


# Muffins Elaborated with Optimized Monoglycerides Oleogels: From Solid Fat Replacer Obtention to Product Quality Evaluation

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**Abstract:** This study demonstrates the effectiveness of using oleogels from high oleic sunflower oil (HOSO) and monoglycerides as solid fat replacers in a sweet bakery product. Firstly, a methodology to obtain oleogels with desired properties based on mathematical models able to describe relationships between process and product characteristics variables followed by multi-objective optimization was applied. Later, muffins were prepared with the optimized oleogels and their physicochemical and textural properties were compared with those of muffins formulated using a commercial margarine (Control) or only HOSO. Furthermore, the amount of oil released from muffins over time (1, 7, and 10 days) was measured to evaluate their stability. The replacement of commercial margarine with the optimized oleogels in muffin formulation led to the obtention of products with greater spreadability, higher specific volume, similar hardness values, and a more connected and homogeneous crumb structure. Moreover, these products showed a reduction of oil migration of around 50% in contrast to the Control muffins after 10 days of storage, which indicated that the optimized oleogels can be used satisfactorily to decrease oil loss in this sweet baked product. Fat replacement with the optimized monoglycerides oleogels not only had a positive impact on the quality of the muffins, but also allowed to improve their nutritional profile (without *trans* fat and low in saturated fat).

**Keywords:** fat replacer, monoglyceride oleogels, muffin, multi-objective optimization, physicochemical properties

**Practical Application:** The food industry demands new ways to reduce the use of saturated and *trans* fats in food formulations. To contribute to this search, oleogels from high oleic sunflower oil and saturated monoglycerides were prepared under optimized conditions in order to obtain a product with similar functionality to margarine, and its potential application as a semisolid fat ingredient in muffins was evaluated. Muffins formulated with oleogels showed an improved quality compare with those obtained using a commercial margarine with the added benefit of a healthier nutritional profile.

## Introduction

Shortenings are edible fats that play a crucial role in the texture and mouthfeel of baked products (Jang, Bae, Hwang, Lee, & Lee, 2015). They confer the structure and geometry of the final products, and also assist in the incorporation of air, heat transfer and extending shelf life (Ghotra, Dyal, & Narine, 2002). Although shortenings are the most selected fats in cooking formulations, they usually have significant amounts of saturated and *trans* fatty acids. In the last years, the intake of saturated and *trans* fatty acids has been associated with deleterious health effects, compelling the food industry into searching for novel products with similar physical and organoleptic characteristics but healthier for consumers (Dhaka, Gulia, Ahlawat, & Khatkar, 2011; Marangoni & Co, 2012). Moreover, the growing awareness of the link between diet and health has led to an increase of concern from the consumers, and hence there is an increasing demand for foods with reduce fat levels.

Among the different approaches that can be explored for *trans* and saturated fat replacement or reduction, oleogelation is one of the up and coming ways to obtain semisolid fat alternatives as well as to develop new products with attractive physical and organoleptic characteristics. An oleogel can be defined as an oil entrapped within a thermoreversible, 3-dimensional gel network, which is formed by the addition of gelator molecules at low concentration and elevated temperatures, followed by cooling (Da Pieve, Calligaris, Co, Nicoli, & Marangoni, 2010; Hughes, Marangoni, Wright, Rogers, & Rush, 2009; Marangoni & Co, 2012; Palla, Giacomozzi, Genovese, & Carrín, 2017). Among the wide number of different molecules which are able to structure oils, monoglycerides (MG) and waxes are some of the most promising alternatives for replacing hard stock structured fats (Fayaz et al., 2017). MG have been identified as efficient gelators in the forming of strong gels at relatively low concentrations and its gelling properties have been extensively studied in various edible oil systems (López-Martínez et al., 2014; Ojijo, Neeman, Eger, & Shimoni, 2004; Da Pieve et al., 2010; Rocha-Amador et al., 2014; Valoppi et al., 2017).

Different studies have demonstrated that not only the chemical nature of oleogel components are important to determine physical properties of the final product, but also the selected preparation conditions of the gels (Blake & Marangoni, 2015; Ojijo

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et al., 2004; Rocha-Amador et al., 2014). In the particular case of oleogels from MG (a commercial mixture called Myverol 18-50 XL PL) and high oleic sunflower oil, we found in a previous work (Palla et al., 2017) that textural and rheological properties were greatly affected by the concentration of Myverol in the oleogels formulation and the cooling temperature. Furthermore, the temperature of preparation or mixing and the stirring speed showed significant effects over some textural properties indicating that their effects should not be ignored. In the same work, we proposed the use of a methodology to obtain a product with desired characteristics based on mathematical models able to describe relationships between variables—associated to the obtention process and product characterization—and their interactions followed by multi-objective optimization. Taking into account that oleogels systems are highly dependent on the chemical nature of their constituting molecules, the mentioned procedure can be applied to develop new oleogels in a very efficient way when their formulation is changed.

The potential use of oleogels, mainly from waxes, as fat replacers in sweet baked products has been studied in recent years (Calligaris, Manzocco, Valoppi, & Nicoli, 2013; Jang et al., 2015; Lim et al., 2017; Manzocco, Calligaris, Da Pieve, Marzona, & Nicoli, 2012; Mert & Demirkesen, 2016; Patel et al., 2014; Yilmaz & Ögütçü, 2015). Calligaris et al. (2013) used MG oleogels of sunflower oil as a shortening alternative in the elaboration of a sweet bread. On the other hand, oleogels have been proposed as possible oil migration inhibitors in food products (Hughes et al., 2009; Marangoni & Co, 2012). However, there is still a lack of knowledge about the use of MG oleogels as replacers for solid fats in baked products and their effects on the products quality attributes. In particular, muffins are sweet baked products highly appreciated by consumers due to their good taste and spongy texture (Martínez-Cervera, de la Hera, Sanz, Gómez, & Salvador, 2013), in which the nature and quantity of the used ingredients determine their quality. Muffins nutritional profile would be greatly improved by substituting their generally high solid fat content.

In this study, oleogels from a commercial mixture of monostearin and monopalmitin (Myverol 18-04 K SG) and high oleic sunflower oil were produced and evaluated as fat replacers in muffins. Firstly, we tested the effect of oleogel preparation variables—the concentration of Myverol, the speed of agitation and the cooling temperature—on selected responses variables—oil binding capacity (OBC) and the most representative rheological and textural parameters—using a three factors Box-Behnken design. In order to find optimal oleogels preparation conditions, a fitting model for each response was obtained and later a multi-response optimization was made. All tests were accomplished in comparison with a commercial margarine in order to determine if these oleogels would be appropriate as an alternative for this product. After that, muffins were prepared with optimized oleogels and their physicochemical properties were compared with those obtained in muffins prepared using the commercial margarine (Control) or only the oil. Furthermore, the amount of oil released from muffins over time was measured to evaluate their stability since it negatively influences the quality of food products. To the author's knowledge, the application of MG oleogels in muffins has not been studied, representing a novel aspect of the present contribution. Moreover, it is important to point out that this study analyses the influence of oleogel incorporation into the food matrix in order to reduce oil migration over time, which has not been considered in similar studies.

## Materials and Methods

### Materials

The high oleic sunflower oil (HOSO) used to prepare the oleogels was purchased from a local grocery store and the Myverol 18-04 K SG (Myv) used as gelator agent was generously donated by Kerry (Ireland). The weight composition of predominant fatty acids in both materials was determined as fatty acid methyl esters (FAME) by GLC analysis according to AOCS Official Methods Ce2-66 and Ce1-62 (AOCS, 2009): HOSO (saturated fatty acids 6.58%, monounsaturated fatty acids 85.25%, and polyunsaturated fatty acids 7.71%) and Myv (stearic acid 43.85% and palmitic acid 53.63%). The Myv melting point (69 °C) was measured by DSC. A commercial semisolid fat product, light margarine (CM) was used to compare the results. Its weight composition, supplied by the manufacturer (Molinos Río de la Plata S.A, Argentina), was 40% of total fat and 23% of saturated fat. The CM and the other ingredients used for muffin formulation—wheat flour, sugar, baking powder, eggs, vanilla essence—were purchased from local stores.

### Experimental design, modeling, and optimization

In this work, response surface methodology was applied to evaluate the influence of selected preparation conditions on response variables. The three selected factors were: the concentration of Myv (C), the speed of agitation (A), and the cooling temperature (T). They were selected according to similar works (Palla et al., 2017; Rocha-Amador et al., 2014; Yilmaz & Ögütçü, 2014) in which these variables had significant effects. The setting of the factors was determined in accordance with previous studies, in addition to practical considerations. MG are very efficient structuring agents because small amounts of them were able to structure large quantities of oil (Da Pieve et al., 2010). From exploratory trials we found that 2 wt% of Myv was the minimum amount necessary for gelation of HOSO, but concentrations higher than 4 wt% were required to achieve rheological and textural properties close to that of the commercial semisolid fat selected as model product. The experimental design was based on Box-Behnken incomplete factorial design of three levels and three factors with three replicates of the central point. Since it is a second order design based on three levels, coded as -1, 0, and +1, the chosen factor levels in terms of actual values were: C (4, 7, 10 wt%), A (200, 600, 1000 rpm) and T (5, 17.5, 30 °C). Matrix design was performed using the Design Expert 7.0 software. Table 1 shows factors and levels of each studied variable and the analyzed responses. The selected responses were: oil binding capacity (OBC), textural properties as hardness (Ha), adhesiveness (Ad) and cohesiveness (Co), and the elastic modulus ( $G'$ ) as rheological parameter. All experiments were performed in completely randomized order. Data were fitted with a second order polynomial equation with the following form:

$$\hat{y} = \beta_0 + \sum_{i=1}^3 \beta_i x_i + \sum_{i=1}^3 \beta_{ii} x_i^2 + \sum_{i=1}^2 \sum_{j=i+1}^3 \beta_{ij} x_i x_j \quad (1)$$

where  $\hat{y}$  is the response being studied,  $\beta_0$  is the intercept,  $\beta_i$  is the main effect coefficient for the  $i$ th factor,  $\beta_{ii}$  is the quadratic effect coefficient for the  $i$ th factor,  $\beta_{ij}$  is the model coefficient for the interaction between factors  $i$ th and  $j$ th, and  $x_i$  and  $x_j$  are each of the considered factors. An ANOVA for the response surface quadratic model was used to determine the statistical significance of the model factors. Those model terms with  $P$  values  $<0.05$  were considered significant and the others were not taken into account, except when they increased the quality of the

**Table 1**—Experimental design and the results obtained for the selected responses for oleogels formulated with high oleic sunflower oil and Myverol.

Exp <sup>a</sup>	Experimental factors <sup>b</sup>			Response variables <sup>c,d</sup>												
	C(wt%)	A(rpm)	T(°C)	log G <sub>0</sub> ' (Pa)	m <sub>G</sub> ' (Pa.s)	n	OBC (wt%)	Ha (N)	Ad (N.s)	Co						
1	4	200	17.5	3.92 (3.91)	0.05 (0.04)	0.63 (0.64)	79.50 (78.42)	0.28 (0.57)	0.59 (1.05)	0.49 (0.45)						
2	4	600	30	3.70 (3.71)	0.04 (0.05)	0.91 (0.89)	81.93 (80.08)	0.22 (0.16)	0.64 (0.34)	0.53 (0.55)						
3	10	200	17.5	4.85 (4.84)	0.05 (0.05)	1.15 (1.15)	91.17 (90.63)	2.63 (2.48)	4.11 (3.66)	0.25 (0.26)						
4	7	600	17.5	4.56 (4.60)	0.05 (0.05)	0.73 (0.74)	90.21 (90.36)	1.41 (1.52)	2.81 (2.70)	0.31 (0.34)						
5	7	200	5	4.36 (4.38)	0.08 (0.08)	0.85 (0.84)	95.88 (97.32)	0.83 (0.82)	1.63 (1.98)	0.29 (0.28)						
6	7	1000	30	4.25 (4.23)	0.06 (0.05)	0.84 (0.85)	88.04 (88.47)	0.77 (0.80)	2.23 (1.99)	0.41 (0.39)						
7	7	600	17.5	4.65 (4.60)	0.05 (0.05)	0.73 (0.74)	91.22 (90.36)	1.50 (1.52)	3.09 (2.70)	0.31 (0.34)						
8	7	1000	5	4.46 (4.46)	0.07 (0.08)	0.85 (0.85)	96.38 (94.33)	0.64 (0.83)	1.48 (1.74)	0.33 (0.30)						
9	10	600	5	4.76 (4.75)	0.09 (0.08)	1.31 (1.33)	96.70 (98.15)	1.70 (1.78)	3.07 (2.70)	0.27 (0.26)						
10	7	200	30	4.36 (4.36)	0.05 (0.05)	0.79 (0.79)	90.51 (91.47)	0.83 (0.79)	2.06 (2.23)	0.39 (0.37)						
11	4	1000	17.5	3.88 (3.89)	0.04 (0.04)	0.78 (0.78)	71.65 (75.42)	0.31 (0.57)	0.73 (0.81)	0.49 (0.47)						
12	7	600	17.5	4.60 (4.60)	0.05 (0.05)	0.74 (0.74)	90.43 (90.36)	1.68 (1.52)	2.73 (2.70)	0.30 (0.34)						
13	10	1000	17.5	4.80 (4.81)	0.06 (0.05)	1.10 (1.08)	89.00 (87.64)	2.87 (2.48)	2.98 (3.42)	0.26 (0.28)						
14	4	600	5	3.66 (3.64)	0.07 (0.07)	0.55 (0.55)	79.68 (78.84)	0.12 (0.13)	0.33 (0.09)	0.35 (0.38)						
15	10	600	30	4.41 (4.43)	0.06 (0.06)	0.94 (0.93)	84.75 (85.20)	1.37 (1.75)	2.59 (2.96)	0.29 (0.28)						
<b>Commercial margarine</b>				4.54 ± 0.01	0.06 ± 5.10 <sup>-3</sup>	0.65 ± 0.03	— <sup>f</sup>	1.60 ± 0.15	5.37 ± 0.29	0.58 ± 0.04						
<b>O-1<sup>e</sup></b>	8.06	1000	25	4.54 (4.55)	0.05 (0.05)	0.84 (0.84)	88.60 (90.37)	1.60 (1.60)	2.81 (2.81)	0.33 (0.28)						
<b>O-2<sup>e</sup></b>	6.60	200	17.5	4.54 (4.57)	0.05 (0.05)	0.73 (0.85)	90.94 (90.97)	1.39 (1.33)	2.63 (2.81)	0.34 (0.33)						

<sup>a</sup>Number of experiment.<sup>b</sup>C, concentration of Myverol; A, speed of agitation; T, cooling temperature.<sup>c</sup>log G<sub>0</sub>', m<sub>G</sub>' and n, parameters of the elastic modulus (G') model; OBC, oil binding capacity; Ha, hardness; Ad, adhesiveness; Co, cohesiveness.<sup>d</sup>Experimental value (predicted value by corresponding model). The experimental values are the average of at least 4 individual replicates.<sup>e</sup>O, Optimization.<sup>f</sup>-, Not determined.

fit. The adequacy of the developed regression models was evaluated by statistical coefficients: *R*-squared (*R*<sup>2</sup>), adjusted *R*-squared (*R*<sup>2</sup>adj), predicted *R*-squared (*R*<sup>2</sup>pred), and adequate precision (AP) and also by testing it for the lack of fit (Ferreira et al., 2007; Montgomery, 2009). Furthermore, in order to validate experimental data, the homoscedasticity was graphically assessed by the examination of residual versus fitted values plot for each response verifying that the residual ones showed a structureless pattern (Granato, de Araújo Calado, & Jarvis, 2014; Montgomery, 2009).

In order to find out the optimal preparation conditions that would allow us to produce oleogels with textural and rheological properties close to the ones of the chosen commercial semisolid fat product, a multi-objective optimization procedure based on the desirability function (*D*) was applied using the Design Expert 7.0 software following the methodology described by Palla et al. (2017). This software uses a numerical optimization algorithm, in which convergence is achieved when the objective function change is less than a 10<sup>-6</sup> ratio (State-Ease, 2018).

### Oleogel preparation

The 160 g mixture used in each experiment design run was prepared by solubilizing the correspondent amount of Myv (4, 7, or 10 wt%) in HOSO previously heated at 80 °C using a glass container system with a temperature-controlled water bath. The mixture was kept under magnetic agitation at the selected speed (200, 600, or 1000 rpm) during 30 min. After that, the molten sample was put into rectangular polyethylene containers allowing it to form gel by cooling at controlled ambient air temperature (5, 17.5, or 30 °C) under static conditions. The obtained oleogels were stored in darkness at 5 °C for 24 hr until analysis.

### Analytical Methods

#### Measurement of oleogel properties

All the techniques described in the following sections were performed using the oleogel samples encompassed in the rect-

angular containers where they were formed. We understand that this methodology allows us to analyze the same material avoiding influences that could be generated by gelifying molten samples in different containers according to each technique.

**Determination of the viscoelastic properties.** All rheological analyses were conducted using a Paar Physica rheometer (model MCR 301, Anton Paar GmbH, Austria) with a parallel plate geometry (50 mm diameter) and a computerized data acquisition system (Rheoplus/32 V3.40). Samples were taken from the container by cutting thin sheets that were used to obtain disk-shaped samples (50 mm diameter, 3 mm height) using a round metallic cutter. Oscillatory frequency (*ω*) sweep test was performed at 20 °C (strain 0.5%) from 10 to 100 rad/s. A sandpaper (grade 360, Norton) was attached to the lower and upper surfaces of the parallel-plate geometry to avoid the samples from slipping. The gap was set to produce a normal force of 10 N (Palla et al., 2017). A number of 4 individual replicates were used for each experimental run. Additionally, an oscillatory stress sweep test was done at a frequency of 10 rad/s with a strain range from 0.01% to 100% in order to ensure that all measurements were carried out in the linear viscoelastic region.

Since an oleogel with viscoelastic properties similar to a commercial margarine in all the studied frequency range was being searched, a mathematical model was used to describe the *G'* behavior. Thus, the obtained experimental curves were adjusted using the following empirical correlation:

$$\log G' = \log G'_0 + m_G \log \omega^n \quad (2)$$

where log *G'*<sub>0</sub>, *m*<sub>*G*</sub>, and *n* are constants to be determined experimentally.

**Oil binding capacity (OBC).** The gel network ability to retain oil was tested using a simple centrifugation method. One gram of oleogel was carefully transferred into an eppendorf tube and centrifuged at 9000 rpm for 15 min at 17 °C using a

**Table 2—Formulations of the different evaluated muffins.**

	Ingredients (wt%) <sup>a</sup>				Water
	CM <sup>b</sup>	HOSO <sup>c</sup>	O1 <sup>d</sup>	O2 <sup>d</sup>	
Control	26.08	—	—	—	—
M1	—	10.43	—	—	15.65
M2	—	—	—	10.43	15.65
M3	—	—	10.43	—	15.65

<sup>a</sup>Remaining ingredients: 26.09 wt% flour, 26.09 wt% sugar, 18.84 wt% homogenized eggs, 1.45 wt% vanilla essence, and 1.45 wt% baking powder.

<sup>b</sup>CM, commercial margarine.

<sup>c</sup>HOSO, high oleic sunflower oil.

<sup>d</sup>Oleogel obtained from O-1 (O1) and O-2 (O2) preparation conditions.

microcentrifuge (Giumelli z-127-D, Argentina). Three replicates of each sample were performed. OBC was calculated as a function of the percentage of oil released from the sample after centrifugation:

$$\text{OBC (\%)} = 100 - \left( \frac{\text{mass of released oil}}{\text{mass of oleogel}} \right) \times 100 \quad (3)$$

**Texture analysis.** The texture analysis measurements were conducted using a Texture Analyzer TA Plus (Lloyd instruments, England) equipped with a 50 N load cell. The texture profile analysis (TPA) test used for textural evaluation consisted of a two-cycle penetration of the sample with 10 s waiting time between the cycles using a cylindrical probe with rounded head (12.5 mm in diameter × 56 mm in length). In each cycle, the sample was penetrated to a depth of 10 mm at a crosshead speed of 1 mm/s, and then the probe was returned to the initial position at the same speed. From the resulting force–time curve, hardness (the maximum force measured during the first penetration cycle), adhesiveness (the negative force area of the first cycle), and cohesiveness (the ratio of the positive force areas under the second and first penetration cycles) were determined (Palla et al., 2017; Yılmaz & Ögütçü, 2014). Measurements were carried out after holding the samples at 20 °C for 1 hr in their original containers. The test was replicated four times for each sample.

### Muffin preparation

A basic muffin formulation, which originally included a 26% of butter, was modified and used to test the ability of optimized oleogels to replace solid fat content. The Control formulation was made with the commercial light margarine. The other formulations were modified by substituting the CM by HOSO (M1), oleogel from O-1 (M2) and oleogel from O-2 (M3) (Table 2). Taking into account that margarine is a water/oil type emulsion, fat replacement was accomplished by considering its total fat and water content. Each batter was prepared in a mixer, in which the eggs were whisked for 2 min at top speed. The fatty ingredient, CM or HOSO/oleogel together with water, was added and mixed for 3 min at intermediate speed. After that, the baking powder and flour were added and the mixture was beaten for a further 3 min at intermediate speed. Lastly, vanilla essence was added and the mixer speed was increased to the top. By weighing in analytical balance, exactly 30 g of batter was dosed into each paper mold (43 mm diameter, 35 mm height). Each batch of batter was used to fill 12 individual silicone baking molds that were arranged in 3 rows of 4 on a baking tray and baked for 20 min at 200 °C in an electric oven. After cooling to room temperature for 1 hr, the muffins were placed into plastic containers and stored at room temperature for 24 hr until analysis, unless specifically stated oth-

erwise. The oven, the tray and all other baking conditions were the same in all cases. In randomized order, each muffin formulation was processed in duplicate.

### Analysis of muffin properties

**Oil loss determination.** Oil migration was determined according to the technique described previously by Dibildox-Alvarado, Rodrigues, Gioielli, Toro-Vazquez, and Marangoni (2004). Muffins were put into cylindrical containers (60 mm diameter) with filter papers (Whatman #4, 110 mm diameter). The amount of oil lost by each sample was determined by the difference in weight of the filter paper before and after placing the muffin on it for 1, 7, and 10 days after muffin preparation at 20 °C. Four replicates were performed and the average and standard deviation were reported. Oil loss (OL) was expressed as % (g oil/100 g muffin).

**Textural properties.** Muffins were cut horizontally at its height, the upper halves were discarded and the 2.5 cm high lower halves were used to perform TPA test using the texture analyzer mentioned above. During this assay, the samples were penetrated twice to a depth of 10 mm at a crosshead speed of 1 mm/s with 10 s waiting time between the two cycles using a cylindrical probe with rounded head (12.5 mm in diameter × 56 mm in length). From each force–time curve, hardness and cohesiveness parameters were calculated to characterize muffins textural properties. Four muffins from each batch were measured.

**Color.** Surface and crumb color of the muffins were measured in a HunterLab UltraScan XE tristimulus colorimeter (Hunter Associates Laboratory, Inc., Reston, VA, U.S.A.). The total color reflected by the sample was expressed in accordance with the CIELAB system at 10° observer angle with D65 illuminant. The parameters measured were  $L^*$  [lightness: 0 = black, 100 = white],  $a^*$  [greenness (–), redness (+)], and  $b^*$  [blueness (–), yellowness (+)]. Four muffins from each batch were measured at 3 different points. The total color difference ( $\Delta E^*$ ) between samples and Control muffins was calculated from the parameters differences as follows:  $\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ . A value of  $\Delta E^* < 3$  means that the differences are not visible to the human eye (Francis & Clydesdale, 1975).

**Weight and dimensional characteristics.** Muffins were removed from the oven and their dimensions were determined after cooling for 1 hr at about 22 °C. Diameter ( $d$ ) and height ( $H$ ) were measured with a digital caliper (327-06-0, ASIMETO, Germany). Weight and dimensions of 4 randomly selected muffins were recorded. Spread ratio (SR) was calculated by dividing the average diameter by the average height of the muffins produced with each formulation.

**Moisture content.** Moisture content was measured with an OHAUS MB35 moisture analyzer (Switzerland). Results were expressed on a wet weight basis. Measurements were done in triplicate.

**Specific volume.** Specific volume ( $\text{cm}^3/\text{g}$ ) was obtained by seed displacement technique according to Approved Method 10-05 (AACC, 2000) using chia seeds. Three replicates of each batch were performed.

**Digital image analysis of crumb structure.** Muffins were cut on a horizontal plane at a distance of 2.5 cm from the base. Color images of freshly cut crumb surfaces were captured using a Nikon D3400 digital camera (55 mm,  $f/3.5-5.6$ , ISO 100), with a magnification of 24x. Digital images of the crumbs were analyzed with the software ImageJ 1.47v (National Institutes of Health, Bethesda, MD, U.S.A.). The center of each image was

cropped to a square field of view and converted to 8-bits with grey levels ranging from 0 to 255. These images were binarized (converted from grey-level to black and white) using an automated fuzzy measure thresholding method to differentiate air cells from noncells. Crumb grain features, that is to say, air cells size distribution and total number of air cells/cm<sup>2</sup>, were chosen for analysis.

### Statistical analysis

All the experiments were performed in a completely randomized design. Statistical differences in muffins properties were determined by one-way ANOVA using the Infostat software (Di Rienzo et al., 2011). Fisher's LSD method was used with a significance level  $P \leq 0.05$ . All the results were expressed as the mean  $\pm$  standard deviation.

## Results and Discussion

### Properties of oleogels obtained by experimental design

Experimental design was applied to investigate the individual and interactive effects of the preparation conditions on the selected responses. Polynomial regression modelling was performed between the response variables and the corresponding coded values of the three factors, and finally, the best-fitted model equation was obtained. The properties of the oleogels obtained under the selected preparation conditions, as well as their corresponding predicted values by each model, are shown in Table 1. Model coefficients (B), indicated in actual variables, and model terms statistical significance are given in Table 3. Also the statistical coefficients, which indicate the accuracy of the regression models, are presented. Obtained values of  $R^2$ , which were quite high ( $>0.91$ ), demonstrated that the experimental data variation was adequately explained by the developed models. The  $R^2_{adj}$  and  $R^2_{pred}$  were in good agreement (difference less than 20%) in all response models, and values of AP were higher than 13.9, which means that the models equations can be used to predict the responses for any value of factors within the range of the experimental design.

As result of the oscillatory frequency sweep tests it was found that all obtained oleogels had a gel-like behavior ( $G' > G''$ ). The experimental  $G'$  data were transformed using logarithmic function and properly fitted by Eq. (2), since all the fits presented  $R^2$  values higher than 0.98 (data not shown). Values for the parameters of this model,  $\log G'_0$ ,  $m_{G'}$  and  $n$  obtained for each condition of oleogel preparation are shown in Table 1. Regarding parameter  $\log G'_0$ , C and T were the most influential factors followed by C-T and A-T interactions to a lesser extent (Table 3). The highest  $\log G'_0$  values were obtained using the highest Myv content and preferentially T below 25 °C (Figure 1A). Since parameter  $\log G'_0$  is directly associated to  $G'$  magnitude, it can be noted that these preparation conditions lead to the obtention of materials with greater elasticity. With regard to  $m_{G'}$  and  $n$  parameters of  $\log G'$  model, the regression model to represent  $m_{G'}$  showed that T was the most significant factor and C in a minor degree and, in the case of  $n$ , C, and C-T were the most influential terms, followed by A, T, and C-A. Taking into account that both parameters indicate how dependent is  $G'$  on the frequency and that the found values were low, it can be claimed that all the obtained oleogels showed a  $G'$  with slight dependence upon frequency. Even so, the approach used to evaluate the preparation conditions effects identified that some of them led to obtain less stable materials. In Figure 1B it can be noticed that the highest  $m_{G'}$  values were ob-

tained at the highest C and the lowest T, which was in agree with the found  $n$  behavior (response surface not shown). This lowest stability in the samples containing the highest number of smaller crystals (Palla et al., 2017) could be attributed to the presence of weaker interactions in crystalline phase over the timescale of the frequency test. Oleogels from Myverol 18-50 XL PL and HOSO prepared in similar conditions (Palla et al., 2017) showed comparable  $\log G'_0$  values and  $m_{G'}$  values higher than those obtained in this work, which indicates that the last ones present a more stable and strengthened network. Last but not least, based on rheological parameter values obtained by modelling oleogel behavior and comparing them to the ones corresponding to the CM, it can be inferred that it would be possible to develop a material able to resemble this semisolid product under the experimental conditions explored in this work.

The formulated oleogels showed OBC values ranged between 71.65% and 96.70%, hereby evidencing that MG crystal networks provide high levels of structuration and oil stabilization. According to the  $P$  value, C was the most significant factor on this response followed by T and C-T interaction, whereas A did not turn out to be a significant factor. The maximum OBC value was reached using almost the highest C ( $\sim 9$  wt%) and the lowest T (Figure 1C). From a structural point of view, this means that lowest T, which led to faster cooling, generates a network of homogeneous small crystals (Palla et al., 2017) that resulted more effective when the amount of them increased (higher C), providing greater surface area and a more tortuous pathway impeding the migration of loose oil. The importance of the cooling rate becomes evident at high levels of C, where the increase in T produced a material with a lower capacity to retain oil, which was associated with the longer crystals formation. A similar result was found by Blake and Marangoni (2015) in the study of cooling rate on wax oleogels. In general terms, the OBC values obtained with oleogels from Myverol 18-04 K SG resulted slightly lower than those found using oleogels from Myverol 18-50 XL PL (Palla et al., 2017), but it is not possible to attribute this difference to the chemical nature of the used MG mixture, since the technique applied to determine OBC was slightly different.

Hardness is one of the most important attributes to characterize hard stock fats. Ha values of oleogels ranged between 0.12 and 2.87 N, which means that some oleogels resulted harder than the CM (Ha = 1.60 N). The ANOVA indicated that the significant factors over Ha were C and T. For all the analyzed cooling temperature range, Ha was found to increase linearly with the increase of C (Figure 1D), which was related to the rise of the crystals number constituting the gel network. Furthermore, an optimum T that allows to obtain the hardest oleogels was identified ( $\sim 17.5$  °C). Related to adhesiveness and cohesiveness attributes of the obtained oleogels, the determined values resulted lower than those of the commercial product chosen for the comparison. It was found that Ad response presented the same significant factors as Ha, as well as a similar trend when these parameters changed (Figure 1E). Concerning to Co parameter, besides C and T, the C-T interaction proved to be significant. As it could be noticed in Figure 1F, oleogels prepared using the highest amount of Myv showed Co values that did not change significantly with the changes in the used T. On the other hand, this response was maximized using the highest T and the lowest C. This opposite effect generated by C on Co and Ha was also found in oleogels from Myverol 18-50 XL PL and HOSO and it has been explained in detail in one of our previous works (Palla et al., 2017). Over each textural response, the speed of agitation was not a significant factor.

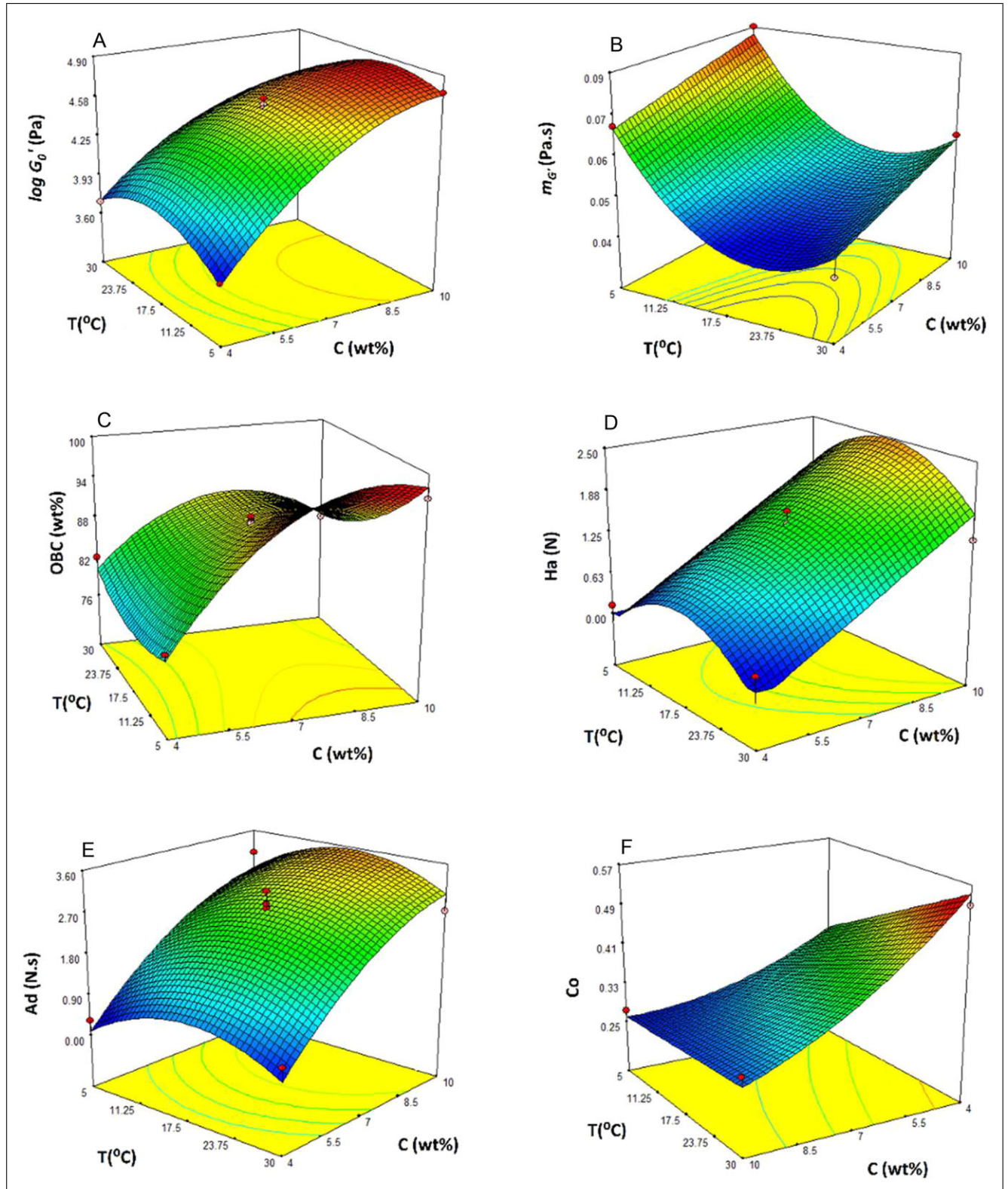


Figure 1–Response surfaces of (A)  $\log G'_0$  and (B)  $m_{G'}$ , parameters of the elastic modulus ( $G'$ ) model; (C) OBC, Oil binding capacity; (D) Hardness, Ha; (F) Cohesiveness, Co; (E) Adhesiveness, Ad. Effect of the concentration of Myverol (C) and the cooling temperature (T) at speed of agitation fixed at central point.

**Table 3—Coefficients of the quadratic model (actual factors) for each response variable, their statistical significance and the model statistical coefficients.**

Model	Response variables <sup>b</sup>							
	Coefficient <sup>a</sup>	log $G_0'$ (Pa)	$m_G'$ (Pa.s)	$n$	OBC (wt%)	Ha (N)	Ad (N.s)	Co
$B_0$		1.442	$8.10 \times 10^{-2}$	$4.25 \times 10^{-1}$	$3.59 \times 10^1$	-2.090	-4.261	$5.22 \times 10^{-1}$
$B_C$		$5.62 \times 10^{-1***}$	$2.25 \times 10^{-3**}$	$-3.0 \times 10^{-2***}$	$1.59 \times 10^{1***}$	$3.18 \times 10^{-1***}$	1.155 <sup>***</sup>	$-6.2 \times 10^{-2***}$
$B_A$		$2.46 \times 10^{-4*}$	$6.25 \times 10^{-7*}$	$-9.58 \times 10^{-7**}$	$-3.75 \times 10^{-3*}$	$8.75 \times 10^{-6*}$	$-3.03 \times 10^{-4*}$	$2.50 \times 10^{-5*}$
$B_T$		$7.3 \times 10^{-2**}$	$-4.78 \times 10^{-3***}$	$2.0 \times 10^{-2*}$	$-1.41 \times 10^{-1**}$	$1.59 \times 10^{-1*}$	$1.69 \times 10^{-1*}$	$1.1 \times 10^{-2**}$
$B_{CA}$		$-2.47 \times 10^{-6*}$	—	$-4.42 \times 10^{-5**}$	—	—	—	—
$B_{CT}$		$-2.62 \times 10^{-3**}$	—	$-4.91 \times 10^{-3***}$	$-9.5 \times 10^{-2**}$	—	—	$-1.06 \times 10^{-3**}$
$B_{AT}$		$-1.09 \times 10^{-5**}$	—	$2.40 \times 10^{-6*}$	—	—	—	—
$B_C^2$		$-2.6 \times 10^{-2***}$	—	$1.5 \times 10^{-2***}$	$-8.15 \times 10^{-1***}$	—	$-5.1 \times 10^{-2*}$	$3.44 \times 10^{-3*}$
$B_A^2$		$-5.79 \times 10^{-8*}$	—	$2.63 \times 10^{-7**}$	—	—	—	—
$B_T^2$		$-1.52 \times 10^{-3***}$	$1.10 \times 10^{-4***}$	$3.46 \times 10^{-4**}$	$1.60 \times 10^{-2**}$	$-4.58 \times 10^{-3**}$	$-4.5 \times 10^{-3**}$	—
<b>Statistics<sup>c</sup></b>								
$R^2$		0.997	0.954	0.998	0.954	0.924	0.917	0.931
$R^2_{adj}$		0.993	0.935	0.993	0.920	0.893	0.871	0.893
$R^2_{pred}$		0.981	0.881	0.962	0.787	0.805	0.749	0.782
AP		44.263	21.021	57.133	16.599	16.612	13.933	15.758

<sup>a</sup>C, concentration of Myverol; A, speed of agitation; T, cooling temperature.

<sup>b</sup>log  $G_0'$ ,  $m_G'$ , and  $n$ , parameters of the elastic modulus ( $G'$ ) model; OBC, oil binding capacity; Ha, hardness; Ad, adhesiveness; Co, cohesiveness.

<sup>c</sup> $R^2$ , R-squared;  $R^2_{adj}$ , adjusted R-squared;  $R^2_{pred}$ , predicted R-squared; AP, adequate precision.

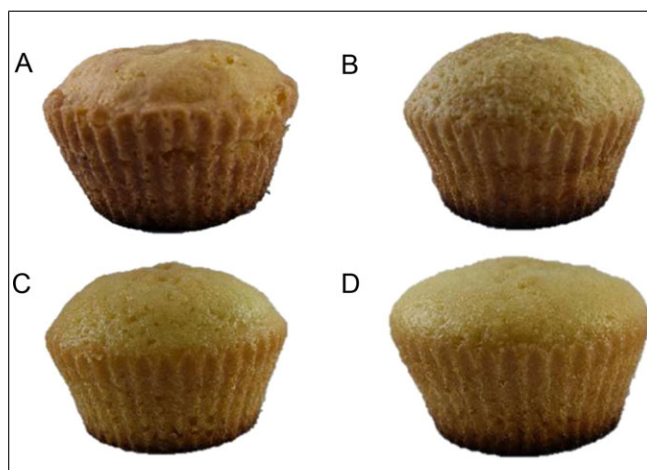
Statistical significance of the model terms: \*\*\* $P \leq 0.0001$ ; \*\* $0.0001 < P \leq 0.05$ ; \* $P > 0.05$ .

### Multiresponse optimization to produce oleogels with desired characteristics

The mathematical models obtained for each response were used to carry out a multi-objective optimization. The goal of the optimization was to determine the best set of preparation conditions that allow us to obtain a stable oleogel with textural and rheological properties close to those of CM. The requirements imposed were: Ha = 1.60 N, log  $G_0' = 4.54$  Pa and OBC = maximum, allowing the rest of responses to take any value within the analyzed ranges. The found optimum result (O-1) was: C = 8.06 wt%, A = 1000 rpm, and T = 25 °C, and the corresponding  $D$  value was 0.810. Another optimization was performed adding to the former criteria the minimization of the amount of Myv. In this case, the found optimum result (O-2) was: C = 6.60 wt%, A = 200 rpm, and T = 17.5 °C with  $D = 0.743$ .

Validation of the predictive models was performed through triplicate experiments at O-1 and O-2 conditions. The experimental response values found in those conditions are shown in Table 1, where it can be noted that the experimental response values were very close to the predicted ones. Moreover, when comparing the properties of these oleogels with the CM ones, it is observed that oleogels obtained under O-1 conditions achieved the same Ha value, whereas by following O-2 conditions the hardness value was slightly lower. With regards to the other imposed responses, both oleogels showed a log  $G_0'$  value close to that of CM and a high oil binding capacity. Co and Ad values resulted lower than the corresponding to the CM, which was expected taking into account the values previously obtained in the experimental design, besides that no restrictions of maximization were established on them. O-1 conditions involve the use of a moderate temperature, which requires less energy consumption being useful from a practical standpoint, whereas the O-2 conditions allow to minimize gelator concentration, which is a desirable characteristic in the development of oleogels. According to these, both preparation conditions were selected to be tested as fat ingredient in the subsequent muffin formulations.

It is important to mention that the composition of the optimized oleogels presented around of 37% less of saturated fats compared to



**Figure 2—Images of muffins formulated with (A) commercial margarine (Control), (B) high oleic sunflower oil (M1), and oleogel from optimization (C) O-1 (M2), and (D) O-2 (M3).**

the CM composition, achieving one of the main goals proposed in this work: the development of a tailored material with a healthier fat composition.

### Physicochemical properties of the muffins

Muffins prepared with the two optimized oleogels (M2 and M3 to O-1, and O-2, respectively), HOSO (M1) and CM (Control) according to the formulations presented in Table 2 are shown in Figure 2. The improvement in the nutritional profile due to the use of these fat replacers was a saturated fat content of 1.29%, 2.1%, and 1.9% in M1, M2 and M3, respectively, respect to the 6.6% in the Control.

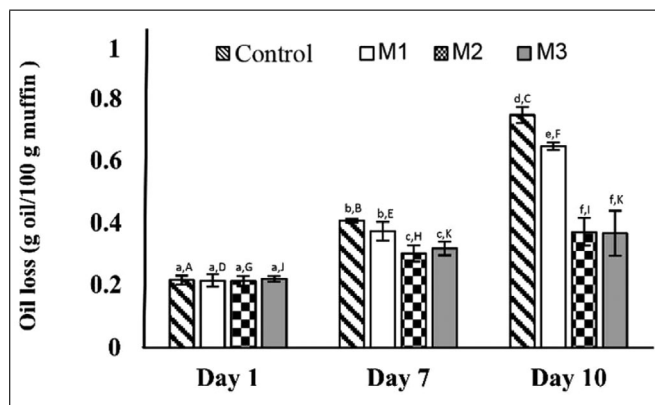
The physicochemical properties determined in the elaborated muffins are presented in Table 4 and discussed in detail in the following sections.

**Oil loss (OL) characterization.** Since an important problem that arises from saturated fat substitution in food products is the oil leakage, resulting in quality defects, the OL from all elaborated

**Table 4—Some physicochemical features of the muffin samples containing commercial margarine (Control), high oleic sunflower oil (M1) and optimized oleogels obtained from O-1 (M2) and O-2 (M3) preparation conditions.**

Sample	Ha (N) <sup>a</sup>	Co <sup>a</sup>	L* <sup>a</sup>	a* <sup>a</sup>	b* <sup>a</sup>	$\Delta E^*$ <sup>a</sup>	W (g) <sup>a</sup>	d (cm) <sup>a</sup>	H (cm) <sup>a</sup>	M (wt%) <sup>a</sup>	V <sub>s</sub> (cm <sup>3</sup> /g) <sup>a</sup>
Control	2.05(0.20) <sup>b</sup>	0.60(0.02) <sup>b</sup>	49.19(0.77) <sup>b</sup>	9.14(0.59) <sup>b</sup>	32.14(0.24) <sup>b</sup>	0.00	25.31(0.41) <sup>b</sup>	5.42(0.02) <sup>b</sup>	3.55(0.02) <sup>b</sup>	22.11(0.56) <sup>bc</sup>	0.35(0.01) <sup>b</sup>
M1	2.34(0.20) <sup>b</sup>	0.61(0.04) <sup>b</sup>	49.78(0.78) <sup>b</sup>	9.66(0.86) <sup>b</sup>	30.56(0.39) <sup>c</sup>	1.76	25.05(0.20) <sup>b</sup>	5.47(0.04) <sup>b</sup>	3.56(0.04) <sup>b</sup>	22.49(0.33) <sup>b</sup>	0.35(0.01) <sup>c</sup>
M2	2.16(0.11) <sup>b</sup>	0.55(0.03) <sup>c</sup>	50.40(1.41) <sup>b</sup>	6.13(0.65) <sup>c</sup>	31.20(0.37) <sup>c</sup>	3.38	24.99(0.03) <sup>b</sup>	5.67(0.01) <sup>c</sup>	3.41(0.04) <sup>c</sup>	21.51(0.47) <sup>bd</sup>	0.38(0.01) <sup>c</sup>
M3	2.13(0.14) <sup>b</sup>	0.51(0.02) <sup>c</sup>	48.39(1.72) <sup>b</sup>	6.94(0.79) <sup>c</sup>	31.06(0.55) <sup>c</sup>	2.58	25.15(0.05) <sup>b</sup>	5.60(0.03) <sup>c</sup>	3.35(0.02) <sup>c</sup>	20.72(0.38) <sup>d</sup>	0.39(0.01) <sup>c</sup>

<sup>a</sup>Ha, hardness; Co, cohesiveness; L\*, a\*, b\*, color parameters corresponding to the CIELAB scale;  $\Delta E^*$ , the total color difference between samples and Control muffins; W, weight; d, diameter; H, height; M, moisture; V<sub>s</sub>, specific volume. Values in parentheses are standard deviation. Means in the same column without a common letter are significantly different ( $P < 0.05$ ).



**Figure 3—Percentage of oil released from muffins as a function of time: muffin samples containing commercial margarine (Control), high oleic sunflower oil (M1), and optimized oleogels obtained from O-1 (M2) and O-2 (M3) preparation conditions. Bars represent means  $\pm$  standard deviation. Significant differences between samples are represented by different letters ( $P < 0.05$ ). The comparison between different muffin formulations for a specific time is indicated with lowercase letters and the comparison over time for a specific muffin formulation is indicated with uppercase letters.**

muffins was evaluated and compared. Taking into account that the two optimized oleogels showed high OBC values, we hypothesized that the introduction of these materials in muffin formulation could reduce their oil loss. The OL from muffins of the four evaluated formulations as a function of a short storage time period (1, 7, and 10 days) is shown in Figure 3. The oil migration of all samples was not significantly different from each other at day 1. After 7 days, the OL values of M2 and M3 were significantly lower than those obtained with CM and HOSO ( $P = 0.0001$ ). At the end of the evaluated time period, the Control muffins showed the highest OL value, followed by muffins from HOSO, whereas those samples prepared with oleogels released the lowest amount of oil and no significant difference between them was found. The incorporation of optimized oleogels in muffins reduced approximately 50% of the OL found in the Control muffins after 10 days of storage, indicating that the developed materials can be used effectively as oil migration inhibitors, which verified the aforementioned hypothesis. It suggests that, although the oleogels were mixed with the remaining ingredients during the batter preparation, their components remained linked by strong interactions between the crystalline network and the oil. The baking process produced the melting of monoglycerides whereas the subsequent cooling generated their recrystallization, therefore the surrounding oil once again should have been trapped by crystals, avoiding their migration. A highlighted aspect is that, although all samples exhibited an increment in OL over time, there was not a large change in M2 and M3 samples between the first day and the last one. This

implies that muffins with an improved short-term stability can be produced using the optimized oleogels as fat replacer.

**Textural parameters, color, weight, dimensional characteristics, moisture, and specific volume.** TPA test of elaborated muffins was performed to obtain their hardness and cohesiveness parameters. All alternative muffin formulations, M1, M2, and M3, showed average Ha values which do not differ significantly from the Control one. Previous studies have shown that Ha can be significantly affected by replacement of shortening in baked products, which makes it one of the most studied attributes (Onacik-Gür, Żbikowska, & Jaroszevska, 2015; Tarancón, Salvador, & Sanz, 2013; Yılmaz & Ögütçü, 2015). Yılmaz and Ögütçü (2015) reported a reduction in the Ha values when they evaluated the replacement of a commercial bakery shortening with sunflower and beeswax oleogels in a cookie formulation. Onacik-Gür et al. (2015) found a significant reduction in hardness of biscuits prepared with HOSO and additives (inulin and lecithin), against those prepared with a commercial shortening. Since oleogels used in this work produced muffins with similar Ha to the Control product, a possible explanation for this might be that these soft materials were designed to have the same Ha than CM. But it is noteworthy that muffins elaborated with HOSO also showed comparable Ha. This could suggest that the different evaluated fats used in a proportion about 10% respect to the total amount of ingredients do not play a determining role over Ha in this type of spongy texture products. Related to cohesiveness parameter, no significant differences were found when CM was replaced with HOSO. However, the Co values decreased significantly when the optimized oleogels were used in the formulation, reflecting an easily deformable crumb structure. Even so, although the optimized oleogels showed Co values about 43% lower than CM, this difference was not kept in equal proportion in the Co of the final product (average difference 13%). From a textural point of view, it may be thought that the use of HOSO as CM replacer would be more effective than the oleogel incorporation since M1 showed comparable Co and Ha values to the Control ones. However, the advantage of structuring HOSO with Myv was the development of a product with the lowest oil migration over time.

Color is an important factor in the determination of the final quality of sweet baked products since it influences their acceptability by consumers. Table 4 shows measurements of crumb color of the elaborated muffins. No significant differences in L\* values were observed among the samples, which means that the fats evaluated as CM replacers did not modify the luminosity of the muffins. Contrarily, the values of a\* and b\* parameters of muffins prepared with oleogels were significantly lower than the Control ones, indicating that M2 and M3 crumbs showed a less orangey and more saturated yellowish color (overall a less brownish tone). On the other hand, muffins obtained with HOSO led to crumbs with a less orange tone respect to the Control one. Since the  $\Delta E^*$



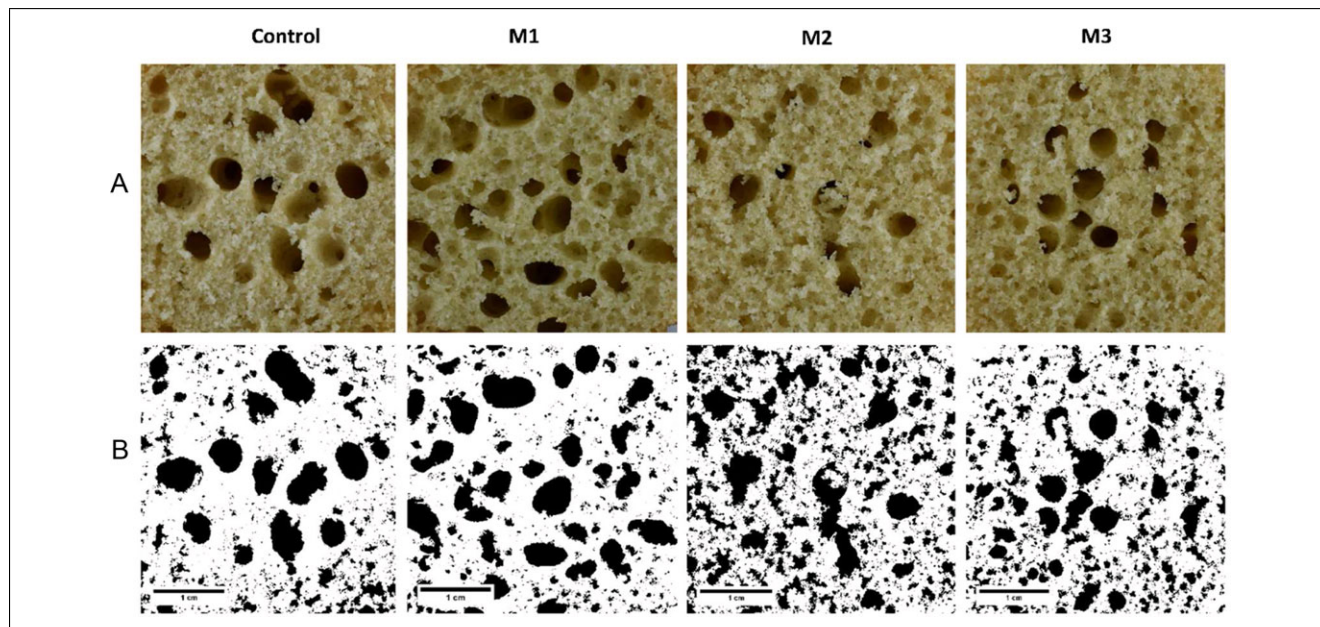


Figure 4—Crumbs of the muffin samples containing commercial margarine (Control), high oleic sunflower oil (M1), and optimized oleogels obtained from O-1 (M2) and O-2 (M3) preparation conditions. (A) The transversal section selected for the image analysis. (B) The corresponding binarized images.

value was higher than three when the M2 was compared with the Control, only color differences between both muffins would be appreciated by the human eye and could affect or not the acceptability of M2 by consumers. Similar tendencies were observed in muffins crust color (data not shown). Their general aspect could be seen in Figure 2.

No significant differences were found in the weight of all samples. The dimensions of the muffins were measured in order to investigate the effect of oleogels on the spreading characteristics. The Control and M1 samples showed greater height and lower diameter than the muffins containing oleogels. As a result, the spread ratio values of the muffins prepared with oleogels were greater compared to the ones made with HOSO and CM, in average about 1.67 compared with 1.53, respectively. Since one of the critical challenges to replace traditional shortenings used in baked products has been to overcome the loss of the spreadability (Jang et al., 2015), this result means an improvement in muffin quality due to the use of optimized oleogels as fat replacers in this food formulation.

Moisture content of M1 and M2 was found statistically equal to that of the Control, whereas M3 presented a slightly lower value (average reduction of 6.3%). The retention of water is the result of different interactions between water and the remaining ingredients that form the batter. Since all the muffins were produced using the same amount of batter and no statistical differences were found in their determined weights after baking, it was expected that the different type of muffins had comparable water contents. The fact that the technique used to determine the moisture of muffins yielded results, which were significantly different could be attributed to the removal of a lower portion of water from muffins containing oleogels during the assay because of a possible stronger retention of water into the structure of muffins elaborated with oleogels.

The specific volume ( $V_s$ ) was raised by the fat replacement with oleogels, while no significant differences were detected between the Control and M1 values. This result was in agreement with those found in the dimensional characteristics evaluation. Taking

into account that the weight of the different muffins was similar, the effect on the  $V_s$  was determined by geometry features of muffins ( $V_s$  is proportional to  $d^2H$ ). Thus, the increase in the diameter showed by M2 and M3 samples respect to the Control (about 4.0%) had a greater influence on  $V_s$  than the decrease in the height experimented by these samples (about 4.8%). Volume is one of the critical quality attributes of aerated baked products that can affect consumer acceptance. Hence, preventing volume loss can be a technological challenge when replacing solid fats in baking industry (Kim, Lim, Lee, Hwang, & Lee, 2017). In this study, the replacement of CM with the optimized monoglycerides oleogels contributed to improve the specific volume of the muffins.

**Crumb structure.** Transversal section images of muffins are shown in Figure 4A, where it can be seen that the crumb structures presented different appearance. Figure 4B present the corresponding binarized images used for analyses.

Table 5 shows the total number of air cells and the number of air cells grouped by size ranges determined in the crumb of the elaborated muffins. A significant decrease in the total number of air cells was observed in muffins formulated with oleogels compared with those produced by CM and HOSO. This decrease generated a more connected structure, which was in agreement with the results found in the cohesiveness evaluation, as discussed above. It is well known that the higher the spread ratio is, the lower the aeration in the batter. Hence, these results were consistent with dimensional characteristics found in muffins obtained with oleogels and reported in a similar study (Yılmaz & Ögütçü, 2015). In that contribution based on the incorporation of wax oleogels in a cookie formulation, it was shown that the aeration was lower in cookies prepared with the oleogels instead of a commercial bakery shortening.

The air cells size is generally important since it can influence the texture of the final product. In spite of the fact that Control and M1 showed a significant higher number of big air cells than M2 and M3, which could lead to a weaker crumb structure, this effect was not evidenced in the  $H_a$  measurements. However, bigger air cells in the crumb structure would cause a more crumbly product and

**Table 5—Air cell number and distribution in muffins prepared with the different type of evaluated fats.**

Sample	Air cells number/cm <sup>2a</sup>			Total number/cm <sup>2</sup>
	Cell area range (cm <sup>2</sup> )			
	10 <sup>-5</sup> – 5 × 10 <sup>-4</sup>	5.1 × 10 <sup>-4</sup> – 10 <sup>-3</sup>	1.1 × 10 <sup>-3</sup> – 5 × 10 <sup>-1</sup>	
Control	1119.0 (35.6) <sup>b</sup>	103.1 (14.3) <sup>b</sup>	15.2 (3.1) <sup>b</sup>	1237.8 (24.4) <sup>b</sup>
M1	1098.2 (42.1) <sup>bc</sup>	88.8 (1.1) <sup>b</sup>	14.2 (3.0) <sup>b</sup>	1201.3 (46.2) <sup>b</sup>
M2	967.1 (82.9) <sup>cd</sup>	67.1 (1.9) <sup>c</sup>	8.2 (0.6) <sup>c</sup>	1042.4 (81.7) <sup>c</sup>
M3	944.9 (4.2) <sup>d</sup>	60.5 (0.7) <sup>c</sup>	8.1 (0.0) <sup>c</sup>	1013.5 (4.9) <sup>c</sup>

Muffins samples containing commercial margarine (Control), high oleic sunflower oil (M1), and optimized oleogels obtained from O-1 (M2) and O-2 (M3) preparation conditions.  
<sup>a</sup>Area of analyzed image. Means in the same column without a common letter are significantly different ( $P < 0.05$ ). Values in parentheses are standard deviations.

the loss of its typical shape, which are recognized as negative quality features. Likewise, a crumb structure showing these characteristics would facilitate the oil migration towards the muffins crust. This fact was verified through the OL assays where muffins obtained with CM and HOSO presented the highest levels of released oil. A greater proportion of small air cells generating a more homogenous crumb in muffins produced with the MG oleogels was also found in the analysis carried out by Calligaris et al. (2013) in sweet bread products in which palm oil or sunflower oil was replaced with oleogels formulated with these oils and MG. It seems that MG aided in the transformation of large air cells in the liquid oil batter into small, uniform cells, which results in a crumb with a finer texture (Zhou, Faubion, & Walker, 2011).

Based on the aforementioned results, it was possible to obtain muffins having similar physicochemical properties independently of the used optimized oleogel, which implies that the preference to use one over another will be given by practical considerations.

## Conclusions

Among the most important results, it was found that by modifying the concentration of Myverol and the cooling temperature, it was possible to change significantly the rheological behavior and the textural properties of oleogels giving rise to the development of a wide range of fat materials with different functionality. The optimization process allowed us to obtain oleogels with high oil binding capacity and with elastic modulus and hardness values very close to the ones of the commercial margarine.

Replacement of commercial margarine with the optimized oleogels in muffin formulation led to the obtention of products with a greater spreadability and a higher specific volume, which represent a desired aspect from an industrial and commercial point of view. Moreover, similar hardness values and more connected and homogeneous crumb structures were reached. Likewise, these products showed a reduction of oil migration of around 50% respect to the Control muffin after 10 days of storage, which indicated that these oleogels can be used satisfactorily to decrease the oil loss in this sweet baked product. In general, fat replacement with the optimized monoglycerides oleogels had a positive impact on the quality and the nutritional profile of elaborated muffins.

In a further study, sensory evaluation and long-term stability analysis will be performed in order to determine if these healthier baked products exhibit consumer preferences and shelf lives similar or better than products containing fats currently commercialized. As well as, a more in-depth research will be conducted in order to explore the complex interactions between all ingredients and their relationship with physical changes occurring during storage in baked products, which incorporated oleogels in their formulation.

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## Conflict of Interest

The authors have no conflict of interest to declare.

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