Summer School on Polymeric Networks for Sustainable Development

July 6th – 12th, 2025

Viscoelasticity of well-characterized Vitrimer Melts

Arcangela Russo[1,2,3], Saibal Bhaumik[4], Konstantinos Ntetsikas[4], Nikos Hadjichristidis[4], Benoit Loppinet[1], Dimitris Vlassopoulos[1,2]

1 FORTH , Institute of Electronic Structure and Laser, Heraklion, Crete, Greece

2 University of Crete, Department of Materials Science and Technology, Heraklion, Crete, Greece

3 Université Catholique de Louvain, Institute of Condensed Matter and Nanosciences, Louvain La Neuve, Belgium

4 King Abdullah University of Science and Technology (KAUST), Physical Sciences and Engineering Division, Kingdom of Saudi Arabia

Vitrimers uniquely combine the advantageous properties of both thermoset and thermoplastic materials. Their defining characteristic is the presence of dynamic covalent bonds, which exhibit an Arrhenius temperature dependence.

This study investigates the linear viscoelastic (LVE) behavior of well-characterized model polyisoprene vitrimers and their solutions, providing insights into the complex interplay between chemical dynamics and mechanical responses. We explore the evolution of storage and loss moduli across various frequencies and temperatures, focusing on the strong time dependence and the role of cluster formation of the bonding motifs. LVE master curves are formed and compared against the respective precursors, revealing the dramatic extent of the plateau region and the distinct signature features of these materials. These features are shown to be tailored by systematically changing the molecular weight of the precursor. The vitrimer networks exhibit a gradual transition from elastic behavior to viscoelastic flow, driven by the activation of bond exchange reactions.

These findings open avenues for the design of next-generation vitrimers and their application as compatibilizers for innovative blends with adjustable mechanical properties, facilitating transformative possibilities in self-healing materials, sustainable manufacturing, and more.