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Peculiar fracture behavior of ultra-soft hydrogel with the effect of strain-induced anisotropic structure

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The stiffness of human tissue and organs varies over a wide range of modules. Deformation and fracture properties of ultra-soft biological material have received less attention when compared to tough materials, which is of vital importance in medical and tissue engineering sciences. In this work, we investigate the fracture behavior of model ultra-soft hydrogels by puncture tests, which are suitable for soft solids. We used chemically cross-linked poly (vinyl alcohol) (PVA) hydrogels as model systems with elastic modulus ranging from 100 – 2000 Pa. We report an unexpected intriguing fracture phenomenon in puncture tests of PVA hydrogel coupled with birefringence observation. We found (1) that the failure initiates above the needle tip for several millimeters, rather than at the needle tip edge where the stress should be highly concentrated, and (2) that there is a permanent birefringence pattern remaining when unloading and pulling out the needle. Also, By the cyclic loading-unloading puncture test, we observed a hysteresis loop (Fig.1a) and a permanent birefringence pattern when large displacement is applied even before the occurrence of fracture (Fig.1b-e)). Interestingly, when we performed the same experiments with a PVA having a low hydrolysis degree, we found that fracture initiates at the needle tip edge and no permanent birefringence structure remains after a puncture. From these results, we conclude that the strain-induced anisotropic structure is mainly from the strong associations of hydrogen bonding between the –OH groups on PVA chains, which strengthen the network around the needle, displacing the crack initiation point from the needle tip edge. However, the fracture resistance (the nominal stress over G') of PVA showing above needle fracture behavior is weakened by the anisotropic structure, compared to PVA showing right-needle fracture behavior. This suggests that the crack initiates at the interface between zone with strong association and without association.

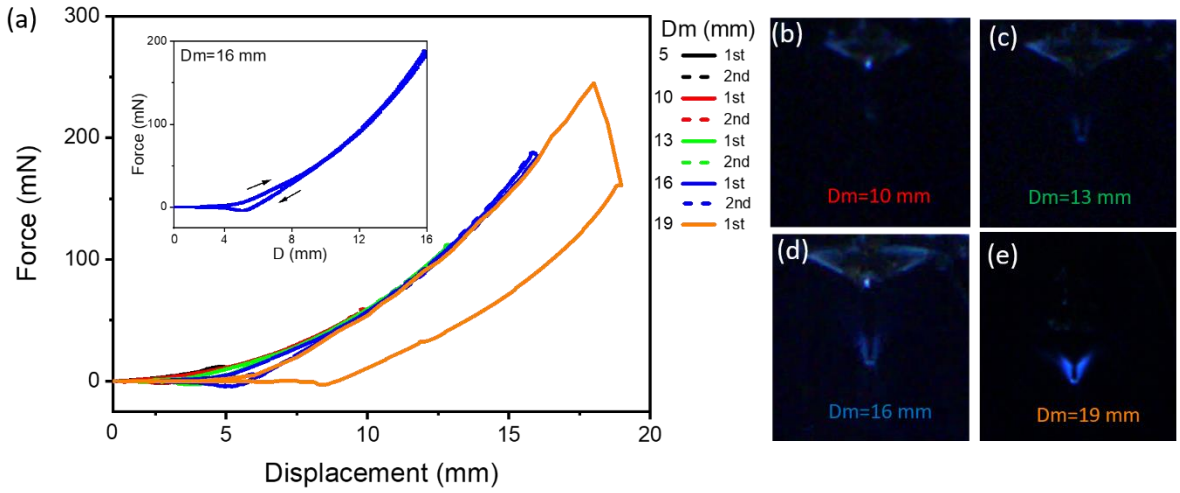

Figure 1. Cyclic loading and unloading test of puncture process. (a): Cyclic loading and unloading process for PVA hydrogel (G'=1000 Pa and R=0.23 mm) with different indentation depths. (b)(c)(d)(e): the birefringence morphology after pulling out the needle.