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| Author(s) | 陶, 真 |
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Abstract of Doctoral Dissertation

Degree requested Doctor of Life Science ✓ / Pharmaceutical Science / Clinical Pharmacy Applicant's name Zhen Tao

Title of Doctoral Dissertation

Fabrication of Tough Hydrogels Composites from Photo-Responsive Polymers to Show Double Network Effect

(光応答性ポリマーを用いた新規高靱性ハイドロゲル複合材料の合成とそのダブルネットワーク効果に関する研究)

Hydrogels reinforced by rigid skeleton could exhibit combined characteristics of each component, which remarkably improve the properties and versatile the application of hydrogels. However, it remains difficult to develop hydrogel composites with synergistically improved mechanical properties as like double network hydrogels. To have the synergistic effect, the matrix component must possess a higher strength than the rigid skeleton. Otherwise, the hydrogel composite will not be capable of sustaining higher stress once the rigid skeleton fractures. Therefore, to date, many efforts have been made to product energy-dissipated tough hydrogel as the soft matrix, by introduction of reversible crosslinkers.

On the other hand, the interface between the soft and rigid phase presents another great challenge. The interface between heterogeneous phases in composite materials plays an essential role in delivery of force and dissipation of energy, resulting in effective reinforcement on the mechanical properties. While, in general, hydrogel composites are formed by polymerization or crosslinking with distrusted phases such as particles, fibers and layer elements as the reinforcing components. The swelling mismatch between soft and rigid phases may lead to surface wrinkle, bulk deformation or even fracture of materials. Another practical strategy is to assemble the rigid surface and hydrogels into hybrids by chemical anchoring or physical adhesion. But it restricts multiple registration steps or precise chemical design.

In this work, I provide a new strategy that integrating the mechanical strong and photo-responsiveness in one hydrogel by copolymerizing hydrophobic stimuli-responsive monomers (2-nitrobenzyloxycarbonylaminoethyl methacrylate, NBOC) into polymer chains. In the gel network, the NBOC residues play two roles, one is working as physical crosslinkers based on their hydrophobic association, which can dissipate large amount of energy during deformation so that toughen the hydrogels; the other is providing photo-responsiveness to further improve the mechanical strength. Specifically, 2-nitrobenzyl functionalities exhibit photo-labile characteristics under UV irradiation, and the resultant amino groups of 2-aminoethyl methacrylate (AMA), which is transformed from NBOC by self-immolative reaction, will further attack the close-by ester bonds, so that replace the original physical crosslinkers by forming new chemical crosslinks, which further increase the modulus of hydrogel.

Our approach is based on copolymers containing pendent photo-responsive, o-nitro benzyl moieties that allow crosslinking at different spatial location tuned by UV irradiation, free of multiple stampings or registration steps. By controlling the position of UV irradiation, the NBOC-based hydrogels can be easily reconstructed into hieratical structures with different modulus regions.

Furthermore, the mechanical response of this hydrogel composites mimics that of double-network (DN) gels, where the hard layers, P(UM-co-AMA), act as the first network, imparting high modulus and breaking to dissipate energy, and the soft layer, P(UM-co-NBOC) or P(HEA-co-NBOC), acts as the second network, stretching to maintain global integrity. By the observation and investigation of mechanical properties of this hydrogel composites, the hypothesis for fracture mechanism of DN gels could be proved and demonstrated, which has not been witnessed before.