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Toughening elastomers with sacrificial bonds and watching them break[1]

COSTANTINO CRETON, ESPCI ParisTech - UPMC - CNRS

Most unfilled elastomers are relatively brittle, in particular when the average molecular weight between crosslinks is lower than the average molecular weight between entanglements. We created a new class of tough elastomers by introducing isotropically prestretched chains inside ordinary acrylic elastomers by successive swelling and polymerization steps. These new materials combine a high entanglement density with a densely crosslinked structure reaching elastic moduli of 4 MPa and fracture strength of 25 MPa. The highly prestretched chains are the minority in the material and can break in the bulk of the material before catastrophic failure occurs, increasing the toughness of the material by two orders of magnitude up to 5 kJ/m^2. To investigate the details of the toughening mechanism we introduced specific sacrificial dioxetane bonds in the prestretched chains that emit light when they break. In uniaxial extension cyclic experiments, we checked that the light emission corresponded exactly and quantitatively to the energy dissipation in each cycle demonstrating that short chains break first and long chains later. We then watched crack propagation in notched samples and mapped spatially the location of bond breakage ahead of the crack tip before and during propagation. This new toughening mechanism for elastomers creates superentangled rubbers and is ideally suited to overcome the trade-off between toughness and stiffness of ordinary elastomers.

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