Applicability of the Shadowgraph Method for the Determination of Diffusion Coefficients

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Accurate knowledge on heat, mass, and momentum transfer is important for the description of industrial and natural processes involving fluid mixtures. Thus, their transport properties, e.g., thermal diffusivity a, Fick diffusion coefficient D_{11}, and kinematic viscosity ν, have to be known at process-relevant conditions. Here, we present a newly developed shadowgraph apparatus for the determination of multiple transport properties. The realization of thermodiffusion experiments in such an apparatus is based on the application of a macroscopic temperature gradient resulting not only in a concentration gradient, but also in intense non-equilibrium fluctuations (NEFs). Retrieving the dynamics of NEFs in temperature, concentration, and fluid velocity gives access to the above-mentioned properties. Besides proving the applicability of the new apparatus for the determination of multiple transport properties over a wide range of thermodynamic states, the contribution also demonstrates that the method allows the accurate measurement of D_{11} for a large variety of binary fluid mixtures.

At first, a reference mixture consisting of 1,2,3,4-tetrahydronaphthalene and *n*-dodecane was investigated up to T = 373 K and p = 40 MPa for validating and probing the limits of the apparatus. For this mixture, D_{11} and a determined with expanded uncertainties of (2.8 and 6.6)% agree with own dynamic light scattering results and literature data. In addition, with help of density data, ν and the Soret coefficient could be determined. Further investigations of binary mixtures consisting of 1-hexanol or *n*-hexane with carbon dioxide as well as of methane and propane demonstrate that D_{11} and a can be simultaneously accessed down to $Le = a/D_{11} \approx 7$ or even for mixtures with minor advection in the case of a negative Soret coefficient. In summary, all the investigations demonstrate the performance of the newly developed apparatus and the applicability of the technique for the routine determination of diffusion coefficients.