

Dynamic Light Scattering for the Determination of Thermal and Mutual Diffusivities of Liquids with Dissolved Gases in Chemical and Energy Engineering

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At present, increasing technical interest in liquids containing dissolved gases can be found in energy technology. For example, ionic liquids (ILs) are potential solvents for the separation of carbon dioxide from flue gas due to their attractive properties. Furthermore, the synthesis of hydrocarbons from hydrogen and carbon monoxide by the Fischer-Tropsch process is used for the production of high-value fuels. The mutual diffusivity in corresponding mixtures strongly affects the selectivity and kinetics, e.g., in separation processes or catalytic reactions. Until now, however, hardly any binary diffusion coefficients can be found for mixtures of ILs with carbon dioxide as well as liquid hydrocarbons with hydrogen and/or carbon monoxide. This is caused by the limitations of available measuring methods. The Taylor dispersion technique can only be applied at low viscosities. For the Loschmidt-cell technique, the slow diffusion processes in liquid systems generally cause extensive measuring times. Other methods based on the analysis of time-dependent absorption of gases in liquids are often affected by additional flow contributions.

The present contribution demonstrates that Dynamic Light Scattering (DLS) can be applied for a reliable simultaneous determination of mutual and thermal diffusivity of liquids containing dissolved gases. The determination of both properties by DLS is based on the temporal analysis of microscopic fluctuations of concentration and temperature present in a fluid mixture in macroscopic thermodynamic equilibrium. Measurement results are presented for mixtures of 1-alkyl-3-methylimidazolium-based ILs having the anions tetracyanoborate and tricyanomethanide with carbon dioxide as well as of different liquid n-alkanes with carbon monoxide or hydrogen over a wide range of temperature and pressure or composition. Here, special attention is paid to the separation of signals from concentration and temperature fluctuations, which match under certain conditions. Results for the mutual diffusivity agree well with experimental data from conventional techniques and data derived from molecular dynamics (MD) simulations.