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Solvent organisation and dynamics in self-assembled molecular gels

Physicals gels formed by low molecular weight organic gelators (LMMOG) are composed of a rigid network formed by the gelators, in which is trapped a large quantity of solvent. Gathering properties of both liquids and solids, they find applications as functionalized nanomaterials in diverse domains, although applications suffer several limitations, such as the unexpected collapse after some time or the difficulty to predict gelation ability of new molecules. The subtle interplay of the different forces exerted between the solvent and gelators enable the gels to reversibly assemble in a restricted temperature range in a complex structure that depends on the solvent. The interface between gelators and solvent is soft and ill-defined. Because of the intrinsic complexity, at the molecular level, of the structural organization and the dynamics, these materials are quantitatively studied with difficulty. In this context, we undertook the study of the microscopic dynamics in a model physical molecular gel, methyl-4,6-O-benzylidene-α-D-mannopyranoside (α-manno) [1] in water and toluene are probed by neutron scattering [2] and Transient Grating Spectroscopy [3]. The α-manno is an amphiphilic gelator that adopts different organizations in both solvents. We were able to distinguish, on a timescale from a few ps to few ns, several dynamical populations of solvent molecules in interaction with the rigid network formed by the gelators. We found that only few toluene molecules per gelator participate to the network which is formed by hydrogen bonding between the gelators' sugar moieties. In water, however, the interactions leading to the solid network assembly are weaker and each gelator forms a tens of hydrogen bonds with the surrounding water molecules, that are stable over few hundreds of ps only. Eventually, the gelator network dynamics can be distinguished just before the melting when its characteristic relaxation time enters the ns timescale. This study shows that quantitative information on the behaviour of solvent confined in a molecular gel can be obtained, also relevant to diffuse interfaces as often encountered in soft matter systems.

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Preferred topic

Characterization - large instruments

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