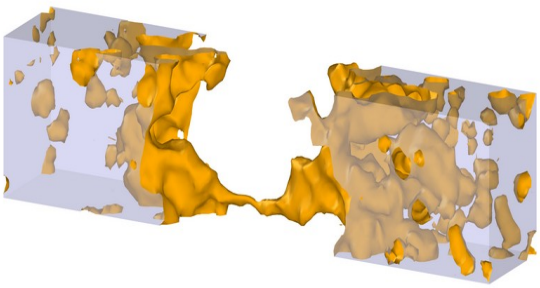
A Multiscale MD-QM/MM Approach for Modeling Fracture Behavior in Epoxy Resins

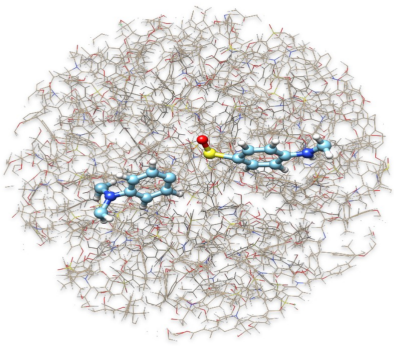
Barışcan Arıcan[1], ChristianWick[1], Ana-Sunčana Smith[1,2]

[1] *PULS group, Institute for Theoretical Physics, IZNF, FAU Erlangen-Nürnberg, 91058 Erlangen, Germany*

[2] *Group of Computational Life Sciences, Division of Physical Chemistry, Ruder Bošković Institute 10000 Zagreb, Croatia*

Epoxy resins are essential in advanced engineering applications due to their superior mechanical properties. However, predicting their fracture behavior at the molecular level remains challenging. This study introduces a multiscale framework that integrates molecular dynamics (MD) simulations with quantum mechanics/molecular mechanics (QM/MM) calculations, specifically employing the ONIOM method[1], to examine bond rupture and crack propagation in epoxy networks subjected to tensile straining. This approach leverages MD to capture the overall mechanical response of the material, while transitioning to QM/MM to accurately describe the inherently quantum mechanical nature of bond breaking. The framework utilizes spin density as a quantum mechanical criterion for bond breakage, where many previous approaches have relied solely on geometric distance criteria.





The MD simulations monitor strain accumulation and identify bonds that have elongated beyond critical thresholds, indicating impending fracture. Upon reaching these thresholds, the simulation transitions to QM/MM calculations. In this phase, bond breakage is dynamically assessed using the spin density, while explicitly considering long-range interactions and charge distributions within the surrounding chemical environment. This approach necessitates updating the molecular topology and recalculating partial atomic charges to accurately represent the evolving chemical structure during fracture events. Our method employs the block chemistry approach[2] to maintain charge-neutrality and ensure a chemically accurate representation of fractured sites. This on-the-fly methodology enables efficient and precise modeling of fracture events and their impact on larger-scale material behavior, providing a foundation for future studies of epoxy resin failure mechanisms.

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[2] M. Livraghi, S. Pahi, P. Nowakowski, D. M. Smith, C. R. Wick, A.-S. Smith, J. Phys. Chem. B, 2023, 127, 7648–7662.