# SUPPORTING INFORMATION

## Dynamics and Rheology of Supramolecular Assemblies at Elevated Pressures

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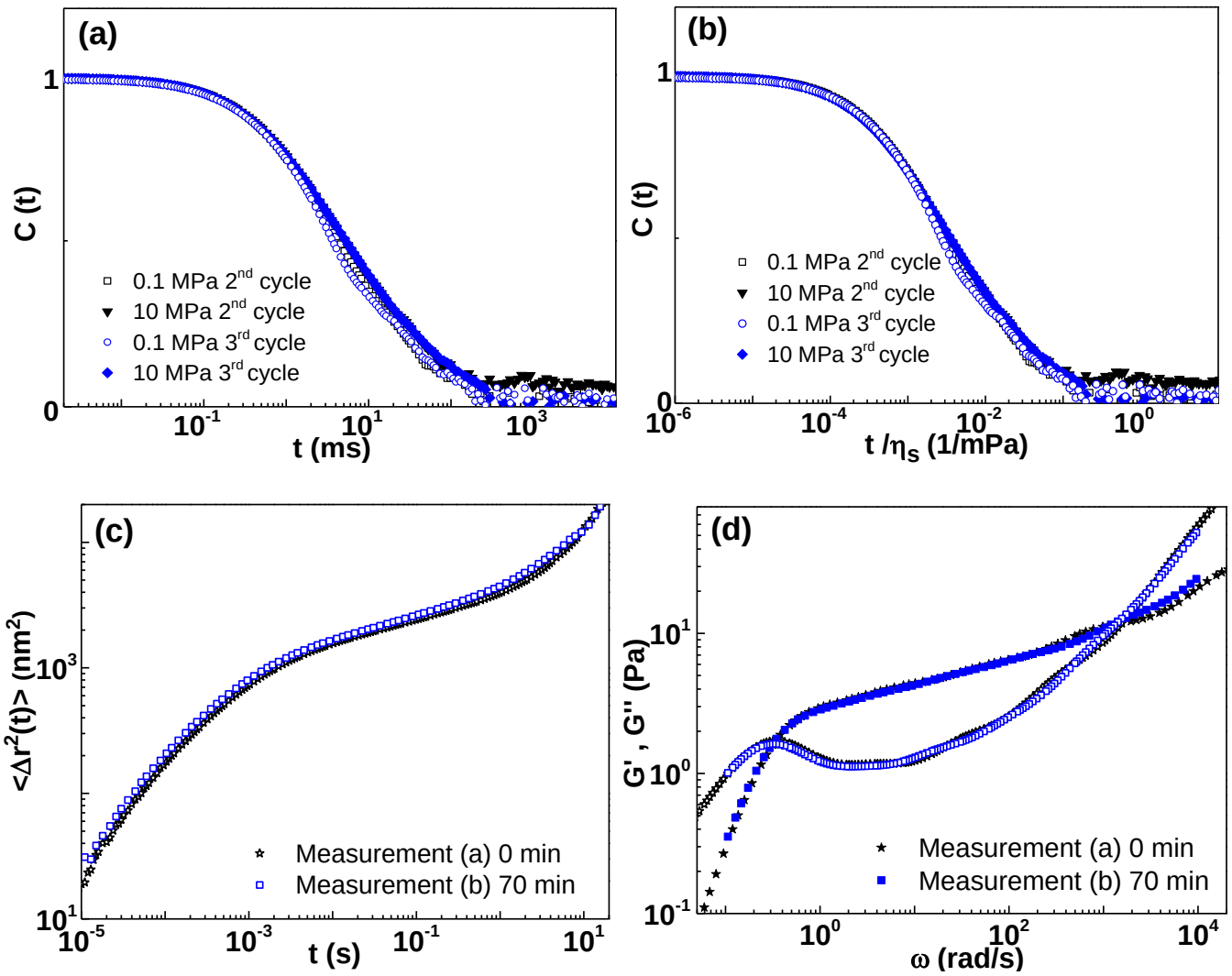



**Figure S1:** Intermediate scattering function C(t) of EHUT/dodecane solutions at 25°C, 1 g/L and two pressures, 0.1 (open symbols) 10 MPa (filled symbols) measured at different waiting time times under pressure conditions. (a) repeat cycles of compression/decompression between 0.1 and 10 MPa (squares and inverted triangles correspond to the 2nd cycle, circles and diamonds to the 3rd cycle). (b) Same data as in (a) but with the time being scaled with the solvent viscosity. (c) Mean square displacement of an EHUT/dodecane solution with added PMMA tracers for a concentration of 3 g/L, at 10 MPa and 25°C. Data are taken at two different times which are 70 min apart. Measurement duration is 4200 s. (d) respective linear viscoelastic spectra.

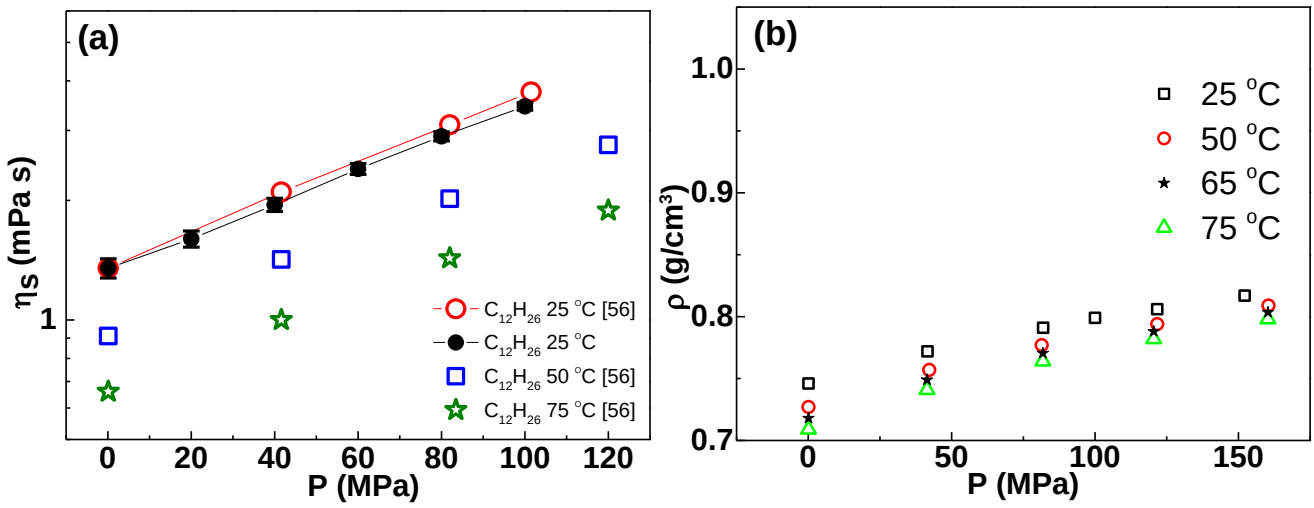



**Figure S2:** (a) Solvent viscosities of dodecane as a function of pressure at different temperatures (see legend). Red open circles, blue squares and green stars correspond to 25°C, 50°C and 75°C, respectively [Ref. 56 of main text]. Black filled circles represent data extracted from microrheology measurements with dilute PMMA suspensions in dodecane (see text). The lines are drawn to guide the eye. (b) Dodecane densities as a function of pressure, extracted from [56].

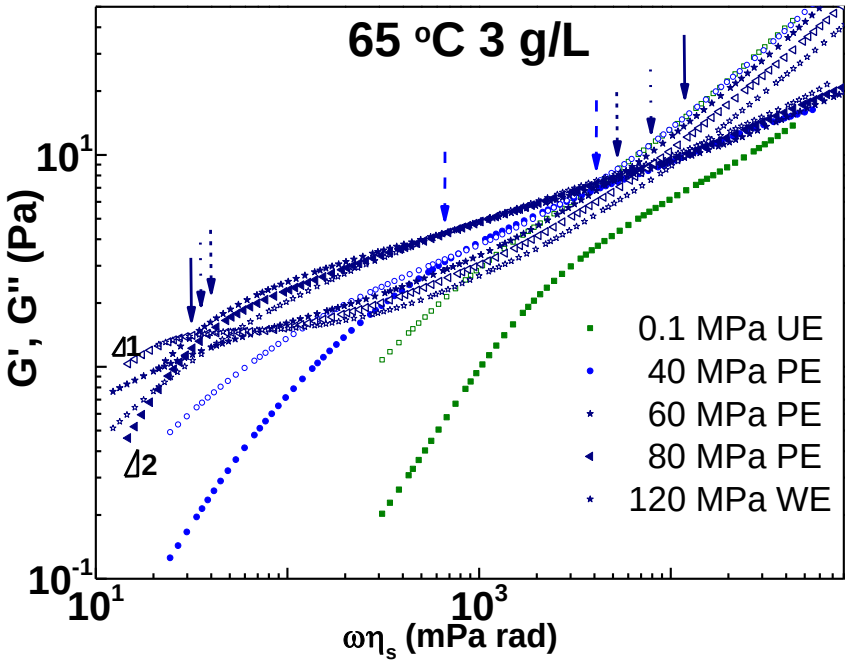



**Figure S3:** Microrheological G' and G'' as a function of frequency (scaled with the solvent viscosity) for EHUT/dodecane at c=3 g/L, T=65°C and different pressures (indicated in the legend). G' data are represented by filled symbols and G'' data by open symbols. Vertical arrows mark the high- and low-frequency moduli crossover. Terminal slopes are also indicated, while the state of the different solutions (UE, PE, WE) is mentioned in the legend (see main text).

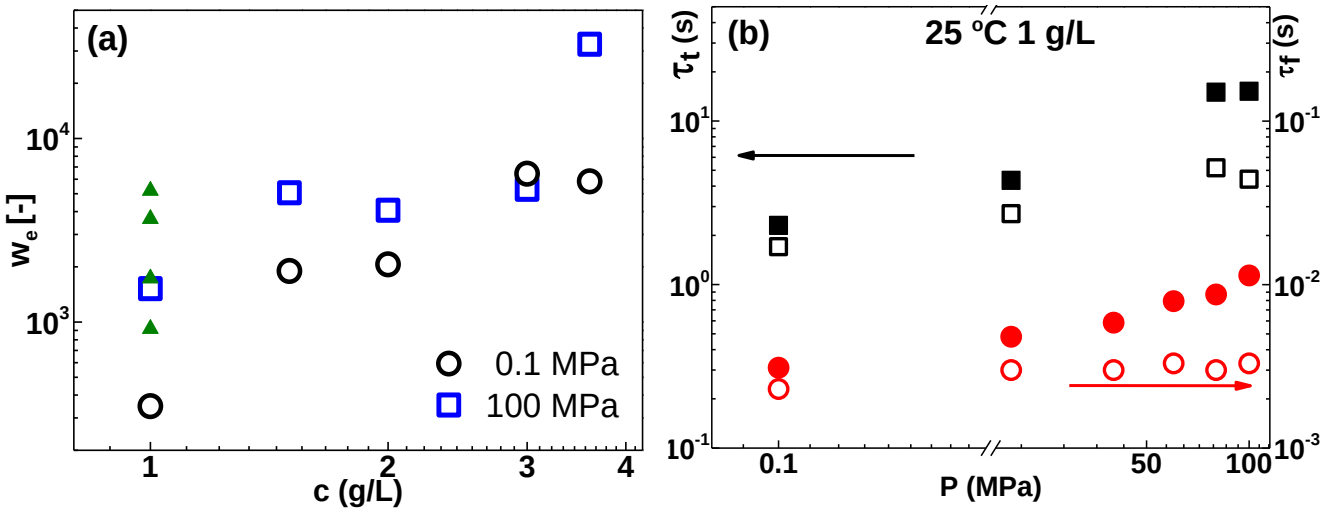
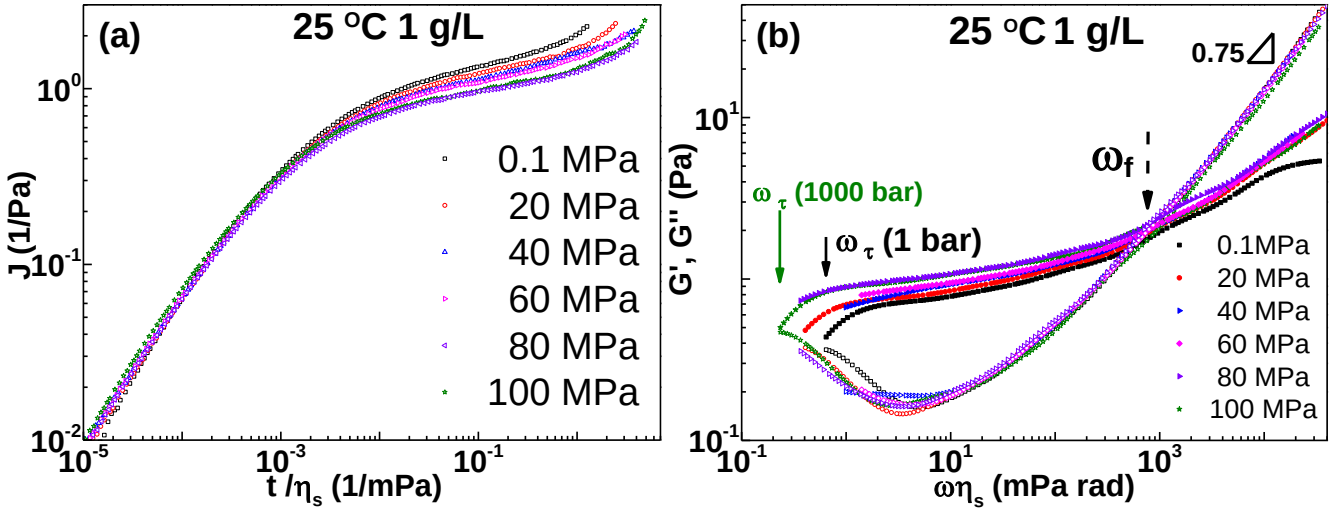



**Figure S4:** (a) Ratio of high- to low-frequency moduli crossover ($w_e$) as a function of concentration for 0.1 MPa (black circles) and 100 MPa (blue squares). The green triangles represent different pressures (0.1, 20, 80, 100 MPa) and are extracted from (b). (b) Terminal time (squares, left axis), fast time (circles, right axis) as a function of pressure of EHUT/dodecane 1 g/L at 25°C. The open symbols are the respective times which have been rescaled by solvent viscosity.



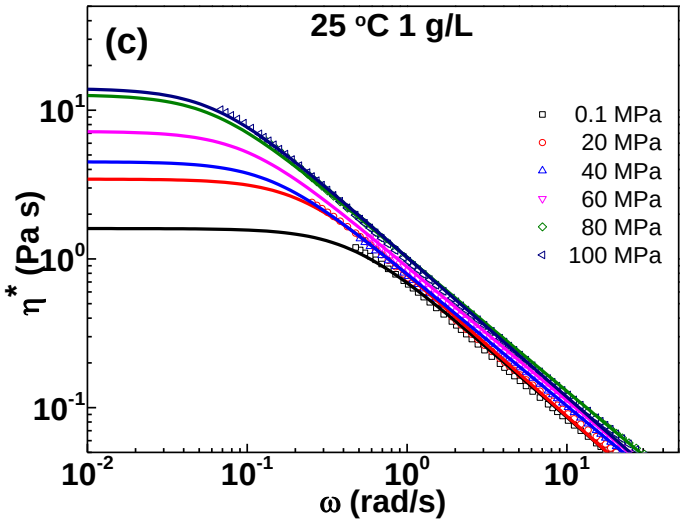



**Figure S5:** (a) Creep compliance and (b) respective frequency-dependent storage G' (filled symbols) and loss G'' moduli (open symbols) for 1 g/L EHUT/dodecane solutions at different pressures (see legend) and 25°C. Times in (a) and frequencies in (b) are scaled with the solvent viscosity. (c) Respective data of complex viscosity along with the Carreau fits (lines) to extract the zero-shear viscosity $\eta_0$ (the infinite-rate viscosity is taken to be zero, $\tau_c$ reflects the time at the onset of shear thinning, i.e., it is essentially the terminal relaxation time, and the parameters $\alpha$ and n adjust the thinning part) reported in the main text:

$$\frac{\eta\left(\dot{\gamma}\right)-\eta_\infty}{\left(\eta_0-\eta_\infty\right)}=\left(1+\left[\dot{\gamma}\,\tau_c\right]^\alpha\right)^{(n-1)/\alpha}$$

Experimental account of humidity effects: In order to assess the role of humidity, we loaded the closed sample cell to a closed beaker (filled with water) and we left it for 36 hours. At this point the relative humidity was constant at about 93%. The first microrheological data under "ambient conditions" are shown in Fig.S5 (bordeaux squares). Then, we removed the cup of the sample cell and measurements took place at different times (3,5,19 hours), corresponding to different humidity levels. During each measurement we added the cup to reduce changes of humidity during the measurement.

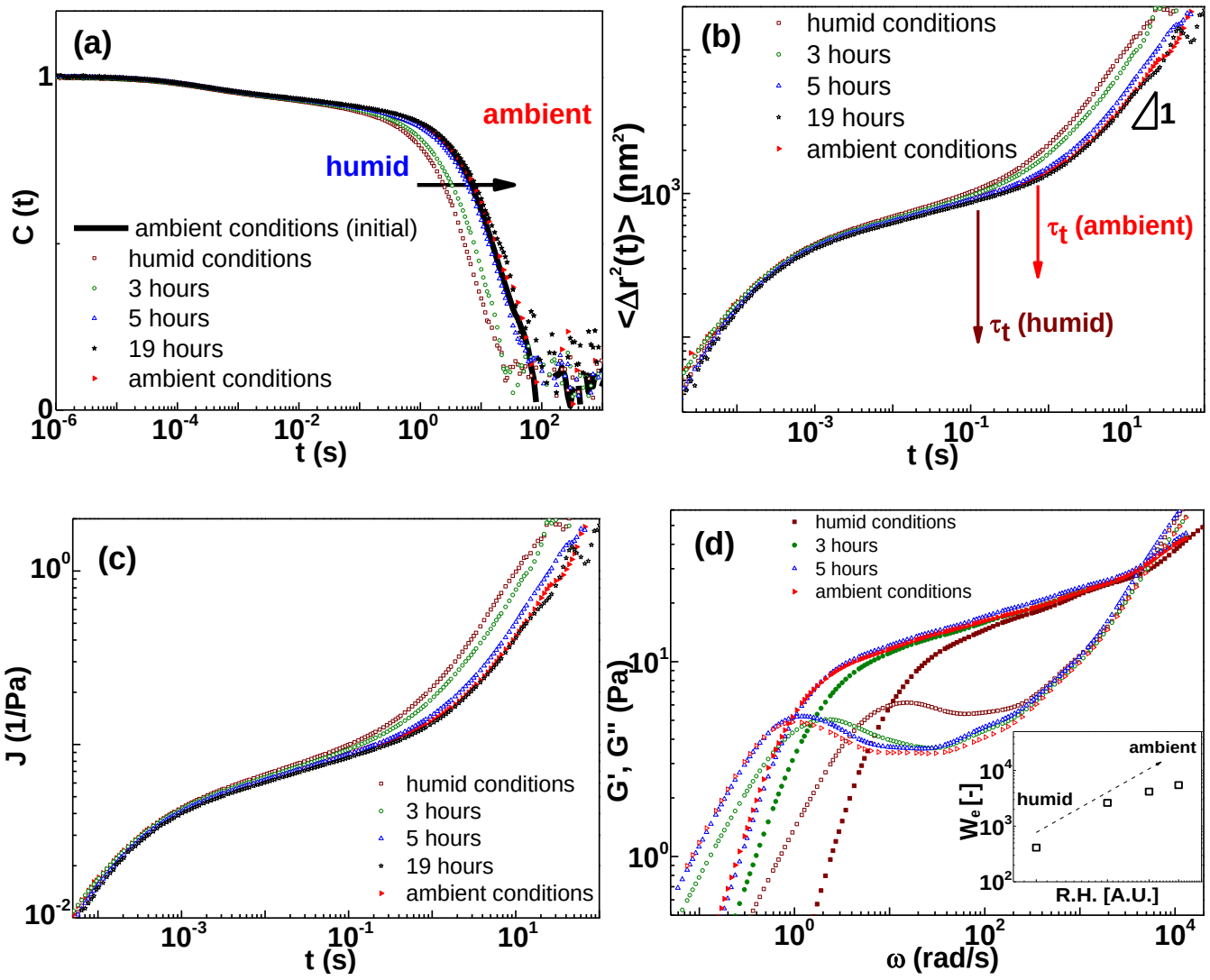



**Figure S6.** Effects of humidity on the dynamics of EHUT/dodecane solutions (with added PMMA probes), at c=3.6 g/L, T=25°C and P=0.1 MPa. Intermediate scattering correlation function (a) and Mean Square Displacement (b) at different humidity contents (corresponding to different time of measurement); $\tau_t$, indicated by arrow, is the terminal relaxation time (marking the onset of diffusion with slope of 1). Ambient conditions (reference measurements) correspond to relative humidity of about 43 %. Humid conditions correspond to relative humidity of about 93 %. Intermediate measurements (t = 3, 5 and 19 hours) were conducted upon removing the top cup of the sample cell (being at ambient conditions) in order to expose the sample to the external environmental conditions. Between measurements at different times the cup was put back, and the sealed sample had different humidity compared to ambient. The black line in (a) corresponds to the reference measurement at ambient conditions before any change of the humidity. Respective creep compliance data (c) and frequency-dependent linear viscoelastic moduli, G' (filled symbols) and G'' (open symbols) (d) are also shown.

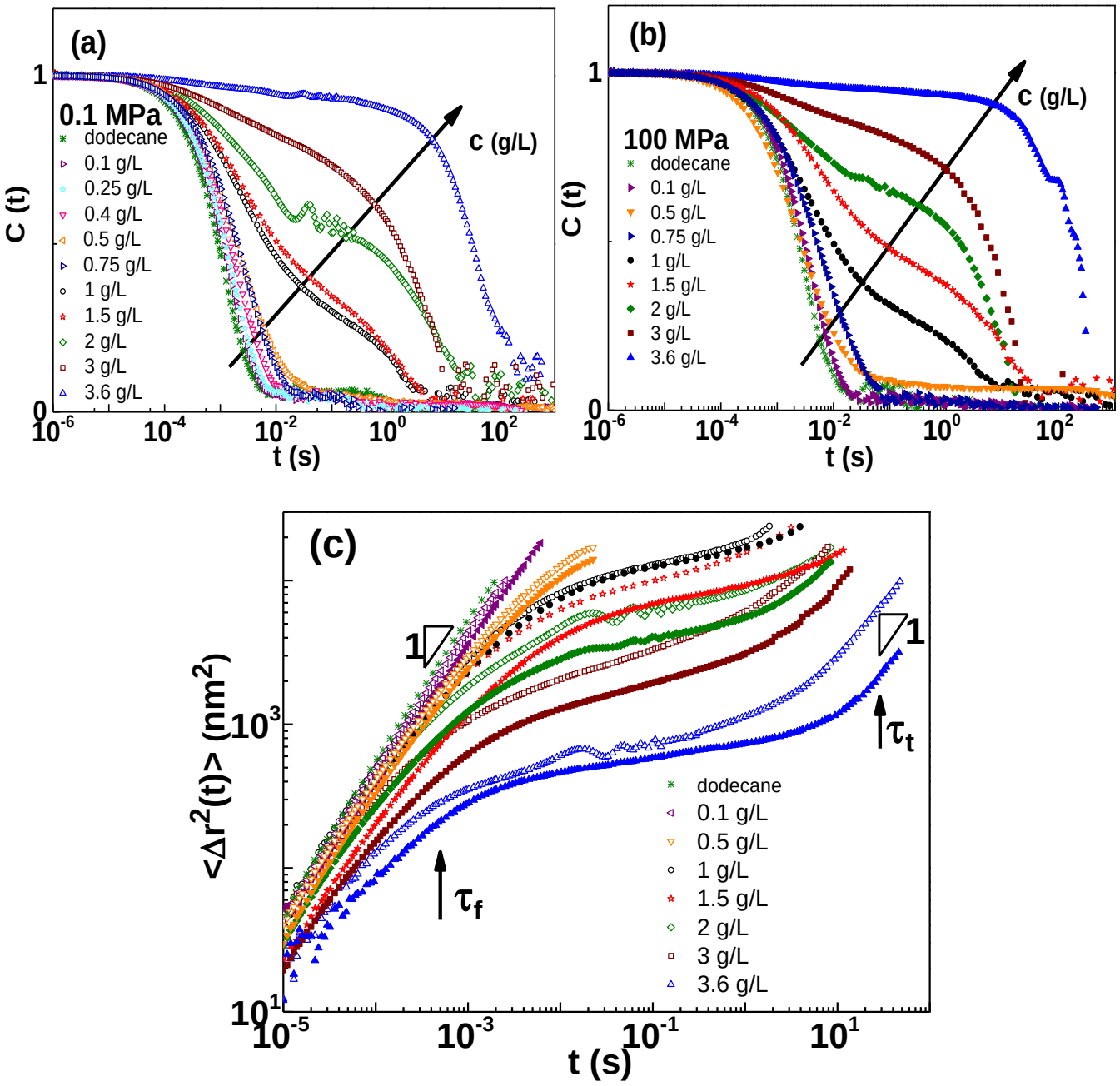



**Figure S7:** Intermediate scattering function C(t) of EHUT/dodecane solutions with added PMMA probes at 25°C and different pressures, (a) 0.1 MPa (open symbols) and (b) 100 MPa (filled symbols), at different concentrations (see legend). (c) Respective mean square displacement data for 0.1 (open symbols) and 100 MPa (filled symbols). The data for the pure solvent (with the PMMA tracer particles) are also shown for reference.

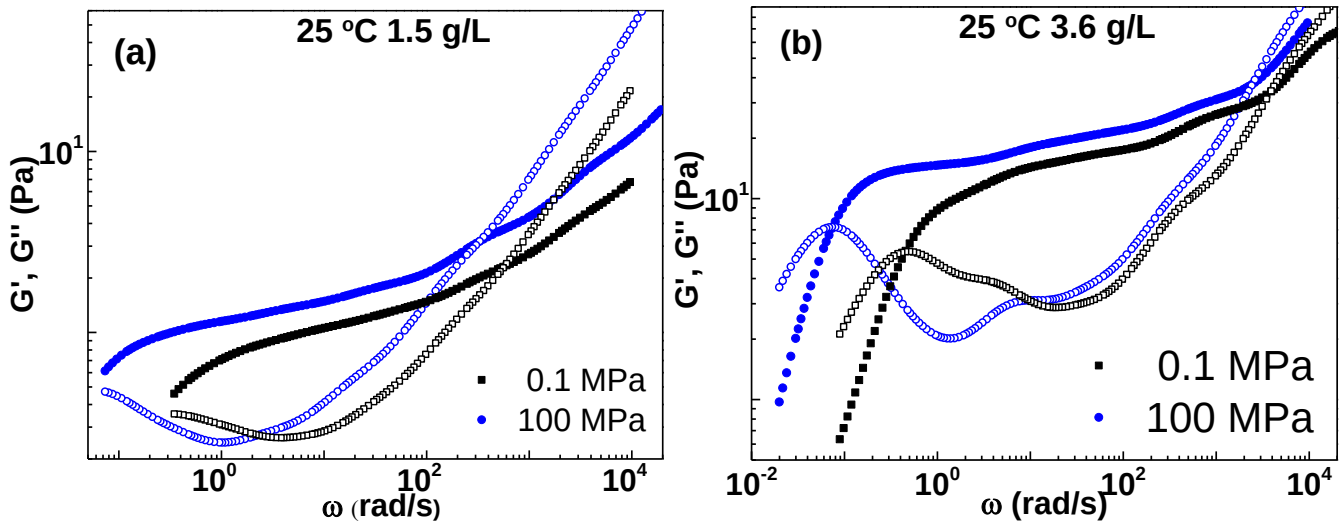



**Figure S8:** Frequency-dependent storage G' (filled symbols) and loss G'' (open symbols) moduli for EHUT/dodecane solutions at 25°C, 0.1 MPa (black squares) and 100 MPa (blue circles) and different concentrations, 1.5 g/L (a) and 3.6 g/L (b).

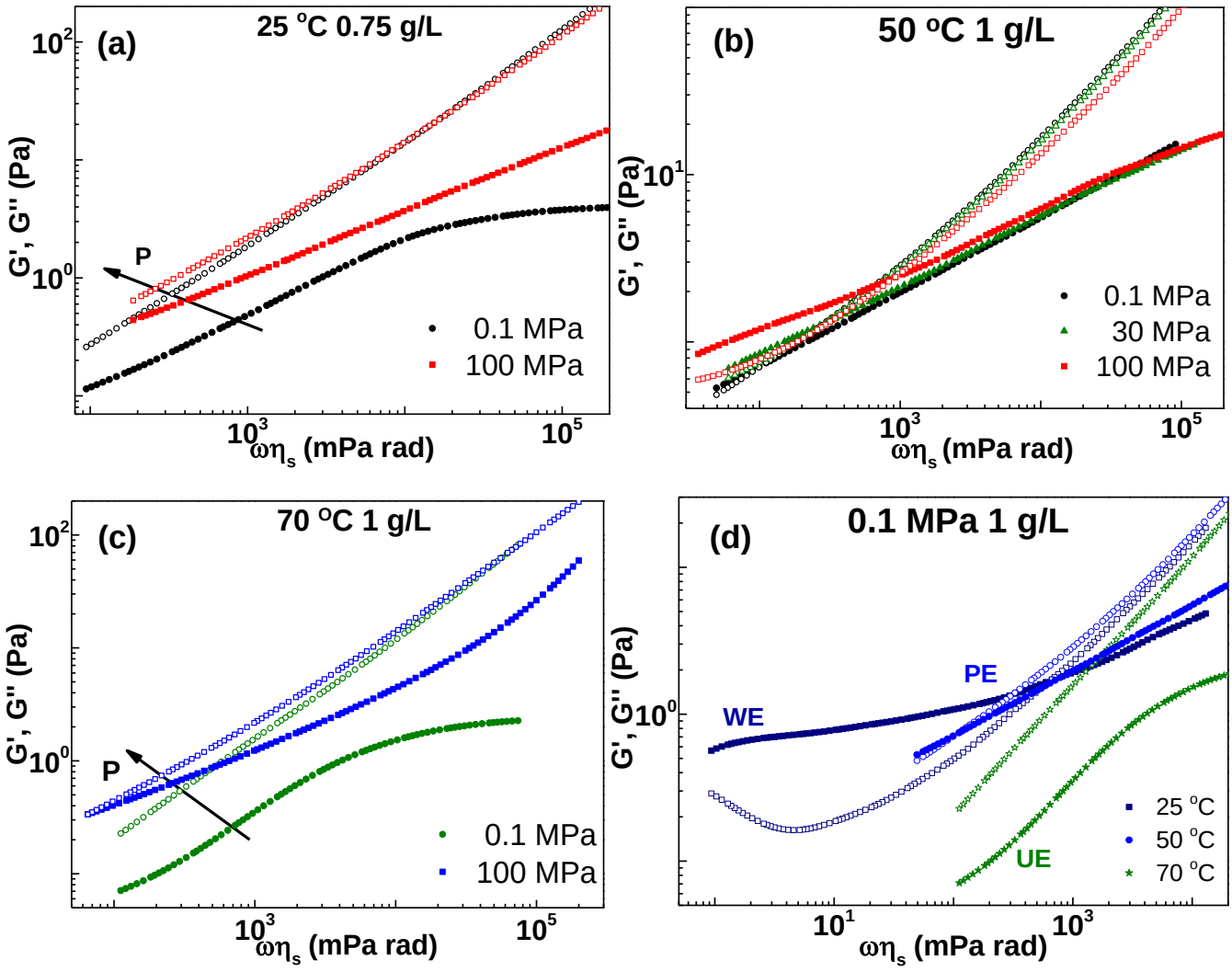



**Figure S9:** Linear viscoelastic spectra of EHUT/dodecane at (a) 25°C, 0.75 g/L, 0.1 MPa (circles) and 100 MPa (squares); (b) 50°C, 1 g/L, 0.1 MPa (circles), 30 MPa (triangles), 100 MPa (squares) ; (c) 70°C, 1 g/L, 0.1 MPa (circles), 120 MPa (squares) ; (d) 1 g/L, 0.1MPa, 25°C (WE state), 50°C (PE state) and 70°C (UE state). The frequencies are scaled with the solvent viscosity.

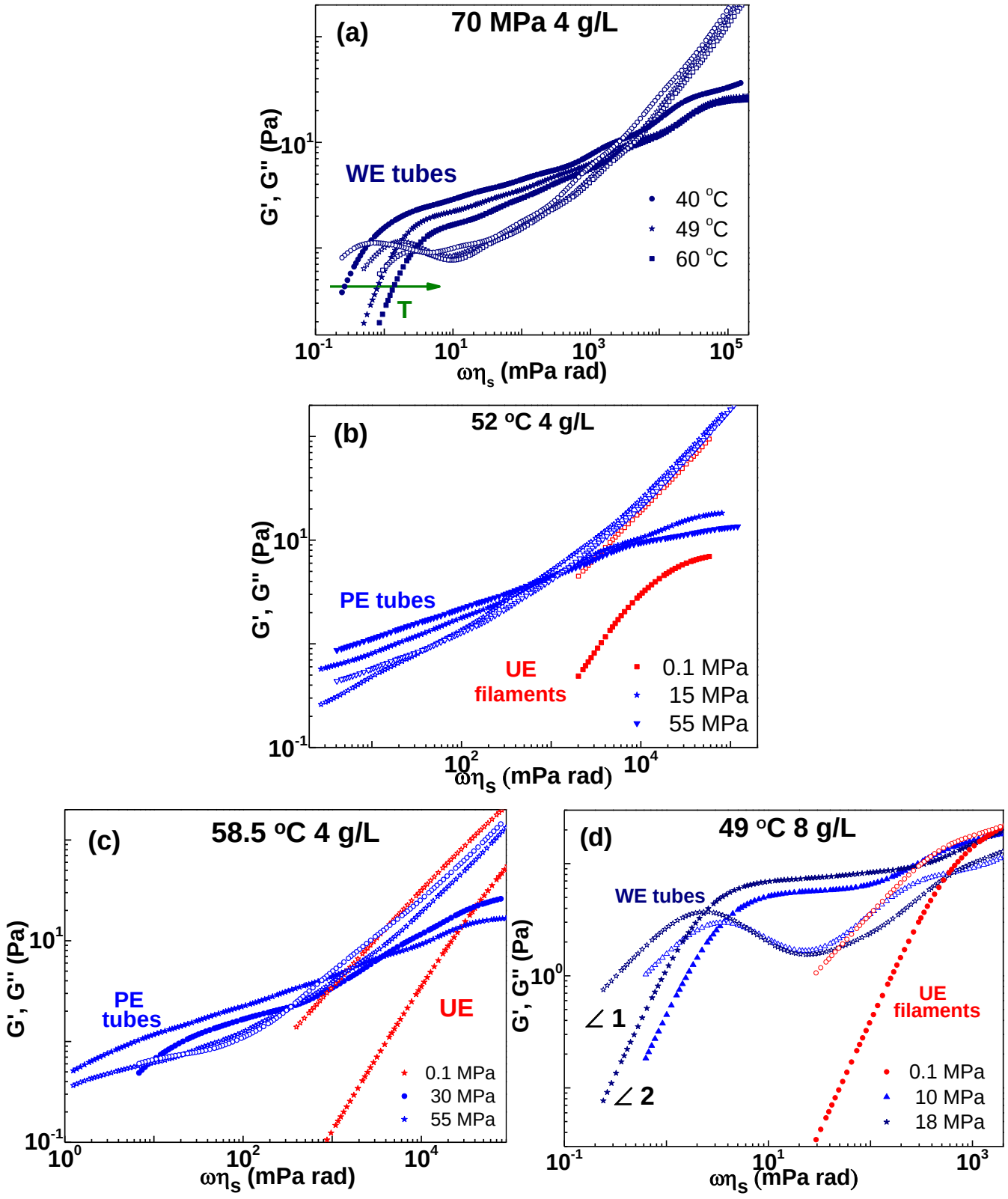



**Figure S10:** Frequency-dependent storage G' (filled symbols) and loss G'' (open symbols) moduli of EHUT/cyclohexane solutions at different pressures and temperatures for: (a) 4 g/L, 70 MPa, at 40°C, 49°C

and 60°C; (b) 4 g/L, 52°C, at 0.1 MPa, 15 MPa and 55 MPa; (c) 4 g/L, 58.5°C, at 0.1 MPa, 30 MPa and 55 MPa; (d) 8 g/L, 49°C, at 0.1 MPa, 10 MPa and 18 MPa. The frequencies are scaled with the solvent viscosity. The data of (a) are taken from Ref. 27.

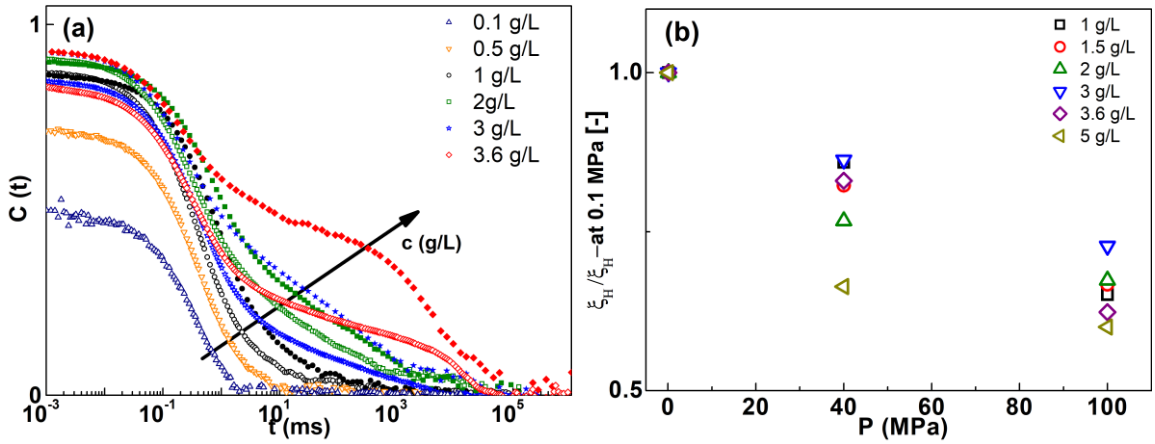



**Figure S11:** (a) Intermediate scattering function of EHUT/dodecane solutions at 25°C, different concentrations ranging from 0.1 to 3.6 g/L (see legend) and two pressures (open symbols correspond to 0.1 MPa, filled symbols to 100 MPa). (b) Hydrodynamic correlation length (mesh size) normalized by its value at 0.1 MPa, as a function of pressure (0.1, 40, 100 MPa) for different EHUT/dodecane concentrations (see legend).

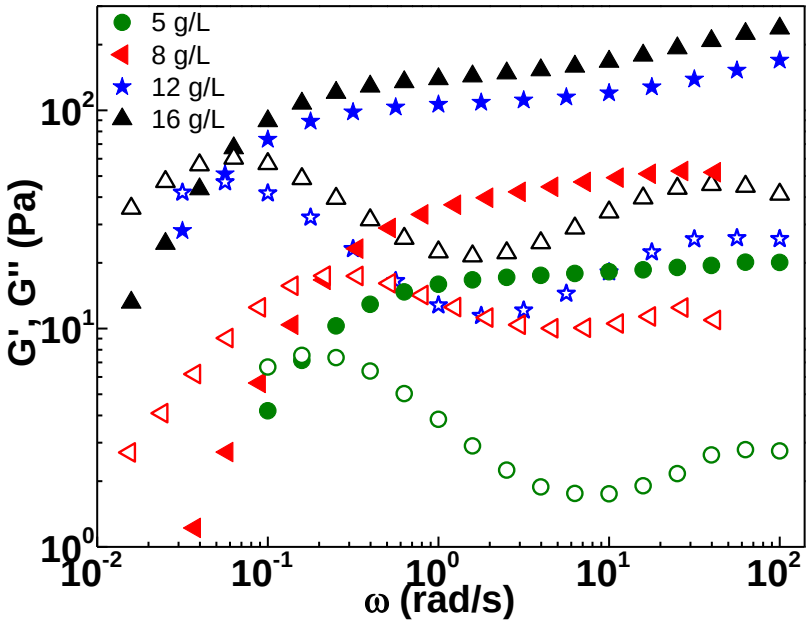



**Figure S12:** Linear viscoelastic spectra of EHUT/dodecane solutions (concentrations are indicated in the legend). Data obtained from bulk (conventional) rheological measurements at ambient conditions (T=25 $^{o}$C, P = 0.1 MPa, humidity ~ 40%) are shown for G' (filled symbols) and G'' (open symbols).

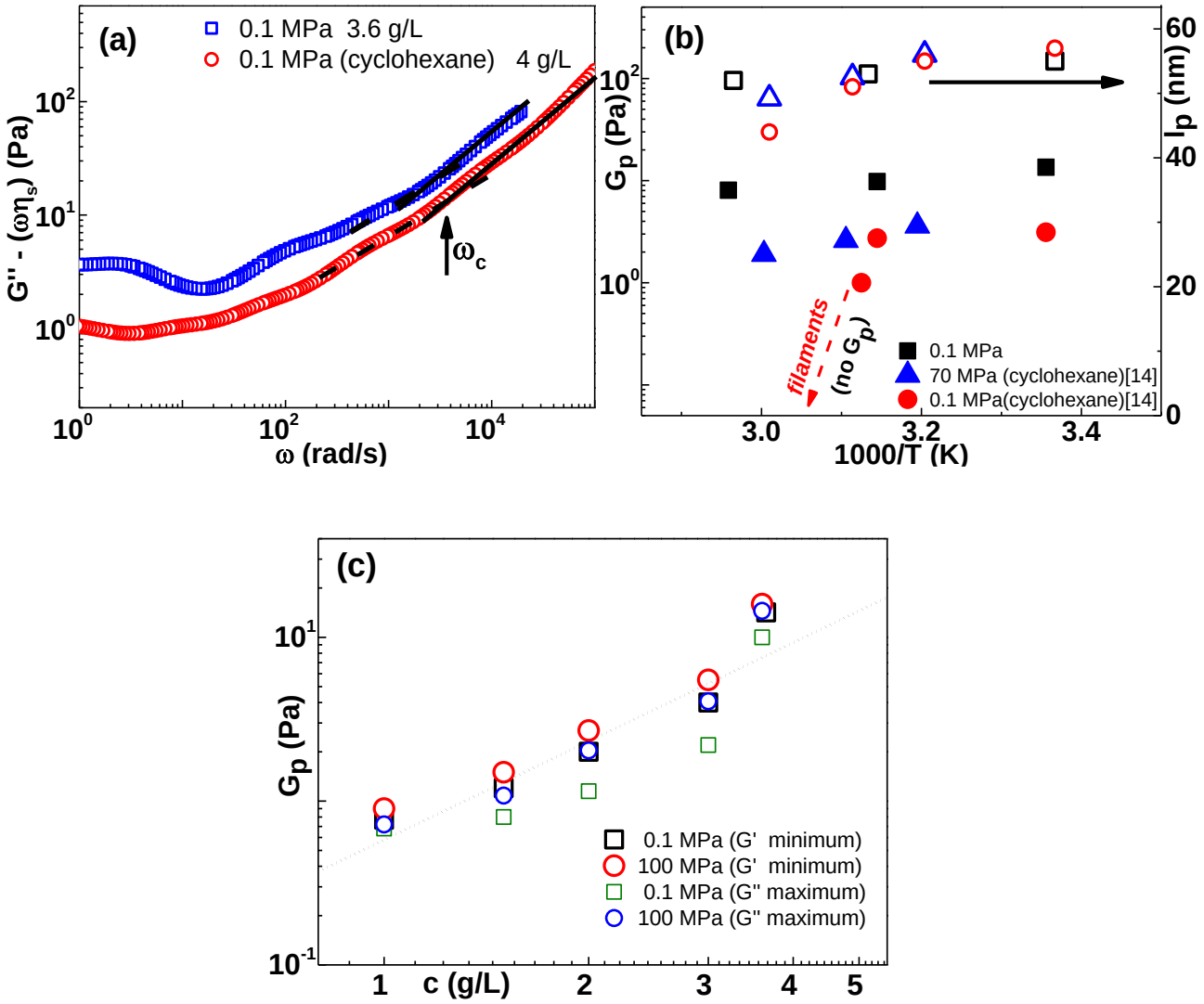



**Figure S13:** (a) Frequency-dependent loss modulus data for EHUT/dodecane solutions (3.6 g/L, blue squares) and EHUT/cyclohexane solutions (4 g/L, red circles) at 25 °C and 0.1 MPa, after having subtracted the solvent contribution ($\omega\eta_s$). The lines have a slope of 0.55 (dashed) and 0.75 (solid). (b) Arrhenius plot of plateau modulus (filled symbols, left axis) and persistence length (open symbols, right axis). The persistence length was extracted from equation S1 (where $\omega_0$ is the frequency marking the onset of the HF regime with a slope of 0.75 [14]. The increase of temperature was followed by a structural transition (tube to filaments), which was marked by the drop in persistence length and plateau modulus (case of EHUT/cyclohecane solution, red circles). (c) concentration dependence of the plateau modulus extracted from the minimum value in G'' and/or the maximum in G'' (in the terminal regime); in the latter case, $G_p = 2G_{max}$.

$$\omega_0 = \frac{k_B T}{8\eta_s {l_p}^3} \qquad \text{(S1)}$$

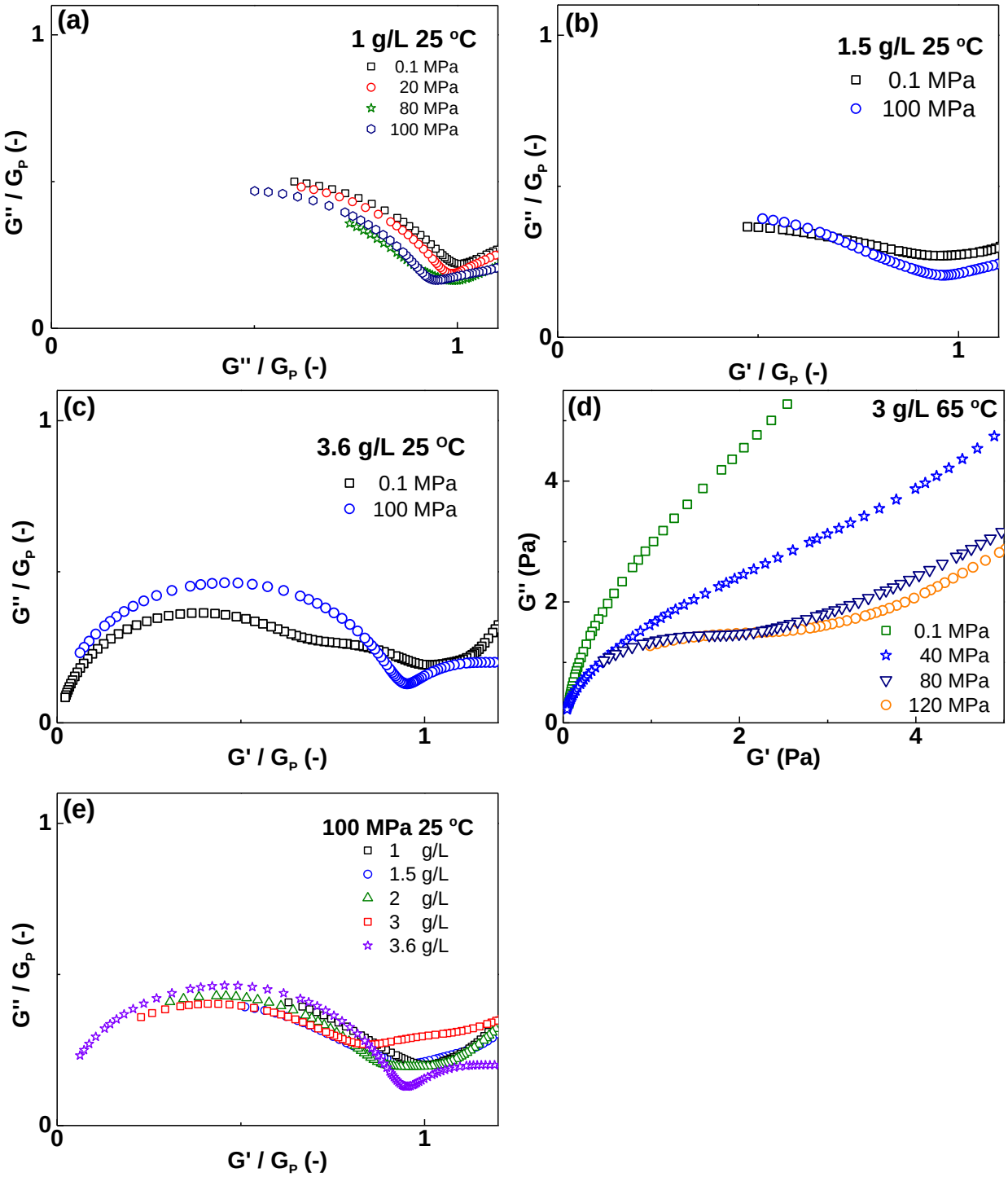



**Figure S14:** Cole-cole plots of EHUT / dodecane solutions at different pressures, temperatures and concentrations (indicated as legends in each plot).