

The role of humidity in the flow properties of supramolecular organogelators

Emmanouil Vereroudakis^{1,2}, N.J. Van Zee^{3,4}, E.W. Meijer³ and Dimitris Vlassopoulos^{1,2}

1 Institute of Electronic Structure and Laser, Foundation for Research and Technology (FORTH), 71110 Heraklion, Crete, Greece

2 Department of Materials Science & Technology, University of Crete, 71003 Heraklion, Crete, Greece

3 Institute for Complex Molecular Systems, Eindhoven University of Technology, PO Box 513, Eindhoven 5600 MB, The Netherlands

4. Matière Molle et Chimie, UMR 7167 CNRS-ESPCI, Ecole Supérieure de Physique et Chimie Industrielles, 10 rue Vauquelin, 75005 Paris, France.

E-mail: mvereroud@iesl.forth.gr

Supramolecular organogelators have been known for more than 30 years, however the extraordinary sensitivity of their self-assembly and viscoelastic properties to humidity have been ignored until recently.^{1,2} Typically, organic oils contain very tiny quantities of water (0.01% by weight) which can influence the self-assembly as well as the flow properties of supramolecular organogelators^{1,2}. Here, we explore the linear and nonlinear viscoelastic properties of biphenyl tricarboxamides (BPTA) in dodecane which exhibit structural transitions with varying temperature. The driving force behind these transitions is the interaction of the supramolecular structure with the water contained within the solvent. The present system is investigated using shear rheology under controlled humidity conditions in both humid (~60% relative humidity) and dry (~5% relative humidity) conditions. We observe that in humid environment the linear and nonlinear rheological properties are temperature-dependent. Alternatively, the humidity content in the self-assembled structure tunes its rheology at constant temperature. At temperatures where the system does not interact with water, we observe higher plateau modulus, slower relaxation time and substantial shear-strain-hardening. On the other hand, at temperatures where the self-assembled structure strongly interacts with water the plateau modulus is lower, the relaxation faster and there is absolutely no sign of strain hardening behaviour. These findings suggest that humidity effects cannot be dismissed in hydrogen-bonded supramolecular structures in oily environments and provide insights into tailoring the flow properties of organogels.

1. A. Louhichi et al., *J. Rheol.* 61, 1173-1182 (2017)
2. N.J. Van Zee et al., *Nature*, 558, 100-103 (2018)