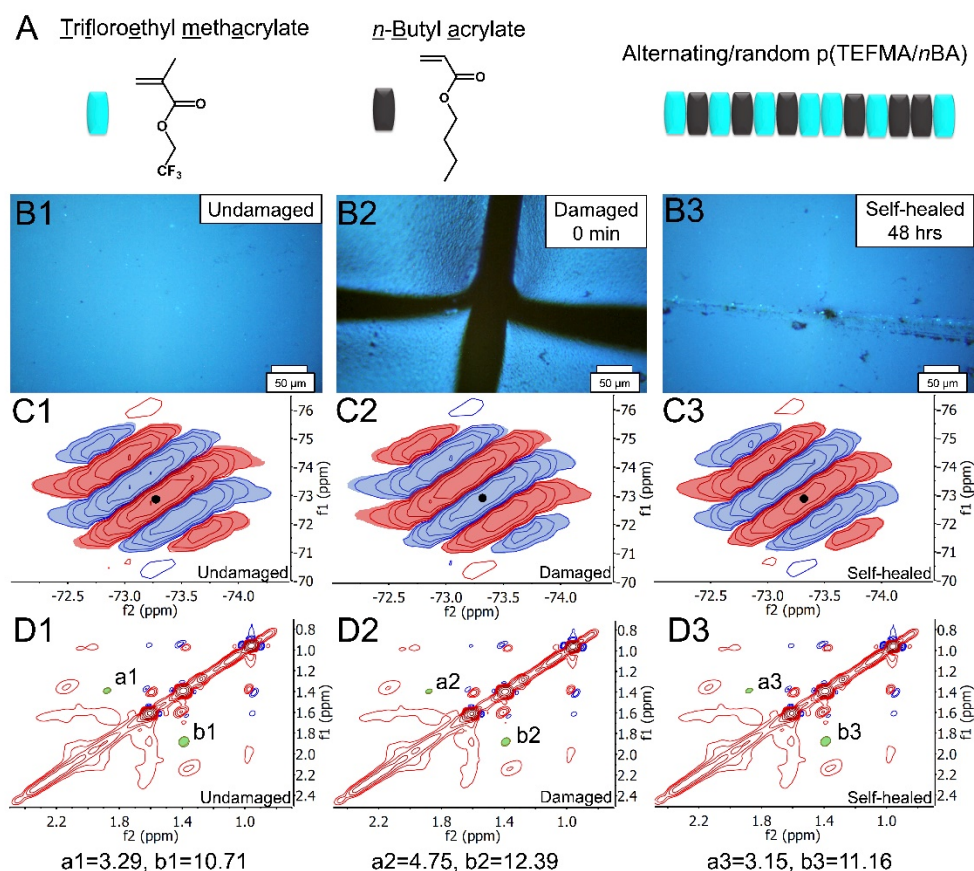


Self-healable Fluorinated copolymers governed by dipolar interactions

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We developed a route to obtain self-healable properties in thermoplastic copolymers that rely on non-covalent dipolar interactions present in essentially all macromolecules, and particularly fluorine-containing copolymers. The combination of dipolar interactions between C-F and C=O bonds as well as CH₂/CH₃ entities facilitates self-healing without external intervention. The presence of dipole-dipole, dipole-induced dipole, and induced-dipole induced dipole interactions lead to a viscoelastic response that controls macroscopic autonomous multicycle self-healing of fluorinated copolymers under ambient conditions. Energetically favorable dipolar forces attributed to monomer sequence and monomer molar ratios induces desirable copolymer tacticities, enabling entropic energy recovery stored during mechanical damage. The use of dipolar forces instead of chemical or physical modifications not only eliminates additional alternations enabling multiple damage-repair cycles but also provides further opportunity for designing self-healable commodity thermoplastics.



Chemical structure of 2,2,2-trifluoroethyl methacrylate (TFEMA) and *n*-butyl acrylate (nBA) as well as p(TFEMA/nBA) copolymers; (A) Optical images of p(TFEMA/nBA) copolymer films: undamaged (B1); damaged (B2) – cut size: ~50 μm; and self-healed (B3). 2D ¹⁹F NOESY NMR spectra of undamaged (C1), damaged (C2) and self-healed (C3); 2D ¹H NOESY NMR spectra of undamaged (D1), damaged (D2) and self-healed (D3) copolymer