Soft thermoplastic silicone elastomer via supramolecular chemistry

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Introduction

Traditionally polydimethylsiloxane (PDMS) elastomers are prepared by chemical crosslinking reactions.¹ As a result, a covalently bonded polymer network is formed, which gives elastomeric properties.² However, since the polymer backbones are covalently interconnected, elastomers are relatively difficult to process and recycle. To overcome these drawbacks, different approaches have been applied to form thermoplastic elastomers (TPEs) including self-assemblies through supramolecular interactions.³ Here, we present an ultra-soft TPE by the combination of urea and 2-ureido-4[1H]-pyrimidone (UPy), possessing two hydrogen bond (H-bond) acceptors and donors.

Design of the TPE

Figure 1 shows the chemical structure of the PDMS-based TPE, where urea groups are inserted in between long PDMS chains via the condensation reaction by amine-terminated PDMS (molecular weight: 21,000 g mol⁻¹) with hexamethylene diisocyanate. Then, the both chain-ends are end-capped by urea-adjacent UPy.

Here the urea-adjacent UPys are known to form laterally stacked UPy structure (**figure 2a, 2d**).⁴ Therefore, the UPy serves as a strong crosslinker between the PDMS chains (**figure 2**), whereas the weak H-bond (urea, **figure 2b**) serves as a sacrificial bond under deformation.

Figure 1. Chemical structure of the PDMS TPE based on urea (grey) and urea-adjacent UPy (red)

Figure 2. (a) schematic structure of the PDMS TPE, hydrogen bond by (b) urea, (c) dimerization of UPy, and (d) laterally stacked dimers of UPys by adjacent urea

Characterization

Figure 3. (a) temperature sweep tests, (b) frequency sweep tests in different temperature, (c) tensile tests in different deformation rate, (d) normalized data to the 1st cycle from successive cyclic tensile tests (deformation rate: 30 mm min⁻¹, 300 % elongation)

Application as dielectric elastomer actuator

Figure 4. (a) schematic image of corrugated dielectric elastomer actuator with silver electrodes and its bilateral deformation, (b) actuation strain under given potentials

Figure 5. (a) the images of DEA before and after the electrical breakdowns, (b) microscopic images of the surface of the electrodes

Figure 6. scheme of autonomous self-healing mechanism of the DEA

Conclusion

The combination of urea and urea-adjacent UPy with PDMS yields the ultra-soft (Young's modulus: 178 kPa) PDMS TPE with 119 °C of melting temperature. The laterally stacked urea-adjacent UPys are intact under deformation and the urea is responsible for the softness of the elastomer.

The electrical breakdown of the DEA is observed on the surface of the electrode yet, it does not cause detrimental failure within the given potentials.

It is known that the electrical breakdown is accompanied by localized electric field and heat (**figure 6**, left). Then, the self-clearing effect of the silver electrode⁵ and thermo-reversibility of the TPE allow the TPE to heal the voids that were formed by the electrical breakdowns.

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