Measurement and Prediction of Multicomponent Diffusion Coefficients in Four Ternary Liquid Systems

Sebastian Rehfeldt [C, S] and Johann Stichlmair
Institute for Plant and Process Engineering, Technische Universität München, Garching, Bavaria, Germany

The physical phenomenon of diffusion is omnipresent in every natural and industrial process involving mass transfer. In many cases diffusion plays an essential role as the rate limiting mechanism. Therefore, the determination of diffusion coefficients in liquids is of paramount interest for the simulation and calculation of mass transfer processes. Most studies on diffusion coefficients focus on binary mixtures. However, real processes mostly deal with mixtures involving more than two components. In this case, both the experimental and the theoretical investigations of diffusion are much more complex. Prediction of multicomponent diffusion coefficients is still at its very beginning due to lack of experimental ternary diffusion data. To overcome this limitation, multicomponent diffusion coefficients were measured by holographic interferometry within the whole concentration space of four non-ideal ternary systems. Experimental Fickian diffusivities are transformed into Maxwell-Stefan diffusivities using the thermodynamic factor. This correction factor is computed from available excess energy models such as Wilson, Uniquac, or NRTL. Existing prediction models for Maxwell-Stefan diffusion coefficients as well as a novel one are tested against more than 100 experimental ternary data points. For the first time, different prediction models are evaluated against experimental data over the whole concentration range of several ternary systems. Our results show that the quality of prediction significantly depends on the quality of the thermodynamic description of the system. While deviations of predicted diffusion coefficients in weakly non-ideal systems are rather small, predictions in strongly non-ideal systems exhibit a large variance. Therefore, prediction models for multicomponent diffusion coefficients should only be tested in systems with accurate thermodynamic description. Apparently, reliable prediction in strongly non-ideal systems is dependent on improvements in modeling the thermodynamic factor.