International Conference on Advanced Optical Technologies
University of Erlangen-Nürnberg, March 13th – 15th 2019

Fick Diffusivity of Binary Fluid Mixtures Consisting of Methane, Propane, and Carbon Dioxide by Optical and Theoretical Methods

U. A. Higgoda, M. Piszko, P. Zangi,
M. H. Rausch, C. Giraudet, T. M. Koller, and A. P. Fröba

Institute of Advanced Optical Technologies - Thermophysical Properties (AOT-TP),
Friedrich-Alexander-University Erlangen-Nürnberg (FAU)
ubaya.higgoda@fau.de

Mixtures containing natural gas components such as methane, propane, and carbon dioxide are important in many areas of process and energy engineering. For the optimum design of apparatuses and processes related to such mixtures, accurate data for the Fick diffusivity are required over a broad range of thermodynamic state. For this, reliable measurement techniques are needed to provide a reference database and to contribute to the development of theoretical approaches including molecular simulations. The latter are helpful to understand the diffusion process from a molecular perspective. In a currently ongoing research project, the aim is to accurately determine the Fick diffusivity of binary fluid mixtures based on methane, propane, and carbon dioxide over a wide density range by optical and theoretical methods.
With holographic interferometry applied for a Loschmidt cell (HILC), the instationary diffusion process in binary mixtures subjected to a macroscopic concentration gradient is investigated. From the analysis of the temporal change in the partial molar density in both half cells applying two interferometers, the concentration dependency of the Fick diffusivity D_{11} was accessed in the superheated vapor and gaseous state at pressures between 0.1 and 0.5 MPa. In dynamic light scattering (DLS), the dynamics of microscopic fluctuations in the hydrodynamic fluid regime is analyzed at macroscopic thermodynamic equilibrium via the study of the temporal behavior of the scattered light intensity. By evaluating the relaxation behavior of microscopic fluctuations in the composition, absolute D_{11} data were determined as a function of concentration in the supercritical and compressed liquid state at pressures between 9 and 12 MPa. The measurement results obtained by HILC and DLS with expanded uncertainties of about 5% served as reference for equilibrium molecular dynamics (MD) simulations calculating thermophysical properties from the statistical motion of molecules. By separating the kinetic and thermodynamic contributions to the Fickian diffusion process, D_{11} values were calculated by MD simulations over the entire concentration range at pressures between 0.1 and 12 MPa with statistical expanded uncertainties of about 10%. For the investigated thermodynamic states at temperatures between 293 and 353 K, agreement between the experimental and simulated Fick diffusivities of the studied binary fluid mixtures within their expanded uncertainties was found.