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Exploring Vitrimer Dynamics: Effect of Cross-Linking Density on the Apparent Activation Energy for Flow

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Vitrimers are a unique class of polymeric networks that incorporate dynamic covalent bonds, enabling them to undergo bond exchange reactions and rearrange their topology in response to external stimuli. These materials have sparked significant interest in the field of material science due to their potential in developing sustainable materials with enhanced mechanical and processing properties. However, a fundamental understanding of their viscoelastic behaviour, in particular the temperature dependence of their storage and loss moduli, remains elusive, and conventional models for supramolecular polymers often fail to accurately describe experimental observations.

Elucidating the temperature-dependent dynamics of vitrimers is critical for optimizing their performance in industrial and commercial applications, supporting broader sustainability goals. Therefore, the development of new approaches to explain the viscoelastic properties of vitrimers is essential.

In this work, we investigate polystyrene-based vitrimers with varying concentrations of dioxaborolane cross-linkers, characterizing their mechanical properties and relaxation dynamics. The Time-Temperature-Superposition (TTS) principle is used to extract the activation energy for flow. The influence of various parameters, including molecular weight and chemistry of the backbone, will be further explored to understand their impact on the material properties.

Building on these experimental data, we adapt the Time-Marching Algorithm (TMA) to incorporate dynamic covalent bond exchange. Our aim is to offer a theoretical foundation for the relationship between cross-linking density and activation energy for flow in vitrimers.