Research Article

Characterization and Stability of Emulsion Gels Based on Acrylamide/Sodium Acryloyldimethyl Taurate Copolymer

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Abstract. Sepineo P 600, a concentrated dispersion of acrylamide/sodium acryloyldimethyl taurate copolymer in isohexadecane, has self-gelling and thickening properties and the ability to emulsify oily phases, which make it easy to use in the formulation of gels and o/w emulsion gels. In this paper, gels were prepared using a Sepineo P 600 concentration between the 0.5% and 5% (w/w), and then emulsion gel was also prepared from the 3% Sepineo gel by adding a specific amount of almond oil. All the prepared systems were analyzed and characterized by oscillation rheology and acoustic spectroscopy. The particle size of the oil droplets and the microrheological extensional moduli (G' and G'') of the systems were determined from acoustic parameters and used together with the classical oscillatory rheological tests to assess the stability of the systems. Classical oscillatory analysis revealed that the dynamic moduli were very dependent on polymer concentration; as this parameter increased, there was progressive improvement in the sample elasticity. In fact, the mechanical spectra of the 0.5% and 1% (w/w) Sepineo samples were characterized by strong frequency dependence and multiple crossover points, typical of dilute polymer solution with no organized structure. On the other hand, the 3-5% (w/w) concentration systems showed typical gel-like spectra, marked by the absence of crossover points between the dynamic moduli and by weak dependence on frequency. Nevertheless, the elastic properties of the gel-like structure even at elevated polymer concentrations were not strongly long-lasting, as demonstrated by the increase of the viscous contribution in the low frequency range during acoustic spectroscopy analysis. This fact could indicate that the gel structure is characterized by weak polymer-polymer interactions, an advantageous characteristic for topical administration, as the sample is thus easier to rub into the skin. Finally, both rheology and acoustic spectroscopy indicated that addition of the oily phase caused minimal changes to the elastic character of the gel. Thus, Sepineo P 600 gel and emulsion gel are very effective systems for use in topical and other types of applications.

KEY WORDS: acoustic spectroscopy; emulsion gel; Sepineo; viscoelasticity.

INTRODUCTION

In recent years, there has been great interest in the use of novel polymers with complex functions as emulsifiers and thickeners because the gelling capacity of these compounds allows the formulation of stable emulsions and creams by decreasing surface and interfacial tension and at the same time increasing the viscosity of the aqueous phase. In fact, the presence of a gelling agent in the water phase converts a classical emulsion into an emulsion gel.

Recently, new research with emulsion gels has focused on the characterization of milk protein (used as surfactant and gellifying agents) o/w emulsions (1–6). Other investigations have examined the thickening and emulsification properties of water-soluble amphiphilic polymers, such as modified hydroxyethylcelluloses (7–8). Gelled emulsions have been described as composite materials in which oil droplets behave as filler particles, and free proteins (referring to protein gel systems) form the gel matrix (9) and also act as emulsifiers. Thus, differences in emulsion gel rheology are due to the differences in interactions across the interfaces between the filler particles and the gel matrix (10). Previous studies demonstrated that the presence of oil droplets could modify the mechanical characteristics of the protein gel by modifying its viscoelastic properties (11), substantially increasing gel strength (12). In fact, oil droplets increase the bulk protein concentration so that the latter can strengthen the gel in the bulk phase of the emulsion (12). Furthermore, the oil droplets create an interface able to interact with the gel matrix (11), and the presence of the proteins adsorbed at the interface assures interactions within the matrix and filler (13)

Investigations of viscoelastic properties have provided fundamental characterization of these systems and made rheology the most widely used technique for such study (1,2,7,10,12).

In this work, the self-gelling properties of the acrylamide/ sodium acryloyldimethyl taurate copolymer (Sepineo P 600),

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both alone and as dispersing phase for the preparation of o/w emulsion gels, have been investigated by oscillatory rheological measurements and acoustic spectroscopy.

Sepineo P 600, based on the concept of droplet hydroswelling, is a concentrated droplet dispersion of acrylamide/ sodium acryloyldimethyl taurate (a viscous liquid at room temperature) in isohexadecane as the oily dispersing phase. The presence in this dispersion of polysorbate 80 is important for keeping the resultant dispersion stable. When water is added, these polymer droplets disappear because the polymer molecules interact with it strongly, instantly forming a stable semisolid system (14).

The possibility of obtaining stiff and stable gelled phases with this polymer makes it a good candidate for the formulation of emulsion gels. Obviously, the presence of the oily phase can influence the viscoelastic behavior of Sepineo P 600 gels, with consequent changes in the characteristics of the final emulsion.

Thus, in this study, rheological oscillatory measurements, in particular, frequency sweep analysis, were used for the mechanical characterization of gels and emulsions, while acoustic spectroscopy provided a better understanding of sample microstructure. This non-destructive technique offers a unique possibility for characterizing concentrated colloidal dispersions while avoiding dilution, which is a limiting step, particularly when the tested sample is highly structured. The parameters defined by acoustic spectroscopy, such as attenuation frequency spectra and sound speed, allow the calculation of particle size from 5 nm to 1,000 μ m (15).

The operating principle of acoustic spectroscopy is based on the generation of sound pulses that pass through a sample and are measured by a receiver. During the passage through the sample, sound is attenuated by the presence of the liquid medium. The energy changes in intensity and phase are measured.

There are six mechanisms of sound interaction with a dispersed system: *viscous* (related to the shear waves generated by the particles oscillating in the acoustic pressure field due to the difference in the densities between particle and medium), *thermal* (related to the temperature gradients generated near the particles' surface), *scattering* (the same principle as light scattering), *intrinsic* (losses of acoustic energy occur when the sound wave interacts with the particles and the medium as a homogeneous phase), *structural* (caused by the oscillation of a network of particles; this mechanism is specific to structured systems), and *electrokinetic* (ultrasound/ double layer interactions). The electrokinetic losses are negligible in terms of the total attenuation, making it possible to separate electroacoustic spectroscopy from acoustic spectroscopy (15).

Through complex processing and modeling of these kinds of energetic contribution to the total acoustic attenuation, it is possible to calculate the particle size and the zeta potential of the dispersed particles (15).

The acoustic spectrometer can also act as a microrheometer, taking into account the fact that, in this case, "longitudinal" viscoelastic properties are measured because the stress is not tangential, as it is in an oscillation experiment in a rotational rheometer, but normal.

It is possible to demonstrate that ultrasonic absorption and velocity are related to the real and imaginary part of the complex modulus (16) and that G' and G'' moduli are related to sound speed (V), sound attenuation (α), and frequency (ω), using the following equations (16):

$$G' = \rho V^2 \tag{1}$$

$$G' = 2\rho V^3 \alpha / \omega \tag{2}$$

Based on these facts, a number of Sepineo P 600 systems were prepared using a Sepineo concentration ranging between 0.5% and 4%, and after accurate characterization of Sepineo P 600's gelling ability, almond oil was added to the 3% Sepineo gel. This oil was chosen because it is widely used in pharmaceutical and cosmetic applications for its practically inexistent toxicity and its high tolerability. The emulsion stability over time was monitored by observing the variation of the rheological parameters and mean diameter of the droplets.

MATERIALS AND METHODS

Sepineo P 600 and Sepicide HB, an anti-microbial agent composed of a mixture of phenoxyethanol, methylparaben, ethylparaben, propylparaben, and butylparaben, were a kind gift of Seppic (Paris). Almond oil USP was obtained from ACEF s.p.a. (Fiorenzuola D'Arda, Italy), and deionized water was obtained from an MF3 ion-exchange system (San Salvatore di Cogorno, Genova, Italy).

Gel Preparation

Gels were prepared for simple dispersion by mechanical stirring (30 min at 300 rpm; Eurostar Digital, IKA Labor-technik) of the Sepineo in the required amount of deionised water. The Sepineo concentration ranged between the 0.5% and 5% (*w/w*).

Systems were then left to rest for 24 h before being analyzed.

Emulsion Preparation

Emulsions were obtained by dispersing the 3% w/w Sepineo P 600 in the required amount (20% w/w) of almond oil and then adding the aqueous phase, containing 0.5% (w/w) of Sepicide HB, and stirring (Eurostar Digital, IKA Labortechnik, 1,200 rpm).

Samples were left at room temperature for 24 h before being analyzed and then stored for 3 months at 40°C to check sample stability over time.

Rheological Characterization

Non-destructive oscillatory measurements made it possible to obtain the principal rheological parameters, such as the storage or elastic modulus (G'), the loss or viscous modulus (G''), and the loss tangent (tan δ). Rheological analyses were performed in triplicate using a stress control rheometer (Stress-Tech, Reologica) equipped with a cone-plate geometry (4/40) operating in the oscillation mode. The gap was

370

 $150 \ \mu m$. Several tests were carried out on gels and emulsion systems:

- Oscillation stress sweep. The sample was exposed to increasing stress (0.05–10 and 0.05–50 Pa) at constant frequency (1 Hz) and temperature (20°C), and the G' values were plotted in logarithmic scale. This test makes it possible to determine the linear viscoelastic regime of the sample and therefore to choose the stress value for the other oscillation tests.
- Temperature sweep. This test was performed to characterize sample behavior at constant frequency and stress in a range of temperatures. The experimental parameters were 1 Hz frequency, 1 Pa stress, a temperature range of 10–60°C and a heating rate of 1°C/min.
- Frequency sweep. The sample was exposed to stepwise increases of frequency (0.01–50 Hz) at constant stress (1 Pa) in the field of linear viscoelasticity, and the average values of G', G" and δ were calculated at 10, 1, and 0.02 Hz. The frequency range, the G', and the G" values were plotted in logarithmic scale.
- Creep/recovery. This test was carried out at 20°C at different values of stress, depending on the system studied. The selected stress was kept constant for 100 s, then instantly removed, followed by a 200-s recovery. The creep compliance J_C (defined as the ratio between the measured strain and the applied stress) was monitored against time. The test was also used to calculate the viscosity of the sample from the linear stress/strain region of the retardation curve.

The rheological behavior of emulsions was also studied by performing the following test:

• Shear test + time sweep. This test was performed by applying a constant stress (200 Pa) for 5 min (shear test) and then observing the sample recovery for 90 min in the oscillation mode at 1 Pa of stress and 1 Hz of frequency at the temperature of 20°C (time sweep test).

Emulsion stability was checked 1 and 3 months after preparation, using stress sweep and creep-recovery tests and shear analysis followed by time sweep.

Acoustic Spectroscopy Measurements

The cell of the DT-1200 acoustic and electroacoustic spectrometer (Dispersion Technology, USA) was filled with

15 ml of Sepineo P 600 gel or the emulsion, and the sound attenuation and speed were monitored. Analyses were performed at the gap interval of 0.325-20 mm, in the frequency range of 3–100 MHz at 20°C. Particle size and rheological G' and G" moduli were calculated (15). The possible variation of droplet size and modification of the microrheological parameters over time were adopted as stability criteria of the semisolid systems.

RESULTS AND DISCUSSION

Rheological Analysis of the Gels

The rheological behavior of the gels was strictly related to polymer concentration. Stress sweep results and viscosity values obtained from the creep recovery test showed increasing elasticity in the samples, as the Sepineo concentration was increased from 0.5% to 3%, as demonstrated by the values of elastic modulus G' and viscosity (Table I). This behavior was also confirmed by the temperature sweep test, which showed higher and very similar values of G' modulus for the 3% and 5% samples. This test also showed that there was no significant variation in G' or G'' moduli at increasing temperatures (Fig. 1).

From the frequency sweep analyses, it was possible to observe that the 3-5% (w/w) concentration samples showed typical gel-like spectra, characterized by the absence of crossover points between the dynamic moduli and by weak dependence on frequency (Fig. 2, Table II). In contrast, as expected, the 0.5% and 1% (w/w) systems presented different viscoelastic behavior with mechanical spectra characterized by stronger frequency-dependence and multiple crossover points. These considerations were confirmed by the slope values of the G'-G'' curves versus frequency (Table III). It is known that the rheological behavior of a polymer in dispersion can show a modification in the slope values of the G'-G'' versus frequency curves at increasing concentrations or molecular weights. In this case, given the strong frequency dependence, the Zimm or Rouse models seemed the best-fitting (17). These models concern the viscoelastic behavior of an isolated chain that relaxes independently of the presence of the others and, for this reason, are usually employed for dilute polymer solutions. However, in this study, they failed to provide correctly fitting results, even for the 0.5% concentration, perhaps because of the co-presence of an entanglement network at this concentration. Certainly, at low polymer concentrations, it is possible to assume the

 Table I. G' and G" Obtained from the Stress Sweep Tests and Calculated at the Stress of 1 Pa and Viscosity Values Obtained from the Creep-Recovery Tests of the 0.5–5% Sepineo Gels and of the Sepineo 3%/Almond Oil Emulsion at Different Storage Times

		G' (Pa)	SD	<i>G</i> " (Pa)	SD	Viscosity (Pa s)	SD
Gels	0.5%	0.630	0.03	1.100	0.03	0.4265	0.045
	1%	6.670	3.17	4.620	0.84	47.180	2.540
	3%	615.7	16.1	90.67	1.98	84,414	2,740
	5%	665.7	44.4	94.73	11.2	112,647	6,215
Emulsion	1 day	619.6	7.70	68.20	12.5	92,346	3,421
	1 month	611.4	4.00	67.80	8.31	92,123	3,836
	3 months	608.6	6.50	60.60	5.76	92,011	4,971

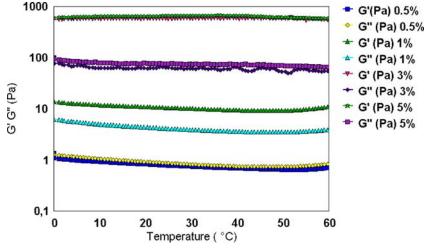


Fig. 1. Temperature sweep test of Sepineo P 600 systems (0.5–5% w/w)

existence of topological constraints and thus to imagine that there are entanglement sites, rather than specific interactions between polymeric chains. More in-depth analysis of the frequency sweep plot of the 0.5% sample revealed that it could easily be fitted with the Doi-Edwards model (17,18) (Fig. 3), which assumes that the motion of the chain is confined to a tube-like region formed by the surrounding polymer molecules. Running along the center of the tube is a primitive chain that represents the shortest path down the tube, and every deviation from this path is considered a defect. The motion of these defects allows the chain to move along the tube with a reptilian motion (reptation). The plateau modulus, which refers to the plateau observed in the plot of G' versus frequency at intermediate frequencies and is related to the relaxation modulus, can be calculated using this type of modeling. In addition, it was possible to obtain the characteristic system relaxation time (0.5185 s.), the zero shear viscosity (2.303 Pa s), and the related steadystate compliance from the values of the plateau modulus and

of the system relaxation time. The low value of zero shear viscosity and plateau modulus (5.338 Pa) obtained from the fitting, and the fact that this model is usually applied to semidilute and concentrated solutions demonstrated that the 0.5% systems did not possess an organized structure. In fact, the Doi–Edwards theory adequately describes the dynamic behavior of ideally flexible polymers in concentrated non-entangled solution, where the polymer chains do not possess permanent cross-links, and the chain is free to move along the "tube."

Slightly different considerations can be made for the 1% concentration sample. In fact, the modeling of its frequency spectra showed better correlation with the "tube linear dilution theory" (17,19,20).

This theory considers the shortcomings of reptation theory applied to linear chains, treating them as two-armed stars (21-23). In this model, relaxation phenomena are related to arm retraction. This means that the retraction motion of the arm (for each star) is very similar to the

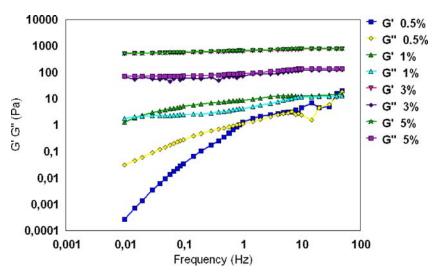


Fig. 2. Frequency sweep test of Sepineo P 600 systems (0.5-5% w/w)

Table II. G' and G" from the Frequency Sweep of Sepineo P 600Gels (0.5%, 3% and 5% w/w) Calculated at Frequencies of 10, 1, and0.02 Hz

% (w/w)	Frequency (Hz)	G' (Pa)	SD	<i>G</i> " (Pa)	SD
0.5	10	1.830	0.93	2.320	0.26
	1	1.257	0.07	1.103	0.05
	0.02	0.001	0.00	0.061	0.00
1	10	11.26	2.84	11.06	0.92
	1	9.130	1.46	4.240	0.21
	0.02	2.680	1.40	2.000	0.13
3	10	760.0	7.21	123.7	5.51
	1	650.7	8.62	78.87	5.80
	0.02	530.0	4.00	62.73	6.40
5	10	811.3	16.8	147.0	1.73
	1	686.6	13.0	90.40	0.95
	0.02	545.0	13.1	73.30	1.97

fluctuation motion of the contour along the linear chain (fluctuation-driven stretching and contraction of the chain along the tube). It should be taken into account that, in linear chains, the tube segments, which do not have retracting ends, will be relaxed by reptation. For these linear chains, the fast retractions will involve motion near the chain end, and thus, this model must consider the center of the chains as fixed. Application of this model and calculation of the relative spectra revealed the increasing elasticity of this system compared to the 0.5% concentration, as confirmed by the higher values of the plateau modulus (491.6 Pa), shear viscosity (870.6 Pa s), and characteristic relaxation time (170.6 s.). This means that at 1% concentration, Sepineo is present in dispersion in a more "organized" structure, which implies more complex mechanisms of relaxation and higher polymer-polymer or polymer-solvent interactions despite the absence of a three-dimensional network (Fig. 4).

Concentrations from 3% to 5% showed very different mechanical spectra. As confirmed by the slope values very close to 0, these systems can be considered as gels from a rheological point of view, and in this case, the spectra were typical of a solid-like sample with a three-dimensional network structure.

Once again, these results confirmed that the rheological behavior depended strongly on polymer concentration. Increased concentrations from 0.5% to 5% gave rise to a progressive change in system mechanical spectra, which could lead one to think that increasing chain interactions and formation of entanglements foster the development of connectivity across the entire system until a network is formed.

Rheological Analysis of the Emulsions

Based on the rheological characterization of the different gelled systems, the 3% (*w/w*) concentration was selected as the external dispersing phase for the preparation of the emulsions. In fact, the 0.5% and 1% samples were not stiff enough, while the considerable elasticity of the 5% system could compromise correct emulsion formation. As mentioned in the "MATERIALS AND METHODS", almond oil was

added in order to assess the influence of an oily phase on the rheology of the dispersing phase and the stability of the emulsions themselves.

The stress sweep test was used to follow emulsion stability because the elastic modulus slowly decreases when the dispersed droplets become bigger but less numerous (24–27).

The stress sweep analyses performed 1 day after the preparation of these emulsions, and then at 1 and 3 months, showed good stability (Table I). It is important to note that the dynamic moduli values were just slightly higher than those found for the corresponding Sepineo gel, proving that the addition of the oily phase did not affect Sepineo rheological behavior too much (Table I).

The time sweep performed to check sample recovery after shear showed that emulsions were unable to recover their structure during the test, as confirmed by the smaller moduli values (data not shown). This indicated a degree of sample deformability after shear, which can improve system spreading and absorption, making it easy to rub the sample into the skin after topical administration (28,29).

The frequency sweep tests (Fig. 5), performed 1 day after preparation, confirmed the behavior previously observed from the stress sweep: The almond oily phase did not substantially affect the mechanical spectrum of the Sepineo aqueous phase, showing a very similar dependence on frequency for both moduli as revealed by the slope values in Table III.

Furthermore, the emulsion behavior with temperature (data not shown) confirmed that the systems were not affected by temperature, as already observed for the Sepineo gel.

Acoustic Spectroscopy Analysis of the Gels

Acoustic spectroscopy afforded a microrheological characterization of the gel samples. It is important to note that the dynamic moduli obtained from acoustic spectroscopy measurements are not comparable with those derived from rotational rheometers working in the oscillation mode, since the applied stress is not tangential, as in an oscillatory experiment, but "normal," and the tested frequencies are much higher.

In any case, analysis of the rheological parameters and, in particular, of the G'' modulus (Fig. 6), which is the frequency-dependent modulus, showed some differences within the various systems, in agreement with the oscillatory results. Basically, the spectra of the 0.5% and 1% Sepineo

Table III. Slope Values Calculated by the Linear Fitting of the G' and G'' Average Curves Obtained from the Frequency Sweep of the 0.5–5% Sepineo Gels and of the Sepineo 3%/Almond Oil Emulsion

	Slopes G'	Slopes G"
0.5% gel	1.568	0.696
1% gel	0.243	0.272
3% gel	0.056	0.115
5% gel	0.060	0.102
Emulsion	0.057	0.120

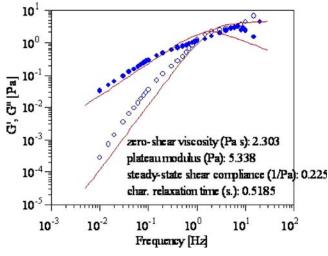


Fig. 3. Frequency sweep test of Sepineo P 600 systems 0.5% (*w/w*) with Doi–Edwards spectra fitting (*continuous line*)

samples were identical and characterized by a linear dependence on frequency. On the other hand, this linear trend was followed by a sharp modulus increase at lower frequency when the Sepineo concentration rose to 3% and 5% (w/w). The behavior of the latter two concentrations may seem unusual compared to the classical rheological trend, due to the fact that during the tests, the polymer chains showed significant modification in their deformability. This greater deformability at low frequency values indicates that the polymer/polymer interactions responsible for the gel-like behavior were not particularly strong. These interactions showed short-lasting elastic properties that probably failed when the system was stressed for a longer time. This change in the system structural characteristics modified the mechanisms of interaction between the sound and the samples (see "MATERIALS AND METHOD") and consequently changed the resultant attenuation parameter. The structural contribution, which is characteristic of gel-like systems, was dominant at higher frequencies. At low frequencies, the

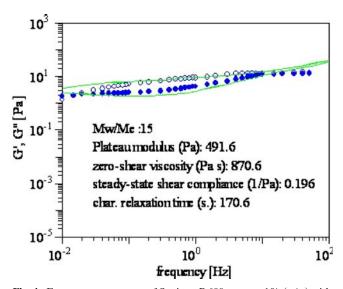


Fig. 4. Frequency sweep test of Sepineo P 600 systems 1% (*w/w*) with tube dilation spectra fitting (*continuous line*)

progressive loss of the network structure made the viscous contributions more important, giving rise to the remarkable increase of the G'' modulus.

On the other hand, the G' modulus did not show significant differences among the various systems, even though the 3% (w/w) and particularly the 5% concentrations showed slightly higher values for this modulus (Fig. 6). In this case, the increase in the density and sound speed of the systems at greater polymer concentration could explain the results obtained (see Eq. 1).

Acoustic Spectroscopy Analysis of the Emulsions

Ultrasound technique was used to monitor emulsion microrheological behavior and stability by considering the variation of both their rheological parameters and particle size (such as mean diameter).

The frequency spectra used to analyze system stability over time (Fig. 7) confirmed the stability of the emulsion. In fact, the G' and G'' plots obtained after 1 day, 1 month, and 3 months were very similar except for the G'' modulus at low frequencies. These results are in agreement with the oscillatory analysis results that revealed good stability for the emulsion.

It is interesting to compare the G'' spectra between the gel and the emulsion. As can be seen from Figs. 6 and 7, the emulsion system in the high frequency range exhibited quite similar behavior, with slightly greater G'' values compared to the gel. The presence of the oily phase caused greater frequency dependence, which can be explained by a more organized structure and consequently greater initial energy dissipation during the frequency sweep. As mentioned in the introduction, the oil in the emulsion–gel formation mainly served as an interface able to interact with the gel matrix, providing general improvement in the system rheological characteristics.

Instead, a different behavior was present in the low frequency region, where the curve related to the system analyzed after 1 day revealed a trend very similar to that of the corresponding Sepineo sample (3% *w/w* concentration), which showed, as previously mentioned, a certain deformability. After 1 and, in particular, after 3 months, this deformability gradually decreased, indicating a progressive interfacial stabilization of the emulsion. On the other hand, the G' modulus can be considered practically identical between gel and emulsion (Figs. 6 and 7).

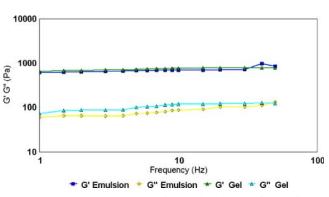


Fig. 5. Frequency sweep of Sepineo P 600/almond oil emulsion

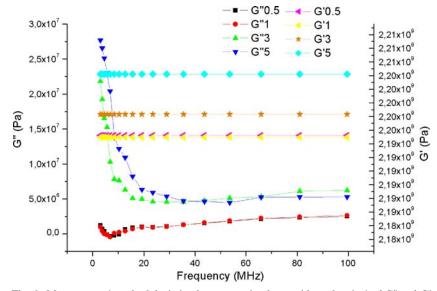


Fig. 6. Mean curves (standard deviation bars are omitted to avoid overlapping) of G'' and G' moduli from the acoustic spectroscopy of the different Sepineo P 600 concentration samples (0.5–5% w/w)

The results obtained from the analysis of the particle size (mean diameter values in micrometer) confirmed the previous statements concerning the stability of the emulsion. Mean diameter values (micrometer) of the dispersed almond oil droplets at the storage time of 1 day, 1 month, and 3 months were, respectively, 1.567 ± 0.14 , 1.592 ± 0.22 , and 1.578 ± 0.25 .

well, a property that depends strongly on polymer concentration. Concentration increases from 0.5% (*w/w*) to 5% (*w/w*) modified the viscoelastic properties of the Sepineo samples, changing the typical behavior of a concentrated non-entangled solution to that of a "gel-like" sample. On the other hand, the microrheological parameters obtained from acoustic spectroscopy showed that the physical interactions forming this gel-like structure were not particularly strong.

CONCLUSION

Oscillatory rheology and acoustic spectroscopy analyses were utilized in conjunction in order to characterize the Sepineo gel systems as well as to study how the addition of oil affects gel characteristics and the final stability of the resultant emulsion. Both techniques revealed that Sepineo P 600 thickens and gels Concerning the emulsions, the most important result is surely the fact that the addition of an oily phase increased system consistency only minimally. The viscoelastic characteristics depended exclusively on the gel structure.

In conclusion, Sepineo P 600 is a prime candidate for use in the formulation of gels and emulsion gels with rheological properties suitable for topical administration.

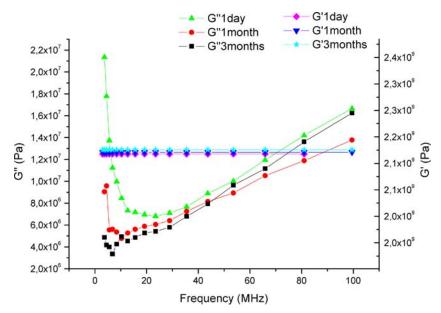


Fig. 7. Mean curves of G" and G' moduli from the acoustic spectroscopy of Sepineo/ almond oil emulsion

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REFERENCES

- Reiffers-Magnani CK, Cuq JL, Watzke HJ. Composite structure formation in whey protein stabilized O/W emulsions. I. Influence of the dispersed phase on the viscoelastic properties. Food Hydrocoll 1997;13:303–16.
- Chen J, Dickinson E, Langton M, Hermansson AM. Mechanical properties and microstructure of heat-set whey protein emulsion gels: effect of emulsifiers. Lebensm-Wiss U-Technol 2000;33:299– 307.
- Dickinson E, Merino LM. Effect of sugars on the rheological properties of acid caseinate-stabilized emulsion gel. Food Hydrocoll 2002;16:321–33.
- Chen J, Dickinson E. Viscoelastic properties of protein-stabilized emulsions: effects of protein-surfactant interactions. J Agric Food Chem 1998;46:91–7.
- Dickinson E. Milk protein interfacial layers and the relationship to emulsion stability and rheology. Colloids Surf B Biointerfaces 2001;20:197–210.
- Kerstens S, Murray BS, Dickinson E. Microstructure of *b*lactoglobulin-stabilized emulsions containing non-ionic surfactant and excess free protein: Influence of heating. J Colloid Interface Sci 2006;296:332–41.
- Akiyama E, Kashimoto A, Fukuda K, Hotta H, Suzuki T, Kitsuki T. Thickening properties and emulsification mechanisms of new derivatives of polysaccharides in aqueous solution. J Colloid Interface Sci 2005;282:448–57.
- Landoll LM. Nonionic polymer surfactants. J Polym Sci Polym Chem Ed 1982;20:443–55.
- Langley KR. Functional aspects of particle-matrix interactions in composite foods. Food Qual Prefer 1990;2:111–5.
- Chen J, Dickinson E. Effect of surface character of filler particles on rheology of heat-set protein emulsion gel. Colloids Surf B Biointerfaces 1999;12:373–81.
- Van Vliet T. Rheological properties of filled gels. Influence of filler matrix interactions. Colloid Polym Sci 1998;266:518–24.
- Dickinson E, Hong ST. Influence of water-soluble non-ionic emulsifier on the rheology of heat-set protein stabilized emulsion gels. J Agric Food Chem 1995;43:2560–6.
- McClements DJ, Monahan FJ, Kinsella JE. Effect of emulsion droplets on the rheology of whey protein isolate gels. J Texture Stud 1993;24:411–22.
- Seppic brochure. Sepineo P600 an emulsifying/thickening polymer for new sensations. http://www.seppic.com

- Dukhin AS, Goetz PJ. Ultrasound for characterizing colloids. Particle sizing, zeta potential, rheology. The Netherlands: Elsevier; 2002. p. 75–144.
- Litovitz TA, Davis CM. Structural and shear relaxation in liquids. In: Mason WP, editor. Physical acoustics. New York: Academic; 1964. p. 285–90.
- 17. Goodwin JW, Hughes RW. Rheology for chemists. Cambridge: The Royal Society of Chemistry; 2000. p. 146–212.
- Doy M, Edwards SF. The theory of polymer dynamics. Oxford: Oxford University Press; 1986.
- 19. De Gennes PG. Scaling concepts in polymer physics. Ithaca, NY: Cornell University Press; 1979.
- Ebert U, Schäfer L, Baumgärtner A. Segment motion in the reptation model of polymer dynamics. I. Analytical investigation. J Stat Phys 1998;90:1325–73.
- Milner ST, McLeish TCB. Reptation and contour length fluctuations in melts of linear polymers. Phys Rev Lett 1998;81:725–8.
- Milner ST, McLeish TCB. Arm-length dependence of stress relaxation in star polymer melts. Macromolecules 1998;31:7479– 82.
- Blottière B, McLeish TCB, Hakiki A, Young RN, Milner ST. Rheology and tube model theory of bimodal blends of star polymer melts. Macromolecules 1998;31:9295–304.
- Tadros T. Application of rheology for assessment and prediction of long-term physical stability of emulsions. Adv Colloid Interface Sci 2004;108-109:227–58.
- Korhonen M, Hellen L, Hirvonen J, Yliruusi J. Rheological properties of creams with four different surfactant combinations effect of storage time and conditions. Int J Pharm 2001;221:187–96.
- Chanamai R, McClements DJ. Dependence of creaming and rheology of monodisperse oil-in-water emulsions on droplet size and concentration. Colloids Surf A Physicochem Eng Asp 2000;172:79–86.
- Masmoudi H, Piccerelle P, Le Dréau Y, Kister J. A rheological method to evaluate the physical stability of highly viscous pharmaceutical oil-in-water emulsions. Pharm Res 2006;23 8:1937–47.
- Jones DB, Woolfson AD, Brown AF. Textural, viscoelastic and mucoadhesive properties of pharmaceutical gels composed of cellulose polymers. Int J Pharm 1997;151:223–33.
- Adeyeye MC, Jain AC, Ghorab MKM, Reilly WJ Jr. Viscoelastic evaluation of topical creams containing microcrystalline cellulose/sodium carboxymethyl cellulose as stabilizer. AAPS PharmSciTech 2002;3 2:1–10.