Data-driven identification and analysis of the glass transition in polymer melts

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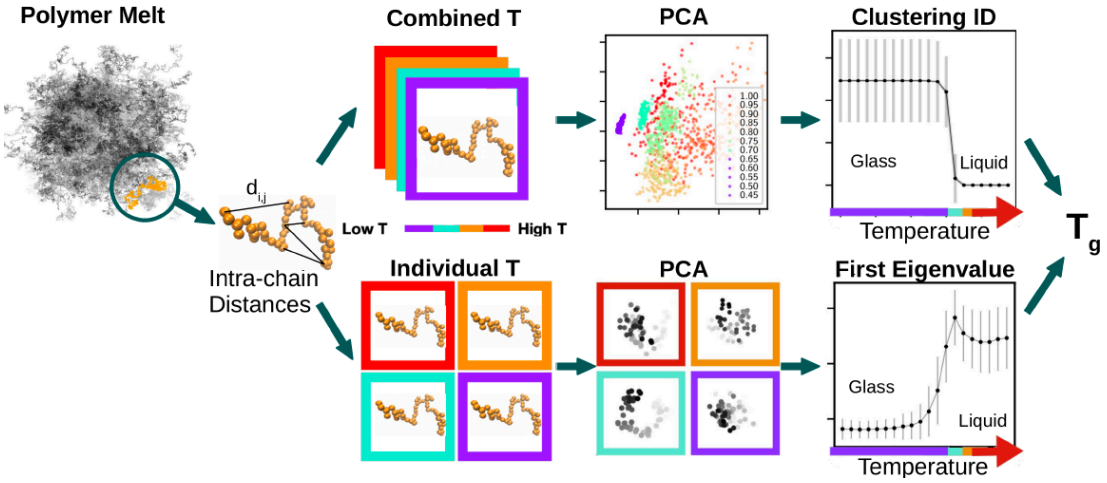
On cooling, the dynamical properties of many polymer melts slow down exponentially, leading to a glassy state without any drastic change in static structure. Understanding the nature of glass transition, as well as precise estimation of the glass transition temperature (Tg) for polymeric materials, remain open questions in both experimental and theoretical polymer sciences. We propose a data-driven approach, which utilises the high-resolution details accessible through the molecular dynamics simulation and considers the structural information of individual chains. It clearly identifies the glass transition temperature of polymer melts of semiflexible chains.
By combining principal component analysis (PCA) and clustering (shown in the schematic), we identify glass transition temperature at the asymptotic limit even from relatively short-time trajectories, which just reach into the Rouse-like monomer



displacement regime [1]. We demonstrate that fluctuations captured by the principal component analysis reflect the change in a chain's behaviour: from conformational rearrangement above to small vibrations below the glass transition temperature. We demonstrate the generality of the approach by using different dimensionality reduction and clustering approaches. The method can be applied to a wide range of systems with microscopic/atomistic information. More recently we applied this methodology to all-atom acrylic paint systems [2]. Our study reveals the explicit role of backbone and side chain residues to determine the glass transition temperature.

[1] Data-driven identification and analysis of the glass transition in polymer melts,
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[2] Defining glass transition temperature in acrylic polymeric melts with machine learning methods,
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