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Communication

Super-tough and rapidly self-recoverable multi-bond network hydrogels facilitated by 2-ureido-4[1*H*]-pyrimidone dimers

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ABSTRACT

Multi-bond network (MBN) hydrogels contain hierarchical dynamic bonds with different bond association energy as energy dissipation units, enabling super-tough mechanical properties. In this work, we copolymerize a protonated 2-ureido-4[1*H*]-pyrimidone (UPy)-contained monomer with acrylic acid in HCl solution. After removing excess HCl, UPy motifs are deprotonated and from dimers, thus generating an UPy-contained MBN hydrogel. The obtained MBN hydrogels (75 wt% water content) exhibit super-tough mechanical properties (0.39 MPa to 2.51 MPa tensile strength), with tremendous amount of energy (1.68 MJ/m^3 to 11.1 MJ/m^3) dissipated by the dissociation of UPy dimers. The introduction of ionic bonds can further improve the mechanical properties. Moreover, owing to their dynamic nature, both UPy dimers and ionic bonds can re-associate after being dissociated, resulting in excellent self-recovery ability (around 90% recovery efficiency within only 1 h). The excellent self-recovery ability mainly originates from the re-association of UPy dimers based on the high dimerization constant of UPy motifs

In recent years, super-tough hydrogels are attracting more and more researching interest due to their various applications in areas such as sensors [1], stretchable capacitors [2,3], wearable devices [4,5], soft actuators [6,7], wound healing [8] and artificial muscles [9]. A common sense of fabricating super-tough hydrogels is to incorporate energy dissipation units into the hydrogel network. These units include covalent bonds [10–13] and dynamic interactions such as nanofillers [14,15], ionic bonds [16,17] and hydrophobic associations [18,19]. Because covalent bonds cannot repeatedly dissipate energy, dynamically crosslinked hydrogel are more applicable in areas that demand self-recovery ability. By combining hierarchical dynamic interactions with different bond association energy in a sparsely chemically crosslinked network, our groups have fabricated a series of super-tough multi-bond network (MBN) hydrogels with self-recovery, self-healing, shape memory, and super water-absorbance in recent years [20–29]. Among the dynamic interactions, ionic bonds demonstrate the highest energy dissipation ability. However, as the ionic bonds are sensitive to pH, ionic MBN hydrogels will become weak under alkaline conditions. Therefore, it is of great importance to incorporate dynamic bonds that can effectively dissipate energy even under alkaline conditions, expanding the application filed of MBN hydrogels.

The 2-ureido-4[1*H*]-pyrimidone (UPy) motifs can self-complementarily dimerize through quadruple hydrogen bonding, which is relatively stable under alkaline conditions. The obtained UPy dimers show a high bond association energy ($\Delta G \approx 70$ kJ/mol) [30] and a high dimerization constant (> 10^5 L/mol in chloroform) [31]. Therefore, by introducing UPy dimers into MBN hydrogels as an energy dissipation unit, one can expect enhancement in both mechanical property and self-recovery ability. However, because of the poor water-solubility of UPy monomers, the UPy-contained hydrogels are usually fabricated using surfactant, or in organic solvents like dimethyl sulfone (DMSO) and then treated with a solvent substitution procedure. The organic compounds cannot be removed completely, and the formation of UPy dimers will be affected by the residuals. As a result, the UPy dimers cannot effectively function as energy dissipation units [32]. Therefore, although many researches have employed UPy motifs as dynamic interaction to endow hydrogels with dynamic properties such as self-healing [33,34], stimuli-responsive [35] and controlled release [36], very few of these works has mentioned good enough mechanical strength enabled by energy dissipation of UPy dimers, which is extremely vital for materials to be applicable. For example, Jeon *et al.* [37] incorporated UPy motifs into a poly (acrylamide) chain and obtained an extremely stretchable hydrogel that could be stretched up to more than 100 times. However,

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the tensile stress was merely 4 kPa even at the strain of 10000%. Guo *et al.* [38] fabricated stimuli-responsive polyurethane-based hydrogels having UPy motifs that exhibited 1.49 MPa tensile strength. However, it is hard to distinguish whether this mechanical strength originates from UPy motifs or from the hydrophobic effect of the hard segments. Chang *et al.* [39] copolymerized acrylic acid (AA), UPy monomer, and an acrylic acid derivative with hydrophobic side chains to achieve high mechanical properties. However, they found that without hydrophobic interactions, the hydrogels solely with UPy motifs were so weak that their strength could not be measured by tensile tests. Therefore, it is of great importance of developing a fabrication method that avoids the effect of organic residuals, facilitating effective energy dissipation by the dissociation of UPy dimers.

In this work, we develop an acid-aided method to avoid the use of organic solvents. At first, a vinyl-hybrid UPy monomer is protonated in a hydrochloride (HCl) solution to enhance its water solubility. After the copolymerization between the protonated UPy monomer and AA, the excess HCl is removed through dialysis in a large amount of water. The influence of UPy motifs on mechanical properties of the obtained MBN hydrogels and the energy dissipation of quadruple hydrogen bonds in UPy dimers are systemically investigated. Moreover, ionic bonds are introduced to the system, in which the energy dissipation mechanism and the dynamic properties of the two dynamic bonds are compared.

The UPy-contained MBN hydrogels are fabricated through a radical copolymerization (experimental details can be found in the Supporting information). Note that since UPy monomer has a poor water solubility, regular aqueous polymerization is not adaptable here. Moreover, hydrogels containing UPy motifs fabricated using DMSO as a solvent are generally too weak to be applied in areas that demand high mechanical properties. Therefore, instead of DMSO, here we use HCl aqueous solution, in which UPy monomers can be protonated and the dimerization can be interrupted, resulting in enhancement in their water-solubility. The success of protonation is demonstrated by the slight shift in the ^{1}H NMR spectrum of UPy motifs (Fig. S1 in Supporting information). The protonation ratio is calculated to be 71%. Next, the protonated UPy monomer is polymerized with AA and a slight amount of BIS (0.1 mol% with respect to AA) as the chemical crosslinker to give a sparsely chemically crosslinked hydrogel. The obtained hydrogels contain protonated UPy motifs on the polymer chains as shown in Fig. 1, which are then immersed in a large amount of water. With the decrease of HCl concentration inside the network, UPy motifs are gradually deprotonated and spontaneously form dimers due to the high dimerization constant, which can act as new dynamic crosslinking points. Due to the high dissociation energy of UPy dimers, the resulted MBN hydrogels are expected to exhibit excellent mechanical behavior because of the energy dissipation enabled by the dissociation of UPy dimers. It is worth noting that the water content of these hydrogels are not the same at this step (Fig. S2 in Supporting information). As is well known, mechanical properties of hydrogels relate very closely to water content [29], so it is vital to compare different hydrogels at the same water content for the sake of investigation of strengthening mechanism. To address this issue, the water content of these hydrogels is adjusted to 75 wt%. These hydrogels are denoted as UPy*x*C*y*, where *x* and *y* stand for the molar ratios (mol%) of UPy monomer and BIS, respectively, with respect to AA. Moreover, ferric citrate (FeCA) can be introduced to the UPy-contained MBN hydrogels to induce homogeneous ionic bonds, which will be discussed later.

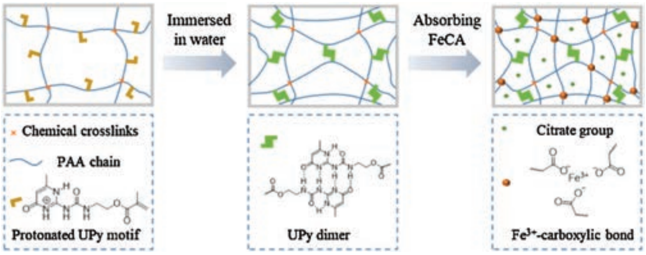

Fig. 1. Synthetic schemes of the UPy-contained MBN hydrogels.

Next, the contribution of UPy dimers to the enhancement of mechanical properties of the UPy-contained MBN hydrogels is investigated (BIS content is 0.1 mol%, water content is fixed at 75 wt%). Note that the mechanical strength of UPy0C0.1 is so weak that it cannot be measured on a tensile tester. This is because that the ordinary hydrogen bonds in PAA hydrogels are weak, resulting in insufficient energy dissipation, which is consistent with our previous works [26,27]. It can be seen in Fig. 2 that as the increase of UPy content from 2 mol% to 10 mol%, the tensile strength increases from 0.39 MPa to 2.51 MPa, while elongation at break decreases from 1101% to 707% (Fig. 2b). This significant enhancement in mechanical strength indicates that the dissociation of UPy dimers have a great contribution to energy dissipation during the stretching process. The dissipated energy, *i.e.*, the fracture energy of the hydrogels drastically increases from 1.68 MJ/m^3 to 11.1 MJ/m^3 as the increase of UPy content from 2 mol % to 10 mol% (red line in Fig. 2d). Furthermore, to further understand the network structures of the UPy-contained MBN hydrogels, the modulus of each stress-strain curve is calculated from the slope and plotted in Fig. 2c, because tensile modulus is closely related to the network crosslinking density. It can be seen that when UPy content is 2 mol% to 6 mol%, the initial modulus of the hydrogels is almost proportional to UPy content. However, when UPy content increases from 6 mol% to 10 mol%, the initial modulus shows a drastic increase (black line in Fig. 2d). In fact, in the UPy-contained hydrogel network structure as shown in Fig. 1, UPy dimers divide the sparsely covalently crosslinked PAA network into smaller networks. Because the dimerized UPy motifs is randomly distributed, the formed network chain length between crosslinking points in the hydrogel is not the same but random. At the beginning of the tensile test, the shorter network chains would firstly be stretched. So the initial modulus mainly

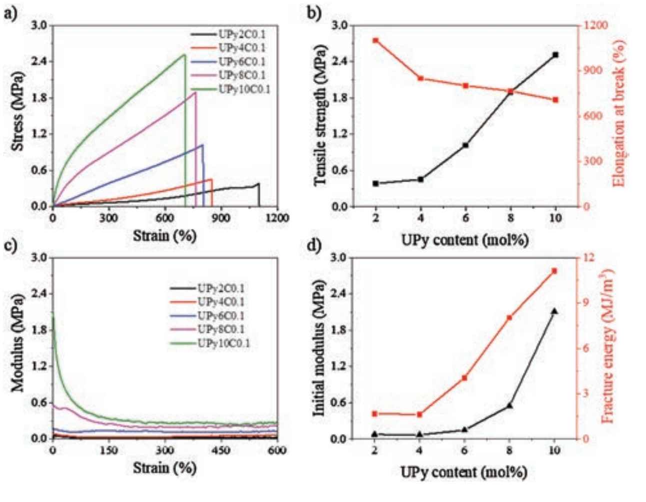

Fig. 2. (a) Stress-strain curves of the UPy-contained MBN hydrogels with different amount of UPy motifs. (b) Tensile strength and elongation at break of the MBN hydrogels as a function of UPy content. (c) Tensile modulus of the MBN hydrogels as a function of tensile strain. (d) Initial modulus and fracture energy of the MBN hydrogels as a function of UPy content.

depends on the number of shorter network chains, which is related to UPy content. The higher the UPy content is, the more the shorter network chains are. As a result, UPy8C0.1 and UPy10C0.1 have much larger initial moduli. As the tensile test proceeds, UPy dimers connecting to shorter network chains are gradually broken, so the moduli of UPy8C0.1 and UPy10C0.1 undergo drastic declines (Fig. 2c). When the strain is between 300% and 600%, the moduli of all samples become constant, and the value is proportional to UPy content (Fig. S3 in Supporting information), indicating that the network structures of all the UPy-contained MBN hydrogels have become similar to each other. Finally, as we stated that the organic residual may decrease the mechanical properties of hydrogels containing UPy motifs, the mechanical properties of a DMSO-treated UPy6C0.1 sample is evaluated (Fig. S4 in Supporting information). The mechanical properties apparently decline after the DMSO treatment, which is consistent with other researchers' results [37–39]. This result clearly demonstrates that the acid-aided dissolution of UPy monomers is an effective way to maintain the energy dissipation ability of UPy dimers in MBN hydrogels.

In MBN hydrogels, the dynamic bonds which are broken to dissipate energy can effectively re-associate after dissociation, resulting in self-recovery of mechanical properties. At first, the dissociation and re-association of UPy dimers during a loading-unloading cycling tensile test are proved by Fourier transform infrared (FT-IR) spectroscopy (Figs. 3a and b). The peak at 1640 cm^{-1} is ascribed to the carboxyl groups of PAA [40], which does not change during the stretching and releasing of the hydrogels. However, both the intensities of the peaks at 2960 cm^{-1} and 1701 cm^{-1} become larger than those of the original sample after stretching to a 300% strain. These peaks are ascribed to the free N—H groups and the C=O groups of dissociated UPy motifs [41], respectively, which form quadruple hydrogen bonds in UPy dimers. Therefore, after UPy dimers being dissociated, these peaks become more intensive. During the releasing process, part of UPy dimers can be re-formed, resulting in weaker peaks of the N—H groups and the C=O groups. Meanwhile, a large residual strain and a big hysteresis loop appear after the first tensile cycle (dash line in Fig. 3c). The dissipated energy of the first cycle $E_{dissipated,1}$ can be evaluated by the area of the hysteresis loop, which is calculated to be 0.325 MJ/m^3. An immediate second loading-unloading cycle (black solid line in Fig. 3c) results in a smaller but still pronounced hysteresis loop, with $E_{dissipated,2} = 0.112$ MJ/m^3 dissipated, demonstrating the partial recovery of UPy dimers with a recovery efficiency of 34.6% (calculated according to Eq. S1 in Supporting information). As time proceeds, the UPy dimers will gradually recover and adjust the network structure, resulting in smaller residual strains, larger hysteresis loops (Fig. 3c), and higher recovery efficiencies (Fig. 3d). After resting for 1 h, the FT-IR spectrum nearly overlaps with that of the original sample (Fig. 3a). Meanwhile, the residual strain becomes ignorable and the recovery efficiency achieves 90.1%. Generally, ionic hydrogels require 4–12 h to achieve 90% recovery efficiency [16,26], but it takes only 30 min for the UPy6C0.1 sample to show a 87.2% recovery efficiency (and 90.1% recovery efficiency after 1 h). This effective recovery ability is ascribed to the high dimerization constant of UPy dimers. Noting that the recovered network cannot be exactly the same as the original one, and a small part of UPy motifs is not able to dimerize, so the full-recovery efficiency cannot achieve 100%. However, this full-recovery efficiency is still higher than those of ionic hydrogels [16,26].

The UPy-contained hydrogel network has a large amount of carboxylic acid groups, which could form ionic bonds with Fe^{3+} as new dynamic crosslinking points, functioning as an additional energy dissipation unit [26–29]. We take UPy6C0.1 as an example to investigate the mechanical properties of PAA-UPy-Fe^{3+} MBN hydrogels, because UPy6C0.1 has less influence from the inhomogeneous distribution of UPy dimers as discussed above. To fabricate the ionic crosslinked hydrogels, the UPy6C0.1 samples are immersed in a given amount of FeCA aqueous solution. As described in one of our previous work [29], Fe^{3+} can disperse homogeneously in the hydrogel network through a controllable permeation process due to competition between CA^- ions and carboxylic ions. These hydrogels are denoted as UPy6C0.1FeCA*z*, where *z* stands for the molar ratios (mol%) of FeCA with respect to AA. The mechanical

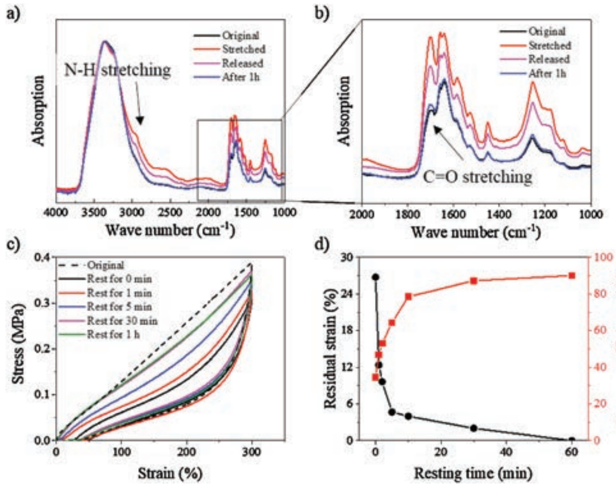

Fig. 3. (a, b) FT-IR spectra of stretched and recovered UPy6C0.1 samples. (c) Self-recovery of UPy6C0.1 and (d) the change of the residual strain and recovery efficiency with resting time.

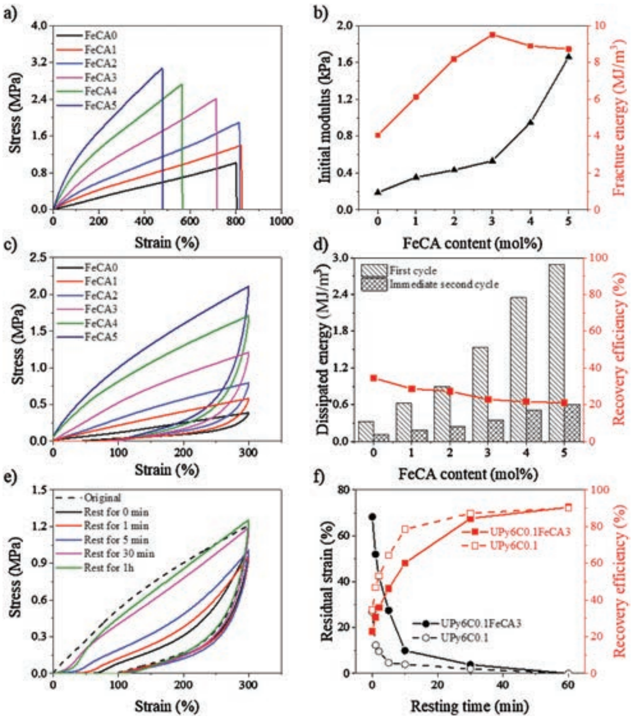

Fig. 4. (a) Stress-strain curves of PAA-UPy-Fe^{3+} MBN hydrogels (UPy6C0.1FeCA*z*) with different amount of FeCA. (b) Initial modulus and fracture energy of the PAA-UPy-Fe^{3+} MBN hydrogels as a function of FeCA content. (c) Cycling tensile tests of UPy6C0.1FeCA*z* hydrogels with different FeCA content. (d) Dissipated energy calculated from the area of the hysteresis loops of the first the immediate second cycle, and the recovery efficiency is calculated by dividing the dissipated energy of the first cycle with that of the second cycle. (e) Self-recovery of UPy6C0.1FeCA3 and (f) the change of residual strain and recovery efficiency as a function of resting time.

properties in Fig. 4 clearly shows that with the addition of ionic bonds, both the tensile modulus and the tensile strength become larger than those of UPy6C0.1 (0 mol% FeCA), which agrees with our expectation that ionic bonds can function as new energy dissipation units and further improve the mechanical properties. The tensile strength and modulus gradually increase with increasing FeCA content due to increasing amount of energy dissipated by ionic bonds. The elongation at break remains constant when FeCA content is low, and begins to decrease when FeCA content is over 2 mol%, indicating that the hydrogels become a little brittle due to increase in crosslinking density. As a result, the fracture energy (dissipated energy) increases to its maximum value at 9.51 MJ/m^3 when FeCA content is 3 mol% and then decreases. Note that in our previous PAA-Fe^{3+} systems [26–29], the dissipated energy is dominantly contributed by the dissociation of ionic bonds. In this work however, the UPy6C0.1 sample with no ionic bonds can dissipate 3.21 MJ/m^3 of energy at the strain of 715% (at which the UPy6C0.1FeCA3 sample is broken), which is more than 30% of the total dissipated energy (9.51 MJ/m^3) in UPy6C0.1FeCA3. This phenomenon again demonstrates that UPy dimers are very effective energy dissipation units for the fabrication of super-tough hydrogels.

Finally, the self-recovery ability of UPy6C0.1FeCA*z* hydrogels is investigated by loading-unloading cycling tensile test. Similar to UPy*x*C0.1 hydrogels, the UPy6C0.1FeCA*z* hydrogels also show large hysteresis loops in the first cycle (Fig. 4c), indicating effective energy dissipation in the MBN hydrogels. The dissipated energy increases as the increase of FeCA content (Fig. 4d). Due to their dynamic nature, both UPy dimers and ionic bonds can partially re-associate, thus the immediate second loading-unloading cycle can also dissipate an adequate amount of energy. The recovery efficiency of the immediate second cycle decreases as the increase of FeCA content (Fig. 4d), which is probably because that the association constant of ionic bonds is lower than that of UPy dimers. Finally, the self-recovery of UPy6C0.1FeCA3 was investigated. Similar to UPy6C0.1 sample, as time proceeds, the network structure recovers and the dynamic interactions (UPy dimers and ionic bonds) gradually re-associate, resulting in decrease of the residual strain and increase of the recovery efficiency (Figs. 4e and f). After resting for 1 h, UPy6C0.1FeCA3 can achieve 90.5% recovery efficiency, which is also faster than common ionic hydrogels [16,26]. However, the recovery rate of UPy6C0.1-FeCA3 sample is apparently lower than that of UPy6C0.1 sample (Fig. 4f), demonstrating that the high dimerization constant of UPy dimers is the main reason that leads to the rapid self-recovery of UPy-contained MBN hydrogels.

In conclusion, we introduced UPy dimers as energy dissipation units to MBN hydrogels using an acid-aided method. By employing HCl solution to protonate UPy-contained monomers, we avoided the use of organic solvent that may weaken the mechanical properties. The obtained UPy-contained MBN hydrogels (75 wt% water content) exhibit increasing tensile strength from 0.39 MPa to 2.51 MPa, and fracture energy from 1.68 MJ/m^3 to 11.1 MJ/m^3 when UPy content increases from 2 mol% to 10 mol %. Furthermore, FeCA is introduced to the system to induce homogeneous ionic bonds, which further improves the mechanical properties of the MBN hydrogels. Moreover, owing to their dynamic nature, both UPy dimers and ionic bonds can re-associate after being dissociated, resulting in an excellent self-recovery ability (around 90% recovery efficiency within 1 h). The excellent self-recovery ability mainly originates from the high dimerization constant of UPy motifs. This work provides an effective energy dissipation unit for fabricating super-tough MBN hydrogels with rapidly self-recovery ability.

Declaration of competing interest

The authors declare that there are no conflicts of interest.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.cclet.2020.04.039.

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