MOLECULAR SIMULATION OF CO_2 SORPTION AND DYNAMICS IN IONIC LIQUIDS UP TO HIGH PRESSURES

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ABSTRACT

Ionic liquids (ILs) are organic salts with melting points usually near room temperature (RTILs). ILs exhibit a unique combination of properties such as extremely low vapor pressures, thermal stability, chemical tunability, good electrolytic, separation and solvation properties, non-flammability and easy recycling. The combination of these properties renders them ideal for use in a number of industrial applications [1] such as solvents and catalysts in synthesis, as lubricants, as electrolytes in electrochemistry and in gas storage and CO_2 capture applications. Molecular simulation methods are proven to be extremely valuable means of reliable property prediction, enabling simultaneously the elucidation of the underlying mechanisms that are responsible for the macroscopic behavior of ILs aiming at the molecular design of materials with controlled properties.

The present work is focuses on the molecular simulation of $[C_4mim^+][TCM^-]$ ionic liquid in mixtures with CO_2 using an optimized and validated classical atomistic force field [2,3]. Molecular simulations have been applied at various thermodynamic conditions and CO_2 concentrations up to high pressures. A wide range of properties such thermodynamic, structure and transport properties have been calculated by performing very long molecular dynamics simulation at various ensembles and the effect of CO_2 concentration and of temperature on the above properties has been thoroughly investigated. Sorption isotherms and associated volumetric effects have been extracted using an multistage iterative scheme [4] that incorporated MD simulations in the NPT ensemble and the Widom particle insertion method. The dynamics is enhanced as the CO_2 concentration is increased while simultaneously the viscosity is decreased in a self-consistent manner. The presence of CO_2 induces a decrease in the system's molar volume, which is in very good agreement with experimental data in the same ionic liquid/CO_2 systems while the local structure remains rather unaffected even at high CO_2 concentrations.

KEYWORDS: Ionic liquids, Molecular Simulation, Carbon Dioxide, Sorption, Diffusion

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