Self-healable supramolecular polymer toughened by ionic cluster

Supported by the National Natural Science Foundation of China, the research group led by Prof. Qu DaHui (曲大辉) at the East China University of Science and Technology, realized a self-healable supramolecular polymer toughened by ionic cluster, which was published in *Angew Chem Int Ed* (2020, 59: 5278—5283).

Supramolecular polymers, a species of polymers formed by dynamic chemical bonds, exhibit unique advantages such as self-healing ability, recyclability, and preparation simplicity. These properties differ supramolecular polymers with traditional covalent polymers, enabling supramolecular plastics to be of great promise for future industrial applications. However, there has been a trade-off between material cost and performance in the design of supramolecular polymers, intrinsically inhibiting the practical application of supramolecular polymeric materials.

Previously, Qu's group pioneeringly exploited a supramolecular polymer from a natural small molecule, thioctic acid (TA) (*Sci Adv*, 2018, 4: eaat8192). This easy-to-make supramolecular material integrated stretchability, self-healing ability, and adhesiveness. However, its softness limited its further application. To surmount this disadvantage, in this work, they took advantage of the high-affinity ionic bonds as the secondary interaction, and thus developed a toughening strategy for this dynamic supramolecular network. By simply increasing the concentration of iron ions and reaction temperature, the primary iron-carboxylate complexes would further aggregate, forming secondary ionic clusters in the dry network. The resulting hierarchically crosslinked supramolecular network exhibited remarkably increased mechanical moduli (63 times higher than the original network). Meanwhile, the dynamic properties of the material were maintained, including high stretchability, self-healing ability, and remoldability. Owing to the presence of ionic cluster interactions, an efficient self-healing process can be also performed under water, showing great potential in the use of water-insensitive soft materials. The remoldability could also facilitate the development of reusable plastics. Most intriguingly, this supramolecular material combining with excellent performances is very easy to prepare. The feedstock of TA is not toxic because it is a commercial antioxidant nourishment. The simultaneous integration of low cost and high performance makes this family of materials truly attractive towards the next generation of supramolecular plastics.

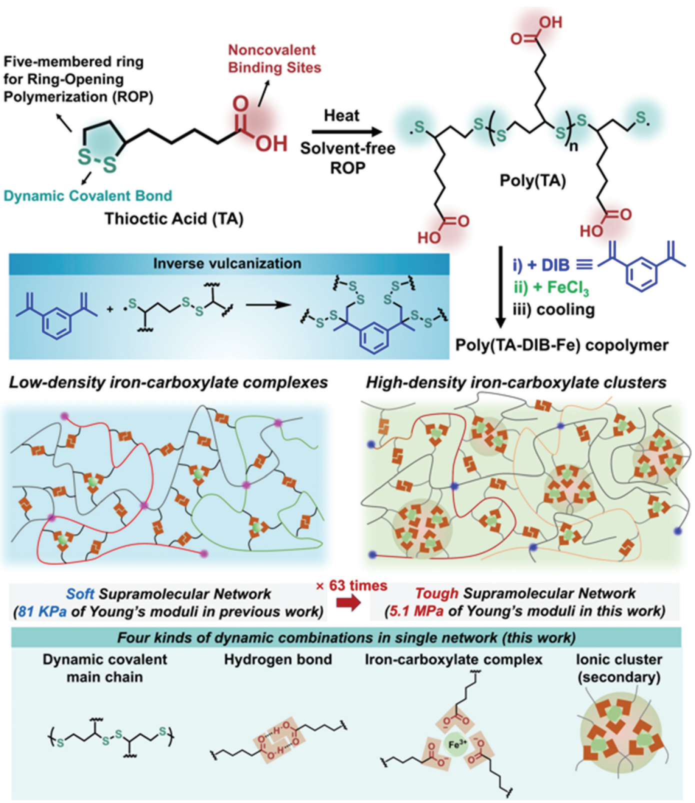

Figure 1 Schematic representation of the TA monomer and poly(TA) polymers. The cartoon representation shows the existing four types of dynamic combinations in the network.