



Rheological properties of emulsion templated oleogels based on xanthan gum and different structuring agents

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ABSTRACT

Oleogel design based on emulsions using food grade polymers is a potential strategy for replacing saturated and *trans* fats. The aim of this paper is the characterization of sunflower oil-based oleogels structured by a non-surface-active polysaccharide, xanthan gum (XG), in combination with different structuring agents, through an emulsion template approach, which consists in the dehydration of the continuous phase of an oil/water emulsion. Four types of molecules with different origins were used: a synthetic one, polysorbate Tween 80, and other three from natural sources: a protein (whey protein, WP), a lipid (soy lecithin, SL) and a polysaccharide (locust bean gum, LBG).

All the emulsions had a high shear thinning character ($s = 0.45$) and a weak gel behaviour ($\tan\delta = 0.2$). Only the LBG emulsions presented significant differences, with higher values of viscosity and viscoelastic moduli. The resulting oleogels showed high gel strength, exhibiting a marked elastic behaviour typical of structured solid systems ($G' > G''$ and $\tan\delta = 0.06$). SL oleogels stood out for presenting the lowest firmness, although internal structure seems to be similar to the rest. Regarding to stability, measurements indicated a great oil binding capacity (90% approx.) after 24 h. It is concluded that obtaining stable and solid-like oleogels with liquid oil using a hydrophilic polymer (XG) combined with different food-grade structuring agents was possible. These systems have great potential in food reformulation, especially for saturated fat substitution.

1. Introduction

There has been concern for years that long-term consumption of saturated fat and especially *trans* fatty acids could cause adverse health effects such as metabolic syndrome, obesity, high risk of cardiovascular disease, type II diabetes (Islam et al., 2019; Wang et al., 2022) and even cognitive disorders (Barnard et al., 2014). For that reason, the World Health Organization has suggested a shift in saturated solid fat consumption to unsaturated liquid oils (World Health Organization, 2018a; World Health Organization, 2018b). Nevertheless, producing solid fats without saturated and *trans* fats has been a big challenge in recent years, as solid fats play critical roles in texture and sensory attributes of foodstuffs (Biguzzi et al., 2014; Drewnowski, 1992; Rios et al., 2014). A promising recent alternative is the technology of vegetable oil oleogelation. Oleogelation is the transformation of liquid oil into a gel-like structure with solid-like properties by using oleogelators agents with

the ability of entrapping liquid oil in a three-dimensional, thermo-reversible gel network (Hinze et al., 1996; Pehlivanoglu et al., 2018).

The most common method of oil gelation is direct gelling, in which a hydrophobic oleogelator agent (commonly waxes, monoacylglycerides or ethyl cellulose) is dispersed in the oil at high temperatures (Martins et al., 2018; Rogers et al., 2009; Patel, 2015). However, legal limitations regarding new ingredients and food additives, the waxy taste and the risk of oil oxidation due to the high temperatures used are major disadvantages when using this method (Abdollahi et al., 2020; Martins et al., 2018). An interesting alternative would be the polymer gelation. Polymers have aroused great concern due to their commercial availability, large production, and lower cost compared to most of the highly purified oleogelators (Meng et al., 2018a). Nevertheless, due the hydrophilic nature they are ineffective in structuring hydrophobic oils, so they must be subjected to indirect methods of oil gelation, such as the emulsion template approach (Romoscanu and Mezzenga, 2006). This

Abbreviations: XG, Xanthan gum; WP, Whey Protein; LBG, locust bean gum; SL, soy lecithin; TW, Tween 80; OL, oil loss; OBC, oil binding capacity.

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multistep approach consists of the preparation of an o/w emulsion with a structuring agent followed by a dehydration process for the elimination of the aqueous phase and subsequent homogenization.

Polysaccharides, because of their hydrophilic nature, can act as thickening agents increasing the viscosity of the aqueous phase and therefore improving the long-term physical stability of the emulsion slowing the movement of the droplets (Zapateiro et al., 2018). Notwithstanding, to obtain kinetically stable emulsion-based oleogels the initial formulation requires a molecule with surface-active properties to disperse and stabilise the oil droplets. Anionic XG is widespread and commercially used non-surface-active polysaccharide in food emulsions to increase viscosity and improve emulsion stability. Combination of XG with other molecules such as amphiphilic polysaccharides, proteins or even synergistic polysaccharide-polysaccharide mixtures has enhanced the stabilization of oil-in-water emulsions for the preparation of oleogels. To this regard, Meng et al. (2018a) investigated the effect of xanthan gum-hydroxypropyl methyl cellulose combination on the physical properties of emulsions and oleogels, obtaining systems with high mechanical strength and great oil binding capacity. Recently, some reports demonstrated the potential of combining XG and proteins (such as gelatin or egg protein) to induce oil structuring, offering promising results in oleogels development (Martins et al., 2018; Abdollahi et al., 2020; Jaber et al., 2020; Patel et al., 2015). The combination of different polysaccharides is also successful in emulsion gelation; it has been found that galactomannans show synergistic interactions with other polysaccharides such as carrageenan and XG (Patel et al., 2014b).

In the current study, sunflower oil/water emulsions were prepared with xanthan gum (XG) as a thickening agent in combination with different types of food-grade structuring agent to obtain oleogels that impart solid-fat functionality to liquid oil. The selected structuring agents were: one of animal nature (whey protein, WP), two of vegetable nature (locust bean gum, LBG and soy lecithin, SL) and one of synthetic nature (polysorbate Tween 80, TW). The effect of the different structuring agents on the rheological properties of emulsions and oleogels and the stability of the oleogels was investigated.

2. Materials and methods

2.1. Materials

High oleic sunflower oil *Capicua* was obtained from Compañía Oléicola SAU (Spain); xanthan gum (XG) (Satiaxane C911, 1500 mPa s viscosity in a 1% aqueous solution at 25 °C) measured by Cargill (France); polysorbate Tween 80® (P1754) from Sigma-Aldrich Chemical Company (St Louis, MO); whey protein from Weider Global Nutrition S.L. (Germany); locust bean gum, LBG (Viscogum® BE, 2900 mPa s viscosity measured by Cargill (France)); and pure soy lecithin, SL, from Diexpa (Spain).

2.2. Preparation of samples

2.2.1. Emulsions

To obtain the oleogel it is necessary to start from the preparation of an oil/water emulsion which will subsequently be subjected to dehydration of the continuous phase. Each emulsion was composed of high oleic sunflower oil (20%), water (73%), XG (2%) and structuring agent (5%) (TW, WP, SL and LBG).

Different preparation procedures were used depending on the hydration/dispersion conditions of each structuring agent.

For XG-TW emulsions: at first, 4 g (2%) of XG was gradually added to the water at room temperature in continuous agitation using a Heidolph RZR 1 stirrer (Heidolph Instruments, Germany) at a speed of 300–500 rpm, for about 10–15 min. Then, 10 g (5%) of structuring agent (TW) was added maintaining the agitation for about 5 min. After that, 40 g (20%) of oil was gradually added by increasing the speed of the agitator

up to 1800 rpm.

For WP emulsions: WP powder required previous hydration by dissolving in water at room temperature in continuous agitation. Following, XG was added to this solution maintaining the agitation for about 10–15 min, increasing the speed around 500 rpm. Finally, the oil was gradually added by increasing the speed of the agitator up to 1800 rpm.

For XG-SL emulsions: SL was added to the oil while stirring. On the other hand, XG was hydrated with the water with the same conditions as the previous emulsions. Then, the oil (which included the SL) was added to the XG solution, increasing the stirring speed.

For XG-LBG emulsions: the powdered gums were mixed and hydrated with water by stirring until a homogeneous gel texture was obtained. The oil was then gradually added by stirring under the above conditions.

As a final step, all the emulsions was homogenized using a high-energy dispersing unit (Ultraturrax T18, IKA, Germany) under the following conditions: 60 s at 6500 rpm, 60 s at 13,500 rpm and 60 s at 17,500 rpm. A total of six emulsions of each type (SL, WP, TW, LBG) were prepared. Three of them were used to carry out the measurements of the emulsion's rheology, and the three other ones were used to obtain the oleogel.

2.2.2. Oleogels

The oleogels were obtained by dehydrating the formulated emulsions. For this purpose, the emulsion was moulded in an aluminium tray and introduced into a forced convection oven (Binder GmbH, Germany) at 60 °C for 24–48 h. Afterwards, the total loss of water from the emulsion was checked by means of a difference in weight, finishing the drying process at 0% water content. Then, a dry and solid film from the dehydrated system was obtained. Using an A320R1 grinder (Moulinex, SEB Grupo Ibérico, Spain), the dry film (composed of the XG, the structuring agent and the oil) was grinded in five cycles of 3 s each one. A ready-made oleogel was obtained, with a more malleable structure. Three oleogels of each type were obtained, made from different batches of emulsions.

2.3. Characterization of the samples

2.3.1. Rheological measurements

A Rheostress HAAKE RS1 (Thermo Scientific, Germany) controlled stress rheometer was used in order to carry out rheological measurements in both emulsions and oleogels. A serrated plate-plate geometry (35 mm diameter) was used to avoid slippage in the samples, with 1 mm gap for emulsions and 1.5 mm gap for oleogels. After loading the sample, a resting time of 900 s was established in order to assure relaxation and thermal equilibrium. Silicone oil was used to cover the edges of the exposed samples in emulsions in order to prevent them from drying out. All tests were conducted at 20 °C, temperature controlled by a thermostatic bath HK10 (Thermo Scientific, Germany).

Rheological tests for emulsions were performed 24 h after preparation while oleogels were measured after 2 h.

In emulsions, stepped flow curves were performed applying shear stresses from 1 Pa to 100 Pa (corresponding to shear rate range of 0.0001–1000 s⁻¹ approx.) in logarithmic distribution (20 points, 60 s/point).

Flow curves were fitted to the simplified Carreau model (Mezger, 2014):

$$\eta = \frac{\eta_0}{\left[1 + \left(\frac{\dot{\gamma}}{\dot{\gamma}_c}\right)^2\right]^s} \quad (3)$$

where η_0 is the zero-shear viscosity, $\dot{\gamma}_c$ is the critical shear rate and s is a non-dimensional parameter.

Stress sweep tests were performed at a constant frequency of 1 Hz in

all systems studied. A range from 0.5 to 100 Pa was applied in emulsions, and 20–10,000 Pa in oleogels. Extent of the Linear Viscoelastic Region, LVR, was determined by calculating (using Haake RheoWin Data Manager 4.87 software) the data point at which G' deviated 5% from the plateau value (Mezger, 2014). Parameters considered for characterization of LVR were the dynamic yield stress, σ_y , and the corresponding shear strain, γ_L . Frequency sweep tests from 0.01 to 10 Hz were performed at a constant stress within the LVR and results were fitted to power law empirical functions (Espert et al., 2020b):

$$G' = G_1' \nu^m \quad (4)$$

$$G'' = G_1'' \nu^m \quad (5)$$

where G_1' and G_1'' are the corresponding modulus at a frequency of 1 Hz, and m is a dimensionless parameter related to the slope on the log-log scale.

All measurements were made in triplicate for each sample (emulsion and oleogel) prepared on different days.

2.3.2. Oil binding capacity of the oleogels

Oleogels oil retention was determined according to a method previously used (Espert et al., 2020a) based on the procedure reported in previous works (Huang et al., 2018; Yi et al., 2017). Briefly, a small amount of the oleogel (about 10 g) was placed on a filter paper at room temperature.

The oil loss (OL) was calculated using the following equation (1):

$$OL = \frac{W_f - W_0}{F_{oleogel}} \quad (1)$$

Where OL is the oil loss, W_f is the final weight of the filter, W_0 is the initial weight of the filter and $F_{oleogel}$ is the oil content of the oleogel.

The filter was weighted after 2 h and 24 h. Therefore, two evaluations of OL were performed in each replication of the different samples.

From the OL data, the Oil Binding Capacity (CBO) was also calculated:

$$\% \text{ OBC} = 100 - \% \text{ OL} \quad (2)$$

Two repetitions of two replicates prepared in different batches were carried out.

2.4. Data analysis

All rheological tests were analyzed using the software RheoWin 4.87 (Haake Thermo Scientific), and the data obtained were processed using Kaleidagraph 4.03 software (Synergy Software, Pennsylvania, USA).

All data were reported as mean value. One-Way analysis of variance (ANOVA) was performed using XLStat 2020 software (Addinsoft, Barcelona, Spain), applying a Tukey's test with a confidence interval of 95% to compare the means.

3. Results and discussion

3.1. Rheological properties of emulsions

3.1.1. Shear thinning behaviour

Fig. 1 shows flow curves for the different emulsions studied. All of them have a clear shear thinning behaviour, with viscosity decreasing when increasing shear rate (Rao, 1999). This fact was to be expected, since XG is a high-molecular-weight polymer that confers pseudoplastic behaviour due to its semi-rigid conformation (Panaras et al., 2011).

This flow behaviour is well predicted by the simplified Carreau model (eq (3)). The constant viscosity of the plateau is η_0 , zero-shear viscosity, $\dot{\gamma}_c$ is the critical shear rate for the onset of the shear-thinning response and s is related to the decrease in viscosity and provides information on the shear thinning character of the sample.

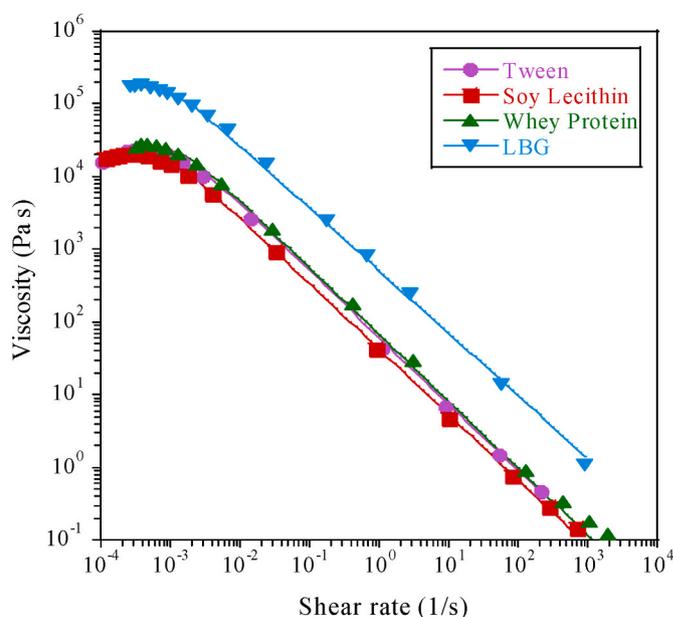


Fig. 1. Flow curves for the different emulsions studied. The continuous lines correspond to simplified Carreau model fits.

The values obtained for the fitting parameters are shown in Table 1 ($R^2 > 0.99$), which reveals that TW, SL and WP emulsions showed zero shear viscosity (η_0) between 20,000 and 30,000 Pa s. However, LBG emulsion had a significantly higher value, one order of magnitude higher than the rest of emulsions. Zero shear viscosity is related to the consistency of the emulsion at rest, inside the recipient. This constant viscosity at low shear rates was maintained at rates lower than the critical shear rate ($\dot{\gamma}_c$). This rate was very similar in all cases, so no significant differences were observed in the values of critical shear rate, $\dot{\gamma}_c$, about 0.001 s^{-1} , suggesting that emulsions will flow easily. Viscosity decreased for increasing shear rates. This shear thinning character was similar for all the emulsions since the values of the slope in log-log plot were approximately the same, except for the very slight difference in LBG emulsion. The increase of η_0 and the decrease of s values in emulsions formulated with LBG can be explained to the synergistic effect between XG and LBG. Mixing XG and LBG results in a dramatic increase in viscosity of aqueous solutions, even at low polysaccharide concentration (Casas and García-Ochoa, 1999). These results were also found in other type of food emulsions containing LBG (Dolz et al., 2007). A high concentration of LBG showed a high viscosity of the water phase and the formation of a three-dimensional network of aggregated fat droplets was produced, which increases the viscosity values significantly (Chung et al., 2013).

3.1.2. Viscoelastic properties of the emulsions

The stress sweeps performed on the different emulsions are plotted in Fig. 2. It is clear that addition of the different structuring agents affected

Table 1

Mean values of parameters obtained for simplified Carreau model fits to flow curves (η_0 , zero shear viscosity; $\dot{\gamma}_c$, critical shear rate; s , shear thinning index) for the different emulsions studied.

Emulsion	η_0 (Pa s)	$\dot{\gamma}_c$ (1/s)	$s \pm 0.01$
TW	24131 ^B ± 3368	0.0014 ^A ± 0.0002	0.46 ^A
SL	19199 ^B ± 295	0.0011 ^A ± 0.0001	0.45 ^A
WP	35124 ^B ± 5022	0.0013 ^A ± 0.0002	0.46 ^A
LBG	151150 ^A ± 42355	0.0014 ^A ± 0.0005	0.43 ^B

TW, Tween 80; SL, soy lecithin; WP, Whey Protein; LBG, locust bean gum.

^{AB} Different letters in each column indicate significant differences between the means ($p < 0.05$) according to Tukey's test.

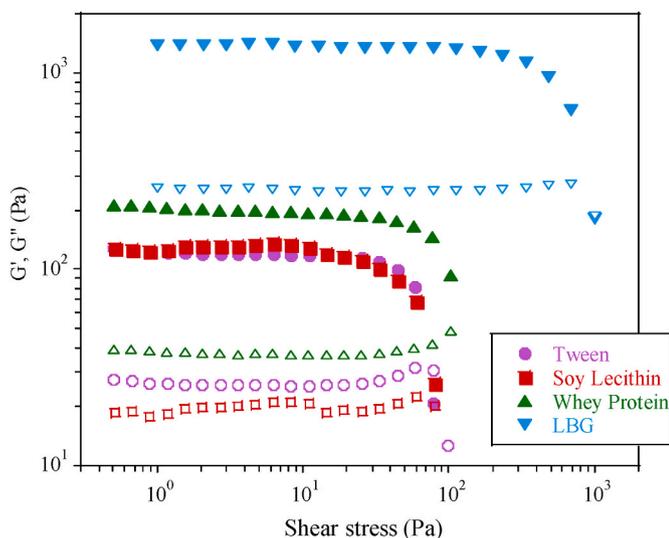


Fig. 2. Stress sweeps for the emulsions studied. G' : storage modulus (filled symbols); G'' : loss modulus (empty symbols).

the extent of the linear viscoelastic region. Values of yield stress, σ_y , and the corresponding strain, γ_L , are shown in Table 2. The emulsions formulated with SL, TW and WP showed higher sensitivity to the applied stress, and lower stress values must be reached to ensure linear viscoelastic properties. On the contrary, LBG emulsions presented a much larger LVR, one order of magnitude higher than the rest when considering shear stresses, although the corresponding shear strain amplitude are similar. Frequency sweep plots (Fig. 3) showed a clear predominance of the elastic behaviour ($G' > G''$), indicating the existence of a firm gel-like network structure (Mezger, 2014). Values of G' and G'' were similar for the emulsions except for the LBG. Viscoelastic moduli were one order of magnitude higher, indicating a stronger internal network in accordance with the greater consistency observed. The synergistic effect between XG and LBG proposed in previous studies was confirmed. Besides producing an increase on viscosity, the binding of XG and LBG forms thermoreversible gels (Ramirez et al., 2002). This fact has been extensively studied in several works which have evidenced a notably effect on the rheological properties, in particular intrinsic viscosity, flow behaviour and viscoelastic properties (Poret et al., 2021). Different mechanisms have been proposed to understand this phenomenon (Grisel et al., 2015; Wang et al., 2002), although the most common scenario is the interaction between XG side chains and LBG backbone (Schreiber et al., 2020).

Despite of LBG provided firmer emulsions, it did not seem the internal structure were different, as the loss tangent values ($\tan \delta = G''/G'$) were similar to the rest of the emulsions (Table 2).

Since the viscoelastic moduli showed a lineal trend in the log-log

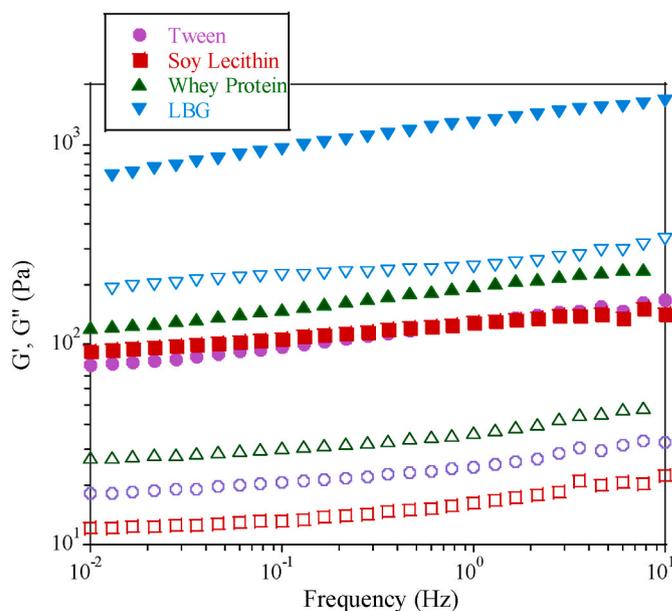


Fig. 3. Frequency sweeps of the different emulsions studied. G' : storage modulus (filled symbols); G'' : loss modulus (empty symbols).

plots, power law empirical functions can be used to reproduce the dependence of both modulus on frequency (eqs. (4) and (5)). The mean values of the parameters obtained after curve fitting ($R^2 > 0.995$) are presented in Table 2. G_1' and G_1'' correspond to the moduli at a frequency of 1 Hz, and m is a dimensionless parameter related to the slope on the log-log scale. The low m values implied a weak frequency dependency of both moduli, which is an important characteristic of structured solid behaviour, indicating a high mechanical strength of the systems. This higher mechanical strength suggests more tight networks of soft solids (Meng et al., 2018a). The analysis of variance confirms what is observed in the graph, since only the values of the modulus in the LBG emulsions were significantly different. So, all the systems provided a positive effect in the stabilization of the O/W emulsion, but it is the LBG emulsion that differs from the others because of the synergy offered between the gum and the glucomannan (Khouryieh et al., 2015). The synergistic effect can be interpreted as an increase in the density of cross-links between the two polysaccharides (Copetti et al., 1997). As the m' values show, the SL emulsion was significantly less dependent on frequency than the other emulsions, although none of them showed the crossover point typical of weaker gels.

Table 2

Mean values of oscillatory tests parameters in emulsions. Amplitude sweeps: end of linear viscoelastic region, LVR (σ_y and γ_L). Frequency sweeps: power law fits parameters (G_1' , G_1'' , m' , m'') and loss tangent at 1 Hz.

Emulsion	LVR		Frequency sweeps				
	σ_y (Pa)	γ_L (%)	G_1' (Pa)	m'	G_1'' (Pa)	m''	$\tan \delta$ (1 Hz)
TW	19.72 ^B ± 0.76	16.86 ^A ± 0.15	118 ^B ± 17	0.113 ^A ± 0.009	24.3 ^B ± 1.7	0.102 ^A ± 0.017	0.200 ^A ± 0.019
SL	10.64 ^B ± 2.03	9.38 ^B ± 0.22	123 ^B ± 6	0.072 ^B ± 0.002	16.8 ^B ± 0.2	0.089 ^A ± 0.004	0.128 ^B ± 0.008
WP	19.20 ^B ± 3.22	9.33 ^B ± 0.89	185 ^B ± 13	0.114 ^A ± 0.007	37.9 ^B ± 0.1	0.080 ^A ± 0.008	0.193 ^A ± 0.012
LBG	102.45 ^A ± 3.18	6.74 ^B ± 1.51	1372 ^A ± 144	0.121 ^A ± 0.012	284 ^A ± 38	0.072 ^A ± 0.002	0.194 ^A ± 0.006

TW, Tween 80; SL, soy lecithin; WP, Whey Protein; LBG, locust bean gum.

^{AB} Different letters in each column indicate significant differences between the means ($p < 0.05$) according to Tukey's test.

3.2. Characterization of the oleogels

3.2.1. Visual appearance of the obtained oleogels

After preparation and refrigeration at 5 °C, the different oleogels were left to stand in an inverted position at room temperature on a flat surface and their appearance was observed (Fig. 4). The photographs revealed that suitable oleogels could be obtained from the designed emulsions. Liquid sunflower oil was successfully structured into solid-like oleogels, remaining retained in the structured network formed by XG-structuring agent, even after drying and shearing. It is important to stand out that the final matrix did not contain any water, as the initial emulsion was dehydrated to get 0% water content. Apparently, oleogel characteristics changed depending on the type of structuring agent used. Oleogels containing TW and WP seemed smooth and homogenous, whereas SL and LBG oleogels presented a non-uniform, grainy appearance.

3.2.2. Viscoelastic properties

For the oleogels, only measurements of viscoelastic properties were performed, since their more solid character made flow experiments not possible. Fig. 5 shows the stress sweeps of the oleogels formulated with the different emulsions. It can be observed that the values of G' were higher than G'' over the whole stress range, indicating that all oleogels exhibited a predominant elastic behaviour. As the stress increased, G' values remained constant and decreased at high stresses, indicating that the structure of the oleogels broke at high stresses. Therefore, it is confirmed that stable oleogels were obtained at a wide range of stresses, up to about 1000 Pa, as it is shown in Table 3. However, SL oleogels showed a lower LVR, indicating that this system was more sensitive to the application of high stresses (Table 3). This is probably attributed the fact that in this system the critical lecithin concentration is exceeded. Increasing the lecithin concentration increases the instability index, which is likely attributed to the fact that the excess lecithin was adsorbed on the oil-water interface, resulting in droplet interaction and thus, instability of the emulsion (Tian et al., 2021). Similar results were obtained in ethylcellulose-lecithin based oleogels (Aguilar-Zárate et al., 2019) in which further addition of lecithin did not promote the increase in moduli, but resulted in gels with lower viscoelastic properties.

Compared to the moduli values obtained for the emulsions, all oleogels exhibited a drastic increase in moduli, indicating that the drying of the initial emulsion allows the achievement of systems with high gel strength. The differentiated increase in consistency caused by the presence of LBG in the emulsions was not observed in the oleogels. Thus, the synergistic effect of XG and LBG was not appreciated in the absence of water (oleogel), but only when the continuous aqueous phase (emulsion) is present.

In frequency sweeps (Fig. 6) elastic modulus (G') and viscous modulus (G'') informed about viscoelasticity and gel strength over the whole frequency range. Expected elastic character of high structured systems is confirmed since G' are greater than G'' with almost not frequency dependence. Moduli values turned to be much higher than the

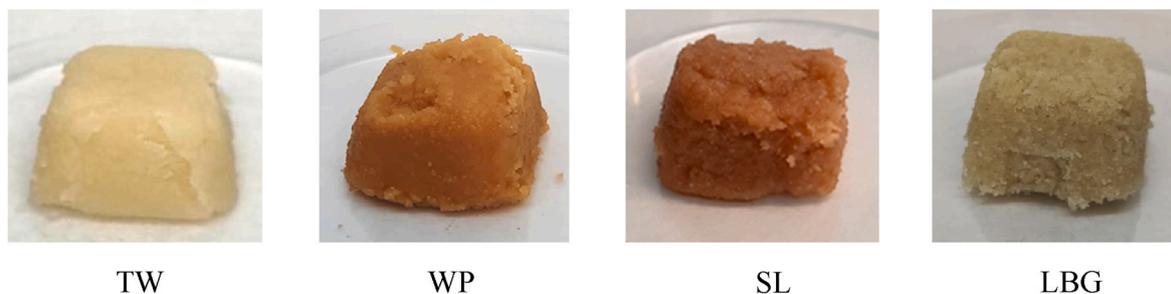


Fig. 4. Visual appearance of the different oleogels obtained from the initial emulsions containing different structuring agents (TW: Tween 80, WP: whey protein, SL: soy lecithin, LBG: locust bean gum).

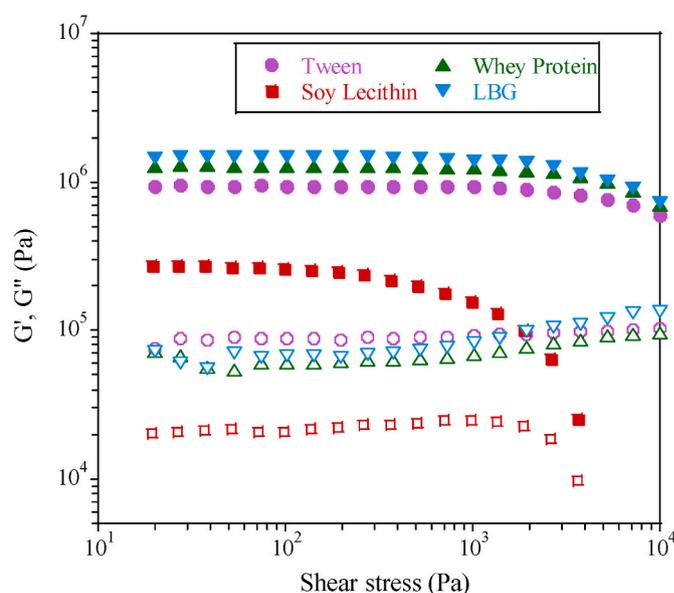


Fig. 5. Stress sweep for the oleogels formulated from the different emulsions. G' : storage modulus (filled symbols); G'' : loss modulus (empty symbols).

emulsions moduli. The high gel strength of these oleogels is outstanding as storage modulus values are above 10^6 Pa. To date, oleogels with these viscoelastic properties have not been obtained using xanthan gum. Using binary or ternary mixtures of XG combined with guar gum and sodium caseinate Abdolmaleki et al. (2020) obtained oleogels with G' values around 300,000 Pa. Another type of oleogel that has not reached these values is the one formulated with isolated cellulose ether (Espert et al., 2020a) or in combination with xanthan at low concentrations (Patel et al., 2014a,b), which yielded G' values of about 500,000 and 4000 Pa respectively.

As in emulsions, power law empirical functions were also used to reproduce the dependence of both moduli on frequency (Eq. (4) and (5)). The rheological behaviour of the oleogels could be corroborated by the values of the parameters obtained from the fits ($R^2 > 0.995$), shown in Table 3. A high mechanical strength of all oleogels is observed not only in the high values of viscoelastic moduli for 1 Hz, but also in their weak dependence of G' on the frequency, typical of solid and strong gel type systems.

The data confirmed significant differences for SL oleogels, which seem to have the lowest firmness. However, values of loss tangent are very low, and similar to the rest of oleogels, what is indicative of a stable internal structure (Table 3).

3.2.3. Oil binding capacity (OBC)

To further investigate the OBC of oleogel gel samples, oil loss (OL) was monitored after 2 h and 24 h, and not statistically significant differences were found between the oleogels studied (Fig. 7).

Table 3

Mean values of oscillatory tests parameters in oleogels. Amplitude sweeps: end of linear viscoelastic region, LVR (σ_y and γ_L). Frequency sweeps: power law fits parameters (G_1' , G_1'' , m' , m'') and loss tangent at 1 Hz.

Oleogel	LVR		Frequency sweeps				
	σ_y (Pa)	γ_L (%)	G_1' (Pa)	m'	G_1'' (Pa)	m''	$\tan \delta$ (1 Hz)
TW	1002 ^A ± 301	0.138 ^A ± 0.042	1101456 ^A ± 131377	0.064 ^A ± 0.011	91944 ^A ± 1994	-0.03 ^{AB} ± 0.045	0.084 ^A ± 0.015
SL	131 ^B ± 11	0.057 ^B ± 0.009	185925 ^B ± 1435	0.037 ^B ± 0.006	13035 ^C ± 1279	-0.009 ^A ± 0.008	0.065 ^{AB} ± 0.007
WP	1352 ^A ± 197	0.097 ^{AB} ± 0.027	1463675 ^A ± 77711	0.036 ^B ± 0.002	73371 ^B ± 88	-0.155 ^C ± 0.016	0.049 ^B ± 0.005
LBG	1232 ^A ± 719	0.074 ^{AB} ± 0.032	1386566 ^A ± 248618	0.047 ^{AB} ± 0.007	78881 ^{AB} ± 2847	-0.123 ^{BC} ± 0.010	0.054 ^B ± 0.006

TW, Tween 80; SL, soy lecithin; WP, Whey Protein; LBG, locust bean gum.

^{ABC} Different letters in each column indicate significant differences between the means ($p < 0.05$) according to Tukey's test.

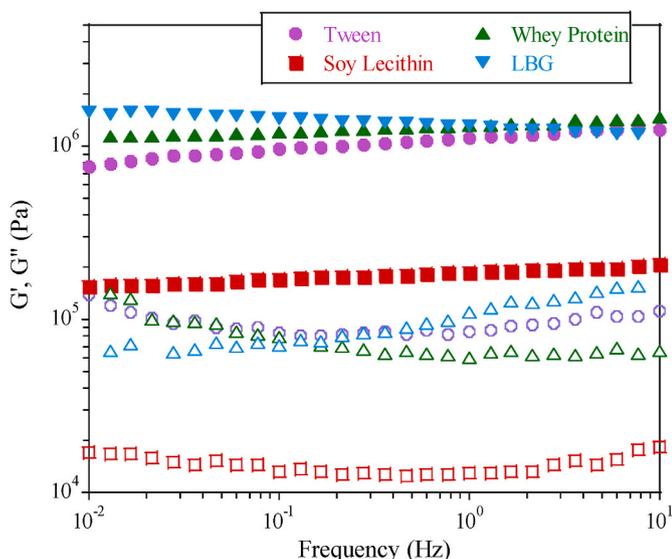


Fig. 6. Frequency sweeps for the different oleogels studied. G' : storage modulus (filled symbols); G'' : loss modulus (empty symbols).

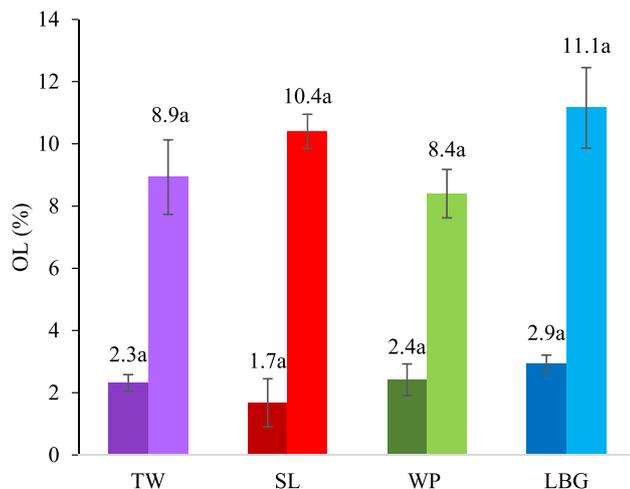


Fig. 7. Oil loss (OL) after 2 h (dark colour) and after 24 h (light colour) for the different oleogels. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

The OBC calculated from the OL values pointed out that all the oleogels had a very similar retention capacity. When performing rheological measurements, after 2 h, the OBC was very high with values

about 98%. After 24 h OBC of approximately 90% were found, being the WP the oleogel with the highest OBC (91.60%), followed by TW (91.07%), SL (89.60%) and LBG (88.84%). Considering these values we can affirm that oil retention capacity of the formulated oleogels is adequate and suitable for their application in food matrices. This high oil retention could be related to the fact that the presence of XG in oleogels provides higher oil retention than other polysaccharides, probably due to the high capacity of XG to form hydrogen bonds and hydrophobic interactions which provides a high stability of the interface improving the stability of the final oleogel (Pan et al., 2021; Sun et al., 2018). Many works add XG to cellulose ether-based oleogels to improve their stability (Meng et al., 2018b), although it is necessary to increase the concentration of cellulose ether in the formulation in order to considerably reduce the oil loss (Meng et al., 2018a).

Despite addition of LBG caused a drastic increase in viscosity and elasticity in the initial emulsion related to the rest, it did not provide the most stable oleogels. On the opposite, although the SL oleogels exhibited lower firmness, the strong internal network structure indicated by the low values of loss tangent ($\tan \delta$) lead to good oil retention properties.

4. Conclusions

Suitable oleogels with adequate mechanical properties and stability were obtained from sunflower based emulsions containing 2% of XG and 5% of different food grade oleogelators. The emulsifiers WP and TW provided similar emulsions as well as oleogels with analogous properties. It is interesting to note that although LBG led to emulsions with higher consistency, the resulting oleogels were similar to the rest. On the other hand, the SL oleogels presented lower firmness although they had a good internal structure. All the oleogels offered a high oil binding capacity, even 24 h after preparation. Therefore, all oleogels obtained would be feasible as substitute systems of conventional saturated fats in solid products such as chocolates or puff pastries. The chosen structuring agent would depend on the interest of the polymer nature required for each specific application.

CRedit authorship contribution statement

M. Espert: Conceptualization, Methodology, Formal analysis, Writing – original draft, preparation, Writing – review & editing. **M.J. Hernández:** Conceptualization, Methodology, Investigation, Funding acquisition, Supervision, Writing – review & editing. **T. Sanz:** Conceptualization, Methodology, Investigation, and, Funding acquisition. **A. Salvador:** Conceptualization, Methodology, Investigation, Funding acquisition, Supervision, Writing – review & editing.

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.

The authors of the article titled “Rheological properties of emulsion templated oleogels based on xanthan gum and different structuring agents” declare that there is no conflict of interest.

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