Role of Damage by Sacrificial Bonds: Intrinsic Differences between Fatigue and Toughness Mechanisms in Multiple Network Elastomers

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Elastomers are ubiquitous in engineering applications that require large reversible deformations (1). Although toughness remains an important consideration to prevent catastrophic failure at large deformations, lifetime often results from the progressive growth of an inherent flaw over 10-100 million cycles of low deformation. This damage mechanism, known as fatigue crack propagation, is usually evaluated by monitoring the crack growth rate of a pre-cracked specimen under cyclic loading over a range of applied energy release rates (2, 3). Typically, elastomers suffer from fatigue crack propagation at low cyclic loads, and exhibit cyclic fatigue thresholds G0 ~0.05-0.1kJ.m-2 (at which the crack is effectively stopped) significantly below the fracture toughness Gc ~1-100kJ.m-2, therefore suffering from fatigue crack propagation over a large experimental conditions window.

Here, we use double-network elastomers as model materials to understand the role of damage by sacrificial bond scision on their mechanical durability and fracture toughness. These elastomers are composed of a pre-stretched, stiff, and continuous filler network embedded in a highly extensible, soft, and incompressible matrix. Even though their elastic properties are controlled by the filler network, similar to filled elastomers, their toughness results from energy dissipation by sacrificial bonds (4, 5). We tagged the filler network with mechanofluorescent probes based on pi-extended anthracene-maleimide adducts. Upon chain elongation, these probes undergo a force-induced cycloreversion reaction that results in pi-extended anthracene moieties of high quantum yield and stability to photobleaching (6). As such, they serve as ideal fluorophores for quantifying polymer chain scission in a post-mortem manner (7). These probes enable reconstruction of 3D damage maps near the crack surface of fractured specimens. This combination of network design and damage quantification provides novel insights on structure-property relationships, and fracture mechanisms under cyclic and monotonic loading. To conclude, we show that optimum durability and fracture toughness result respectively from different mechanisms of damage accumulation and localization ahead of the crack tip in multiple network elastomers.

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References

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