3SR Seminars

21/06/2024 at 10:30 a.m. | Thibaut Divoux | Galilée room 015

Unveiling Hydrophobic-Driven Gelation: Exploring the case of Carboxymethylcellulose (CMC) Hydrogels

The sodium salt of carboxymethylcellulose (NaCMC) is a water-soluble derivative of cellulose, which is broadly used for industrial applications such as food, pharmaceuticals, paints, etc., and serves as a thickener and water retention. The properties of NaCMC can be exquisitely tuned via their degree of substitution (DS), which corresponds to the average number of carboxymethyl groups per repeating glucose unit and varies between 0 and 3. Highly substituted polymers, i.e., for DS >1, are hydrophilic and disperse easily in water, yielding rheological features typical of polyelectrolyte solutions. In contrast, weakly substituted polymers, i.e., for DS<0.9, contain hydrophobic regions, which favor interchain aggregation and the formation of so-called ``fringed micelles'' yielding thixotropic and even gel-like properties at high enough concentrations. Here we show that gelation of NaCMC solution can be induced by lowering the pH, which decreases the charge density along the CMC chain and promotes the formation of multichain aggregates.

This presentation will offer a comprehensive description of the acid-induced gelation of carboxymethylcellulose (CMC). Linear viscoelastic properties measured at various pH and CMC contents allow us to build a sol-gel phase diagram, and show that CMC gels exhibit broad power-law viscoelastic spectra that can be rescaled onto a master curve following a time-composition superposition principle. These results demonstrate the microstructural self-similarity of CMC gels, and inspire a mean-field model based on hydrophobic inter-chain association that accounts for the sol-gel boundary over the entire range of CMC content under study. Neutron scattering experiments further confirm this picture and suggest that CMC gels comprise a fibrous network crosslinked by aggregates. This picture is further tested by examining the gel non-linear mechanical response during large-amplitude oscillatory shear experiments. Finally, low-field NMR measurements offer an original signature of acid-induced gelation from the solvent perspective. Altogether, these results open avenues for precise manipulation and control of CMC-based hydrogels.