

Structuring olive oil with ethyl cellulose of different molecular weights: Influence of concentration, viscosity and mixtures

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ABSTRACT

This study explored the impact of ethyl cellulose (EC) of different viscosities (22, 46, and 100 mPa·s) and concentration (8, 10, 12, and 15% w/w) on the color, oil binding capacity (OBC), textural properties (hardness, elasticity, cohesiveness, adhesiveness), rheological properties and oil oxidation of olive oil oleogels. These properties were also measured in oleogels using EC mixtures (50:50 (T), 25:75 (A), and 75:25 (B) ratios (w/w)) by combining 22 and 100 mPa·s EC maintaining a total polymer content of 10% (w/w).

In those oleogels elaborated with a single EC, the findings demonstrated that increasing the EC content significantly enhanced OBC, hardness, and elasticity. The viscosity had less impact, but oleogel hardness decreased with increasing EC viscosity. Oleogels formulated with the highest EC content showed reduced primary and secondary oxidation products, likely by lower oxygen diffusion rate within the sample due to increased viscosity. Oleogels formulated from EC mixtures required less EC polymer to achieve similar textural and rheological properties as those using only 46 mPa·s EC. Additionally, increasing the proportion of 22 mPa·s EC in blends reduced primary and secondary oxidation, indicating this EC's protective role against oleogel oxidation.

Overall, this study demonstrates that blending ECs can produce oleogels with properties comparable to those made with a single EC, but requiring less organogelator. This approach offers significant economic advantages for the food industry.

1. Introduction

In recent years, numerous studies have identified a strong link between the consumption of foods high in saturated fatty acids (SFAs) and trans-fatty acids (TFAs) with chronic diseases (e.g. cardiovascular diseases and type 2 diabetes) as major causes of death (WHO, 2023). In response, several Western countries have enacted policies aimed at reducing or eliminating industrially produced TFAs and SFAs from food products (Nutrition and Food Safety, 2021). Indeed, since 2018, the U.S. Food and Drug Administration (FDA) has banned the use of TFAs in manufactured foods, removing them from the list of ingredients classified as GRAS “Generally Recognize as Safe” (Co and Marangoni, 2018).

To replace these solid fats in foods, several studies have proposed innovative strategies for structuring triacylglycerols using colloidal networks, like those achieved through industrial partial hydrogenation, but with the aid of structuring agents known as oleogelators. These approaches aim to replicate the physical qualities of traditional solid fats, such as hardness, spreadability, elasticity, and mouthfeel, yet face important challenges in fully matching these organoleptic properties

essential to the food industry. The direct method is the simplest and most economical procedure, as it avoids the dehydration operation required when using hydrophilic organogelators. This method entails dispersing the structuring agent in oil at elevated temperatures, with gel formation occurring during cooling and ethyl cellulose (EC)-based oleogels are the most extensively studied (Patel, 2018; Sabet et al., 2023). EC is a linear polysaccharide made of glucose units connected by β -1,4 glycosidic bonds, originating from cellulosic sources (Davidovich-Pinhas et al., 2014). The EC oleogels are physically cross-linked via hydrogen bonds with their properties influenced by factors such as molecular weight, polymer concentration, and the degree of oil unsaturation (Ahmadi et al., 2020).

Commercial EC is available in viscosities ranging from 4 to 300 mPa·s, with higher viscosities associated with increased polymer chain length and molecular weight (M_w), typically from 20 to 220 kDa. Studies indicate that higher M_w of EC strengthens oleogels by creating additional bonding zones, due to its capacity to create a higher number of junction zones with other polymer strands (Davidovich-Pinhas et al., 2016; Ahmadi et al., 2020). Other important characteristics of EC,

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making it an organogelator of interest for the food industry, are those related to thermoreversible transitions during heating, such as glass transition point (T_g : 120-135°C), melting point, (T_m : 165-173°C), as well as during cooling, crystallization (185°C) and vitrification (120°C). However, due to the high temperatures required to structure oils with EC, it is essential to consider the oxidation and stability of vegetable oils, as these factors impact the final quality of EC-based oleogels. Several options are available, including antioxidant compounds that also act as structuring agents or mixtures of organogelators with lower gel temperatures. Indeed, in the last five years, most studies conducted on oleogels EC have also been based on the use of other structural gelling agents. For instance, corn oil oleogels formulated with EC-10 mPa·s, and sorbitan monopalmitate (Liu et al., 2020); sunflower oil oleogels formulated with EC-100 mPa·s and monoacylglycerol (Naeli et al., 2020); canola oil oleogels formulated with EC-20 mPa·s with three types of surfactants as glycerol monostearate, stearyl alcohol/stearic acid, and soybean lecithin (Woern et al., 2021); rapeseed oil oleogels formulated with EC-100 mPa·s and mixture of sunflower wax/monoacylglycerol (Schubert et al., 2022); canola, chia and linseed oleogels formulated with EC of 100 mPa·s and sorbitan monostearate (Millao et al., 2023). Therefore, EC stands out as a highly promising organogelator (i.e. food-grade quality, wide commercial availability, it can be directly dispersed in oil and the EC's structure responds effectively to various functional modifications, making it an ideal choice for the oleogels customization in industrial applications).

However, although extensive studies have been conducted on EC oleogel mixtures, no research has yet explored the use of mixed ECs with different molecular weights (or viscosities). To the best of our knowledge, only two studies on mixtures of cellulose derivatives (i.e. EC, MC, and HPMC) would be related to research focused on EC mixtures (Li et al., 2021; Sánchez et al., 2011). To date, limited attention in the literature has been given to quantifying the impact of temperature on the final oxidation of EC oleogels. Considering the high temperatures required for producing oleogels via the direct method, managing oil oxidation is crucial for the oleogels application in the food industry. In this sense, olive oil is characterized by its notable resistance to oxidation a quality attributed to its fatty acid composition and its high content of natural antioxidants including polyphenols, tocopherols, and carotenoids, that play a crucial role in enhancing the oil's stability. However, despite its greater nutritional fatty acid profile as well as its stability against oxidation very few studies can be found on olive oil gelation (Giacintucci et al., 2018; Lupi et al., 2015). A innovative study was recently published in which ethyl cellulose was used following the emulsion template method to avoid the high temperatures required in the direct method. This highlights the interest in ethyl cellulose as a fat organogelator in the food industry (de Freitas Santos et al., 2024).

This study aims to structure olive oil using EC with different viscosities and mixtures, while examining the effect of these variables on oil binding capacity, texture, rheology, and oxidation level. Considering that within commercial ECs there are significant price differences and that those with intermediate viscosity are the most expensive, a strategy based on mixing ECs of different viscosities, that do not compromise the mechanical properties of the oleogel would be interesting to study.

2. Materials and methods

2.1. Materials and reagents

Refined olive oil with acidity percentage less than 1% from Aceites Abril, S.L. (Ourense, Spain) was purchased in a local supermarket. Ethylcellulose polymers with viscosities of 22, 46 and 100 mPa·s were used, evaluated in 5% toluene:ethanol solution, 80:20 v/v, and ethoxyl content of 48% according to specifications of supplier (Sigma-Aldrich, St. Louis, MO, USA). Isooctane, chloroform, pentahydrate sodium thiosulfate (Panreac, Barcelona, Spain), potassium iodide (VWR Chemicals, Leuven, Belgium), p-anisidine (Thermo Scientific, Branchburg, New

Jersey, USA) and potato starch (Sigma Aldrich, St. Louis, MO, USA) were employed in the experiments related with oil oxidation.

2.2. Preparation of oleogels

The oleogels were elaborated according to the experimental design shown in Fig. 1. This process involved two stages, (1) the characterization of single viscosity oleogels, where 12 oleogels were formulated using 3 different viscosities (22, 46, and 100 mPa·s) and 4 EC concentrations (8, 10, 12, and 15%, w/w). Subsequently, (2) oleogels at 10% w/w EC content made from mixtures of EC with viscosities of 22 and 100 mPa·s, were characterized, using 3 different proportional mixtures: Mixture A consists of 25% 22 mPa·s EC and 75% 100 mPa·s EC (25:75 ratio). Mixture B follows the same order but with a 75:25 ratio, while Mixture T has a balanced 50:50 ratio (Fig. 1). These ratios were arbitrarily selected, while proportions of Mixture T were calculated employing equation [1], which establishes a weighted logarithmic relationship between the viscosities of the components, μ_1 (22 mPa·s) and μ_2 (100 mPa·s) to obtain a theoretical viscosity of 46 mPa·s (μ) as a function of the proportions of each EC.

$$\ln(\mu) = x_1 \ln(\mu_1) + x_2 \ln(\mu_2) \quad (1)$$

The resulting ratio was 50:50, being this blend the most adequate to compare the obtained results with those corresponding to the single EC-46 mPa·s oleogel

For the preparation of olive oil oleogels, with EC as structuring agent, the direct method described by Zetzl et al. (2012) with slight modifications was followed. Briefly, 40 g of olive oil was weighed into an Erlenmeyer flask, covered with aluminium foil to minimize heat loss and exposure to air and light. The flask was placed on a heating plate with constant stirring (300 rpm) and heated to 180°C for 10 min, monitoring the temperature with a thermocouple. Subsequently, the corresponding mass of single or blends of ECs was added at the selected content (and ratios for each mixture). To ensure full ECs solubilization, the temperature (180°C) was maintained for 1 h under constant stirring from the start of oil heating.

Without cooling, the EC-oil mixture was transferred to an aluminium-coated silicone cuvette, dosing 5 g of sample into each compartment. Samples were cooled at room temperature for 20 min, covered with polyethylene film to avoid exposure to any contaminant, and then stored at 4°C for 24 h before analysis.

2.3. Oleogel characterization

2.3.1. Color

Colour parameters [CIELAB trichromatic coordinates: lightness, (L^*); redness, (a^*); yellowness, (b^*)] of mixtures and single viscosity EC oleogels were measured by a portable colorimeter (Konica Minolta CR-400, Osaka, Japan). Before the measurements, the colorimeter was adjusted using a white ceramic tile. The colour measures were done in five different points per oleogel and the average value of the six replicates was calculated.

2.3.2. Oil binding capacity

Oil binding capacity of oleogels was determined according to Morales et al. (2023), with minor modifications. Oleogels (1 g) were introduced in Eppendorf tubes previously weighed. The tubes were centrifuged (HWLAB, HW12, Shiley, NW, USA) at 13,500g for 25 min at 20°C. Supernatant oil was removed with a Pasteur pipette after centrifugation, and the Eppendorf tubes were weighed again. The OBC (%) was calculated according to equation [2]:

$$OBC (\%) = \left(\frac{m_2 - m}{m_1 - m} \right) 100 \quad (2)$$

where m (g) is the weight of the empty Eppendorf tube, m_1 (g) and m_2 (g)

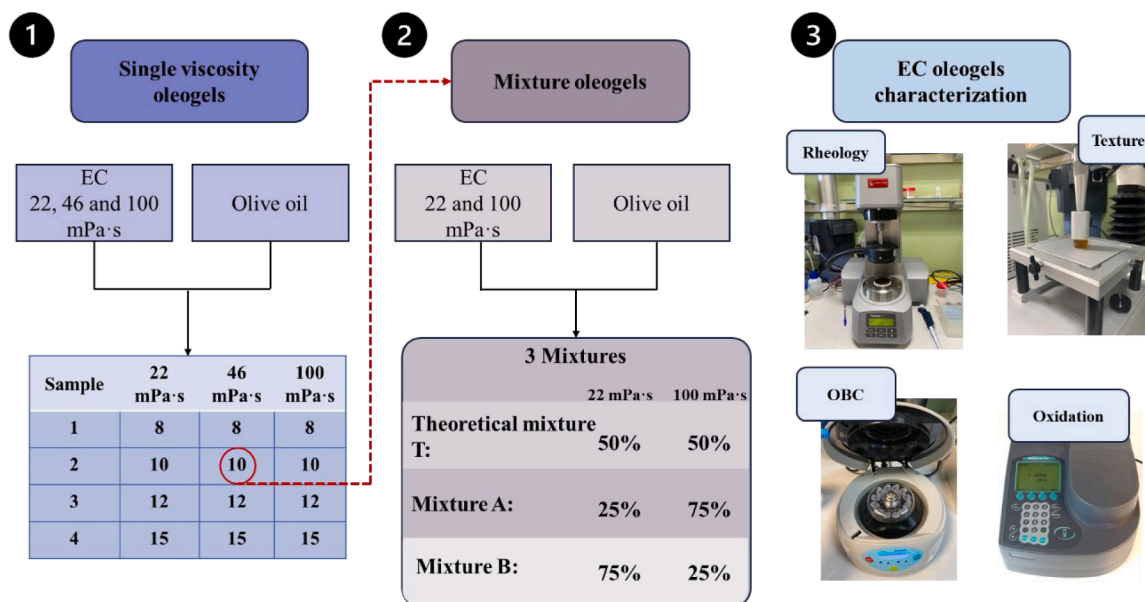


Fig. 1. Scheme of the experimental design and methodology employed for the elaboration and characterization of EC oleogels.

are the weight of the Eppendorf tube with oleogel before and after centrifugation, respectively.

2.3.3. Rheological characterization

The rheological characterization of oleogels was performed with a stress-controlled rheometer (Physica MCR 301, Anton Paar, Austria) with plate-plate geometry (diameter 50 mm) and a gap of 1.0 mm at constant temperature of 25°C, according to Lama et al. (2024) with the modification that frequency sweep was performed up to 100 Hz. Briefly, the linear viscoelasticity range (LVR) was determined with a strain sweep from 0.01 to 10% at constant frequency of 1 Hz. After, a frequency sweep was performed from 0.1 to 100 Hz at 0.1% of strain (within LVR) to determine the viscoelasticity of samples.

2.3.4. Textural properties

Oleogels (19 mm diameter and 9 mm height) were compressed in a texturometer (TA.XT Plus, Stable Micro Systems, Surrey, UK) fitted with a cylindrical probe of 25 mm diameter (SMS P/25). Three (minimum) sample replicates were analyzed for each oleogel batch. Tests were performed with a 50% compression, being the remaining parameters based on Qiu et al (2023). Initial force applied was 0.3 N, and the pre-test, test and post-test speeds were fixed at 5 mm/s.

2.3.5. Oxidation degree

Determination of the oleogel oxidation degree involved extracting oil from the oleogel by centrifugation at 9,600g for 20 min. Primary oxidation was quantified by the determination of the peroxides index (PV) and of conjugated dienes and trienes, meanwhile secondary oxidation was determined as the *p*-anisidine value. PV was determined according to the method CD-8b90 proposed by the American Oil Chemists' Society (AOCS, 2017a), with some modifications. Briefly, extracted oil from oleogel samples (0.3-0.5 g) was dissolved in chloroform, adding afterwards glacial acetic acid, water, potassium iodide and potato starch (this latter as indicator, giving to the solution a blue coloration). Titration with sodium thiosulfate until the solution turned transparent allowed to calculate the PV in meqO₂·kg⁻¹. Conjugated dienes (CD) and trienes (CT) were determined following ISO 3656:2011 with slight modifications (ISO, 2011). Oil samples were weighed (0.01-0.03 g) in a volumetric flask (25 mL) and level up with isoctane. The flask was introduced in an ultrasonic bath for 5 min and the resulting solution was poured into quartz cuvettes and measured for

absorbance (233 and 268 nm for CD and CT, respectively) in a spectrophotometer (Genesys 10 UV, Thermo Spectronic, Menlo Park, CA, USA). The *p*-anisidine test was performed based on the method CD 18-90 of the AOCS (2017b) with some modifications. Briefly, 0.2 mL of anisidine solution in glacial acetic acid was added to 1 mL of oil dissolved in isoctane (same procedure as the previous method) and to 1 mL of isoctane (blank), being homogenized in a vortex (Velp Scientifica, ZX3, Usmate, Italy) for 2 s, and left in darkness for 10 min. Afterwards, the absorbance of the samples was measured in quartz cuvettes at 350 nm.

2.4. Statistical analysis

The effect of EC concentration and viscosity on oil binding capacity, textural, rheological parameters and oxidation values was examined using an analysis of variance (ANOVA) using the General Linear Model (GLM), for all dependent variables considered in the study (OBC, textural parameters and oxidation indices). The fixed effect of viscosity and EC concentration and their interaction were included in the model. The least squares mean (LSM) were separated using Duncan's *t*-test. All statistical tests of LSM were performed for a significance level $\alpha < 0.05$. Correlations between independent and dependent variables ($P < 0.05$) were determined by correlation analyses using Pearson's linear correlation coefficient with a minimum of $n = 35$. All statistical analysis was performed using the software IBM SPSS Statistics 29 (IBM Corp, Armonk, NY, USA).

3. Results and discussion

3.1. Color and oil binding capacity of oleogels

Colour is a key parameter in the sensory evaluation of foods and is often examined in studies focused on replacing SFAs, as this substitution usually alters the final colour, impacting consumer acceptance. Furthermore, the oleogels oxidation can be monitored through color changes that may occur during processing, especially at elevated temperatures (Silva et al., 2022).

Colour parameters were measured for both single viscosity and blends of EC oleogels (Table 1). The lightness (L^*), ranged from 34.9 ± 2.4 to 47.6 ± 4.5 . For oleogels formulated with EC of 22 mPa·s, an increase in EC concentration led to darker oleogels, a trend that was also observed for oleogels containing 100 mPa·s EC. Conversely, oleogels

Table 1
Colour coordinates for single viscosity oleogels and blends of ECs of different molecular weights.

| Viscosity (mPa-s) | EC content (% w/w) | L* | a* | b* |
|--------------------------------------|--------------------|---------------------------|-----------------------------|-----------------------------|
| Single oleogels | | | | |
| 22 | 8 | 46.37 ± 2.71 ^b | -0.57 ± 0.66 ^a | 26.39 ± 2.42 ^a |
| | 10 | 43.41 ± 2.88 ^b | 3.01 ± 1.90 ^b | 28.27 ± 2.57 ^a |
| | 12 | 44.49 ± 3.53 ^b | -0.69 ± 0.89 ^a | 26.51 ± 4.52 ^a |
| | 15 | 34.92 ± 2.37 ^a | 5.08 ± 0.79 ^c | 25.44 ± 5.19 ^a |
| 46 | 8 | 41.57 ± 2.85 ^A | -0.37 ± 0.24 ^{B,C} | 25.18 ± 3.54 ^{A,B} |
| | 10 | 44.15 ± 1.89 ^B | -2.78 ± 0.14 ^A | 25.76 ± 5.02 ^A |
| | 12 | 44.99 ± 2.17 ^B | -1.04 ± 1.21 ^B | 27.19 ± 3.22 ^B |
| | 15 | 47.63 ± 4.48 ^B | 0.76 ± 0.49 ^C | 34.12 ± 4.54 ^C |
| 100 | 8 | 42.36 ± 1.11 ^Y | 0.08 ± 0.68 ^X | 25.68 ± 1.85 ^Y |
| | 10 | 38.62 ± 3.32 ^X | 3.57 ± 1.70 ^Y | 25.59 ± 3.63 ^Y |
| | 12 | 35.92 ± 2.37 ^X | 6.90 ± 1.01 ^Z | 21.85 ± 3.20 ^{X,Y} |
| | 15 | 35.94 ± 3.11 ^X | 5.22 ± 2.02 ^{Y,Z} | 19.41 ± 7.36 ^X |
| Mixture oleogels (10% w/w EC) | | | | |
| A | | 40.23 ± 0.54 ^b | 4.37 ± 0.66 ^a | 28.19 ± 1.36 ^b |
| B | | 33.25 ± 1.22 ^a | 4.95 ± 0.60 ^{a,b} | 18.83 ± 2.34 ^a |
| T | | 32.56 ± 1.27 ^a | 5.66 ± 0.81 ^b | 20.69 ± 1.08 ^a |

Different superscripts (a, b, c, for 22 mPa-s and mixture oleogels, A, B, C for 46 mPa-s, and X, Y, Z for 100 mPa-s) indicate significant differences ($P < 0.05$) among mean values at different EC concentration according to Duncan's test.

with 46 mPa-s EC exhibited the opposite trend, becoming lighter as the concentration increased. Concerning redness (a^*) and yellowness (b^*) parameters, no significant ($P > 0.05$) trends were observed in relation to EC content. However, oleogels with viscosities of 22 and 46 mPa-s exhibited lower a^* values (1.71 vs. -0.9 for 22 and 46 mPa-s, respectively) and higher b^* values (26.7 vs. 28.1 for 22 and 46 mPa-s, respectively), creating a more pronounced yellow hue. In the blends, T and B mixtures appeared darker than mixture A (32.9 vs 40.2; $P < 0.05$ for T and B and for A, respectively). Concerning a^* and b^* mixture T showed a higher redness intensity compared to mixtures A and B, while mixture A exhibited a stronger yellowness index than mixtures B and T. When comparing the color of mixture T oleogel with the simple one of 46 mPa-s formulated at 10%, it is observed that the mixture T was darker and less yellowish.

The ability to maintain a liquid phase within a solid matrix property (OBC) is attributed to the phenomenon of oil packing in the three-dimensional network of the structuring agent. High values of this parameter are desirable because it is related to the quality and functionality of the oleogel. Poor OBC values give as result unpleasant appearances and significant nutritional losses in foods. (Liu et al., 2020). Fig. 2 shows the OBC of the single viscosity oleogels at different EC concentrations. The OBC (%) values ranged from 71.8-99.4, 82.3-99.4, and 84.4-99.8 for the oleogels with EC viscosities of 22, 46 and 100 mPa-s, respectively. The maximum values (> 97.0) were obtained at the highest EC content tested (15% w/w), with no significant ($P > 0.05$)

influence of EC viscosity (Fig. 2A). On the contrary, significant differences ($P < 0.05$) were found regarding to concentration effect (Fig. 2B). A positive correlation between OBC and EC concentration was observed ($r = 0.875$, $P < 0.01$, $n = 51$). For example, differences in the OBC value of up to 25% were found increasing the EC concentration from 8 to 15% w/w. These results agreed with previously reported data in which higher OBC values have been found at high EC concentrations (Shao, et al., 2023; Ursachi, et al., 2023; Yang, et al., 2024). For instance, Shao et al. (2023) obtained OBC values of 99.4% in peanut oil oleogels formulated with 12% w/w EC (50 mPa-s). Similarly, Yang, et al. (2024) reported the same effect for coconut oil oleogels made with EC (100 mPa-s) at concentrations of 10, 15 and 20% w/w, with OBC value of 99% at the highest EC concentration. However, the concentration of the structuring agent is not the only factor influencing OBC. The type of oil, particularly its degree of unsaturation, along with the presence of surfactants, also play crucial roles (Davidovich-Pinhas et al., 2016). This finding was corroborated by Millao et al. (2023) who studied oleogels elaborated from various oilseed (canola, chia and linseed) at three EC concentrations (5, 7.5 and 10% w/w) and three surfactant concentrations (0, 1, and 2% w/w). These authors observed that chia and linseed oleogels exhibited higher OBC values of 96.0 and 97.4%, respectively. Moreover, a strong positive correlation ($r = 0.994$) was reported between the polyunsaturated fatty acid content and the OBC parameter. Specifically, the linolenic acid (C18:3) content in chia and linseed, which exceeds 50% of total fatty acids, was significantly higher than in canola,

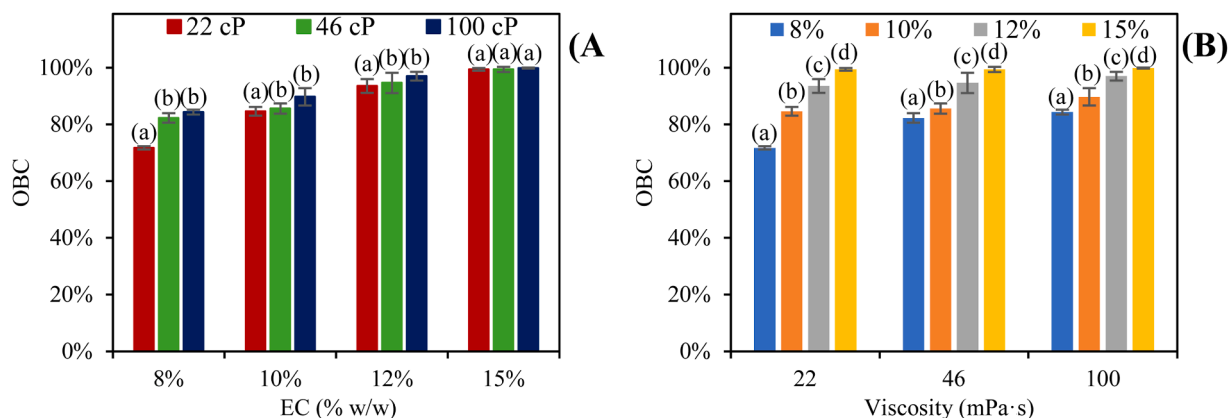


Fig. 2. Oil binding capacity (OBC) of single viscosity oleogels. (A) Effect of EC concentration (w/w); (B) Effect of EC viscosity. Different letters (a, b, c) indicate significant differences ($P < 0.05$) among mean values according to Duncan's post hoc test.

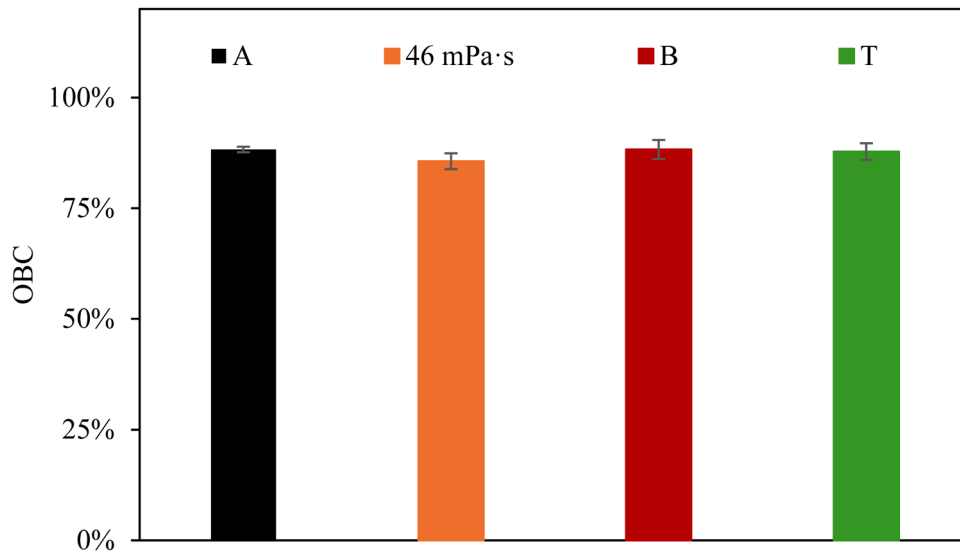
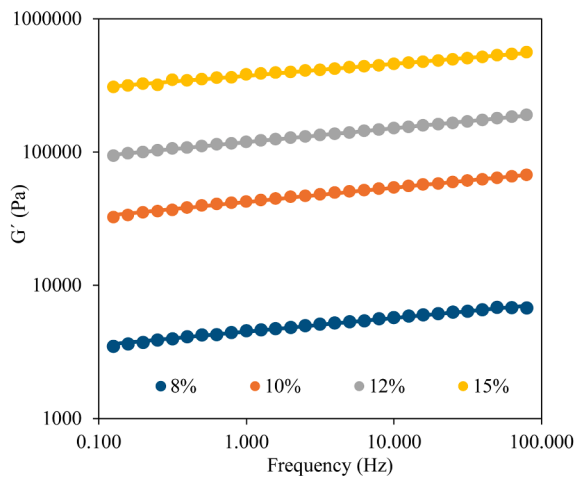


Fig. 3. Oil binding capacity (OBC) of oleogels of different viscosity EC mixtures (A, B, and T) compared to single viscosity oleogel at 10% w/w of EC.

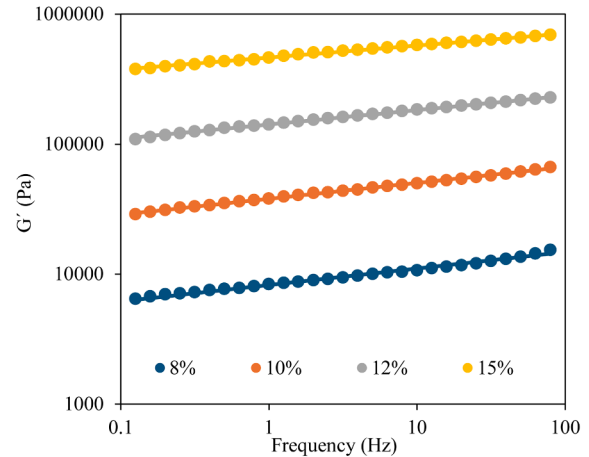
explaining the OBC values. Supporting this hypothesis, chia and linseed oils contain linolenic acid levels (C18:3) that are over 97% higher than those in olive oil (Aparicio and Harwood, 2013), which may further

explain the differences in OBC values with respect to our study.

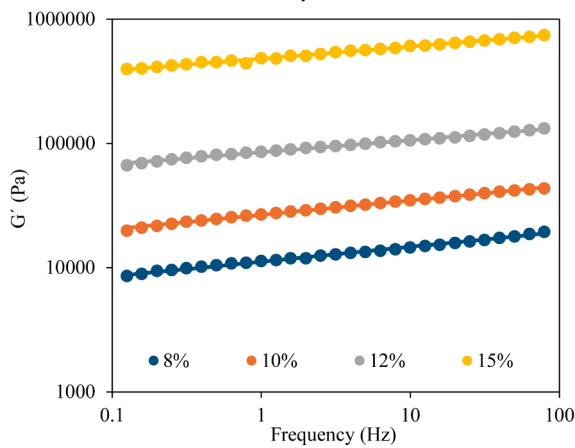
Fig. 3 shows the OBC values obtained for oleogels made with EC mixtures of different viscosities. The samples had an average OBC value



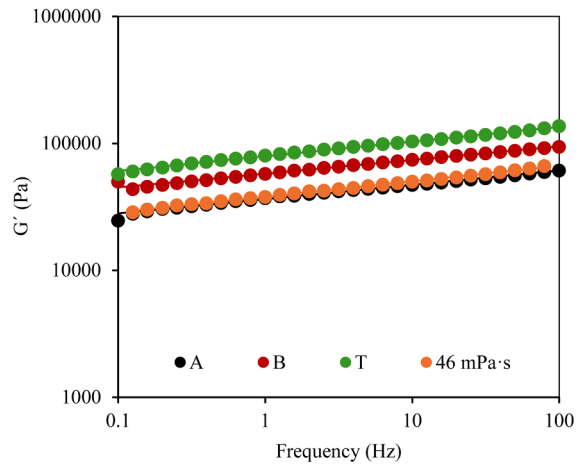
A)



B)



C)



D)

Fig. 4. Elastic modulus, G' , trend with angular frequency of oleogels elaborated with single EC of different viscosity (mPa·s): 22 (A), 46 (B) and 100 (C), at different EC concentration (% w/w) and mixtures, A, B, T and single viscosity oleogel of 46 mPa·s (D).

of $87.5 \pm 2.0\%$, and no significant differences ($P > 0.05$) were observed. Thus, no effect of the EC mixture of different viscosities on the OBC of the oleogels was observed. This finding corroborated data about single oleogel systems, where no trends between OBC and EC viscosity were observed (Fig. 2).

3.2. Rheological properties of oleogels

Figs 4 A, B and C show the elastic moduli (G') of oleogels made with a single viscosity at different EC content. As expected, the frequency sweeps showed a predominantly elastic (solid) behavior in tested oleogels, with the elastic modulus (G') higher than the viscous modulus (G'') in the whole frequency range studied. The damping factors ($\tan \delta = G''/G'$) ranged from 0.11 to 0.27 across all samples, meaning well-structured gels, regardless of EC concentration and viscosity, but no clear trend was observed for the damping factor with both variables. This behavior was also observed in a 16% w/w EC soybean oil oleogel with a viscosity range of 45–55 mPa·s, used by Qiu et al. (2023).

Significant differences ($P < 0.05$) were found between G' of oleogels with different EC content. Gels formulated with 15% w/w EC exhibited the highest G' values, while those with 8% w/w EC showed the lowest. This finding is consistent with results reported by Shao et al. (2023), a significant increase in G' of oleogels as EC concentration increased from 6 to 12% w/w. Similarly, Yang et al. (2024) working with oleogels of coconut oil determined a rise in both dynamic moduli (G' and G'') increasing the EC concentration (from 10 to 20% w/w). These results were explained by the high structural stability of the interfacial network formed by the crystals and the oil. Indeed, a higher EC concentration offers more hydrogen bonding sites, increasing the likelihood of junction zones. In line with this, Zetzel et al., 2014, reported a negative linear correlation between pore diameter and the increase in EC concentration in the range of 10–14% w/w. Thus, as the number of crystals increases, the strength of the network structure increases, forming a gel with high mechanical stability, confirmed by OBC and hardness values. Supporting this, Table 2 shows the linear modelling fit parameters of G' , where the slope (n) varied in a narrow range of 0.089 and 0.127, decreasing in magnitude with increasing EC content. All batches showed a strong positive correlation between EC concentration and G' values ($r = 0.93$, $P < 0.01$, $n = 116$). Furthermore, for oleogels formulated with 8% w/w EC content a positive correlation was found between G' and EC viscosity ($r = 0.78$, $P < 0.01$, $n = 87$). The same trend was found for oleogels formulated at 15% w/w EC concentration ($r = 0.44$, $P < 0.01$, $n = 87$). The increase of G' with EC viscosity of oleogels has been previously

Table 2

Fitting parameters of the elastic modulus, G' , model, for single viscosity oleogels and for oleogels elaborated from mixtures of EC of different viscosity.

| Viscosity (mPa·s) | EC (% w/w) | Model G' (kPa): $G' = k f^n$ | | |
|--------------------------------------|------------|--------------------------------|------|-------|
| | | k (G' at 1 Hz) | n | R^2 |
| Single oleogels | | | | |
| 22 | 8 | 4.5 | 0.10 | 0.995 |
| | 10 | 42.2 | 0.11 | 0.998 |
| | 12 | 118.7 | 0.10 | 0.999 |
| | 15 | 374.2 | 0.09 | 0.996 |
| 46 | 8 | 8.2 | 0.13 | 0.988 |
| | 10 | 37.9 | 0.12 | 0.998 |
| | 12 | 141.6 | 0.11 | 0.998 |
| | 15 | 463.5 | 0.09 | 0.997 |
| 100 | 8 | 11.2 | 0.12 | 0.996 |
| | 10 | 26.6 | 0.11 | 0.998 |
| | 12 | 84.9 | 0.10 | 0.997 |
| | 15 | 481.1 | 0.10 | 0.995 |
| Mixture oleogels (10% w/w EC) | | | | |
| A | | 36.4 | 0.11 | 0.994 |
| B | | 57.7 | 0.10 | 0.994 |
| T | | 79.2 | 0.12 | 0.997 |

reported (Davidovich-Pinhas et al., 2015; Sánchez et al., 2011; Zetzel et al., 2012). This proportional increase was not observed in oleogels formulated at 10 and 12% w/w EC concentration.

The Fig. 4 D shows the G' of oleogels made from EC mixtures of different viscosities at 10% w/w. Similarly to single oleogels, it was observed that in all the blends G' was predominant versus G'' . Significant differences ($P < 0.05$) were observed between G' of blends, where the maximum G' was observed in the T mixture, with an average value of 79.2 ± 2.0 Pa (at 1 Hz). The lowest value, similar to that measured in the single viscosity oleogel of 46 mPa·s, belongs to A mixture (36.4 ± 0.6 Pa, at 1 Hz). An intermediate value was obtained for mixture B (57.7 ± 1.7 Pa, at 1 Hz). Damping factor values of oleogels formed with mixture of ECs were constant (0.11–0.12) and like the value (0.12) of oleogel with only EC-46 mPa·s at the same content. These results seem to indicate that elastic behaviour was enhanced with the presence of high proportions of EC-100 mPa·s (mixtures B and T). In line with this, Wang et al. (2023), observed that in oleogels made from mixtures of EC and beeswax (BW), at the same EC/BW mass ratio resulted in a significantly higher G' than that obtained with an oleogel made only with EC (at the same total organogelator content) suggesting that the use of two gelling agents enhanced a synergistic effect.

3.3. Textural properties of oleogels

The oleogels hardness varied between 2.9 and 66.7 N, with significant differences ($P < 0.05$) for both studied traits, viscosity and content of EC (Fig. 5A). Concerning viscosity, the oleogels made with EC of 100 mPa·s showed a lower hardness, up to 34% to those oleogels elaborated with EC of lower viscosity (22 and 46 mPa·s). This was only achieved for the highest concentrations (12–15% w/w), as no significant differences were found for lower concentrations (8–10% w/w) because of the EC viscosity. This finding was unexpected, as a polymer with a higher molecular weight typically forms more interactions, which would be expected to increase hardness. In line with this, Zetzel et al. (2012) observed that oleogels made with higher viscosity EC (100 mPa·s) exhibited significantly greater hardness (9 N) compared to oleogels with viscosities of 10 and 45 mPa·s, which showed much lower hardness values (1–2 N). However, these authors and others (Gravelle et al., 2014; Zetzel et al., 2012) also reported that differences could be explained because larger polymers may present solubility problems. In line with this, it has also been reported that the preparation process of oleogels plays a crucial role in their textural properties, particularly the cooling rate. The cooling process is critical for the formation of the polymer network through hydrogen bonds between polymer strands. The physical entrapment of oil within the polymer strands occurs during the cooling phase, leading to the formation of a gel-like structure. A slower cooling rate allows more time for polymer reorganization, resulting in the formation of more crystals or bonding zones, which ultimately leads to stronger gels (Davidovich-Pinhas et al., 2015).

Similarly, significant differences ($P < 0.05$) were observed with EC concentration, finding a strong correlation between EC content and the oleogel hardness ($r = 0.935$, $P < 0.01$, $n = 48$). This finding agreed with those reported by several authors (Davidovich-Pinhas et al., 2015; Shao et al., 2023; Yang et al., 2024; Zetzel et al., 2012). For instance, Shao et al. (2023) reported an increase in hardness in oleogels with EC content (from 6 to 12% w/w) across different oil types. Similarly, Yang et al. (2024) observed this trend, noting that oleogels of coconut oil made with different EC content (from 10 to 20% w/w) displayed particularly high hardness levels, exceeding 5,000 N.

Concerning the other textural parameters, cohesiveness (Fig. 5B) did not exhibit a clear trend with EC content. In contrast, elasticity (Fig. 5C) showed significant differences ($P < 0.05$) with EC content, following a negative correlation ($r = -0.469$, $P < 0.01$, $n = 48$). Finally, adhesiveness (Fig. 5D) increased with EC viscosity, giving a positive correlation ($r = 0.641$, $P < 0.01$, $n = 48$). Significant differences ($P < 0.05$) in adhesiveness were also noted between the 8, 10, and 12% w/w EC oleogels in

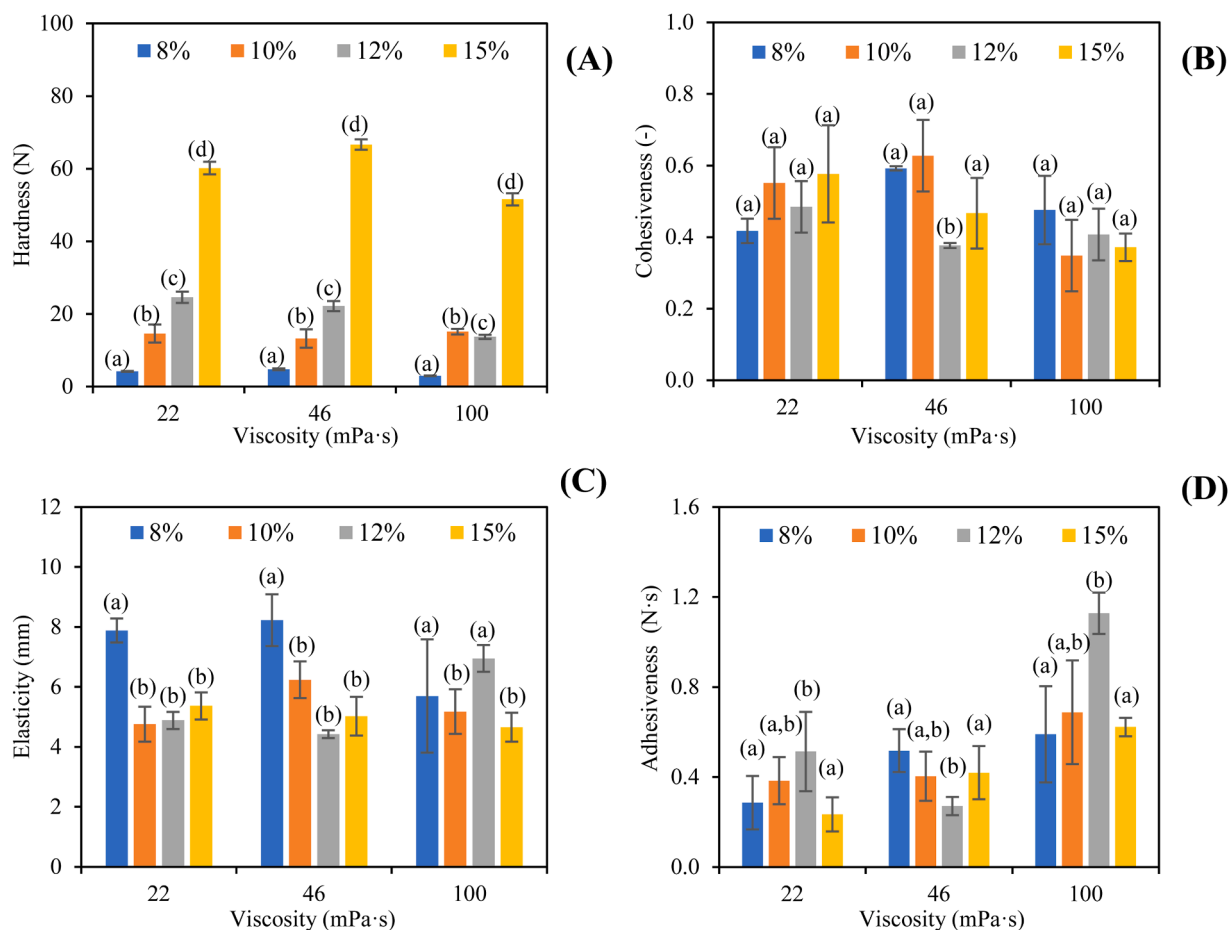


Fig. 5. Texture properties of tested oleogels. Effect of EC Content (% w/w) and viscosity on hardness (A), cohesiveness (B), elasticity (C) and adhesiveness (D). Different letters indicate significant differences ($P < 0.05$) among mean values according to Duncan's post hoc test.

comparison to the highest concentrated samples.

Fig. 6 shows the textural parameters of the oleogels elaborated from mixtures of ECs, compared to the oleogel formulated with only EC of 46 mPa·s. The hardness (Fig. 6A) varied significantly ($P < 0.05$) among these oleogels. The A mixture exhibited the lowest hardness, comparable to the single viscosity oleogel. In contrast, the B and T mixtures showed higher hardness with an increase of up to 123%. This result is relevant since suggests that the blending of EC with different viscosity can achieve hardness comparable to oleogels elaborated only with a single viscosity, but employing lower EC amounts. Additionally, formulations with less than 50% w/w EC of 22 mPa·s decreased both cohesiveness (Fig. 6B) and adhesiveness (Fig. 6D). Main conclusion are that oleogels with higher hardness (at the same EC content) could be produced without significant changes in cohesiveness, elasticity, or adhesiveness. In contrast, the blend with a higher proportion of low-viscosity EC led to a reduction in all textural parameters. When structuring agents of different natures are combined to modify the texture of oleogels, the results are often controversial. For example, Wang et al. (2023) did not observe synergistic effects on the textural parameters of oleogels formulated with EC and vegetable waxes, meanwhile Schubert et al. (2022), demonstrated significantly higher hardness in oleogels formulated with 7.5% w/w EC and 2.5% w/w monoacylglycerides compared to other blends.

3.4. Oxidative stability of oleogels

Oil oxidation is a key parameter for defining oleogel quality. The peroxide value (PV), indicative of primary oxidation, ranged from 1.6 to 15.8 meqO₂·kg⁻¹. No significant differences ($P > 0.05$) were observed

with the effect of concentration and viscosity of EC among studied batches (Table 3). It was observed that all tested oleogels exhibited significantly lower levels of oxidation compared to the blank sample ($P < 0.05$), with oxidation reductions reaching up to 97%. These results could suggest that the presence of EC inhibits primary oxidation, probably due to polymer-solvent interactions that reduce the diffusion of oxygen and free radicals' presence (Martinović et al., 2020). Similar findings have been reported in previous studies. Fu et al. (2020) observed nonlinear oxidation behaviour in EC-based oleogels with soybean oil. Their tests showed that PV increased at low EC content (w/w) below 3% but decreased from 3 to 6%. A similar trend, but in another range was observed by Millao et al. (2023) who observed a decrease in PV for oleogels containing 5 to 7.5% w/w EC, followed by a subsequent increase as the concentration increased to 10% w/w. This behaviour may be attributed to the oxidation of iodine ions by the ethoxyl groups of ECs under acidic conditions. In any case, for oleogels tested, PV values were within the limits established by European regulations (EU, 2011).

The CD values ranged from 10.1 to 19.3 $\mu\text{mol}\cdot\text{g}^{-1}$ and significant differences ($P < 0.05$) were observed in relation to viscosity and content of EC (Table 3). However, no clear trend emerged concerning the viscosity effect. On the contrary, CD values decreased as EC content increased. This suggests that at high EC content the formation of conjugated products is less pronounced, indicating greater oxidative stability in the tested oleogels in agreement with PV results.

Overall, our findings reinforce the idea that EC acts as a physical barrier in primary oxidation, which could have important implications in the food industry to extend the shelf life of oleogel-based products. However, to confirm this protective effect, it is necessary to assess

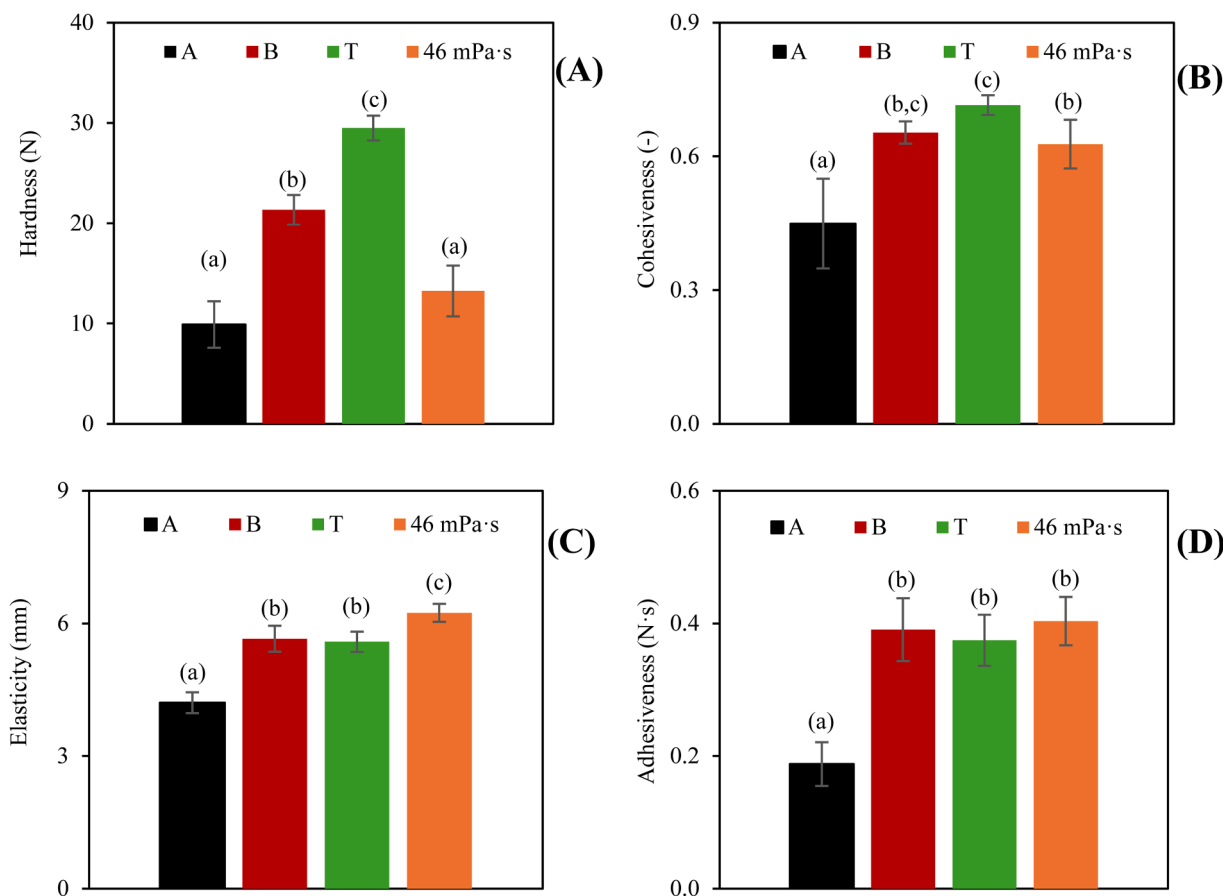


Fig. 6. Texture properties of tested oleogels. Effect of mixtures on hardness (A), cohesiveness (B), elasticity (C) and adhesiveness (D). Different letters indicate significant differences ($P < 0.05$) among mean values according to Duncan's post hoc test.

Table 3

Values of peroxide value, PV ($\text{meqO}_2\cdot\text{kg}^{-1}$), anisidine value (AV) and conjugated dienes (CD) of tested oleogels of single viscosity systems.

| Viscosity (mPa·s) | EC (% w/w) | PV | CD | AV |
|-------------------|------------|------------------|------------------|-------------------|
| Blank | 0 | 53.9 ± 2.1^b | 33.3 ± 1.1^d | 345.4 ± 5.8^d |
| 22 | 8 | 1.6 ± 3.8^a | 19.3 ± 0.7^a | 70.5 ± 3.6^a |
| | 10 | 6.2 ± 3.2^a | 11.7 ± 0.6^b | 65.1 ± 3.7^a |
| | 12 | 8.6 ± 6.6^a | 10.3 ± 0.1^c | 62.9 ± 2.3^b |
| | 15 | 2.0 ± 1.7^a | 10.1 ± 0.1^c | 30.3 ± 1.4^c |
| 46 | 8 | 15.8 ± 4.5^a | 17.0 ± 1.0^b | 40.6 ± 2.4^a |
| | 10 | 4.4 ± 5.2^a | 18.4 ± 0.7^b | 42.8 ± 2.4^a |
| | 12 | 8.4 ± 3.8^a | 11.4 ± 2.2^c | 30.5 ± 5.7^b |
| | 15 | 1.6 ± 1.5^a | 11.6 ± 0.7^c | 16.7 ± 5.2^c |
| 100 | 8 | 6.6 ± 4.5^a | 13.7 ± 0.1^a | 33.5 ± 0.5^a |
| | 10 | 1.8 ± 0.4^a | 13.1 ± 0.9^b | 31.1 ± 3.9^a |
| | 12 | 4.9 ± 3.8^a | 10.6 ± 0.6^c | 29.5 ± 4.3^b |
| | 15 | 2.1 ± 0.4^a | 10.8 ± 0.6^c | 10.1 ± 2.6^c |

Different superscripts a, b, c indicate significant differences ($P < 0.05$) among mean values at different EC concentrations (%w/w) according to Duncan's test.

secondary oxidation, as low PV could also indicate advanced oxidation since peroxides are decomposing and therefore not detected. Hence, anisidine values (AV) in the range between 10.1 and 70.5 were measured (Table 3). Regarding the effect of viscosity, significant differences ($P < 0.05$) were observed, with a negative correlation between AV and EC viscosity ($r = -0.592$, $P < 0.01$, $n = 37$). This suggests that secondary oxidation decreased moderately as EC viscosity increased. Regarding EC content effect on secondary oxidation, significant decreases in AV values were found as the EC concentration increased, with a strong negative correlation ($r = -0.885$, $P < 0.01$, $n = 37$). These findings are consistent with those reported by Millao et al. (2023), who

reported similar behaviour in oleogels formed with EC-100 mPa·s in the interval content from 5.75 to 10% w/w.

On the other hand, Casal et al. (2010) reported maximum AV of 61 for olive oils subjected to 24 h of frying at 170°C , which are comparable to those obtained for oleogels with EC-22 mPa·s at content below 15% w/w (Table 3). For the other two oleogels batches, AV was reduced at least 50% with the most significant reduction, over 83%, in oleogels formulated with EC-100 mPa·s at 15% w/w. This suggests that one hour of heating can generate oxidation levels in the oleogels like those found in frying oils under extreme conditions. However, it should be noted that in open frying systems, primary oxidation products tend to volatilize, resulting in lower levels of secondary oxidation (Aparicio and Harwood, 2013). Furthermore, the tested oleogels were exposed to higher temperatures (180°C), and small variations at high temperatures lead to more pronounced, nonlinear oxidative degradation

Fig. 7 presents the values of the primary (PV, CD and CT) and secondary (AV) oxidation parameters of the oleogels made from mixtures of ECs of different viscosities. Regarding PV, it was observed that, among the blends, A presented the highest value ($1.6 \text{ meqO}_2\cdot\text{kg}^{-1}$), while the lowest value ($0.8 \text{ meqO}_2\cdot\text{kg}^{-1}$) was found in B. However, oleogel made with single-viscosity EC presented a relevant high degree of oxidation ($4.4 \text{ meqO}_2\cdot\text{kg}^{-1}$). No significant differences were observed among the oleogels from mixtures of ECs ($P > 0.05$), but it was noted that a higher proportion of EC-100 mPa·s in the blends contributed to reduce the PV, suggesting a possible protective effect against primary oxidation. The same effect of EC-100 mPa·s in the mixtures was observed for the primary oxidation measured by CD and CT.

Regarding secondary oxidation (p-anisidine values), A presented the highest value ($31.7 \mu\text{mol}\cdot\text{g}^{-1}$), while B and T showed lower values ($< 26.6 \mu\text{mol}\cdot\text{g}^{-1}$). The oleogel made with single-viscosity EC presented

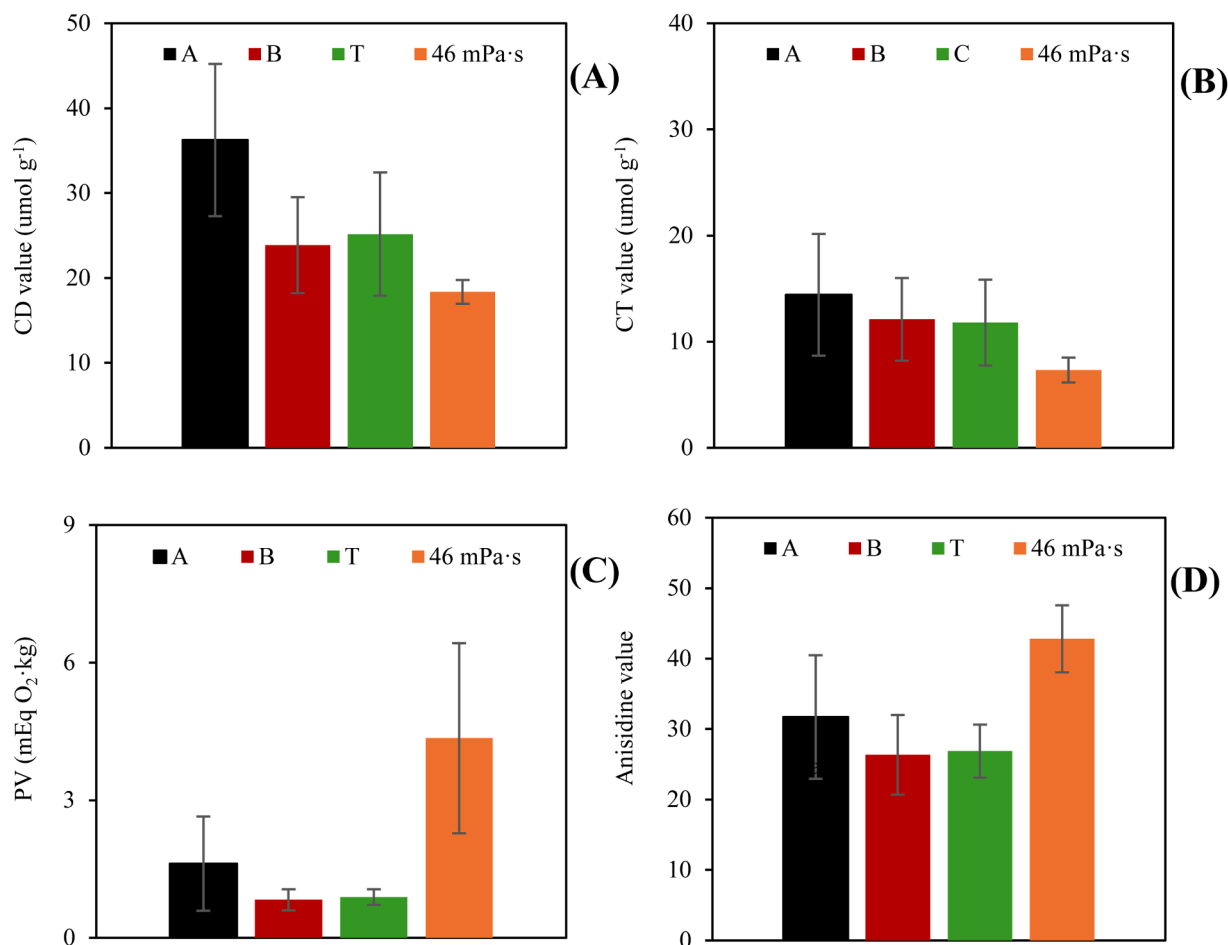


Fig. 7. Oxidation parameters for oleogels mixtures A, B, T and single viscosity oleogel of 46 mPa·s.

significantly higher oxidation values ($P < 0.05$). As with PV, a higher EC content of 22 mPa·s in the mixture contributed to increased secondary oxidation, which reinforces the idea that EC with higher viscosity (100 cP) has a protective effect.

This study demonstrates that mixing 22 cP-EC and 100 cP-EC at a 10% concentration can produce oleogels with rheological, textural, and oil retention properties comparable to those obtained using only 46 cP-EC. While not conducting a comprehensive economic analysis, considering the current prices of 22 cP-EC (\$162.6), 46 cP-EC (\$394), and 100 cP-EC (\$339) (Sigma-Aldrich, 2025), all with a similar degree of substitution or ethoxyl content of 48%, using different mixtures can lead to cost savings of 25.2, 36.39, and 47.61% for A, T, and B blends, respectively.

4. Conclusions

Within single oleogels, the results of this study show that increasing the EC content significantly improves the mechanical properties of oleogels, enhancing OBC, hardness, and elastic character, likely due to a reduction in pore size within the oleogel microstructure. Conversely, oleogel hardness decreased with increasing EC viscosity, possibly due to reduced polymer-solvent interactions related to the solubility limitation of high-viscosity EC. Oleogels with the highest EC content exhibited low primary oxidation products (peroxide values and conjugated dienes) and secondary oxidation (anisidine values), likely due to restricted oxygen diffusion in the oil and increased viscosity.

The study on EC oleogel mixtures found no significant differences in OBC. Oleogels elaborated with single-viscosity EC (46 mPa·s) and mixture A exhibited similar hardness, while blends B and T were

significantly harder and showed increased cohesiveness and adhesiveness but decreased elasticity than single ones. Regarding rheological analysis, the blending EC can replicate the rheological properties of single-EC oleogels with reduced structuring agent requirements.

This study demonstrates that the blending of ECs can be used to create oleogels with similar properties to those produced by a single EC type, while reducing the amount of organogelator needed. On the other hand, using the same EC concentration (i.e. 10%), the blends can give cost savings up to 47%. However, further studies are needed to evaluate EC with other types of oils and across different concentration and viscosity ranges.

Ethical Statement - Studies in humans and animals

Not applicable.

CRediT authorship contribution statement

Claudia Armijo: Writing – original draft, Investigation, Formal analysis. **Leticia Montes:** Investigation, Formal analysis. **Ramón Moreira:** Writing – review & editing, Validation, Methodology, Conceptualization. **Daniel Franco:** Writing – review & editing, Validation, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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