Mass Diffusion Coefficients of Binary Gas Mixtures by Theoretical and Experimental Methods

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Reliable mass diffusivity data of gas mixtures are required for the optimum design of apparatus and processes. As diffusion measurements cannot be performed for all relevant systems at each thermodynamic state, experiments need to be accompanied by reliable theoretical methods. Vice versa, theoretical calculations can often only be as accurate as the experimental data used for their validation. In a related research project, theoretical and experimental methods are combined for obtaining a detailed understanding about mass diffusion in gaseous systems at densities ranging between the dilute and dense gas regime. The objective of the present study is to characterize mass diffusion in gaseous systems by means of molecular dynamics (MD) simulations using new molecular models derived from *ab initio* calculations. As model systems, binary gas mixtures containing methane, propane, and carbon dioxide were investigated at temperatures from (293 to 355) K over the entire concentration range. While from holographic interferometry applied to a Loschmidt cell the Fick diffusivity is obtained in the low-density regime at pressures up to 1 MPa, dynamic light scattering gives access to this property in the dense gas regime up to 10 MPa. The measured data with typical expanded uncertainties smaller than 5 % serve as references for equilibrium MD simulations. Despite the relatively low densities, the self-diffusivities and Fick diffusivity in the binary systems could be calculated with typical expanded statistical uncertainties of 3 % and 10 % for pressures between (0.1 and 10) MPa. Here, the Fick diffusivity was determined by independent calculations of the Maxwell-Stefan diffusivity and the thermodynamic factor. The MD results from common molecular models are compared with those from new pair-specific models obtained by *ab initio* calculations at the proximity of zero density. Agreement within combined expanded uncertainties was found between the simulated and experimental Fick diffusivities at the studied state points.