

Ultra-stretchable vitrimers with tuneable damping and mechanical response

Jiaxin Zhao^{1,2}, Nicholas Warren^{3,4}, Richard Mandle^{1,2}, Peter Hine¹, Daniel J. Read⁵, Andrew J Wilson^{2,6}, Johan Mattsson^{*1}

¹ School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, United Kingdom

² School of Chemistry, University of Leeds, Leeds LS2 9JT, United Kingdom

³ School of Chemical and Process Engineering, University of Leeds, Leeds LS2 9JT, United Kingdom

⁴ School of Chemical, Materials and Biological Engineering, University of Sheffield, Sheffield S10 2TN, United Kingdom

⁵ School of Mathematics, University of Leeds, Leeds LS2 9JT, United Kingdom

⁶ School of Chemistry, University of Birmingham, Birmingham B15 2TT, United Kingdom

*k.j.l.mattsson@leeds.ac.uk

Elastomers that combine considerable stretchability with high mechanical strength and toughness are important for applications such as actuators, sensors, soft robotics, or wearable electronics. Standard elastomers can readily achieve tensile strains of several 100% combined with tensile strengths of ≥ 10 MPa. However, elastomers that can sustain strains of several 1000% combined with a high tensile strength are more challenging to produce and often show significant strain-softening upon yielding.

We demonstrate that dynamically cross-linked networks, characterised by associative exchange reactions, so-called **vitrimers**, can achieve these requirements, while also providing other attractive properties such as excellent mechanical damping, thermo-mechanical and chemical re-processability, and effective self-healing.

Our vitrimers are based on poly(methyl acrylate) with dynamic crosslinks utilising the associative exchange reactions of dioxaboralane metathesis [1]. These vitrimers demonstrate an excellent combination of ultra-stretchability (up to 8000 %), mechanical toughness (20–55 MJ/m³), and thermal stability up to $T \sim 250^\circ\text{C}$. They also show excellent mechanical damping properties with a maximum mechanical loss factor $\tan(\delta)$ value of ~ 2 -3 and a $\tan(\delta) > 0.3$ across a wide (and for application relevant) frequency range of ~ 5 decades (0.001–100 Hz). The balance between the specific material properties is tuneable by variation of both the crosslink density and the processing conditions.

We present a thorough characterisation of the vitrimer material properties including oscillatory rheology and tensile deformation to determine the relevant mechanical response, and calorimetry and broadband dielectric spectroscopy to map out the relaxation dynamics.

Reference:

- [1] Zhao, J., Warren, N.J., Mandle, R., Hine, P., Read, D.J., Wilson, A.J., Mattsson, J., "Ultra-stretchable and self-healable vitrimers with tuneable damping and mechanical response", *arXiv:2503.03701*.