Multiscale imaging reveals four distinct hierarchical networks in supramolecular/polymer hydrogel composites

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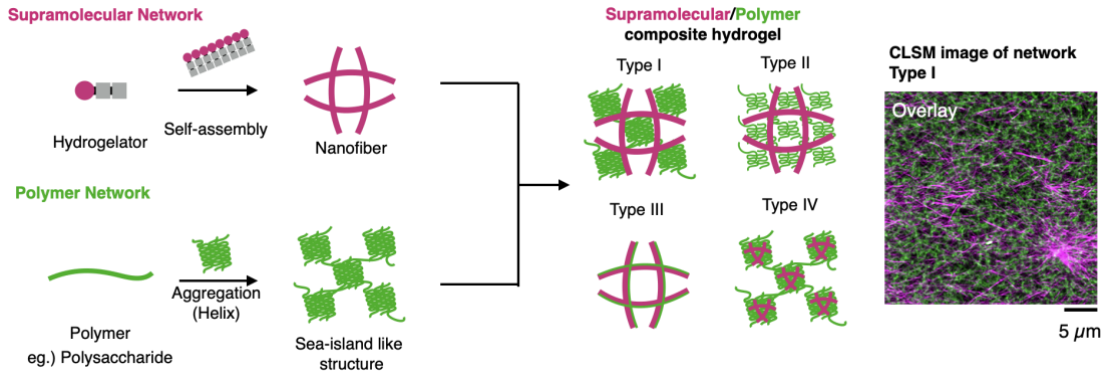
Supramolecular hydrogels formed by self-assembly of small molecules (hydrogelators) via noncovalent interactions have been developed as unique and promising stimuli-responsive soft materials. Composite materials consisting of supramolecular and polymer hydrogels were recently proposed to overcome the mechanical weakness of supramolecular hydrogels[1]. However, the relationships between the network structure and mechanical properties of the composite hydrogels are still unclear due to lack of reliable imaging methods. To date, structural analysis of the composite hydrogels has been mainly conducted by electron microscopy (EM), which can visualize the nanoscale structure of the network. However, EM is not suitable for analyzing the composite network due to a poor capability to discriminate the distinct networks. Recently, confocal laser scanning microscopy (CLSM) has emerged as a powerful tool for structural analysis of multicomponent supramolecular hydrogel systems[2]. CLSM not only allows imaging of supramolecular nanofibers and hydrogels without the need for a drying process, but can distinguish between chemical species through the use of appropriately designed fluorescent probes.

In this work, we sought to evaluate the network structure of various kinds of composite hydrogels by both SEM and CLSM imaging. Multiscale imaging reveals that these are classified into four distinct hierarchical structures based on the morphology and spatial distribution of each network. To investigate structure-mechanical properties relationships of the composite hydrogels, rheological experiments are also conducted.



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