

Chemical Reaction Modelling of Carbon Fiber Production for MC/MD Hybrid Simulation

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Extended Abstract

Carbon fibers (CF) are widely utilized in composite materials because of their excellent stiffness and tensile strength. For further improvement, it is essential to understand the formation mechanism of defects and to properly control the fabrication process with suppressing the defect formation. Since these defects are often of nano-meter scale, numerical simulations with molecular dynamics (MD) methods provide good tools for investigation. However, the gap in time scale between this kind of simulations and the real systems is huge. In this research, we have focused on important chemical reactions, i.e., cyclization, dehydrogenation, and oxidation, which occur mainly at the first stage of CF production in polyacrylonitrile (PAN) precursor assembly under elevated temperature and tensile stress. In normal situations, it typically takes $10\sim 10^3$ s for these chemical reactions to be completed, which is well beyond the time scale of conventional molecular simulations. To address this issue, we have proposed a novel MC (Monte Carlo) / MD hybrid scheme with a simple stochastic reaction model, which makes it possible to deal with the evolution of the target system on a much larger time scale [1].

In the hybrid scheme chemical reactions stochastically occur by search of possible reaction sites and judgment on reaction rate models. We have picked up the thermal stabilization process which consist of seven relevant reactions, for each of which we made a reaction template. We have developed a python-base package combined with LAMMPS [2]. When we a priori assume a simple reaction path for each reaction and empirically give its MC parameters (e.g., atomic distance criteria and reaction rate), we successfully traced the structural change for 10^3 s. Comparison with FTIR experiments confirms a similar trend in functional group changes.

For further improvement of models, we are investigating each chemical reaction in some detail by quantum calculations with DFTB+ [3]. For the cyclization reactions which form ladder structures, we have chosen two atomic distances as the reaction coordinates and the potential energy surface (PES) is investigated, from which the reaction path and the activation energy are modelled. Investigation on the dehydrogenation and oxidation are also in progress.

References

- [1] Mitsuhiro Matsumoto, Bunta Kishimoto, Hiroki Hayami, Tomoyuki Kotani, Daisuke Yuhara, and Ryuichi Hiraiwa, "MD/MC Hybrid Simulation Scheme for Carbon Fiber Fabrication Process," in *Proc. XXXV IUPAP Conf. Comp. Phys.*, Thessaloniki, Greece, 2024, No. 78.
- [2] LAMMPS webpage, https://docs.lammps.org/Python_module.html
- [3] DFTB+ webpage, <https://dftbplus.org/index.html>