Responsive supramolecular cross-links for healable double polymer networks

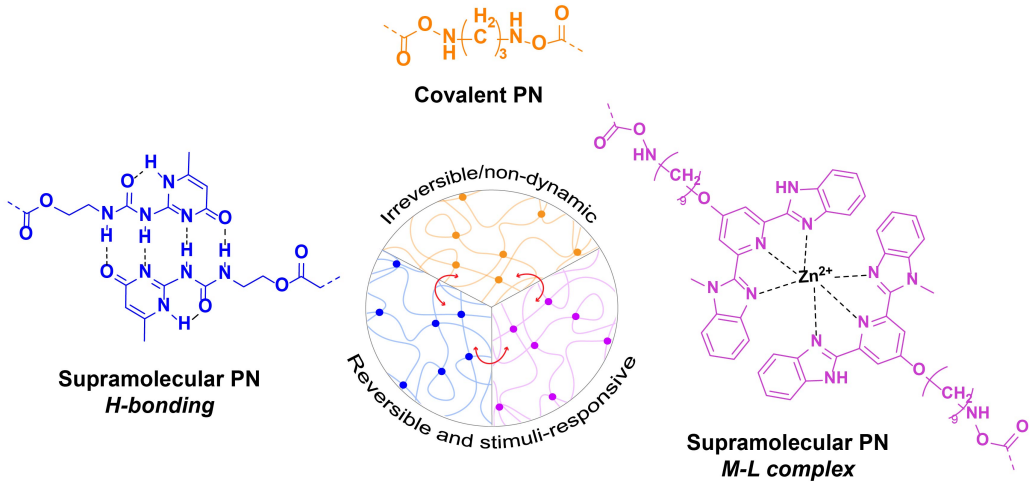
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Double polymer networks (DNs) consist of two polymer networks that usually exhibit different physical properties. These materials are of great interest thanks to the remarkable mechanical properties (toughness, elasticity) that one can achieve, and they can find direct applications in engineering or the medical field.[1]

DNs comprising covalent cross-links synthesized either in the presence or absence of a solvent have been deeply investigated.[2][3] little attention has been dedicated to DNs containing *supramolecular* cross-links. Supramolecular bonds can be highly dynamic and DNs containing such motifs can offer ease of (re)processing and recyclability as well as the possibility of being healable.

Here, we report the synthesis of different DN elastomers based on poly(butyl acrylate) (**PBA**) that comprise covalent and dynamic cross-links in different molar feeds (**Fig. 1**). The supramolecular polymer networks are made by reversible addition−fragmentation chain-transfer (RAFT) co-polymerization of butyl acrylate (**BA**) with either 2-ureido-4[1H]pyrimidinone (**UPy**) motif, which dimerizes by quadrupole hydrogen bonding interaction, or the 2,6-bis(1′-methylbenzimidazolyl)pyridine (**MeBip**) tridentate ligand, which forms complexes with Zn^{2+} ions in a 2:1 stoichiometry.[5][6][7] The covalent polymer network (**PBA-BDA**) comprises 1,4-butanediol diacrylate (**BDA**) as a covalent cross-link, and is prepared by UV-initiated free-radical polymerization (UV curing).[8] The thermomechanical properties of the individual networks and their multiple combinations are investigated. In addition, we investigated the responsiveness of **PNs** comprising dynamic cross-links by healing studies.



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