

Investigation of Mutual Diffusivities in Ionic Liquid Solvent Mixtures by Dynamic Light Scattering (DLS)

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The binary diffusion coefficient is an important transport property for the design of many processes where ionic liquid (IL) solvent mixtures are involved, e.g., separations or catalytic reactions. Nevertheless, hardly any mutual diffusivity data are available for these systems because most conventional measuring methods are restricted to limited concentration ranges or by extensive measurement times. With binary mixtures consisting of the ILs [EMIM][EtSO₄] (1-ethyl-3-methylimidazolium ethylsulfate) or [EMIM][MeSO₃] (1-ethyl-3-methylimidazolium methanesulfonate) and six molecular solvents, however, we recently proved that dynamic light scattering (DLS) is a suitable technique for measuring accurate mutual diffusivities for such systems over the entire concentration range. Following up this work, the influence of different cations, anions, and solvents on the diffusion process is investigated in the present study. DLS was used for the measurement of the mutual diffusivities of two further ILs based on the [EMIM]-cation, [EMIM][N(CN)₂] (1-ethyl-3-methylimidazolium dicyanide) and [EMIM][NTf₂] (1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide), and another IL based on the [NTf₂]-anion, [HMIM][NTf₂] (1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide), in combination with ethanol and acetone. For an optimization of the DLS method, the influence of the measuring time and laser power on the uncertainty of the diffusivity data was studied. Measurements for solvent mole fractions ranging from 0.1 to 0.95 were performed at atmospheric pressure for temperatures between 283.15 K and 323.15 K. The results show that, e.g., the mutual diffusivities for the ethanol mixtures are smaller than those for the mixtures with acetone, which may be related to the formation of hydrogen bonds between ethanol and the ions. Most of the investigated mixtures exhibit increasing binary diffusion coefficients with increasing solvent concentration. In contrast, a minimum of the mutual diffusivities was found for the [EMIM][NTf₂] ethanol mixtures at ethanol mole fractions between 0.7 and 0.8, which may be caused by the experimental conditions being close to a critical demixing point.