Characterization of Mutual Diffusion in Liquids with Dissolved Gases

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Systems based on liquids containing dissolved gases are of interest in many areas of chemical and energy engineering. Well-known examples include hydrocarbons from synthesis gas via the Fischer-Topsch process, liquid organic hydrogen carriers for the storage of hydrogen, and ionic liquids for carbon dioxide sequestration from flue gas. One key property required for the optimum design of corresponding processes is the Fick diffusivity (D_{11}). Therefore, a fundamental understanding of the influence of varying thermodynamic states including the composition on the diffusive mass transport in liquids with dissolved gases is necessary.

The objective of this presentation is to summarize investigations of binary diffusion coefficients of liquids with dissolved gases performed at AOT-TP. In order to gain a fundamental understanding of the influence of the molecular characteristics on the diffusive mass transport, dynamic light scattering (DLS) experiments as well as equilibrium molecular dynamics (EMD) simulations were used to access D_{11}. The systematic studies include alkanes, alcohols, and ionic liquids as solvents as well as 13 different gases with varying molecular characteristics. From DLS, studying microscopic concentration fluctuations at macroscopic thermodynamic equilibrium, D_{11} can be accessed with typical expanded uncertainties of 5%. These results serve as a reference for the validation of EMD simulations, which give access to the self-diffusivities of both components and the Maxwell-Stefan diffusivity. The latter property is combined with the thermodynamic factor, obtained from simulations or equations of state, to determine D_{11} with typical expanded uncertainties of 10%. Agreement within combined uncertainties is generally found between the simulated and experimental diffusivities. The results from experiments and simulations, are used to develop a simple prediction model, based solely on the thermophysical properties of the pure components, for D_{11} at infinite dilution of the dissolved gas. Good agreement between the predicted and measured diffusivities is found with average deviations below 15%.