Modeling Properties of Ionic Liquid Systems based on ePC-SAFT

Zhida Zuo[1], Xiaoyan Ji[1]

[1]Division of Energy Science/Energy Engineering, Luleå University of Technology, Luleå, Sweden, xiaoyan.ji@ltu.se.

Ionic liquids (ILs) have attracted considerable attention as promising alternatives to conventional solvents because of their negligible volatility, high thermal stability, and tunable physicochemical properties.[1] For the design and scale-up of IL-based processes, transport properties such as self-diffusion coefficients (SDCs) and viscosities are of particular importance because they govern fluid flow as well as heat and mass transfer. However, the vast number of possible ILs and the limited availability of experimental data necessitate the development of reliable predictive models.

Although SDCs and viscosities both characterize molecular mobility, they are typically modeled independently. Establishing a consistent theoretical framework that links these properties would significantly improve predictive capabilities. In our previous studies,[2–4] we employed the Einstein relation to connect diffusion and viscosity through molecular parameters within the electrolyte Perturbed-Chain Statistical Associating Fluid Theory (ePC-SAFT) framework.

A predictive SDC model for chain-like molecules was first developed by introducing a correction function to an existing model for spherical Lennard–Jones fluids.[5] Combined with the Stokes–Einstein equation, this approach enables the prediction of viscosities from diffusion data using a universal parameter set. The framework was successfully validated for various alkane families and subsequently extended to ionic liquids.

In parallel, an ion-based ePC-SAFT free volume theory (ePC-SAFT-FVT) model was developed for IL viscosities. Compared with conventional molecule-based approaches, the ion-based approach substantially reduces the number of adjustable parameters while improving predictive flexibility. By coupling the viscosity model with the Einstein relation, SDCs of ionic liquids can be predicted directly from viscosity-derived parameters. The resulting framework accurately describes overall as well as individual cationic and anionic diffusion coefficients using a universal parameter set.

To further broaden the applicability of the model, a Born-term contribution was incorporated into ePC-SAFT to describe (IL + co-solvent) systems. The resulting framework provides a unified description of densities, osmotic coefficients, vapor-liquid equilibria, water activities, and CO_2 solubilities of both pure ILs and (IL + co-solvent) mixtures.[6]

Overall, this work establishes a unified ePC-SAFT-based framework that bridges transport and thermodynamic properties of ionic liquids. By linking viscosity and diffusion through the Einstein relation and extending the model to IL-containing mixtures, the approach offers a predictive tool for the design and optimization of IL-based processes. Figure 1 illustrates the procedure of modeling properties of IL-based systems.

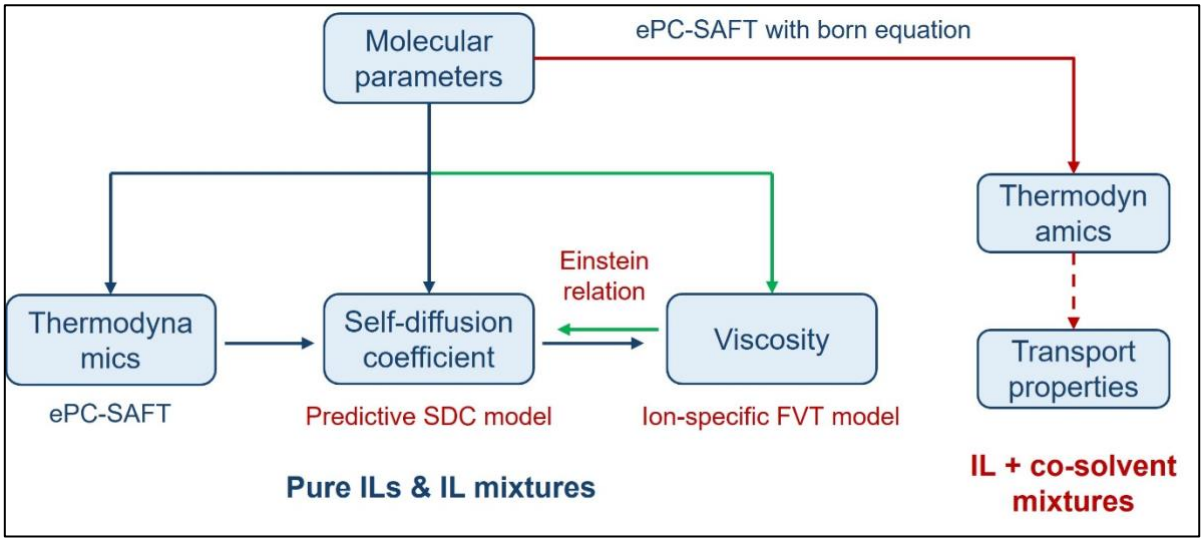

Figure 1 – Procedure for modeling the properties of IL systems based on ePC-SAFT

REFERENCES

[1] Ma, C.; Laaksonen, A.; Liu, C.; Lu, X.; Ji, X., The peculiar effect of water on ionic liquids and deep eutectic solvents. *Chem. Soc. Rev.* 2018, 47, (23), 8685-8720.

[2] Zuo, Z.; Lu, X.; Ji, X., Modeling Self-Diffusion Coefficient and Viscosity of Chain-like Fluids Based on ePC-SAFT. *J. Chem. Eng. Data* 2024, 69, (2), 348-362.

[3] Zuo, Z.; Lu, X.; Ji, X., Modeling self-diffusion coefficients of ionic liquids using ePC-SAFT and FVT combined with Einstein relation. *AIChE J.* 2024; 70(8):e18468.

[4] Zuo, Z.; Lu, X.; Ji, X., Modeling the Viscosity of Ionic Liquids and Their Mixtures Using ePC-SAFT and Free Volume Theory with an Ion-Based Approach. *Ind. Eng. Chem. Res.* 2025, 64, (4), 2446-2464.

[5] Zhu, Y.; Lu, X.; Zhou, J.; Wang, Y.; Shi, J. Prediction of diffusion coefficients for gas, liquid, and supercritical fluid: application to pure real fluids and infinite dilute binary solutions based on the simulation of Lennard-Jones fluid. *Fluid Phase Equilib.* 2002, 194–197, 1141–1159

[6] Zuo, Z.; Figiel P.; Held C.; Liang X.; Kontogeorgis G.; Ji X.; Thermodynamic Modeling of Aqueous Ionic Liquids Using ePC-SAFT with a Born Solvation Term., in preparation, 2026