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Coiled coils as mechanoresponsive material building blocks

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Coiled coils are highly abundant folding motifs in structural proteins. Consisting of two (or more) α -helices wound around each other in a superhelical fashion, they represent essential structural elements of the cytoskeleton and the extracellular matrix. Synthetic coiled coil sequences are further used as dynamo-mechanical supramolecular crosslinks in biomimetic hydrogels. Considering their function as mechanical building blocks, little is still known about the structural determinants that define the molecular mechanical properties of coiled coils and how these affect the viscoelastic properties of coiled coil-based self-assembled materials.

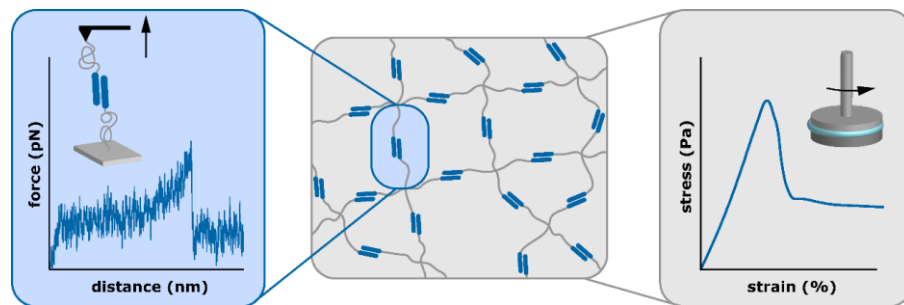


Figure 1. Structure and characterization of coiled coil-crosslinked star-PEG hydrogels. Single-molecule force spectroscopy yields information about the dynamic mechanical behaviour of individual crosslinked chains, whereas rheology provides complementary information about material failure and self-healing at different strains and strain rates.

Using atomic force microscope (AFM)-based single molecule force spectroscopy (Figure 1), we have established the sequence-structure-MECHANICS relationships of a series of synthetic coiled coils [1-4]. We show that the molecular rupture forces of coiled coils depend on coiled coil length, helix propensity and hydrophobic core packing as well as on the pulling geometry.

Based on this knowledge, we have developed a library of coiled coils with tuneable mechanical properties and synthesized a series of poly(ethylene glycol)-based hydrogels using these coiled coils as dynamic physical crosslinks [4-6]. The resulting hydrogels consist entirely of mechanically characterized molecular building blocks and allow for establishing a direct relationship between molecular and bulk mechanics (Figure 1). Using theoretical models, we obtain a direct correlation between the single-molecule parameters and the bulk mechanical response of the hydrogel, as determined with rheology. Serving as a new platform of extracellular matrix mimic, these hydrogels will allow for dissecting local and global mechanical processes that determine cellular mechanosensing [7].

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