Determination of Mutual Diffusivities by the Shadowgraph Method

Patrick S. Schmidt[1, S], Wenchang Wu[1], Xiaohan Huang[1], Michael H. Rausch[1] and Andreas P. Fröba[1, C]

[1]*Institute of Advanced Optical Technologies – Thermophysical Properties (AOT-TP), Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Erlangen, Germany*
andreas.p.froeba@fau.de

In chemical and energy engineering, the design and optimization of processes and apparatus is often based on an exact description and modeling of heat, mass, and momentum transfer. For this, accurate information on transport properties of the involved working fluids at process-relevant temperatures and pressures is required. In this context, the shadowgraph method is a promising technique which allows, in principle, a simultaneous determination of the mutual diffusivity D_{11}, the thermal diffusivity a, the kinematic viscosity ν, and the thermodiffusion coefficient D_T. It relies on experiments where a horizontal layer of a fluid mixture is subjected to a stationary macroscopic temperature gradient that induces a concentration gradient due to thermodiffusion. Both gradients give rise to non-equilibrium fluctuations whose dynamics are associated with the transport properties of interest. In shadowgraph experiments, challenges arise, for instance, in the form of advection occurring in mixtures with negative Soret coefficients $S_T = D_T/D_{11}$ or insufficient signal intensity resulting from too small concentration gradients or from too small refractive index differences Δn of the mixture components.

In this contribution, approaches to overcome these challenges are presented, aiming at the accurate determination of D_{11} in arbitrary fluid mixtures using the shadowgraph method. Based on the results from systematic investigations, it is demonstrated that advection can be sufficiently suppressed by a proper adjustment of the fluid layer thickness and the direction and magnitude of the applied gradients. Furthermore, the influence of signal intensities on the measurement uncertainty is shown by studying mixtures with varying magnitudes of the applied concentration gradients and Δn. By applying the approaches to investigate D_{11} for systematically varied binary hydrocarbon mixtures consisting of, e.g., alkanes and/or aromatics including liquid organic hydrogen carriers over a wide range of temperatures from (298 to 423) K and pressures from (0.1 to 30) MPa, structure-property relationships are derived.