A3-10 Water-Mediated Crosslinking for Designing Tough and Adaptive Polymer Networks

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Reversible polymer networks with dynamic cross-links have drawn increasing attention for their potential in creating tough, self-healing, and environmentally adaptive materials. I will present a series of studies demonstrating how host–guest interactions can be harnessed to design polymer networks with water-responsive mechanical properties. (**Figure 1**).

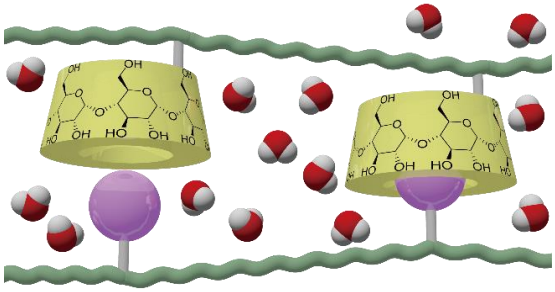

Figure 1. Schematics of a hydrogel cross-linked with host-guest interaction in the presence of waters.

The first study focuses on hydrogels formed via β-cyclodextrin (βCD) and adamantane (Ad) host–guest pairs. These reversible cross-links allowed the network to adapt to hydration changes. Mechanical toughness improved significantly with higher non-freezing water (NFW) and intermediated water (IMW) content, owing to enhanced mobility of the dynamic junctions and efficient stress relaxation. The results reveal how the presence and behavior of NFW and IMW modulate supramolecular dynamics under hydrated conditions.

We developed movable cross-linked polymers where guest units were covalently tethered to the network. This design further enhanced cross-link mobility in hydrated conditions, improving mechanical flexibility and toughness. The networks showed hydration-dependent relaxation behaviors, suggesting that IMW facilitates not only plasticization but also dynamic rearrangement (**Figure 2**).

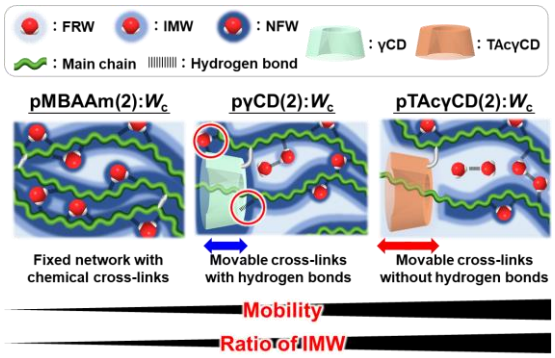

Figure 2. Proposed structures of the hydration in movable cross-linked polymers. Relationship between the mobility and ratio of IMW.

We also examined how the phase state of NFW, IMW vs. free water (FRW) affects biocompatibility. By tuning water distribution in the hydrogel matrix, we could control protein adsorption and promote cell adhesion, with reduced protein denaturation. This underscores the impact of water structuring in designing functional biointerfaces.

These studies offer a coherent strategy for tuning hydrogel mechanics and biofunctionality through molecular mobility and hydration control, with applications in biomedicine, soft robotics, and sustainable materials.

1) M. Osaki, M. Tanaka, Y. Takashima, et al *Macromolecules* **2021**, *54*, 8067-8076.
2) K. Nishida, R. Ikura, T. Inoue, G. Matsuba, M. Tanaka, Y. Takashima, *Macromolecules* **2024**, *57*, 7745-7754.
3) Y. Kawai, Y. Ikemoto, M. Tanaka, T. Kato, Y. Takashima, Y. *ACS Appl. Polym. Mater.* **2025**, *7*, 7767-7776.

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