Reliable calculation of thermophysical lubricant properties from equilibrium molecular dynamics simulations

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Many machine components, such as bearings and gears, operate ideally under the so-called Thermo-Elasto-Hydrodynamic (TEHL) lubrication regime where the surfaces that are in relative motion are completely separated by a thin lubricant film (< 1 μm). In this specific regime, lubricants can experience ultra-high pressures, very high shear rates and high temperatures. These state variables, heavily influence the rheological properties of the lubricants due to intensified interaction between molecules. As it is very difficult to perform experiments at high pressures, Molecular Dynamics (MD) simulations offer a promising alternative to study lubricant rheology at high pressures to extract thermomechanical properties of lubricants at relevant TEHL conditions. However, the accuracy with which the properties are obtained from MD simulations is determined by 1) the accuracy of the force fields used for describing the inter-atomic interactions and 2) the specific way of postprocessing the MD-trajectories to reliably calculate the variables of interest. The latter is not standardized and may have a serious impact on the reliability of the calculated properties, also the former introduces potential errors that are not clear *a priori*.

In this work, Equilibrium Molecular Dynamics (EMD) simulations are considered to investigate the complex nanoscale behavior of 2,2,4-trimethlyhexane and to calculate all its thermo-mechanical properties relevant to TEHL, i.e. density, viscosity, thermal conductivity and heat capacity, as a function of temperature and hydrostatic pressures up to 1 GPa. A reliable postprocessing procedure is developed to analyze EMD trajectories using Green-Kubo and Einstein formalisms for the calculations of viscosity and thermal conductivity. Simulated properties for three common all-atom force fields, namely OPLS-AA, COMPASS and AIREBO-m, are compared to experimental data. EMD results with COMPASS force field lied close to experimental data up to 500 MPa pressures. At higher pressures, especially the viscosity was significantly underestimated. On the other hand, the other force fields overestimated simulated properties even at ambient conditions. Our preliminary results showed that conventional force fields are not appropriate for high pressures as the non-bonded terms in these force fields are not parameterized to account for ultra-high pressure conditions and requires further parametrization from ab-initio calculations to account for extreme conditions.

Keywords: Lubricant modelling, Molecular Dynamics, Thermo-Elastrohydronamic Lubrication, Force Fields

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