal scanning laser microscope (CLSM) of the different chocolate spreads. Staining with Nile Red and FITC (a–e); staining with Nile Red and Calcofluor (f–j). (a) and (f): Control spread made with coconut fat (C); (b) and (g): spread made with 50% coconut fat and 50% olive oleogel (CO); (c) and (h): spread made with 50% coconut fat and 50% sunflower oleogel (CS); (d) and (i): spread made with olive oleogel (O); (e) and (j): spread made with sunflower oleogel (S). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

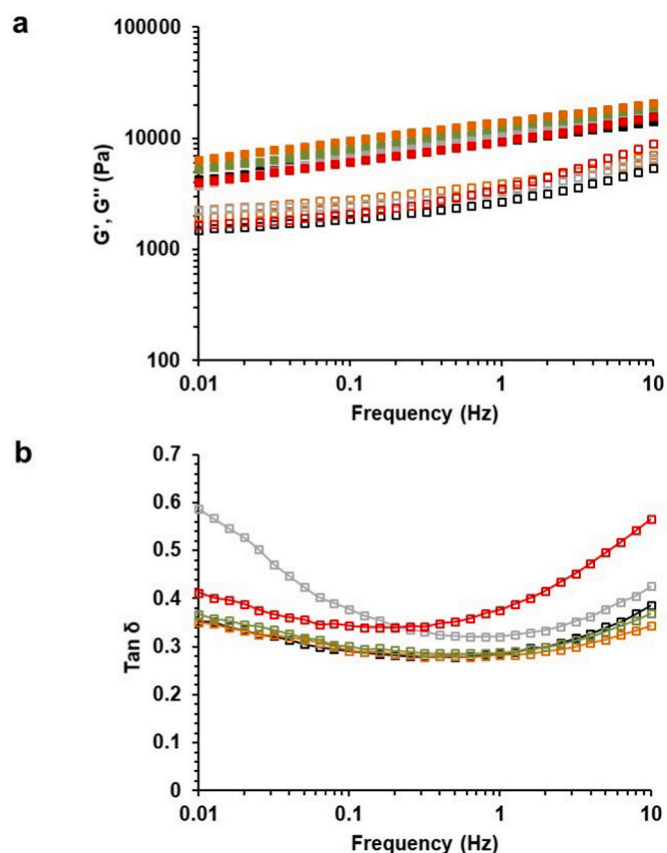


Fig. 2. Frequency sweep (a) and variation of $\tan \delta$ with frequency (b) of the different chocolate spreads. G' : closed symbols; G'' : empty symbols. Control spread made with coconut fat (■ C); spread made with 50% coconut fat and 50% olive oleogel (■ CO); spread made with 50% coconut fat and 50% sunflower oleogel (■ CS); spread made with olive oleogel (■ O); spread made with sunflower oleogel (■ S).

Table 2

Rheological values (G' , G'' , and $\tan \delta$) (mean \pm standard deviation) of the different chocolate spreads at the frequency of 1 Hz.

	G'	G''	$\tan \delta$
C	8396.47 ^a \pm 1507.43	2467.045 ^a \pm 333.80	0.30 ^{ab} \pm 0.01
CO	14,507.25 ^d \pm 693.60	3985.795 ^c \pm 37.70	0.27 ^a \pm 0.01
CS	11,146.3 ^c \pm 542.30	3239.25 ^b \pm 138.44	0.29 ^a \pm 0.01
O	10,370.15 ^{bc} \pm 46.17	3295.63 ^{bc} \pm 31.75	0.32 ^{ab} \pm 0.01
S	9194.455 ^{ab} \pm 322.36	3138.93 ^{ab} \pm 576.31	0.34 ^b \pm 0.05

Control spread made with coconut fat (C); spread made with 50% coconut fat and 50% olive oleogel (CO); spread made with 50% coconut fat and 50% sunflower oleogel (CS); spread made with olive oleogel (O); spread made with sunflower oleogel (S). Values with different lowercase letters (a, b, ...z) within the same column are significantly different ($p < 0.05$) according to the LSD multiple range test.

(Pal, 1996). Fayaz et al. (2020b) reported good mechanical properties in chocolate spreads made with a mixture of palm fat and monoglyceride oleogels, attributed to the good chemical compatibility between the palm fat and the oleogel, and to a strengthening of the structure because of the formation of lamellar structures stabilised by hydrogen bonds.

3.1.3. Texture measurements

The texture of spreads is one of the most influential factors for sensory acceptance. With increased firmness or maximum force of penetration, the sample is perceived as less wet or oily and has a lower spread, meaning it is more stable. Fig. 3 shows the profiles of the texture

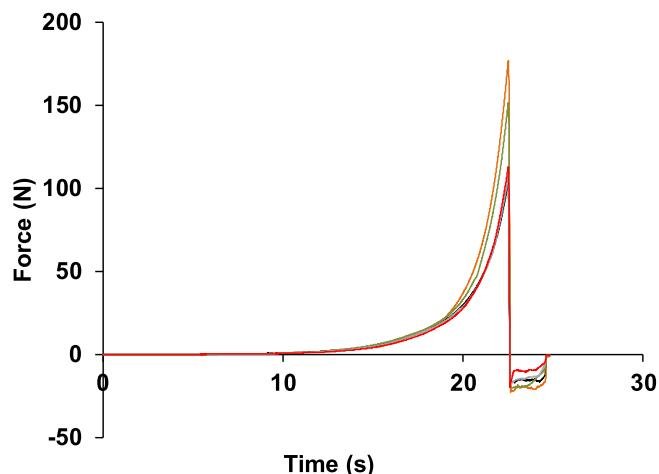


Fig. 3. Texture profile of the different chocolate spreads. Control spread made with coconut fat (— C); spread made with 50% coconut fat and 50% olive oleogel (— CO); spread made with 50% coconut fat and 50% sunflower oleogel (— CS); spread made with olive oleogel (— O); spread made with sunflower oleogel (— S).

Table 3

Peak values of maximum force and areas under the curve (mean \pm standard deviation) obtained by the spreadability test for the different chocolate spreads.

	Maximum Force (N)	Area under the curve (N s)
C	107.01 ^a \pm 6.67	221.57 ^a \pm 7.16
CO	168.23 ^b \pm 12.59	291.38 ^c \pm 12.99
CS	140.71 ^b \pm 14.96	255.94 ^b \pm 22.86
O	104.88 ^a \pm 2.74	213.25 ^a \pm 5.27
S	109.34 ^a \pm 10.56	216.66 ^a \pm 2.90

Control spread made with coconut fat (C); spread made with 50% coconut fat and 50% olive oleogel (CO); spread made with 50% coconut fat and 50% sunflower oleogel (CS); spread made with olive oleogel (O); spread made with sunflower oleogel (S). Values with different lowercase letters (a, b, ...z) within the same column are significantly different ($p < 0.05$) according to the LSD multiple range test.

curves corresponding to the different spreads studied. Similar texture profiles can be seen between the samples, but with greater firmness seen in the CO and CS spreads, which were prepared with partial fat replacement with oleogel.

Table 3 shows the differences in the maximum peak force (N) and AUC (N s) values, calculated for each spread studied. The maximum force is related to the firmness of the samples and the AUC with the spreadability. As it can be seen, the CO and CS spreads presented significantly higher values both in firmness and AUC, which indicates they are samples with greater firmness, while being more spreadable than spreads C, O and S. However, no significant differences were found between the spread C and O or S. These results are consistent with the rheology results, where the CO and CS spreads exhibited a more solid/viscoelastic behaviour.

Nevertheless, this demonstrates the ability of oleogels formulated with HPMC and XG to impart a similar structure to spread C, formulated with a saturated fat, while improving the lipid profile of the fat.

3.2. Sensory analysis

Free Choice Profile (FCP) analysis was performed to determine the attributes that describe the chocolate spreads. With this analysis, information on the spontaneous sensations that occur when the product is consumed is obtained; this information could have been lost using conventional descriptive analysis (Espert, Bresciani, Sanz, & Salvador,

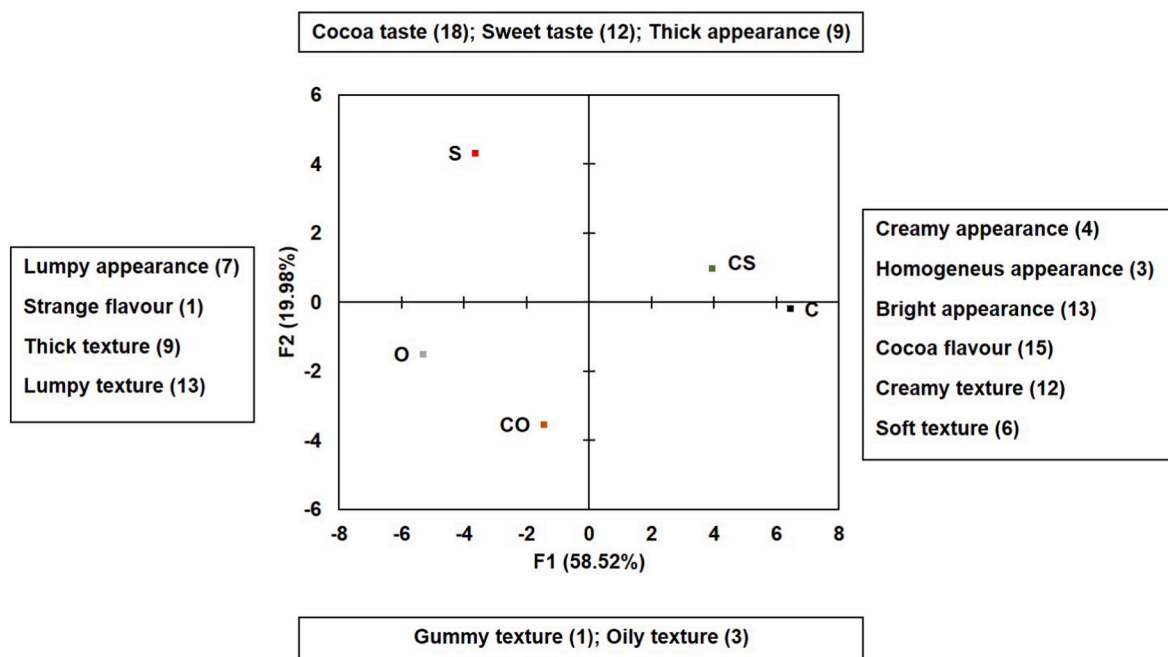


Fig. 5. Representation of the samples in the two-dimensional GPA diagram where the main descriptors correlated with the first two dimensions of the average space are reflected. They are cited in the tables and the number of times each descriptor was mentioned. Control spread made with coconut fat (■ C); spread made with 50% coconut fat and 50% olive oleogel (■ CO); spread made with 50% coconut fat and 50% sunflower oleogel (■ CS); spread made with olive oleogel (■ O); spread made with sunflower oleogel (■ S).

CRedit authorship contribution statement

Santiago Bascuas: Investigation, Validation, Writing - original draft. **María Espert:** Investigation, Validation. **Empar Llorca:** Methodology, Investigation. **Amparo Quiles:** Methodology, Writing - review & editing, Funding acquisition. **Ana Salvador:** Methodology, Investigation, Writing - original draft. **Isabel Hernando:** Supervision, Writing - review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that there is no conflict of interest.

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