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From Molecular Mobility to Macroscopic Recyclability – A Multiscale Study on Vitrimers

Vitrimers are redefining the boundaries between thermosets and thermoplastics by combining permanent network integrity with true recyclability enabled by dynamic covalent chemistry. Yet, the key question remains: how does molecular mobility at the smallest length and time scales translate into macroscopic reprocessability? In this work, we unravel this connection through a multiscale dynamics driven investigation of a fully bio based vitrimer composed of glycerol triglycidyl ether and a vanillin derived imine hardener operating via catalyst free imine metathesis.

Molecular dynamics spanning from localized vibrational motions to cooperative segmental relaxations are captured by a powerful combination of broadband dielectric spectroscopy, mechanical spectroscopy, and neutron scattering. Dielectric relaxation fingerprints are directly correlated with viscoelastic response and neutron derived vibrational and quasielastic dynamics, allowing us to trace how microscopic mobility governs network rearrangements under thermal and mechanical activation. Special emphasis is placed on the low frequency vibrational density of states and the Boson peak as ultra sensitive probes of local elasticity and network integrity during recycling.

By correlating molecular scale mobility with macroscopic thermomechanical performance before and after reprocessing, this study provides direct experimental evidence for the preservation of dynamic and mechanical functionality upon vitrimer recycling. The results establish a fundamental structure dynamics property relationship for vitrimers and highlight the decisive role of molecular mobility in enabling sustainable, recyclable thermosetting polymers for high performance applications.

Session

Primary authors: SZYMONIAK, Paulina (Bundesanstalt für Materialforschung und –prüfung (BAM)); KRUTEVA, Margarita (Forschungszentrum Jülich); ZORN, Reiner; SCHOENHALS, Andreas

Presenter: SZYMONIAK, Paulina (Bundesanstalt für Materialforschung und –prüfung (BAM))

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