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Different approaches for the formation of synthetic hydrogels based on hybrid physically-chemically cross-linked networks

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Many polymers with different types of cross-linking have been designed to develop various hydrogels for industrial and fundamental applications. In particular, they have attracted significant interest in the field of regenerative medicine, as drug delivery vehicles or scaffolds for tissue engineering. However, the main challenging limitation for their extensive use is their low mechanical stability. Many recent efforts have been made to overcome this burden, in particular by the elaboration of double network (DN) hydrogels. Despite the promising results of the few existing physically-chemically cross-linked DN hydrogels, the preparation procedures require long photopolymerization time and toxic precursor materials. Moreover, only the comparison of the mechanical properties between the hybrid DN hydrogel made in very specific conditions with the corresponding two separate networks is studied. The fundamental understanding of the combination of two different cross-links for a rational design of new hybrid DN hydrogels is a challenge.
Herein, we provide synthetic strategies to tailor in-situ formation of DN hydrogels based on physically-chemically cross-linked networks. Three approaches to combine catalyst-free click reaction and hydrogen bonding for in-situ hydrogel formation will be described (see Figure). More in detail, the covalent cross-linking consists of the click reaction between azides and cyclooctyne derivatives, referred to as strain-promoted azide-alkyne coupling (SPAAC), and the non-covalent cross-linking is based on supramolecular interactions via a four-fold hydrogen bonding motif, the ureido-pyrimidinone (UPy) unit.
The mechanical properties of these new hydrogels are studied by rheology and their morphology are analyzed by confocal microscopy. A detailed comparison of the hydrogel properties is discussed according to the approach selected and the ratio of the two types of cross-linking. In particular, this work shows the benefits to associate the dynamicity afforded by supramolecular interactions for gel recovering with the strength provided by the covalent cross-links for enhanced material stiffness. We propose that this work offers both a novel versatile platform for the easy preparation of DN hydrogels under physiological conditions with tunable properties, and one more step in the fundamental understanding of structure-property relationship of physically-chemically cross-linked hydrogels.