

Effect of oil type and concentration on solid fat contents and rheological properties of watery oleogels

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This study aims at evaluating the effect of oil type and concentration on solid fat contents and rheological properties of oleogels (OGs) prepared using different types of oils; namely, HOSO (High oleic sunflower oil), HO (Hazelnut oil), OO (Olive oil) and BF (Blend Fat) as well as food-grade carnauba wax (CW) as an oleogelator at different concentrations (5 and 7%). The rheological analysis showed that all OGs samples exhibited viscoelastic solid-like gel behaviour. Both oil and wax concentrations significantly ($p < 0.05$) affected the viscoelastic rheological properties. K' values of the samples increased with increased wax and aqueous phase concentration. The type of oils and their ratio in an oil phase could significantly ($p < 0.05$) affect the K' values and solid fat content (SFC). At 5% of wax concentration, the sample A4 prepared with 100% HOSO had the lowest SFC at all temperature levels. At 7% of wax concentration, the sample C3 prepared with 100% HOSO had the lowest SFC at the temperature levels ranging between 10 and 30°C; however, the sample (coded as B4) prepared with 100% HOSO had the lowest SFC at 35°C. These results suggest that wax/OGs blends could be produced by using wax and different types of oils at different concentrations. The use of oleogels could be further suggested in terms of healthy nutrition given the possibility to decrease the level of saturated fatty acids in people's diet.

Keywords: Oleogels, viscoelastic properties, margarine, wax

INTRODUCTION

Fats and oils are important bioactive compounds and flavour carriers and play an important role as an energy source and as solvent for valuable products [1]. Therefore, fats and oils have a significant place in food industry. Modified oils such as shortenings and margarine are also extremely popular in the food industry. They have been used for many years in the production of cookies, pastry and cakes, and are also directly used as a spread. These products provide good mouthfeel, palatability, tenderness and long shelf life. However, it is well known that using margarine at high amounts leads to an increased risk of cardiovascular disease, obesity and diabetes due to their high contents of saturated fatty acids and trans-fatty acids [2]. Therefore, many studies have been conducted to investigate replacements for the reduction of saturated fatty acid in the formulations of margarines and shortenings.

In recent years, there has been an increasing trend in the use of vegetable oils in margarine and shortening formulations. However, the direct use of vegetable oil without modification is known to cause several problems, leading to greasy and less crispy products having lower stability and short shelf life due to higher lipid oxidation rate. Therefore, this reveals the need to develop a product formulation free of stability and textural problems. In this respect, organogelation

could be proposed as a novel solution to overcome such problems. Organogelation is a process in which the liquid oils could gain a gel-like structure by trapping the liquid phase into the gel network, which exhibits some thermo-reversible and three dimensional characteristics [3]. Oleogels (OGs) are structured oils prepared by gelation of oil using oleogelators like vegetable waxes, mono-diglycerides, alcohols or esters of fatty acids, phospholipids and phytosterols [3]. Several types of vegetable oils such as sunflower oil, corn oil, olive oil, canola oil, hazelnut oil could be utilised to prepare OGs. The studies conducted on OGs demonstrated that they could be used in food products like cakes [4], cookies [5], meat products [6] and chocolate [7].

In food industry, OGs prepared by different oil and waxes with different concentrations were used in food products such as cookies [8] cakes/muffins [9, 10], chocolate [11], milk-based products [12] and bread [13] in place of shortenings. In literature, there are several studies in which candelilla wax, monoglycerides, and hydroxypropyl methylcellulose, shellac wax, rice bran wax, and ethylcellulose were tested as the oleogelators. These results revealed that the structure and properties of OGs were affected by oil and the wax type at different concentrations. Therefore, further investigations are necessary to optimise the OGs formulation to achieve the best quality properties of shortenings or margarine.

Rheological properties are among the quality properties reflecting the quality of final products. In this respect, mechanical properties of margarine are of great importance to acquire information about spreadability and viscoelastic characteristics [14]. Rheological analyses such as small amplitude oscillatory shear tests (SAOS involved with frequency and amplitude sweep tests) are some of the main test tools to evaluate mechanical properties of margarine-like products. In literature, some studies have studied the rheological properties of different types of oleogels [15-18]. However, limited studies have been carried out studying the rheological properties of the oleogel-based margarine.

This study aimed at evaluating the effect of oil type and concentration on solid fat contents and rheological properties of oleogels (OGs) prepared using different types of oils; namely, HOSO (High oleic sunflower oil), HO (Hazelnut oil), OO (Olive oil) and BF (Blend Fat) as well as food-grade carnauba wax (CW) as an oleogelator at different concentrations (5 and 7%).

MATERIAL AND METHODS

MATERIALS

HOSO, OO, HO and BF (produced by interesterification technology (palm oil (50%), palm kernel (10%),

palm stearin (20%) + cotton oil (20%)), were bought from a local market in Istanbul. Food grade Carnauba wax (2442, Kahlwax; Kahl GmbH & Co. KG) was obtained from Ejder Chemical Inc. The other components of the margarine formulation were supplied from a company in Istanbul, Turkey.

METHODS

Water quantities of OGs were determined as a 16% which is the same amount of water in the commercial margarine sold in Turkey. 5% and 7% CW were used in all the oleogel formulation and the types and composition of oils used in the production of OGs are different in this study.

PRODUCTION OF OGs

Before the preparation of OGs, oils and waxes were embedded into a water bath set at 90°C. Then, for stronger water and oil phases stability, emulgators, β -carotene, Vitamin A, and D₃ were solved in the oil phase, while stabilisers citric acid (0.08%, potassium sorbate 0.10%, and NaCl 0.15%) were solved in the water phase. OGs were manufactured as follows: firstly, the water phase (W) and the oil phase (O) were stirred. For better emulsion, the two-phase mix was exposed to sonication for 10 minutes at 100% amplitude by ultrasonic homogeniser (Hielscher-UP200 Ht, Germany). After sonication, emulsion samples (W/O) were heated for the second time. When the carnauba wax exactly melted, and the emulsion samples reached a similar temperature when the wax was completely melted; it was added into the emulsion samples. To provide the same emulsion quality, the wax/emulsion (W/O/W) mixture was mixed with Ultra Turraks for 10 minutes. Finally, oleogel samples prepared by using this procedure [19].

SOLID FAT CONTENT OF OGs

The solid fat content of the samples was specified using NMR (Bruker Minispec 7.5 MHz, USA) at three different temperatures (10, 20, 30°C) and two different wax concentrations (5% and 7%). 3.5 ml of samples, which was non-inclusive of water, were added into NMR glass tubes and kept at 60°C for 5 min. Then waiting at 0°C for 1 h. Afterward, they were maintained in water-bath at 20°C for 30 min. The hardened sample was then put into the NMR device.

RHEOLOGICAL PROPERTIES OF OGs

Dynamic shear rheological properties of the oleogel samples were determined by using a Stress/strain-controlled rheometer (Anton Paar, MCR 302, Australia) equipped with a Peltier heating/cooling system and parallel plate configuration (diameter = 5 mm). The gap was adjusted to 1 mm and the analyses were performed at 5°C. Firstly, the amplitude sweep test was performed between 0.1 and 100%

strain values to determine the linear viscoelastic region (LVR). According to the LVR results, the strain value applied at the frequency sweep test was selected to be 0.04%. According to the results, the frequency sweep test was conducted at 0.04%. Dynamic rheological analysis parameters which are storage (G'), loss (G'') modulus complex modulus (G^*), complex viscosity (η^*) and $\tan \delta$ were calculated using the Power Law model and nonlinear regression and represented viscoelastic properties of the OGs and measured as a function of angular velocity. Following models (Eqns 1-3) were fitted to the viscoelastic parameters mentioned above to calculate the model parameters which are intercepts (K' , K'' , and K^*), and slopes (n' , n'' and n^*) according to the following equations [20, 21].

$$G' = K'(\omega)^{n'} \quad (1)$$

$$G'' = K''(\omega)^{n''} \quad (2)$$

STATISTICAL ANALYSIS

Statistical analyses were performed using Statistica (StatSoft, Tulsa, USA) software program.

The mean and standard deviation were presented. Duncan multiple comparison tests were used to compare the samples, and the difference between the samples was determined at a confidence interval of 0.05. All measurements were carried out by triplicate and mean and the standard deviation was presented.

RESULTS AND DISCUSSION

Since OGs are two-phase structures consisting of oils and organogelators, the physical and chemical properties of OGs depend on the type and amount of organogelators used. Different types of organogelators used to give a 3-dimensional gel structure to oils. Among these, waxes have low polarization value, long-chain structure, and high melting point components, giving excellent crystallization properties to oils. Therefore, waxes are easily trapped in a 3-dimensional mesh structure with strong oil-binding properties and form durable gel structures. In this study, four different types of oil with different percentages used forming OGs at different wax concentrations.

RHEOLOGICAL PROPERTIES OF OGs

Dynamic rheological properties of oleogel samples which is prepared by different oil type and different wax concentrations are presented in Table I, II and Figure 1, 2. Viscoelastic properties of OGs were determined by frequency sweep test in the LVR limit

to characterised frequency parameters like storage modulus (G'), loss modulus (G'') complex modulus (G^*) and complex viscosity (η^*). These parameters are presented in Figure 1 and Figure 2 as a function of angular velocity. Oleogel samples which are prepared by 5% and 7% wax concentrations respectively presented in Figure 1 and Figure 2 showed that G' values of all samples were dramatically higher than G'' values at all angular velocity values. Similar results were obtained by [5, 22-24]. These results indicated that all OGs samples showed viscoelastic solid character like gel behaviour. Also, both concentrations of oleogel samples displayed no crossover observed between G' and G'' values meaning that solid character was dominant at all oleogel samples. A similar pattern had reported by [19]. In Figure 1 and 2, the G' and G'' value showed a positive slope which means that limited rearrangement of the gel structure [25].

The results showed that G' and G'' values of all oleogel samples slightly increased with angular velocity exhibiting frequency dependence in Figure 1 and 2. [26] mentioned that when the G' and G'' displays slightly increase with angular frequency, this mechanical spectrum represents solid-like gels. This situation is called the 'plateau region', where is typically found in highly entangled polymeric systems [27]. The plateau region was utilised to represent LVR. The critical stress value in the LVR stability of OGs. Stress applied at under the critical stress value the curves of G' and G'' demonstrate plateau region and indicated that zero permanent deformation [28]. Therefore, strain value was chosen in the LVR for the frequency sweep test for all oleogel samples. The results showed that all oleogel samples exhibited plateau region behaviour at low strain (0.03%). The similar result obtained by [25]. On the other hand, this behaviour displays similar properties like a weak gel. The 5% wax concentrations had higher frequency dependency than the 7% wax concentrations of oleogel samples that was made of a stronger gel network and durable to deformation. All oleogel samples with both wax concentrations showed the highest G' values suggesting a harsher and frangible structure. Similar results were obtained by [7, 28]. The last parameter of the dynamic rheological properties is $\tan \alpha$ (G''/G') and is presented in Figure 1 and 2. A4 and B1 had the highest \tan value at 5 and 7% wax concentrations while C8 and B2 had the lowest \tan value at 5 and 7% wax concentrations, respectively. Also, the \tan value of the oleogel samples was found as within this result is another significant indicator of its solid nature. Similar results obtained by [25]. \tan values of the oleogel samples were lower than 1, indicated that oleogel samples demonstrate gel-like consistency and similar elastic dominant characteristics. \tan value can be utilised to obtain

information on the relative elasticity of substance [29]. According to the acquired dynamic viscoelastic measurements, the G' and G'' values were fitted to the Power Law model. In Table I R^2 of G' and G'' value were found to be between at 5% wax concentrations respectively 0,77- 0,99 and 0,94-0,99. Also, in Table II R^2 of G' and G'' values found to be between 0,94-0,99 and 0,98-0,99 at 7% wax concentrations, respectively. These results showed that the power Law model well fitted and was compatible with our results. In addition, these findings showed that the relationship between G' , G'' and angular velocity can be explained by the power-law model except A4 samples that contains 5% wax concentrations and showed less R^2 (0,77) value than the other R^2 value for G' values.

K' values of all oleogel samples were found to be higher than the K'' values indicating that all oleogel samples that are prepared with a different oil type and different wax concentrations demonstrated to have a solid-like behaviour than the viscous behaviour expected from G' and G'' values mentioned above. The similar result obtained by [30].

In Table I Dynamic rheological properties of oleogel samples wax showed C4 and C7 had the highest K' , K'' value at 5% and 7% wax concentrations, respectively. However, A4 and A2 had the lowest K' , K'' values of oleogel samples respectively at 5 and 7% wax concentrations. The A4 and A2 oleogel samples respectively showed the weakest gel network at 5% and 7% wax concentrations of oleogel with a higher frequency dependency than the other oleogel sample. The similar result obtained by [28]. In addition, as clearly seen there was a significant relationship between water content and K' and K'' values among samples with the same oil formulation. In K' , K'' values, while the first letter (upper case letters) of statistical analysis represents the differences between all samples, second letter (lower case letters) represents the differences between samples with the same amount of water.

The effect of oil content on watery oleogel samples was examined in Table I and II. Firstly, at 5% wax concentration, when the oil types were kept constant and the oil content decreased led to K' , K'' values increased in oleogel samples which are containing 100% HOSO and 75% HOSO, 25% BF, except 25% (HOSO, HO, OO, BF). However, same conditions applied as mentioned above for 7% wax concentration K' , K'' value values increased in all samples containing 25% (HOSO, HO, OO, BF) 100% HOSO and 75% HOSO, 25% BF, and were higher than K' , K'' values of samples containing 5% wax concentrations of oleogel sample. When the K' , K'' values of oleogel samples compared by wax concentrations, the results showed that there was a significant relationship between water content, wax

concentrations and K' , K'' values among samples with the same oil formulation.

Especially in oleogel formation, the decrease in oil ratio and an increase in wax concentration leads to a stronger gel formation. This situation could be explained by the decrease in the oil ratio leads to an increase in the K' , K'' values due to increasing the amount of water in the oleogel composition. Another important reason is related that carnauba wax has a higher solid component and viscosity than the oil types used.

In Figure 1 and 2, all oleogel samples which are prepared by different wax concentrations and oil types and amount, G' value was higher than the G'' value which can be related to the degree of (un) saturated fatty acids. The higher saturated fatty acid could be indicated by the large conformational degree of the solvent and facilitate the creation of the gelatine network, therefore producing a stronger gel [31]. The similar result obtained by [32].

The effect of oil types on watery oleogel samples was examined in Table I and II. While 5% and 7% wax concentration used oil ratios were kept constant, 25% (HOSO, HO, OO, BF) values of K' , K'' values of samples A and B were higher than the values of other used oil types and percentage. 75% HOSO, 25% BF has the highest K' , K'' value among C oleogel samples. Especially with the decrease of the used oil ratio and the change of the used oil types and percentage, the highest values of K' , K'' values were obtained as a result of 5% and 7% wax concentration (75% HOSO, 25% BF). In addition, this can be explained the decrease in the oil ratio leads to an increase in the K' , K'' values due to increasing the amount of water in the oleogels composition and occurring compact network by packing of water droplets in an oil continuous phase. HOSO contains more unsaturated fatty acids than OO and HO and among C samples with 60% oil content, C4 oleogel samples containing 5% wax concentration and C7 oleogel sample containing 7% wax concentration were found to have higher G' and G'' values than all other oleogel samples. This result explained by the low oil content, the increased wax concentration, and the unsaturated fat content of HOSO and BF higher than OO and HO.

However, K' , K'' values obtained at 7% wax concentration were higher than those obtained at 5% wax concentration. This is due to the increased wax concentration resulting in the formation of a more stable and solid oleogel, forming a dual-phase gel structure with the oil. As clearly seen in Tables I and II, increasing in wax concentrations of all oleogel samples led to an increase in K' , K'' values. This result can be explained by an increase in wax concentrations led to the increase of SFC of oleogel samples. The similar result obtained by [8].

Table I - Viscoelastic properties of watery oleogels containing 5% wax

Sample	Content	K' (Pa)	n'	R ²	K'' (Pa)	n''	R ²
A4	100% HOSO	13848.66 ^{Cc}	0.049	0.7750581	1246.96 ^{Cc}	0.301	0.94226924
B6	100% HOSO	25949.57 ^{Bc}	0.11	0.9902554	3262 ^{Bc}	0.237	0.97746826
C8	100% HOSO	34933 ^{Ac}	0.11	0.9725213	4434.42 ^{Ac}	0.202	0.98510369
A5	75% HOSO, 25% BF	141763.5 ^{Cb}	0.1	0.9820772	12902.48 ^{Cb}	0.29	0.98422854
B7	75% HOSO, 25% BF	158743.4 ^{Bb}	0.085	0.9855083	13753.79 ^{Ba}	0.27	0.98749093
C4	75% HOSO, 25% BF	240738.75 ^{Aa}	0.1	0.974545	19206.67 ^{Aa}	0.32	0.99138121
A6	25% (HOSO, HO, OO, BF)	193241.76 ^{Aa}	0.073	0.9956184	13411.59 ^{Ba}	0.31	0.98385385
B5	25% (HOSO, HO, OO, BF)	178350.47 ^{Ca}	0.089	0.9640612	13481.27 ^{Bb}	0.31	0.98826603
C6	25% (HOSO, HO, OO, BF)	186450.76 ^{Bb}	0.096	0.9942617	18198.35 ^{Ab}	0.29	0.98930802

HOSO: High oleic sunflower oil; HO: Hazelnut oil; OO: Olive oil; BF: Blend Fat. The different uppercase letter in the same column shows effect of fat content on K' and K'' for same oil. The different lowercase letter in the same column shows effect of oil type on K' and K'' for same fat content.

Table II - Viscoelastic properties of watery oleogels containing 7% wax

Sample	Content	K' (Pa)	n'	R ²	K'' (Pa)	n''	R ²
A2	100% HOSO	62755.99 ^{Cc}	0.094	0.9979416	6549.12 ^{Cc}	0.269	0.98883989
B4	100% HOSO	76330.39 ^{Bc}	0.11	0.9971393	9249.11 ^{Bc}	0.251	0.98261231
C3	100% HOSO	138396.89 ^{Ac}	0.124	0.9934201	18575.11 ^{Ac}	0.25	0.98925904
A3	75% HOSO, 25% BF	234488.63 ^{Cb}	0.095	0.9857926	19784.29 ^{Cb}	0.293	0.99074079
B2	75% HOSO, 25% BF	236429.1 ^{Bb}	0.087	0.9925463	23311.91 ^{Bb}	0.254	0.98461836
C7	75% HOSO, 25% BF	387345.37 ^{Aa}	0.147	0.9457811	56380.13 ^{Aa}	0.307	0.99811252
A1	25% (HOSO, HO, OO, BF)	266873.97 ^{Ca}	0.105	0.9821113	24294.55 ^{Ca}	0.317	0.99407834
B1	25% (HOSO, HO, OO, BF)	294973.89 ^{Ba}	0.12	0.9820318	33452.12 ^{Ba}	0.307	0.99693752
C2	25% (HOSO, HO, OO, BF)	371849.08 ^{Ab}	0.111	0.9926979	46995.15 ^{Ab}	0.249	0.99562597

HOSO: High oleic sunflower oil; HO: Hazelnut oil; OO: Olive oil; BF: Blend Fat. The different uppercase letter in the same column shows effect of fat content on K' and K'' for same oil. The different lowercase letter in the same column shows effect of oil type on K' and K'' for same fat content.

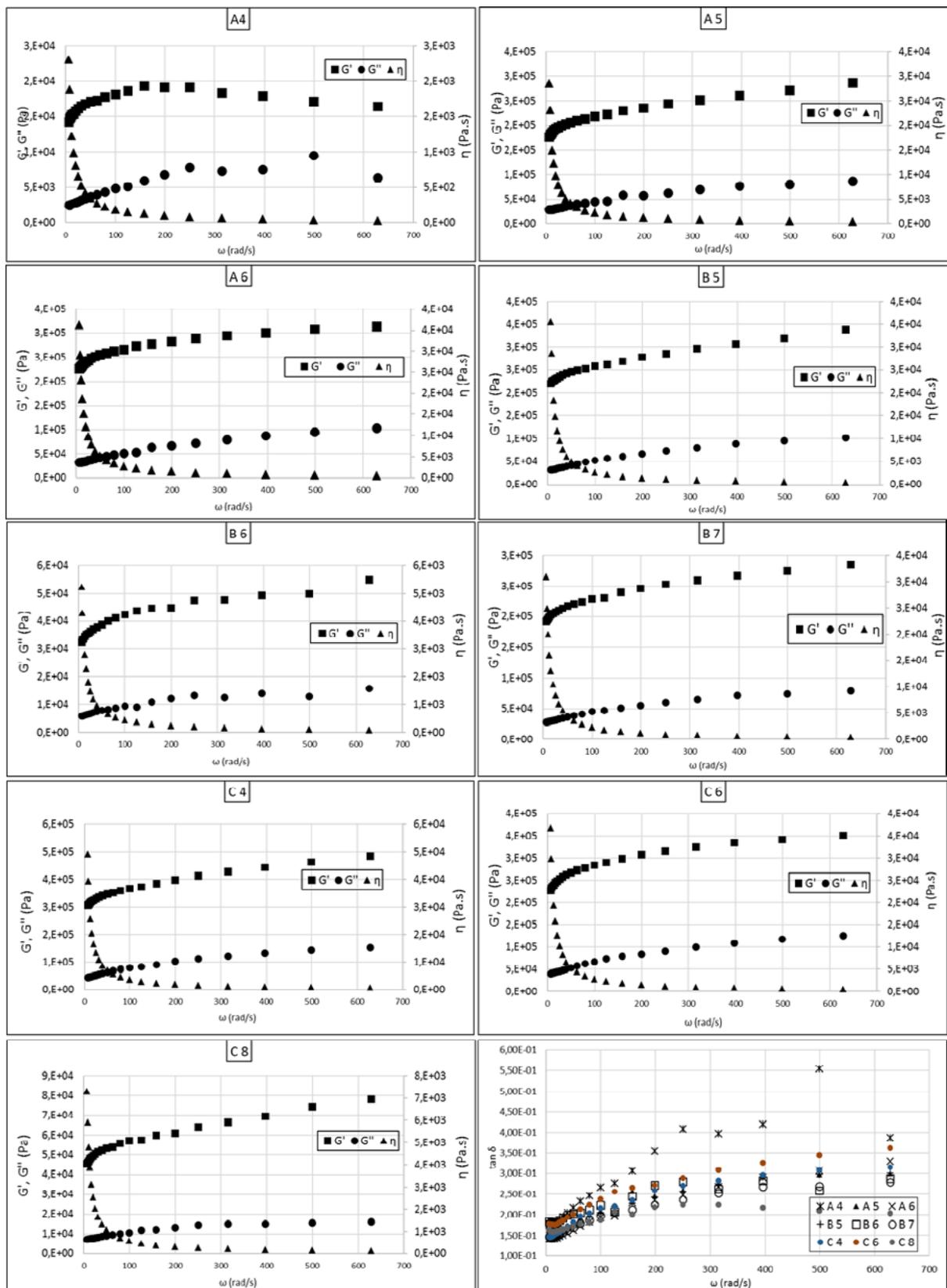


Figure 1 - Viscoelastic properties of watery oleogels containing 5% wax

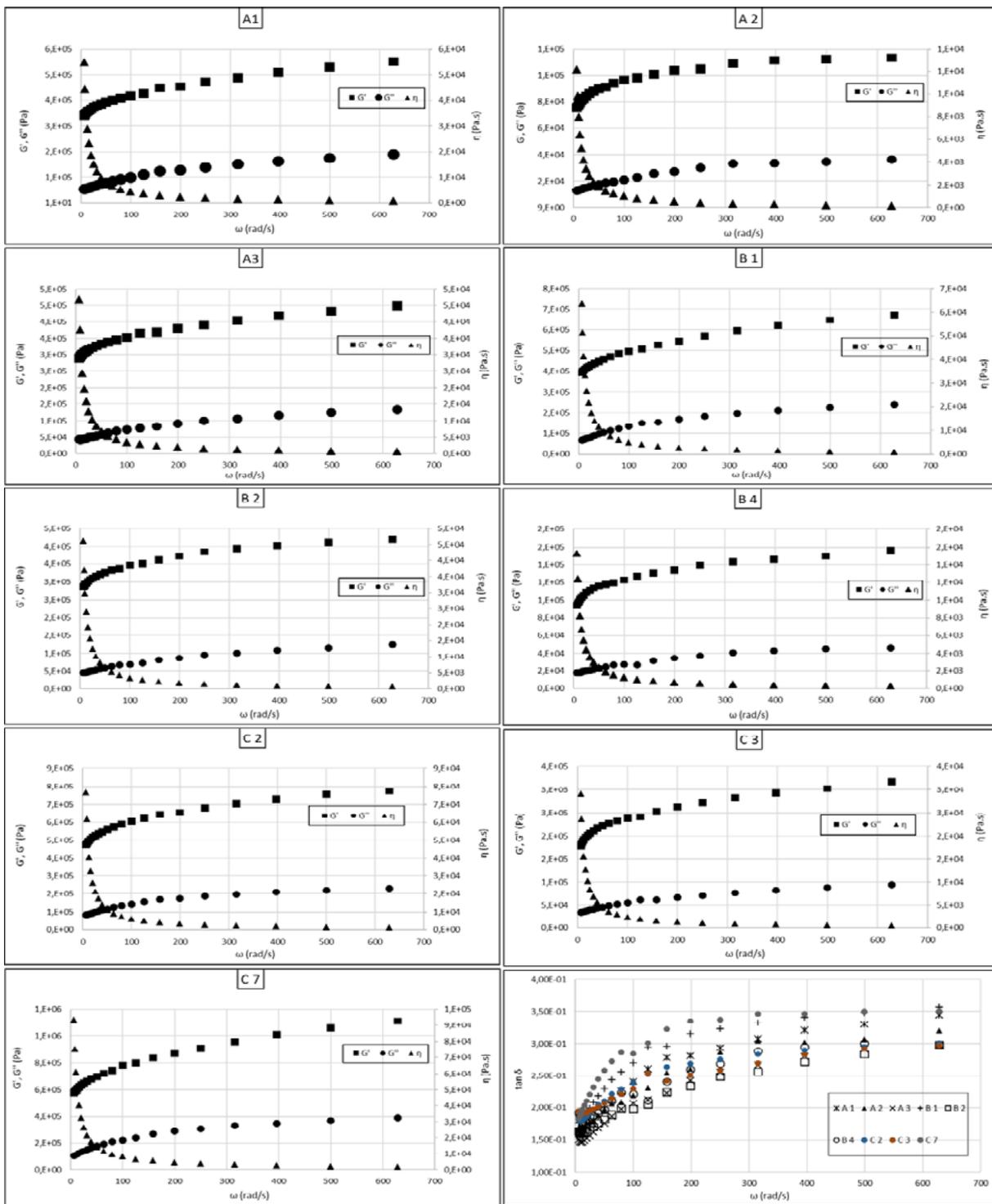


Figure 2 - Viscoelastic properties of watery oleogels containing 7% wax

SOLID FAT CONTENT OF OGs

SFC is described as the ratio of solid content to total matrix at a given temperature and is a very significant parameter for oil or fat [33]. The effect of oleogel samples which is containing different types of oil and composition at two different wax concentrations on solid fat content as a function of temperature is shown in Figure 1. In addition, Solid fat content values of oleogel samples are given in Table III. SFC of oleogel samples was affected by the fatty acid profile and oleogel formulation [19]. SFC is exhibiting changes in consistency and plasticity of OGs [34].

As clearly seen in Table III. At 5% and 7% wax concentrations of oleogel samples, C8 (100% HOSO) and C7 sample (75% HOSO, %25 BF) had the highest SFC respectively at 30°C and 35°C. At 5% and 7% wax concentration oleogel samples, C6 (25% (HOSO, HO, OO, BF)) and B1 (25% (HOSO, HO, OO, BF)) had the highest SFC, at 10°C and 20°C, respectively. This result can be explained by the fatty acid profile and using in the formulation of oleogel samples.

At 5% wax concentration, A4 (100% HOSO) had the lowest SFC at all temperature levels. At % 7 wax concentration, C3 (100% HOSO) had the lowest SFC at 10°C - 20°C - 30°C however at 35°C, B4 (100% HOSO) had the lowest SFC. All these results indicated that using just HOSO in oleogel samples led to decreased SFC content of oleogel samples. This

result seemed a negative point however SFC of oleogel samples can be regulated by raising the amount of saturated fatty acid in the oleogel formulation.

In Figure 3 the SFC of C4, A6, A4, and B6 showed quite different SFC values with flowing between

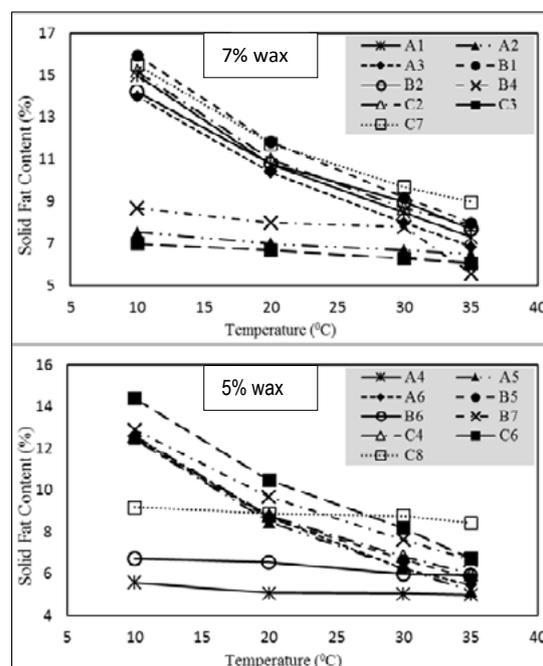


Figure 3 - Solid fat content of oleogel samples

Table III - SFC values of the watery oleogels containing 5% and 7% wax at different temperature levels.

	Sample	10 (°C)	20 (°C)	30 (°C)	35 (°C)	
7% wax	100% HOSO	A2	7.55a	7.00b	6.70c	6.50d
	100% HOSO	B4	8.70a	8.00b	7.80c	5.60d
	100% HOSO	C3	7.00a	6.70b	6.30c	6.10d
	75% HOSO, 25% BF	A3	14.00a	10.40b	8.00c	6.85d
	75% HOSO, 25% BF	B2	14.20a	10.80b	9.00c	7.70d
	75% HOSO, 25% BF	C7	15.50a	11.75b	9.70c	9.00d
	25% (HOSO, HO, OO, BF)	A1	15.00a	10.75b	8.50c	7.30d
	25% (HOSO, HO, OO, BF)	B1	15.95a	11.85b	9.20c	8.00d
	25% (HOSO, HO, OO, BF)	C2	15.20a	11.00b	8.70c	7.90d
5% wax	100% HOSO	A4	5.60a	5.10b	5.05c	5.00d
	100% HOSO	B6	6.75a	6.55b	6.00c	5.95c
	100% HOSO	C8	9.20a	8.90b	8.80b	8.45b
	75% HOSO, 25% BF	A5	12.45a	8.50b	6.25c	5.20d
	75% HOSO, 25% BF	B7	12.90a	9.70b	7.65c	6.65d
	75% HOSO, 25% BF	C4	12.60a	8.80b	6.85c	6.00d
	25% (HOSO, HO, OO, BF)	A6	12.55a	8.75b	6.25c	5.50d
	25% (HOSO, HO, OO, BF)	B5	12.45a	8.70b	6.70c	5.80d
	25% (HOSO, HO, OO, BF)	C6	14.40a	10.50b	8.20c	6.75d

Different lowercase letter in the same line shows effect of temperature on SFC values.

10 and 30. At 5% wax concentrations, the SFC of C4 and A6 ranged from 6.00-12.60 and 5.50 and 12.55 respectively, while A4 and B6 kept stable around 5-6 addition, all types of oil used to prepare oleogel formulation showed that more stable. It was an expected behaviour due to the mechanism of oleogel, which was trapped into the crystalline network by liquid oil created by oleogelators [28]. For this reason, the solid content in OGs primarily came from oleogelators [35].

Although all oleogel samples had lower SFC than the margarine, they were able to create self-standing structures. Similar results reported by [36].

CONCLUSION

The results showed that wax concentration and the oil type significantly affected the rheological properties of the wax-OGs. This study also demonstrated that wax-OGs could be produced by wax and different oil types at various concentrations and the developed OGs can replace margarine in the food industry. Another advantage of the use of oleogels could be its contribution to healthy nutrition in terms of decreased levels of saturated fatty acids in people's diet.

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