Nonlinear shear response of a supramolecular organogelator: Dependence on temperature and humidity

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Recently it was shown that the self-assembly and viscoelastic properties of organogelators in apolar solvents can vary due to the presence of trace amounts of water molecules, which are dispersed in monomeric state [1,2]. The emerging scenario is that water molecules interact with the supramolecular polymer building blocks through strong hydrogen bonding and take part in the self-assembly process affecting the overall properties of the material [3]. Herein, we investigate how such interactions affect the nonlinear shear response of these systems. Firstly, depending on the temperature and the level of humidity the system behaves in two different ways. For low temperatures (< 20^{o}C) and/or high levels of relative humidity (>60%) the supramolecular organogelator strain softens, shear thins and appears to exhibit shear banding instabilities at large Weissenberg numbers. In contrast, at high temperatures (>30^{o}C) and/or low levels of relative humidity (<30%) the system strain hardens while still exhibiting shear thinning and shear banding. For intermediate values of temperature and humidity the system seems to be at an intermediate state characterized by transient hardening and much faster relaxation than the dry state. Changes in the response seem coupled to temperatures at which structural transitions have been observed [1,2] as well as changes in the linear viscoelastic properties [3] and seem to suggest that the driving force is the way water molecules interact with the "monomeric" moieties and ultimately affect the self-assembly.

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[3] Vereroudakis, E. Viscoelastic Properties of Supramolecular Gels Based on Hydrogen Bonding. Master's Thesis, University of Crete, Greece, 2020.