

Benchmarking Transport Properties of Ionic Liquids with a Universal Machine Learning Force Field: SO3LR vs. Classical Force Fields for EMIM NTf₂

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Universal machine learning force fields (MLFFs) trained on large quantum chemistry datasets promise transferable, DFT-quality accuracy without system-specific parametrization. SO3LR [1], trained on ~4 million structures at the PBE0+MBD level, incorporates long-range electrostatics and dispersion and has shown strong performance on biomolecular systems. Systematic benchmarks of universal MLFFs for bulk ionic liquid (IL) properties remain, however, largely absent. ILs present a stringent test: their physical properties are governed by Coulombic interactions, hydrogen bonding, conformational flexibility, and collective ion dynamics. Classical non-polarizable force fields systematically underestimate transport properties in ILs, typically requiring empirical charge scaling to partially compensate for missing polarization effects [2,6]. While polarizable force fields such as APPLE&P achieve good accuracy for specific ILs [3], their parametrization is system-specific and does not transfer across ion families, limiting their utility for high-throughput screening of the vast IL chemical space. Here we benchmark SO3LR against the established classical force fields OPLS [2] and APPLE&P [3] for 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM NTf₂) across 290–330 K, comparing density, liquid structure, ion cage lifetimes and transport properties against experiment. SO3LR overestimates the experimental density by ~9% (~1.66 vs. ~1.52 g/cm³ at 300 K), unlike OPLS which reproduces it to within 1%. However, radial distribution function analysis of C–H···O and C–H···F hydrogen bond contacts shows that SO3LR correctly captures the short-range geometry — with first-shell peak positions and heights consistent with AIMD reference calculations [4] — while OPLS and APPLE&P underestimate the oxygen contact peak. For the C–H···F contact, SO3LR deviates from AIMD, which we attribute to incorrect NTf₂ conformer populations confirmed by dihedral distribution analysis. This indicates that the density failure does not originate from misrepresented local H-bond structure but from deficiencies in intermediate-range or many-body ion–ion interactions. Transport properties (self-diffusion coefficients and ionic conductivity) and ion cage lifetimes are discussed in the context of the structural findings.

References

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Figures

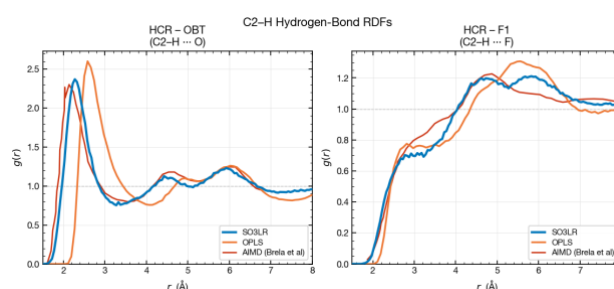


Figure 1. Radial distribution functions for (a) HCR-OBT (C2–H···O) and (b) HCR-F1 (C2–H···F) at 300 K. SO3LR reproduces the short-range oxygen contact consistent with AIMD [4], while OPLS underestimates the first-shell peak. For the C2–H···F contact, SO3LR deviates from the AIMD reference, which we attribute to incorrect NTf₂ conformer populations — the cis/trans dihedral distribution in SO3LR differs from both OPLS and DFT reference calculations.