# Vapor-Liquid Interfacial Properties of Mixtures: Experiment, Theory, and Molecular Simulation

Florian Fleckenstein[1, S], Stefan Becker[2], Hans Hasse[2] and Simon Stephan[2, C]

[1]*Laboratory of Engineering Thermodynamics, RPTU Kaiserslautern, Kaiserslautern, Germany*
[2]*RPTU Kaiserslautern, Kaiserslautern, Germany*
*simon.stephan@rptu.de*

Vapor-liquid interfacial properties of mixtures are of significant importance in various engineering applications as well as in nature. While interfaces are considered as two-dimensional objects on a macroscopic level, there is an interfacial region on the atomistic level, where properties continuously change between the two bulk phase values. While there is plenty of data on the surface tension of pure substances, significantly less data are available for mixtures. For nanoscopic interfacial properties describing the structure of the interface, no information is available today for most systems. The structure of the interface can, however, affect transport processes across the interface. Quantitative information on the density profiles of molecular fluids in the interfacial region cannot be obtained by experiments today. However, both molecular dynamics (MD) simulations based on classical force fields and density gradient theory (DGT) or density functional theory (DFT) combined with an equation of state (EOS) can provide such data.

In this work, we studied vapor-liquid interfacial properties by pendant drop experiments, MD, and DGT+EOS. The PCP-SAFT EOS was used in combination with DGT. Data on the surface tension and the relative adsorption were obtained from pendant drop experiments as well as from MD simulations and DGT + PCP-SAFT. The interfacial thickness and enrichment of low-boiling components were investigated solely through MD simulations and DGT + PCP-SAFT.

Various binary systems were studied – parts of the results were published in Refs. [1-3]. We have studied binary mixtures of different high-boiling components (cyclohexane, toluene, ethanol, isopropyl alcohol, and acetone) with different low-boiling components (carbon dioxide and nitrogen). Additionally, selected binary systems alcohol + water were studied. In all cases, multiple temperatures and pressures were studied by all three methods. For all studied systems, the results from the independent methods are overall in good agreement. Additionally, the comparison of the results from the different mixtures provide insight into the link between the phase behavior, interfacial properties, and molecular interactions. Based on that, a simple heuristic model was developed for describing the enrichment of components at vapor-liquid interfaces based on bulk phase equilibrium properties alone [4].

## References

1. Becker et al., Fluid Phase Equilib. 427 (2016) 476.
2. Stephan et al., Fluid Phase Equilib. 518 (2020) 112583.
3. Stephan et al., J. Chem. Eng. Data (2023) in press.
4. Stephan & Hasse, Int. Rev. Phys. Chem. 39, 3 (2020) 319.