Formation and Novel Thermomechanical Processing of Biocompatible Soft Materials

Prashant D. Sawant and Xiang-Yang Liu*

Department of Physics, Lower Kent Ridge Crescent, National University of Singapore, Singapore, 119 260

Received December 11, 2001. Revised Manuscript Received April 22, 2002

We demonstrated for the first time the thermo-mechanical processing of the organogels of dibutyllauroyl glutamide as a small molecule gelling agent (SMGA) in a biocompatible solvent, isostearyl alcohol. The study focused on the impact of novel thermomechanical processing parameters such as temperature, strain, and the cooling rate on the microstructure and macroscopic properties of gels. The properties of the gels were analyzed by dynamic light scattering and by rheological and electron microscopy methods. The modification of the thermoreversibility and viscoelastic properties of the gels can be associated with the changes in the micro-/nanofiber network structures. The micro-/nanofiber network structures are stable over a wide range of frequencies. Two temperature domains associated with distinct gelation behaviors and rheological properties were identified for the first time in the gelation of organic solvents using SMGA. The transition from one temperature domain to the next can be linked to the changes in the micro-/nanostructure.

1. Introduction

Organogels are of great interest to materials scientists and industries. These soft materials find many applications in the areas of cosmetics, structured materials, and drug delivery[1] due to their thermoreversible[2] and viscoelastic nature. Other potential applications of organogels include media for fragrance delivery agents, inks, paints, and various displays (where ferroelectric or cholesteric liquid crystals are used).[3] The increasing demand for organogels has attracted significant attention in identifying new small-molecule gelling agent (SMGA) molecules.[4−6]

SMGA or low-mass gelling agents (LMGA) (of molecular weight ≤3000) can immobilize organic solvents so as to form organogels.[1,7−18] SMGA can be classified chemically in the following groups: fatty acids, steroids, and their derivatives, anthracene derivatives, cyclo-(dipeptides), and sorbitols.[19] These SMGA molecules can be used as gelling agents for almost all kinds of polar and nonpolar liquids. However, to the best our knowledge, the gelation of the long-chain alcohols such as the isostearyl alcohol by means of SMGA has so far not been reported. The gel formed in long-chain fatty alcohols can be used as a medium to deliver drugs; this has turned out to be of significant relevance in this field. Apart from this, the solvent, isostearyl alcohol, is found to be biocompatible, and since it is not harmful to the skin, it is used in skin care products.[20] Therefore, gelation of this biocompatible solvent using SMGA is an important step for forming novel skin care products based on gels of isostearyl alcohol or other long-chain, biocompatible solvents.

The inherent properties of gels such as hardness, elasticity, clarity, and liquid-carrying capacity are dependent on the microstructure of the fiber network

(1) (a) Fukasawa, J. I.; Tsutsumi, H.; Ishida, A. *Int. J. Cosmet. Sci.* **1989**, *11*, 153. (b) Ono, Y.; Nakajima, K.; Sano, M.; Kaneyiko, Y.; Inoue, K.; Hojo, J.; Shinkai, S. *Chem. Commun.* **1998**, 1477.

(2) Furman, I.; Weiss, R. G. *Langmuir* **1993**, *9*, 2084.

(3) (a) *Polymeric Gels: Fundamentals and Biomedical Applications*; Dessori, D., Kajiwara, K., Osada, Y., Yamauchi, A., Eds.; Plenum Press: New York, 1991. (b) Gunet, J.-M. *Thermoreversible Gelation of Polymers and Biopolymers*; Academic Press: London, 1992. (c) Lagaly, G., Ed. *Prog. Colloid. Polym. Sci.* **1996**, *102*. (d) Corriu, R. J. P.; Leclercq, D. *Angew. Chem.* **1996**, *108*, 1524; *Angew. Chem.* **1996**, *35*, 1420. (e) Reetz, M. T. *Adv. Mater.* **1997**, *9*, 943. (f) Osada, Y.; Gong, J.-P. *Adv. Mater.* **1998**, *10*, 827.

(4) Jung, J. H.; Shinkai, S. *J. Chem. Soc., Perkin Trans.* **2000**, *2*, 2393.

(5) Brotin, T.; Utermohlen, R.; Fages, F.; Bousa-Laurent, H.; Desvergne, J. P. *J. Chem. Soc., Chem. Commun.* **1991**, 417.

(6) Oda, R.; Huc, I.; Candau, S. J. *Angew. Chem., Int. Ed. Engl.* **1989**, *28*, 2689.

(7) Terech, P.; Pasquier, D.; Bordas, V.; Rossat, C. *Langmuir*, in press (acquired by ASAP Alert Services).

(8) Amaike, M.; Kobayashi, H.; Shinkai S. *Bull. Chem. Soc. Jpn* **2000**, *73*, 2553.

(9) Terech, P.; Rodrigez, V.; Barnes, J. D.; Mckenna, G. D. *Langmuir* **1994**, *10*, 3406.

(10) Terech P.; Weiss R. G. *Chem. Rev.* **1997**, *97* (8), 3133.

(11) Terech, P.; Furman, I.; Weiss, R. G.; Bounas-Laurent, H.; Desvergne, J. P.; Ramasseul, R. *Faraday Discuss.* **1995**, *101*, 345.

(12) (a) de Vries, E. J.; Kellog, R. M. *J. Chem. Soc., Chem. Commun.* **1993**, 238. (b) Sohna, J.-E., S.; Fages, F. *Chem. Commun.* **1997**, 327. (c) Hanabusa, K.; Maesaka, Y.; Kimura, M.; Shirai, H. *Tetrahedron Lett.* **1999**, *40*, 2385. (d) Kimura, T.; Yamashita, T.; Koumoto, K.; Shinkai, S. *Tetrahedron Lett.* **1999**, *40*, 6631, (e) Bhattacharya, S.; Acharya, S. N. G. *Chem. Mater.* **1999**, *11*, 3121.

(13) Abdallah, D. J.; Weiss, R. G. *Langmuir* **2000**, *16*, 352.

(14) Van der Schoot, P. *J. Phys. Chem. B* **1999**, *103*, 8804.

(15) Terech, P.; Furman, I.; Weiss, R. G. *J. Phys. Chem.* **1995**, *99*, 9558.

(16) Ilzhoefer, J. R.; Spontak, R. J. *Langmuir* **1995**, *11*, 3288.

(17) (a) Bhattacharya, S.; Acharyam S. N. G. *Chem. Mater.* **1999**, *11*, 3121. (b) Schoonbeek, F. S.; Van Esch, J. H.; Hulst, R.; Kellogg, R. M.; Feringa, B. L. *Chem.—Eur. J.* **2000**, *6* (14), 2633.

(18) Liu, X. Y.; Sawant, P. D. *App. Phys. Lett.* **2001**, *79*, 3518.

(19) Lin, Y. C.; Kachar, B.; Weiss, *J. Am. Chem. Soc.* **1989**, *111*, 5542.

(20) (a) Madhaven B. M. *Int. J. Toxicol.* **1999**, (Suppl. 3) *18*, 41. (b) Schwartzmiller, D. H.; Randen; N. A. U.S. Patent 6,200,596, Appl. No.: 186226, March 13, 2001. (c) Gagnebien, D.; Simon, P. U.S. Patent 6,150,422**,** Appl. No.: 815428, March 11, 1997.

10.1021/cm0116875 CCC: $22.00
Published on Web 08/27/2002

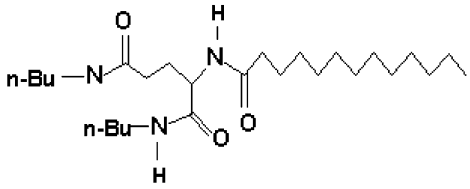

Figure 1. Chemical structure of *N*-lauroyl-l-glutamicacidd-ibutylamide or dibutyllauroylglutamic acid (DBLGA).

structure of SMGA, which in turn is determined by the mutual interactions of SMGA molecules, and organic solvent, supersaturation, and impurities. Recently, we have shown that the gelation of SMGA is controlled by a crystallographic mismatch branching that leads to the formation of the Caley fractal-like interconnecting fiber network structures in the liquid.[18] These networks form highly porous superstructures and can immobilize a large volume of liquid efficiently via capillary and other related forces.[14] It is known[21] that a SMGA can form a gel in one solvent, but may fail to form a gel in other isomeric solvents, or if formed, the network structure and properties may differ.

The three-dimensional micro-/nanostructures of the gels are quite sensitive to the stress, strain, temperature, and so forth. The thermomechanical processing conditions involving these parameters have influence on the microstructure formation and the macroscopic properties of the gels. Although a given gelator−solvent combination may or may not form a good gel, the emerging micro-/nanostructures and their inherent macroscopic properties can be modified by applying novel thermomechanical processing conditions. Thus, two key factors that control the fiber network structures and the macroscopic properties are the gelator−solvent match and the processing conditions.

To improve the gel properties by controlling the formation of fibers and fiber network structures via novel processing is a challenging task to both physicists and chemists. To the best of our knowledge, this task together with the gelation of long-chain alcohols have thus far not been not undertaken. Therefore, the objectives of this paper are as follows:

(a) To demonstrate the thermo-mechanical gelation of biocompatible, long-chain alcohols, such as stearyl alcohol, with SMGA and to study their characteristic thermomechanical properties.

(b) To study the effect of temperature, cooling rates, substrates, and shear on the gelation and properties of organogels.

To achieve these goals, we selected an isostearyl alcohol, which is a long-chain, biocompatible (skin-compatible)[20] solvent.

2. Experimental: Materials and Methods

N-Lauroyl-L-glutamic acid dibutylamide or dibutyllauroyl glutamide (DBLGA) (Figure 1), >99% pure, is purchased from the Ajinomoto Company, Japan. Isostearyl alcohol, (ISA), >99% pure, is from Cognis, U.S.A.

A strain-controlled dynamic mechanical spectroscopy method (model ARES, Rheometric Inc., U.S.A.) was used for the linear viscoelastic measurements. A parallel plate fixture of 25 mm with an in-between gap of 1.5 mm was used throughout all the experiments. A mixture of air and liquid nitrogen was used to control the cooling rate and the temperature. The concentration of DBLGA used was in the range of 4−10% w/v. The experimental errors in these experiments are found to be 1%.

Light-scattering experiments were carried out in a Brookhaven Dynamic/Static light-scattering system (model BI-200MS). A mixture of DBGLA and ISA is heated at 120 °C in an oven to obtain a liquid and immediately transferred to the light-scattering cell chamber, which was stabilized at the given temperature. The count rate obtained as a function of time is used to deduce the time of gelation (t_g). The experimental errors are found to be 1%.

A supercritical CO_2 ($Sc.CO_2$) extraction instrument from Thar Designs was employed to remove the organic solvent from the organogels without disturbing the fiber network structure.[16] The flow rate of $Sc.CO_2$ was set at 20 g/min at a temperature of 35 °C, for the duration of 1 h, to complete the extraction. These conditions are found to be optimal for the purpose of retaining the fiber network structures. The resultant dry fiber network powder, that is, xerogel, was obtained in the vessel and the solvent was separated from CO_2 in the cyclone separator.

Scanning electron microscopy (SEM) from JEOL JSM-5600LV was employed to examine the microstructure of the fiber networks. Fiber network structures obtained from $Sc.CO_2$ were coated with metal titanium (available source) for 60 s to achieve a better contrast.

3. Results and Discussion

3.1. Characteristic Thermomechanical Properties of DBLGA/ISA Gels. In this section, we study the gelation of long-chain alcohol and compare its properties with other existing organogels (particularly made of alcohols). We used the rheological methods, as they are fast and reliable in determining the viscoelastic properties and the time of gelation under different conditions.[22] The thermomechanical conditions used in these tests are optimized based on various preliminary tests (not discussed here).

The gelation time is identified from rheological and light-scattering methods. In dynamic rheology, a sudden increase in the elastic (G') and the viscous (G'') moduli of gels[23,24] and, in light scattering, an increase in count rates immediately after gel formation are considered to be the onset of the gelation process. The elastic modulus is found to be $\approx 10^7$ dyn/cm^2 for a gel of 6.7% w/v DBLGA in ISA, which is in accordance with other reported organogels.[11]

3.1.1. Strain Analysis of Gels. The effect of strain (γ) on G' for different concentrations of SMGAs is shown in Figure 2. DBLGA gels show nonlinear viscoelasticity and can withstand 0.5% of the applied strain (γ_o). This means that the three-dimensional microstructure can withstand a strain of 0.5%, showing no change in elasticity. However, the three-dimensional network cannot withstand a further increase in the applied strain and eventually it collapses. This is reflected in the decrease in the elastic modulus of the gel. This value of γ_o is comparable with reported values in organogels of other alcohols (with chain length less than 10 hydrocarbon units).[2,4,5,8,11,15] Thus, the gelation of DBLGA in ISA is competitive with other gels.

(21) Geiger, C.; Stanescu, M.; Chen, L.; Whitten, D. G. *Langmuir* **1999**, *15*, 2241.

(22) Terech, P.; Rossat, C.; Volino, F. *J. Colloid. Interface. Sci.* **2000**, *227*(2), 363.

(23) Almdal, K.; Dyre, J.; Hvidtand, S.; Cramer, O. *Polym. Gels Networks* **1993**, *1*, 5.

(24) *Viscoelastic Properties of Polymers*; Ferry, J. D., Ed.; Wiley: New York, 1980.

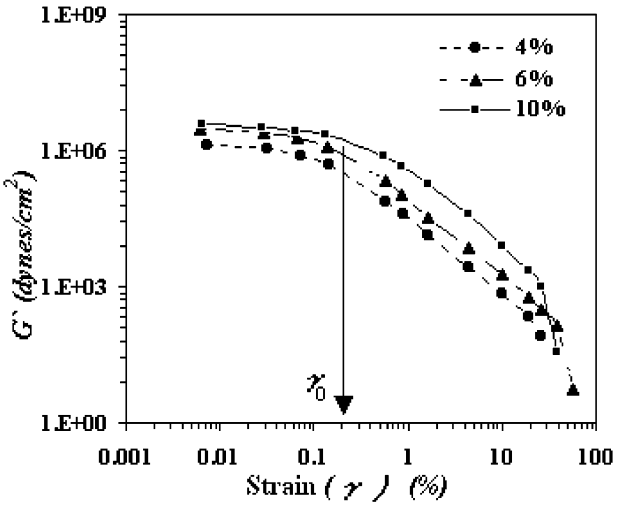

Figure 2. Log−log plot of shear moduli G' vs strain (γ), for 4, 6, and 10% (w/v) of DBLGA in ISA (frequency = 1 Hz, temperature = 20 °C).

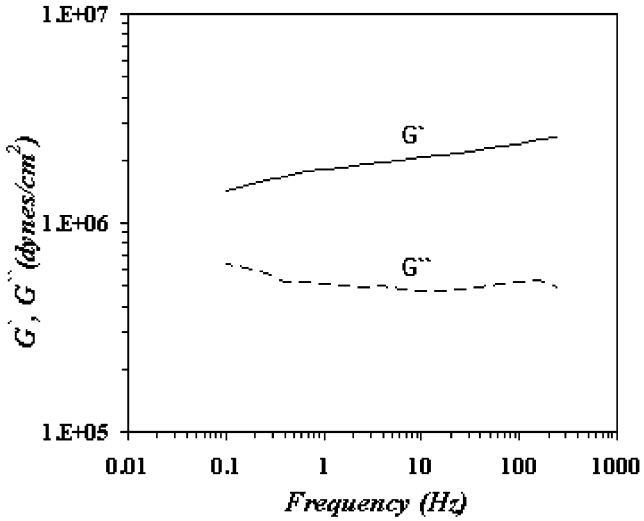

Figure 3. Log−log plot of shear moduli G' and G'' vs frequency (f), for 6.7% DBLGA in ISA obtained by the dynamic frequency test (strain = 0.1%, temperature = 20 °C.)

3.1.2. Effect of Frequency on the Gel Properties. The G' and G'' of the gels are fairly independent of frequency over a wide range of frequencies (Figure 3), indicating the near perfect Newtonian behavior of the solution and the strong elastic properties of the gel.[25] Therefore, DBLGA/ISA gels can be regarded as "true" gels.[25] The gel composed of DBLGA embedded in the long-chain alcohol is equally good compared with the gels made from other SMGA molecules employing relatively shorter chain length alcohols and other solvents.[1,7−17] The initial increase in frequency may be attributed to the structural rearrangement (of microdomains) until this adjusted system reaches a steady state.

The gelation of isostearyl alcohol with SMGA could be useful in biomedical and personal care products where medicines can be dissolved in ISA gels and applied on the skin or penetrate inside the skin. A detailed biocompatibility study of the present gel is to be published.

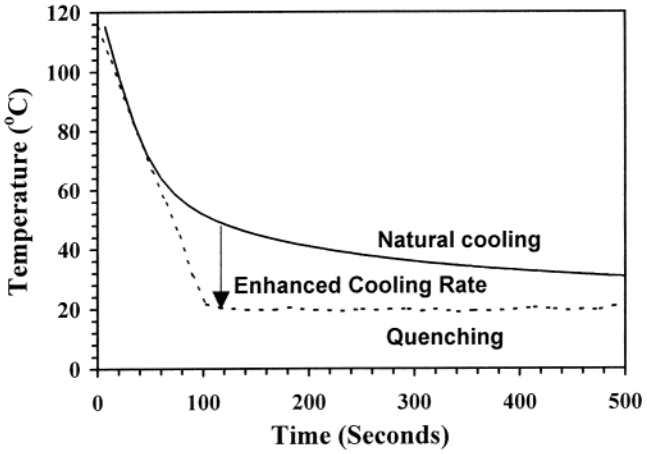

Figure 4. A quantitative comparison of cooling rates was deduced from natural cooling to room temperature, quenching to ambient temperature at the rate of 50 °C/min in situ rheometry.

Table 1. Effect of Cooling Rate on the Gelation Properties (Strain: 0.01%)

cooling rate (°C/min)	elastic modulus (G') (dyn/cm²)	time of gelation (t_g) (s)
10	665 810	235
50	2 300 000	198

The decrease in the elasticity (≈1−2%) observed in the successive cycles can be attributed to the same effect.

3.1.3. Crystallinity of Gels. The crystalline nature of the fiber network structures prepared under different thermomechanical conditions is deduced from X-ray diffraction.[28] The peaks obtained from powdered fiber network structures are similar to those obtained in natural DBLGA powder. Thus, the crystalline structure was maintained in all conditions.

3.2. Effect of Different Thermomechanical Conditions. *3.2.1. Effect of Cooling Rate.* Gels of 6.7% DBLGA in ISA are prepared at two different cooling rates, 10 and 50 °C/min at a frequency of 1 Hz and strain of 0.01%. We found that the the dynamic rheology reveal an increase in t_g and a decrease in elasticity, with an increase in the cooling rate (Table 1). This rheometric analysis shows that the cooling rate has a significant impact on the gelation and aesthetic properties, as described below.

The effect of the cooling rate on the appearance of the gels becomes apparent from the change in the clarity of the gels, by comparing the two gel bottles: naturally cooled to room temperature and quenched to attain 20 °C, respectively. The cooling rates obtained from natural cooling and quenching by rheometry are depicted in Figure 4.

The t_g for a naturally cooled gel is 1100 s, which lasts much longer than the 200 s obtained by quenching of the gel.

The improvement in the above properties can be understood as follows:

(25) (a) Morris, E. R.; Ross-Murphy, S. B. In *Techniques in Life Sciences*; Elsevier/North-Holland: Amsterdam, 1981; Vol. 3, Chapter B310. (b) Ross-Murphy, S. B. *J. Rheol.* **1995**, *39* (5), 1451.

(26) De Gennes, P. G. *Scaling Concept in Polymer Physics*; Cornell University Press: Ithaca, NY, 1979.

(27) Liu, X. Y. *J. Chem. Phys.* **2000**, *112*, 9949.

(28) Liu, X. Y.; Sawant, P. D. *Adv. Mater.* **2002**, *14*, 421.

As mentioned earlier, the formation of the interconnecting fiber network of SMGA is due to a crystallographic mismatch branching.[18] According to this mechanism,[18] the average branching distance $\langle\xi\rangle$ decreases as supercooling increases. A large instantaneous supercooling is obtained by the quenching of the gels (Figure 4), which leads to a denser and a more uniform branching of the fiber network, as predicted in ref 28 and shown in Figure 7b. Conversely, thicker and less extensively branched networks are obtained in the naturally cooled gel (see Figure 7a). Obviously, thinner and uniformly branched gels will scatter less light and consequently give rise to clarity.

3.2.2. Effect of Strain on Gelation. The applied strain is an important parameter in the process of gelation as it can affect the microstructure and macroscopic properties of the gels. The gelation of DBLGA in ISA is carried out at three different strains: 0.01%, 0.05%, and 1%. According to Figure 2, the gel structure can withstand an applied strain up to 0.5%. It was found that gelation continues to occur at a higher strain (1%); however, the network structures then become unstable. It is noteworthy that the gelation at a strain of 1% resulted in a longer t_g and a reduced elasticity (G') as compared to a strain of 0.05% (Table 2). Therefore, gel formation in the presence of high strain could be attributed to partial formation of the network structures. On the other hand, the gel formed at a very low strain of 0.01% does not show any substantial change in its properties when compared to gel formed at a strain of 0.05%. Therefore, it is advisable to carry out the gelation of DBGLA in ISA below the applied strain of 0.5%.

3.2.3. Effect of Processing Temperature on Gelation. Organogels of 6.7% w/v DBLGA in ISA were formed at different temperatures ranging from 20 to 40 °C. It is found that the time of gelation increased with temperature (Figure 5). Two temperature domains were observed, below and above 35 °C. The gelling time increased drastically above 40 °C. The elasticity (G') of the gels decreased by 44 and 73% when the temperature of gelation increased from 20 to 30 °C and 35 °C, respectively. This is attributed to the formation of more flexible fibers and loosely aggregated microstructures at higher temperatures and is consistent with the micro-/nanostructure of the gel formed at 40 °C (Figure 7c).

The change in the microstructure of the gels formed at different gelling temperatures is examined by dynamic stress relaxation tests (Figure 6). In general, strong gels do not withstand the applied strain. In contrast, an extremely weak gel or gel solution is not affected by the applied strain due to the so-called "thixotropic effect".[29] In the case of viscoelastic gels, the fiber network structures that become stretched by the applied strain are temporarily oriented in the direction of the strain and slowly regain their initial orientation when the strain is released. This effect is called "thixoelasticity". Therefore, thixoelasticity and thixotropy are a measure of the strength of the fiber network structures.

A strain of 1% is applied on the gels formed at 20 and 40 °C. We found that a slow decay to a constant value of an instantaneous stress undergone by a gel prepared at 20 °C is evidence of thixoelasticity.[29] The abrupt increase in stress is caused by stretching of the fiber networks, which subsequently slowly relax back to the original position ,exhibiting an exponential decay behavior in stress. Therefore, the instantaneous rise and subsequent decay of the stress with time reflect the viscoelastic nature of the gel.

In contrast, for gels formed at 40 °C, no prominent change was observed in the stress after applying strain (thixotropic effect). This thixotropic effect can be explained by the occurrence of more flexible fibers and fragile networks or the formation of loose aggregates of microdomains at high temperatures, as can be seen in the SEM micrograph (Figure 7c). These results are consistent with the change in elasticity as the temperature of gelation increases. It can be concluded from this analysis that the gel preparation at ambient tempera-

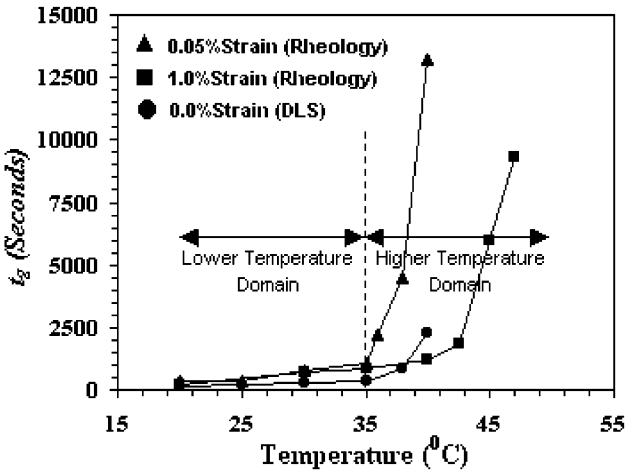

Figure 5. Effect of strains/temperatures on the gelation of DBLGA in ISA. The gelation in situ dynamic light scattering (DLS) is assumed to be free from the strain effect. However, the effect of the substrate must be present in the t_g measurements obtained from DLS and rheometry.

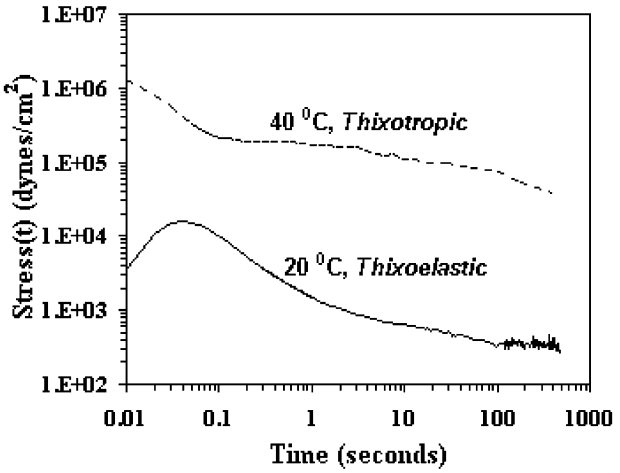

Figure 6. A log−log plot of the stress as a function of time obtained from the dynamic stress relaxation test carried out for 6.7% w/v DBLGA in ISA at different temperatures.

Table 2. Effect of Strain on the Gelation Properties of 6.7% w/v of DBLGA in ISA

strain (%)	G' (dyn/cm^2)	t_g (SEC)
0.01	2 300 000	198
0.05	2 300 000	200
0.10	670 000	185

(29) Quemada, D. *Eur. Phys. J.: Appl. Phys.* **1999**, *5*, 191.

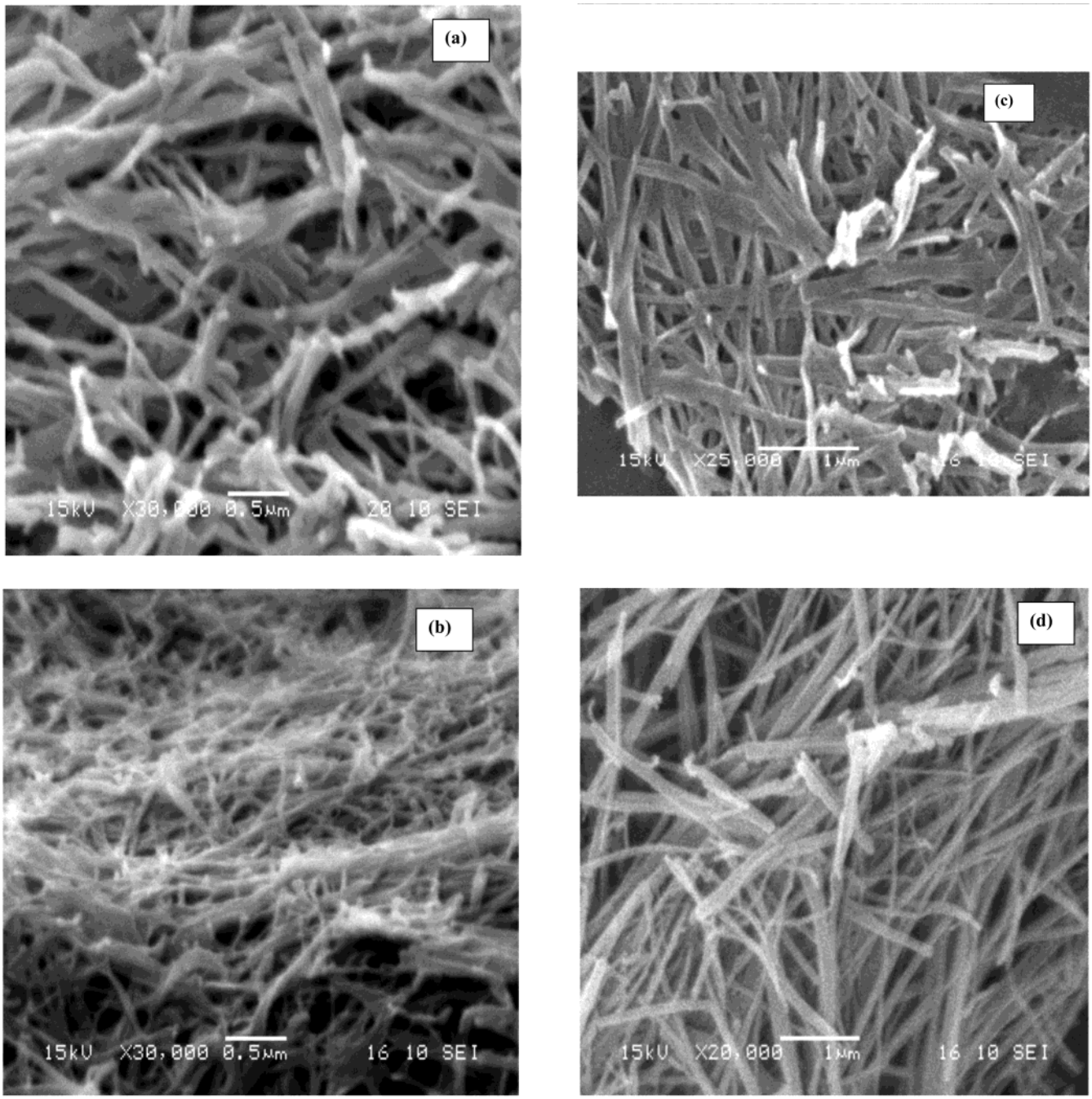

Figure 7. Electron micrographs of fiber network structures obtained under different thermomechanical conditions: (a) 6.7% GP-1/ISA, natural cool, bar = 0.5 μm, ×30 000, cooling rate = 50 °C/min; (b) 6.7% GP-1/ISA, 20 °C, 0.05% strain, bar = 0.5 μm, ×30 000, cooling rate = 50 °C/min; (c) 6.7% GP-1/ISA, 40 °C, 0.05% strain, bar = 1 μm, ×25 000, cooling rate = 50 °C/min; (d) 6.7% GP-1/ISA, 20 °C, 0.01% strain, cooling rate = 5 °C/min, bar = 1 μm, ×20 000.

ture is preferable when good viscoelastic properties are sought.

3.2.4. Effect of Substrate on Gelation. The effect of substrate on the gelation is deduced from the data given in Table 2 and Figure 5. Table 2 shows that the G' and t_g values did not change significantly for strains of 0.05 and 0.01%. Therefore, the t_g obtained at the strain of 0.05% from the rheology should be equivalent to that obtained by the light-scattering method in the lower temperature domain. The strain in the light-scattering method is considered to be zero. However, the t_g obtained from the light-scattering method (at 0% strain) was found to be lower compared to that with the rheology (at 0.05% strain). This is attributed to the substrate effect only. A glass cell used in the light-scattering method is different from the metal plates commonly used in the rheology. Such a difference will have a direct impact on the nucleation rate and consequently on the t_g measurements. It follows from Figure 5 that the wall of the glass cell turns out to be a better substrate for the nucleation of DBGLA fibers.

The substrate effect in the higher temperature domain is more complicated and difficult to explain both qualitatively and quantitatively because the thermal expansion coefficient of these substrates is different at higher temperatures.

3.3. Micro-/Nanostructure Analysis. Since gelation is the process of formation of a three-dimensional

network,[18] the network formation will determine the rheological and aesthetic properties of the gels. The thermomechanical parameters used in the process of gelation will affect the microstructure. The microstructure of organogels obtained under the following thermomechanical conditions is depicted in Figure 7. The concentration of DBLGA is fixed at 6.7% w/v for the microstructure analysis under different thermomechanical conditions. All these micrographs show the occurrence of micro-/nanometer fibers under different processing conditions.

Naturally cooled samples (Figure 7a) of 6.7% w/v DBLGA in ISA exhibit thicker fibers and a lower degree of network branching than the quenched samples (Figure 7b). On the other hand, quenching improves the thixoelasticity and the clarity of the gel due to formation of thinner fibers and more densely branched three-dimensional network structures. The microstructure of the gels formed at 40 °C (Figure 7c) shows short and thicker fiber network structures in combination with thixotropic properties.

The gel quenched to 20 °C at a low cooling rate (5 °C/min) also shows fibers rather similar to those observed in the naturally cooled gel (Figure 7d). This again confirms that the quenching at high cooling rates promotes the formation of thinner fibers, the characteristics of the three-dimensional microstructure and macroscopic properties.

In a nutshell, the microstructure properties and the macroscopic properties are the complementary aspects of the gels. The gelation at ambient temperatures and high cooling rates gives rise to more highly interconnecting three-dimensional network structures and, consequently, shows desirable macroscopic properties. This study also shows that selection of the gelator−solvent chemical match is not the sole parameter for deciding the properties of the gels. The novel thermomechanical controlling factors introduced in this work enable us to modify the microstructure and macroscopic rheological and aesthetic properties.

IV. Conclusions

The DBLGA can form a gel in the biocompatible, long-chain alcohols, showing all the characteristic properties of organogels such as thermoreversibility and nonlinear viscoelasticity. Gels are also formed at temperatures higher than room temperature and higher strains. Two temperature domains were observed, below and above 35 °C. Gels formed in the lower temperature domain possess better rheological and aesthetic properties. All gels contain fibrous network structures. However, the network structures formed in the high-temperature domain are not as strong as those formed at ambient temperatures. The differences in the measurement of time of gelation by dynamic light scattering and rheometry show the effects of the substrate and shear on the gelation. It is possible to modify the microstructure and in turn enhance the rheological and aesthetic properties of the gels.

CM0116875