Deliverable report

Deliverable D1.1: Validated methods for simulating solvent selection (M08)

WP1- Computational modeling and liquid sorbent preparation

Project Information	
Grant Agreement No.	**101192964**
Project Dates	**January 1st 2025 – December 31st 2027**

Funded by the European Union

Project funded by

Schweizerische Eidgenossenschaft
Confédération suisse
Confederazione Svizzera
Confederaziun svizra

Swiss Confederation

Federal Department of Economic Affairs, Education and Research EAER
State Secretariat for Education, Research and Innovation SERI

Document Information

Title of deliverable	D1.1: Validated methods for simulating solvent selection
Work Package	WP1: Computational modeling and liquid sorbent preparation
Responsible Partners	UA (WP Leader), EPFL, and NIC)
Authors	Murilo L. Alcantara, João Fabian, Filipe Sosa, Jan Gerald Rittig, Hanzhang Gong, Zhaofu Fei, Philippe Schwaller, Anže Prašnikar, João Coutinho
Submission date	29/08/25
Due date	31/08/2025

Dissemination Level

PU	Public	X
SEN	Sensitive	

Grant Agreement: **101192964** | 2025– 2027 | Duration: 36 months
Type of Action: Research and Innovation Action (RIA). Topic: Horizon Europe Research and Innovation Actions programme

Document History

Version	Date	Modification	Authors
V1.1	01/08/25	Draft Original	Murilo L. Alcantara, João Fabian, Filipe Sosa, João Coutinho
V1.2	20/08/25	Review	Jan Gerald Rittig and Philippe Schwaller
V1.3	22/08/25	Review	Anže Prašnikar
V1.4	25/08/25	Review	Murilo Alcantara, João Coutinho
VF	29/08/25	Review and submission	Anže Prašnikar

Legend:

V0 - Table of Contents (successive V0.N versions may be used)

V1 – Version submitted to reviewers (successive V1.N versions may be used)

V2 - Version approved after review (successive V2.N versions may be used)

VF – Version submitted to EC

Word name:

ILIMITED_D1.1_Version

Nature of Deliverable

Nature of Deliverable	Description	Select the type (X)
R	Document, report, deliverable (excluding the periodic and final reports)	X
DEC	Websites, patents filing, press & media actions, videos, etc.	
DEM	Demonstrator, pilot, prototype, plan designs	
OTHER	Software, technical diagram, algorithms, models, etc.	
ETHICS	Deliverables related to ethics issues.	
DATA	Data sets, microdata, etc.	
DMP	Data Management Plan	

Acronym/Abbreviations

Acronym/Abbreviations	
CA	Consortium Agreement (contractual document between members of the consortium)
DoA	Description of Action (technical annex to the Grant Agreement)
EC	European Commission
EU	European Union
GA	Grant Agreement (contractual document between EC and beneficiaries)
WP	Work Package
UA	University of Aveiro
NIC	National Institute of Chemistry of Slovenia.
EPFL	Swiss Federal Institutes of Technology
MeOH	Methanol
T_d	Short term thermal decomposition temperature
T_m	Melting temperature (melting point)
μ	Viscosity
γ^{∞}_{MeOH}	Infinite dilution activity coefficient of methanol
γ^{∞}_{water}	Infinite dilution activity coefficient of water
IL	Ionic Liquid
GNN	Graph neural network

Table of contents

List of Figures

1 Executive summary

1.1 Description of the deliverable content and purpose

This deliverable presents the validation of three computational models, ILBERT (ai4solvent), COnductor like Screening MOdel for Realistic Solvents (COSMO-RS), and GNN-IL (graph neural network for ionic liquids) used for screening solvent candidates within the ILIMITED project (Work Package 1). The overarching goal is to identify solvents capable of selectively absorbing methanol and water under the demanding conditions of the CO_2 hydrogenation reaction, particularly at elevated temperatures. To this end, five key physicochemical properties were selected as predictive metrics to evaluate solvent suitability. These include thermal decomposition temperature (T_d), melting temperature (T_m), viscosity (μ), and the infinite dilution activity coefficients of methanol (γ^{∞}_{MeOH}) and water (γ^{∞}_{Water}). ILBERT was used to predict thermal and transport properties, while COSMO-RS and GNN-IL were employed to estimate solvation behavior. Given the unconventionally high temperatures required for the reaction path designed on the ILIMITED project, the reliability of the models under project-specific conditions remains uncertain. This report aims to validate methods for simulating solvent selection by comparing model predictions with available experimental data for selected ionic liquids (ILs). The analysis provides a basis for assessing the applicability of ILBERT, COSMO-RS, and GNN-IL supporting the subsequent stages of solvent screening and experimental validation.

2 Description of the deliverable objective and the content

2.1 Problem description

The ILIMITED project aims to develop a breakthrough process for the hydrogenation of CO_2 to methanol with significantly enhanced single-pass conversion efficiency. While high temperatures (i.e. 513 K) favor the reaction kinetics, they still yield limited conversion rates (<20%) under conventional conditions (Kiss et al., 2016). A promising strategy to overcome this thermodynamic limitation involves the in situ removal of reaction products (methanol and water) through selective liquid sorbents, thereby shifting the equilibrium toward greater methanol formation and potentially achieving conversions above 80%.

To meet the requirements of this strategy, liquid sorbents must possess both high thermal and chemical stability at elevated temperatures and strong selectivity for methanol and water. However, only a few solvents exhibit these properties under the demanding reaction conditions. Among the most promising candidates are ionic liquids (ILs), due to their low vapor pressure, tunable molecular structure, and well-known thermal resilience. The combination of a broad variety of cations and anions leads to a theoretically possible number of 10^{18} Ionic Liquids (Merck, 2025). Within this vast chemical space of ILs, at least 8 mi are considered to be synthesizable (Qiu et al., 2025; Venkatraman et al., n.d.). The experimental screening of ILs for most application can be exhaustive, expensive, and unfeasible if performed via experimental testing. Therefore, reliable computational screening tools are essential for narrowing down viable candidates.

To identify suitable ILs, five key properties must be considered:

- **Thermal decomposition temperature (T_d)** – to ensure short-term stability at >513 K.
- **Melting point (T_m)** – to guarantee a liquid phase at room temperature or startup conditions.

- **Viscosity (μ)** – to ensure process operability, particularly during startup.
- **Infinite dilution activity coefficients** of methanol (γ^{∞}_{MeOH}) and water ($\gamma^{\infty}_{H_2O}$) – to assess solubility and selectivity under operational conditions.

These properties are predicted using two advanced computational models:

- **ILBERT**, a transformer-based deep learning model trained on ~8.5 million IL structures, predicts Td, Tm, and μ.
- **COSMO-RS**, a quantum chemistry-based thermodynamic model, estimates γ^{∞}_{MeOH} and $\gamma^{\infty}_{H_2O}$.
- **GNN-IL**, a machine learning model trained on experimental data, predicting γ^{∞}_{MeOH} and $\gamma^{\infty}_{H_2O}$.

2.2 Objective

The objective of this deliverable is to assess the reliability of two computational models ILBERT, COSMO-RS, and GNN-IL in predicting key physicochemical and solubility data required for the screening of solvent candidates within the ILIMITED project. Specifically, ILBERT is evaluated for its ability to estimate the thermal decomposition temperature (T_d), melting temperature (T_m), and viscosity (μ) of ionic liquids, while COSMO-RS and GNN-IL are assessed and compared for their accuracy in predicting the infinite dilution activity coefficients of methanol (γ^{∞}_{MeOH}) and water ($\gamma^{\infty}_{H_2O}$). The validation is performed through comparison with experimental data, aiming to determine the suitability of these models for guiding the selection of thermally stable, selective sorbents capable of operating under the high-temperature conditions targeted in ILIMITED.

2.3 Methods

2.3.1 ILBERT model

To assist in the selection of thermally stable and low-viscosity ILs suitable for methanol and water sorption at elevated temperatures (up to 513 K), we employed the ILBERT (Ionic Liquid Bidirectional Encoder Representations from Transformers) model (Qiu et al., 2025) via the ai4Solvent.com web platform (Cchen et al., 2025). It is worth mentioning that ILBERT was not developed on this study, but it was applied for being the state-of-the-art chemical language model specifically designed for predicting multiple physicochemical and thermodynamic properties of ionic liquids.

The ILBERT model was pre-trained on a dataset comprising over 8.3 million IL-like molecular structures and subsequently fine-tuned on curated datasets for twelve key IL properties, including melting point, thermal decomposition temperature, and viscosity. The model utilizes transformer-based deep learning architecture, optimized through SMILES-based chemical representations and augmented with advanced tokenization schemes to enhance property-specific predictions (Qiu et al., 2025).

The ILs' SMILES were inserted within the ai4Solvent interface and the model provided rapid estimations of the short-term thermal decomposition temperature, normal melting point, and viscosity at room temperature. These data were compared with available experimental data to evaluate their suitability for use in IL

screening. The current models do not provide estimations for chemical stability (e.g., reactivity) or long-term thermal stability. These aspects are expected to be addressed in future stages of the ILIMITED project. This is particularly challenging since experimental data is limited for these properties, in the order of less than a hundred data points. We are currently exploring multi-task machine learning approaches to enable predictions that can be used as first indicators even in such low-data regimes.

2.3.2 COSMO-RS

The COSMO-RS (Conductor-like Screening Model for Real Solvents) model (Diedenhofen & Klamt, 2010; Eckert & Klamt, 2002; Klamt, 1995) was employed in this study to predict infinite dilution activity coefficients of methanol (γ^{∞}_{MeOH}) and water ($\gamma^{\infty}_{H_2O}$). This model combines quantum chemical calculations with statistical thermodynamics to estimate thermodynamic properties of liquid mixtures, treating molecular interactions through the concept of surface charge densities (σ-profiles). This information represents the polarity distribution over ions' surface. COSMO-RS uses these profiles to compute interaction energies between solute and solvent surfaces. By statistically averaging these interactions, the model estimates the chemical potential of the solute, from which the activity coefficient γ^{∞} is derived. Lower γ^{∞} values correspond to higher solubility, enabling efficient screening of selective solvents.

All geometry optimizations and charge density calculations were performed with the software package TURBOMOLE (TURBOMOLE, 2019), through its interface TMoleX v.4.5 (Steffen et al., 2010), using density functional theory (DFT) with the BP86 functional and the triple zeta valence polarization (def-TZVP) basis set (Steffen et al., 2010). The configuration of those quantum chemistry calculations corresponds to that of the "BP-TZVP_25" parametrization of COSMO-RS, available in the software package COSMOTherm v.25 (Dassault Systèmes., 2021). This parametrization presents other advantages over classical COSMO-RS level DMOL^3, such as improvements in the hydrogen bonding and van der Waals dispersion term and a correction for residual dielectric charge (RDC) (Grimme et al., 2010; Paduszyński, 2018).

The charge density distribution for ions was calculated separately for each ionic species. This means that sigma profiles for neutral and ionic species were determined individually. This approach reduces the number of possible conformers and generally yields high accuracy, especially for dissociated species (Klamt et al., 2010).

The COSMO-RS model relies on a set of universal parameters derived from the regression of large datasets, primarily based on room-temperature properties of molecular systems. Although the model includes temperature-dependent terms, its reliability is inherently linked to the domain of the data used for parameter fitting. Therefore, when extrapolated to high temperatures, the model may exhibit increased deviations due to the lack of high-temperature reference data in its parametrization.

For this reason, the present study critically evaluates the applicability and accuracy of COSMO-RS in predicting methanol and water activity coefficients at elevated temperatures. By benchmarking model predictions against experimental or reference data where available, we aim to assess the extent to which COSMO-RS can be reliably used for solvent screening in high-temperature catalytic systems.

2.3.3 GNN-IL

The GNN-IL (graph neural network for ionic liquids) model (Rittig et al., 2023) was further developed and used in this study to predict infinite dilution activity coefficients of methanol (γ^{∞}_{MeOH}) and water ($\gamma^{\infty}_{H_2O}$). The model thus serves as an alternative to COSMO-RS.

GNN-IL is a machine learning model that predicts the properties of interest directly from a graph representation of ionic liquids and solutes. Specifically, the molecules are represented as graphs with atoms a as nodes and bonds as edges. Then, the GNN extracts structural information from the molecular graph by applying graph convolutional layers. This results in a continuous vector representation of molecules, referred to as molecular fingerprints, which can then be used as input to a multilayer perceptron for property prediction. In the GNN-IL, the molecular fingerprint of the IL and of the solute (i.e., water or ethanol) are concatenated together with the value for the temperature, and then the multilayer perceptron provides infinite dilution activity coefficient predictions.

The GNN-IL model learns directly from experimental data. Here, we used the data set of infinite dilution activity coefficients at varying temperatures of solutes in ILs and deep eutectic solvents published by (Li et al., 2022), which comprises about 52,000 experimental data points for 252 ILs and 112 solvents in a temperature range from 288.15 K to 428.15 K. The GNN-IL model thus learns from a diverse set of ILs.

2.4 Validation of ILBERT

2.4.1 Short-term thermal decomposition temperature (Td)

The performance of ILBERT in predicting the short-term thermal decomposition temperature of ionic liquids was evaluated against experimental data from the literature (Cao & Mu, 2014), as shown in Figure 1. The model exhibits a strong correlation with the reference values, indicating its general reliability in estimating thermal stability.

However, a tendency to underpredict Td values is observed, as most data points lie below the identity line. This systematic deviation suggests that, in practical scenarios, the actual thermal stability of candidate ILs may be higher than the predicted values. From a screening perspective, this conservative bias may be advantageous: ILs predicted to meet a minimum T_d threshold are likely to perform even better experimentally. Therefore, while this deviation should be acknowledged, it does not disqualify ILBERT for screening purposes.

Moreover, the error bars generated by the model appear underestimated or overly narrow, as a substantial number of experimental values fall outside the reported confidence intervals. Despite this, the majority of predictions remain within ±50 K of the experimental values, which can be considered acceptable for early-stage screening. Larger deviations were primarily associated with ILs containing less common functional groups, such as amino-carboxylate moieties (Figure 1), highlighting a possible limitation in the training data coverage of the model.

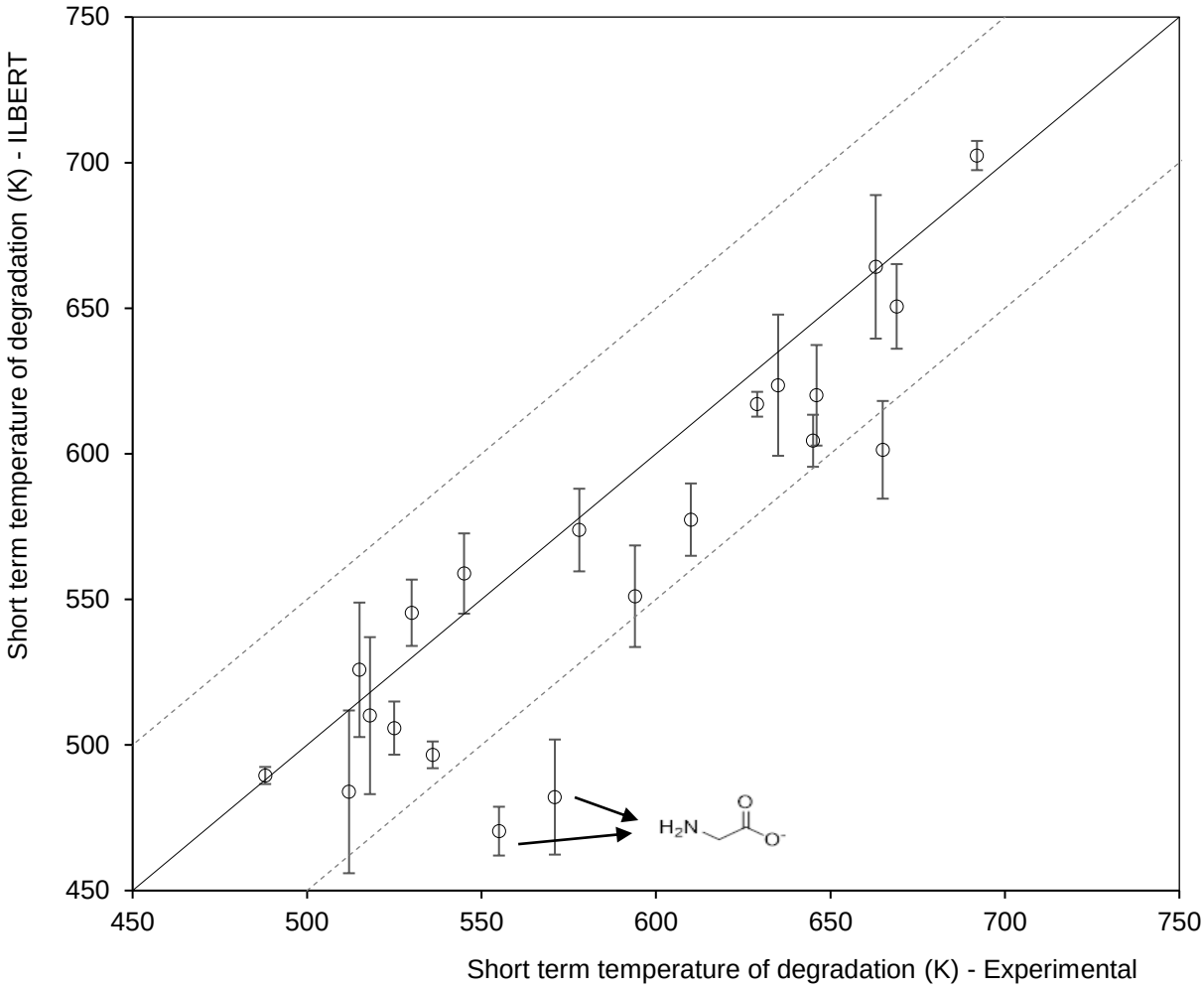

Figure 1. Validation of ILBERT AI for the prediction of ILs' short term thermal decomposition temperature.

2.4.2 Validation of melting temperature (Tm)

Figure 2 presents the comparison between ILBERT-predicted melting temperatures and experimental data compiled from a range of sources (Abranches et al., 2019; Ahmed et al., 2018; Bouarab et al., 2020; Calvar et al., 2013; Domańska et al., 2012; Domańska, Okuniewska, et al., 2015; Domańska, Roguszewska, et al., 2015; Fadeeva et al., 2020; Fernandez et al., 2017; Huang et al., 2018; Khan et al., 2017; Królikowska et al., 2019; Kurzin et al., 2016; Lazzús, 2012; Nakamoto & Watanabe, 2007; Neves et al., 2013; Parajó et al., 2018; Parsana et al., 2023; Simulations, n.d.). Unlike the T_d predictions, ILBERT does not exhibit a systematic bias for melting temperature. Again, predicted values typically fall within a ±50 K range from experimental measurements, which is acceptable for high-throughput screening purposes. Nonetheless, the narrow error margins may not fully reflect the actual prediction uncertainty, particularly for ionic liquids with amino carboxylate structures.

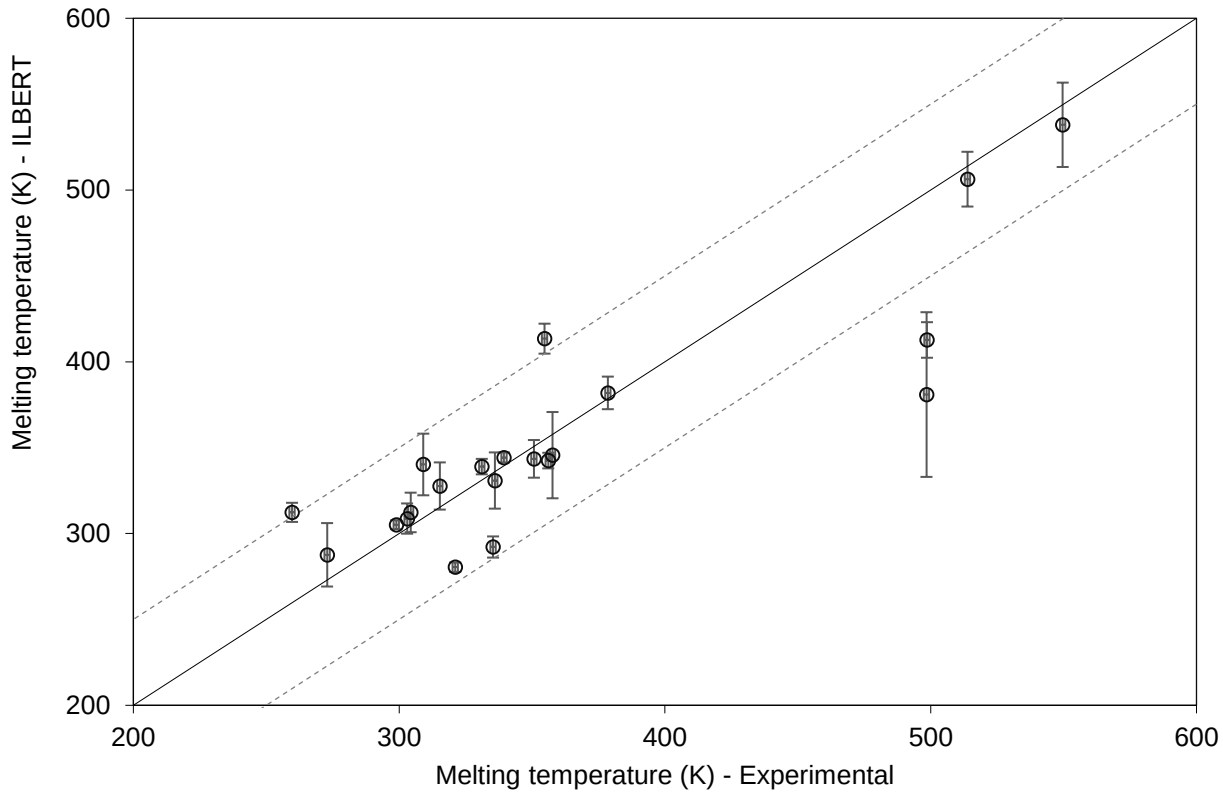

Figure 2: Validation of ILBERT AI for the prediction of ILs' melting point.

2.4.3 Validation of viscosity (μ)

The model's ability to estimate room-temperature viscosities of ionic liquids is shown in Figure 3. ILBERT adequately reproduces the overall trend in the experimental data, confirming its utility for screening ILs with desirable viscosity ranges.

While the agreement is reasonable for ILs with moderate viscosities, the model tends to show higher deviations in the high-viscosity regime. This is expected, as viscosity is highly sensitive to subtle structural features, and the range of reported viscosities spans several orders of magnitude. Despite this, the model's predictions remain qualitatively useful, enabling the identification of ILs that are significantly too viscous or, conversely, promising in terms of fluidity.

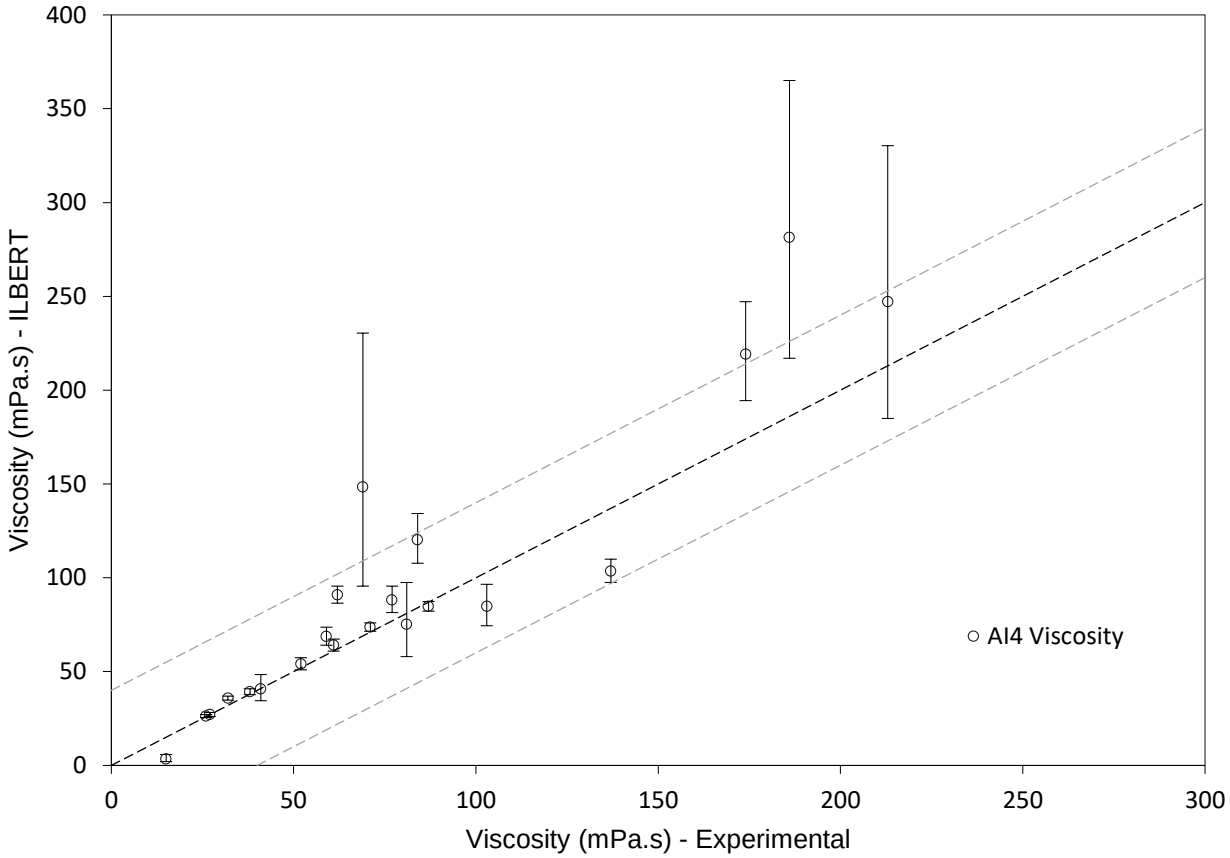

Figure 3: Validation of ILBERT AI for the prediction of ILs' viscosity.

2.5 Validation of COSMO-RS

To validate the activity coefficient predictions of COSMO-RS and GNN-IL for the application at hand, about 1,500 experimental data points were acquired from the dataset published by (Li et al., 2022); notably, most of these data points originate from the NIST ILThermo databank (Dong et al., 2007). Specifically, infinite dilution activity coefficients of methanol or water in several ILs were collected at temperatures ranging from 283 K to 423 K. Figure 4 shows that most data acquired are within 313 K and 373 K range. Less than 4.5% of experimental data were found at temperatures above 383 K, indicating that it is difficult to validate the model at the operational temperature (513 K).

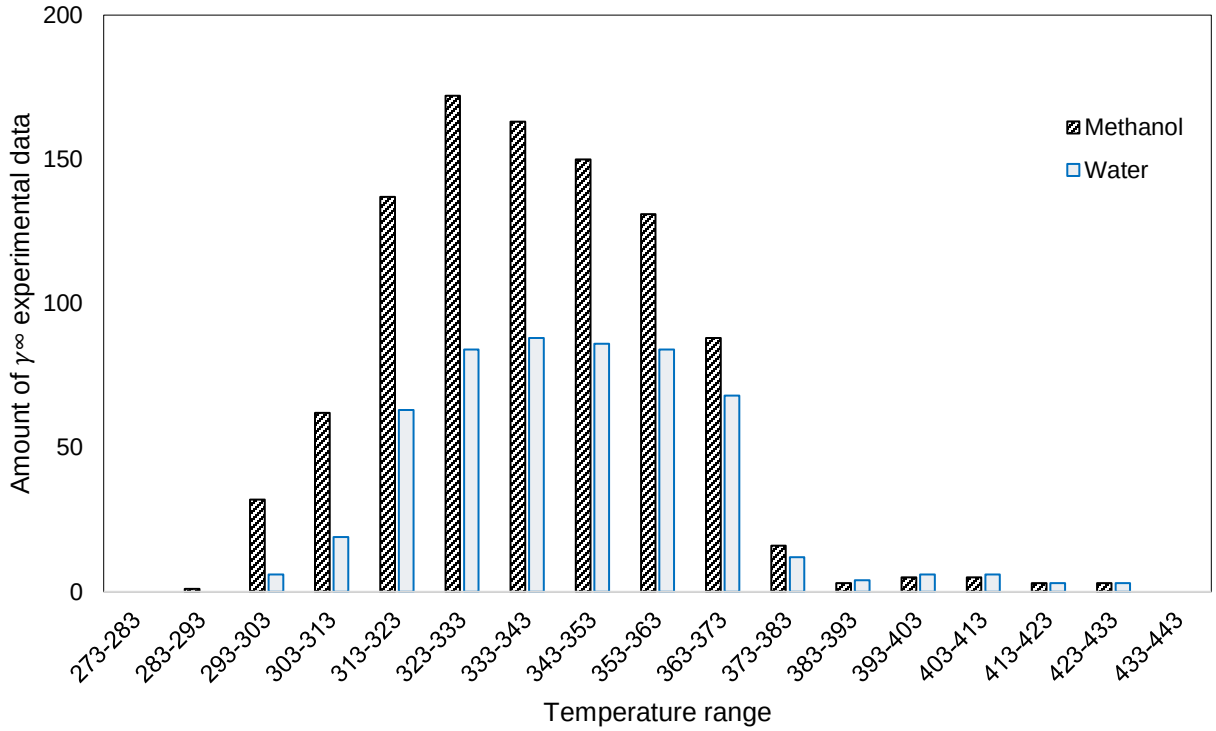

Figure 4. Activity coefficient data dispersion throughout different temperature ranges.

Figure 5 and Figure 6 present the comparison between experimental data and COSMO-RS and GNN-IL predictions for the infinite dilution activity coefficients of methanol (γ^{∞}_{MeOH}) and water ($\gamma^{\infty}_{H_2O}$) in various ionic liquids over a range of temperatures. COSMO-RS systematically overestimated γ^{∞}_{MeOH} and $\gamma^{\infty}_{H_2O}$, which resulted in an overall high quantitative deviation of roughly one order of magnitude compared to experimental data. Since the model was consistent on this overestimation, it is able to capture the relative trends of IDACS across different ILs. This behavior holds for both solutes, with water predictions showing slightly lower variance and stronger correlation. The ability of COSMO-RS to reproduce trend behavior trend across temperatures ranging from 283 K to 433 K (~150 K) suggests that the model can be reasonably extrapolated to the ILIMITED operational temperature of 513 K. Consequently, COSMO-RS should be considered at least as a **qualitative suitable** tool for the screening ionic liquids, as solvents for methanol and water, expected reaction condition of ILIMITED project.

As part of the experimental data was used for training the GNN-IL, we distinguish between training and validation (Train/Val) data and test data, which was not used during training and thus allows to assess the predictive capabilities of the model. Figure 5 and Figure 6 show that GNN-IL provides highly accurate predictions, as is also emphasized by the mean absolute errors (MAEs) and coefficients of determination (R^2) shown in the respective figures. For methanol, we find that these accuracy differences are also reflected in the Spearman rank correlation coefficients (ρ), which indicate the model capabilities to correctly rank ILs against each other and is thus highly relevant for screening purposes. Specifically, the GNN-IL exhibits a ρ value of 0.98 compared to a ρ value of 0.87 of COSMO-RS. Interestingly, for water, the GNN-IL and COSMO-RS models have similar ρ values of 0.97 versus 0.96, although the GNN is much more accurate in absolute values. Thus, for water, COSMO-RS consistently captures the relative trends across different ILs with predictions mostly within the same order of magnitude of experimental results. For methanol, in contrast, the ranking capabilities of COSMO-RS are lower, however, still in a reasonable range for screening. The GNN-IL shows high ranking capabilities for both water and methanol, indicating it can be a **quantitative suitable** tool for the screening ionic liquids.

Both models still require to be validated at the operational temperatures of the Ilimited project (513 K) of which very few data are available in the open literature. However, the ability of COSMO-RS and GNN-IL to reproduce trend behavior across temperatures ranging from 283 K to 433 K (~150 K) suggests that these models can provide highly useful predictions for screening ionic liquids for the ILIMITED project. It should be noted that extrapolation to the expected ILIMITED reactor temperature of 513 K is required, so experimental validation of the identified IL candidates – as planned in the upcoming work packages – will be crucial.

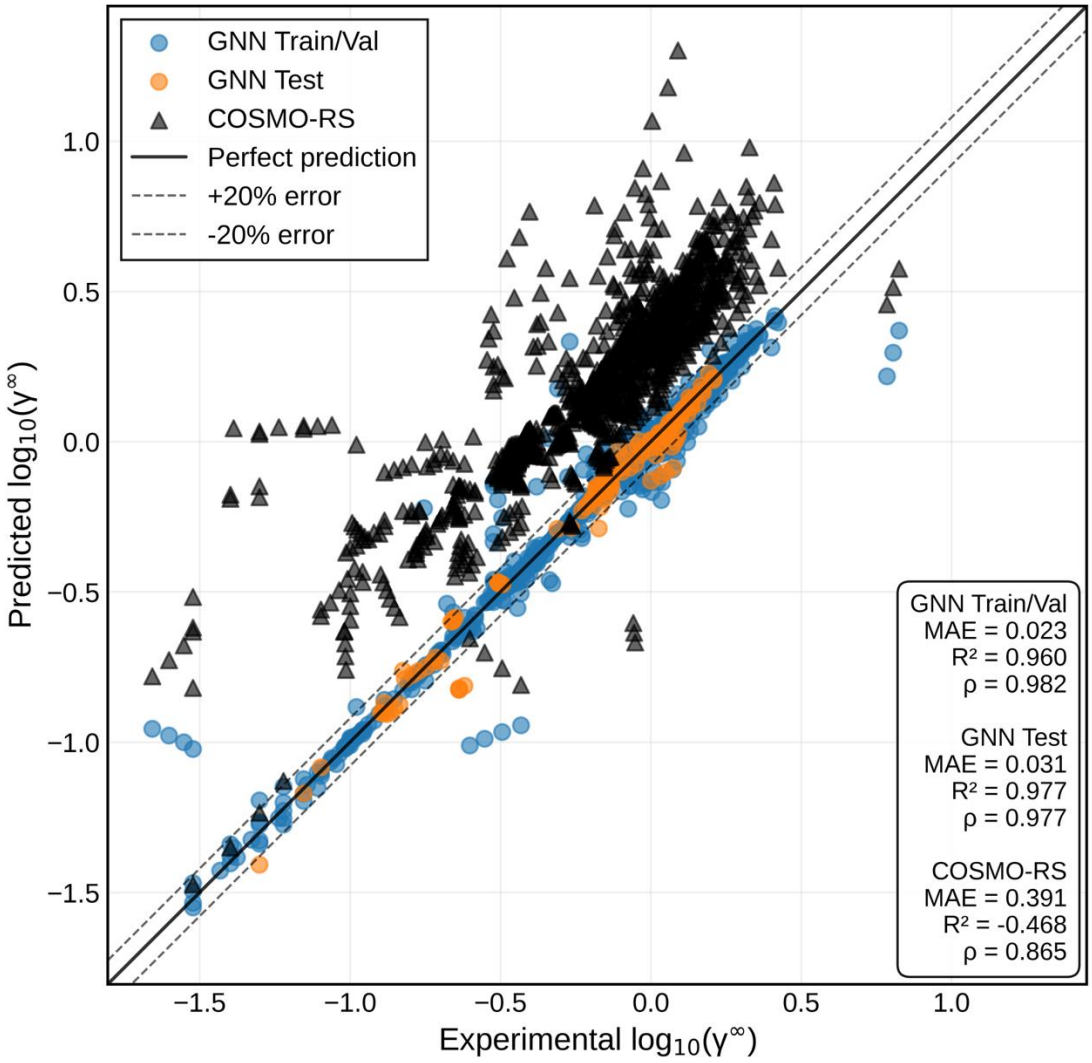

Figure 5: Validation of COSMO-RS and GNN-IL for the prediction of methanol infinite dilution activity coefficients in ILs (γ^{∞}_{MeOH}) at temperatures ranging from (283 K – 433 K)

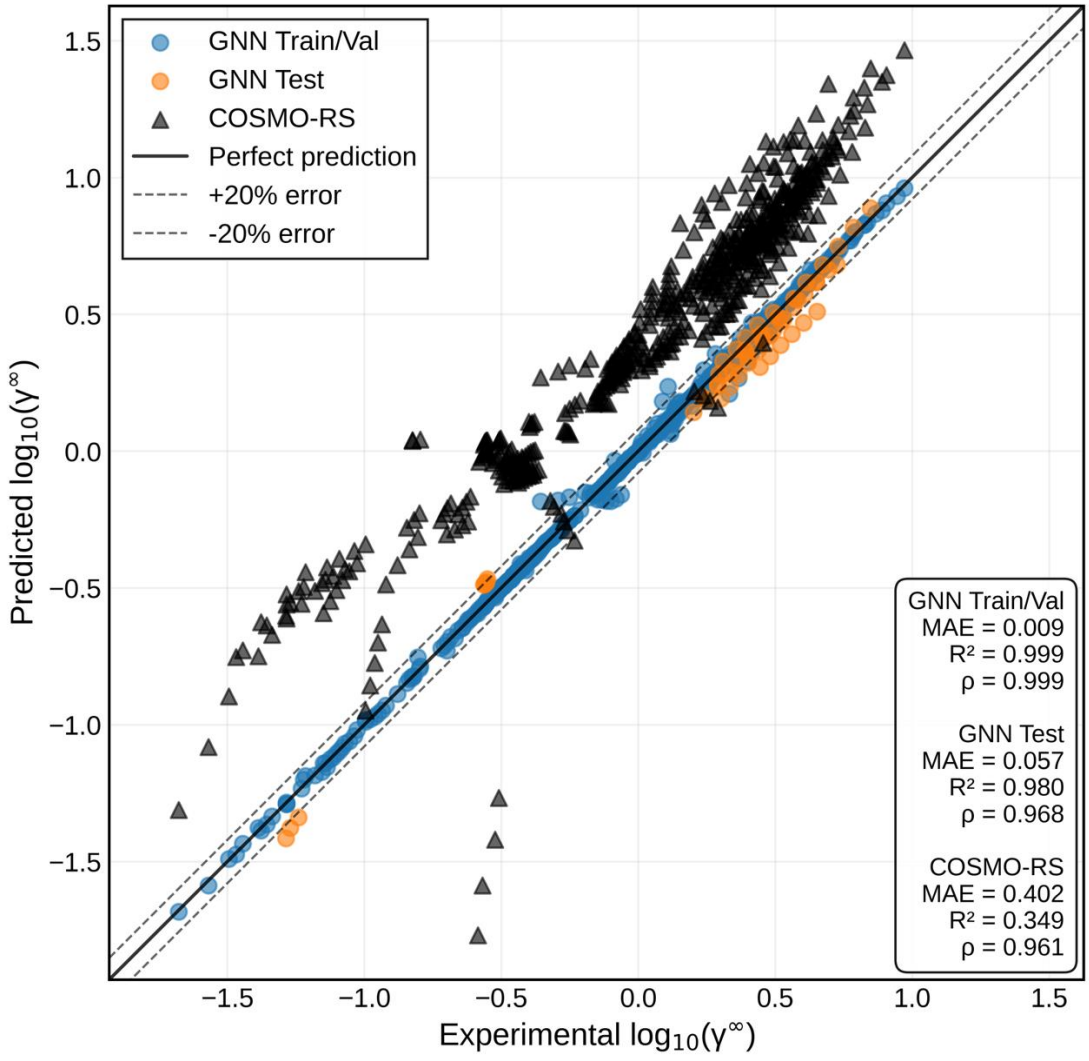

Figure 6: Validation of COSMO-RS and GNN-IL for the prediction of water infinite dilution activity coefficients in ILs ($\gamma^{\infty}_{H_2O}$) at temperatures ranging from 293 K to 433 K.

Conclusion

This deliverable 1.1 (WP1) presents the validation of ILBERT, COSMO-RS, and GNN-IL as predictive tools for screening ionic liquid under the specific requirements of the ILIMITED project. Despite the challenging operational conditions, particularly high temperatures, the models demonstrated consistent trends and reasonable agreement with experimental data across the selected properties.

ILBERT showed adequate predictive capability for thermal decomposition temperature, melting point, and viscosity, with deviations within acceptable ranges for early-stage screening. COSMO-RS, while systematically overestimating the infinite dilution activity coefficients of methanol and water, maintained strong qualitative alignment with experimental trends at a reasonable temperature range (~150 K), GNN-IL exhibits high qualitative accuracies for activity coefficient predictions and enables ranking of ILs, which is highly useful for IL screening as solvents for methanol and water at operational temperature (513 K).

Overall, the models are considered sufficiently reliable for supporting the identification and prioritization of IL candidates within WP1. The application of ILBERT, COSMO-R, and GNN-IL on IL screening should reduce experimental workload, and optimum selection of best candidates, which should be experimentally assessed on the ILIMITED, contributing meaningfully to the broader objectives of ILIMITED's work packages.

Reference

Abranches, D. O., Silva, L. P., Martins, M. A. R., Fernandez, L., Pinho, S. P., & Coutinho, J. A. P. (2019). Can cholinium chloride form eutectic solvents with organic chloride-based salts? *Fluid Phase Equilibria*, *493*, 120–126. https://doi.org/10.1016/j.fluid.2019.04.019

Ahmed, O. U., Farouq, •, Mjalli, S., Al-Wahaibi Talal, •, Al-Wahaibi, Y., & Al Nashef, I. M. (2018). Extractive Desulfurization of Liquid Fuel using Modified Pyrollidinium and Phosphonium Based Ionic Liquid Solvents. *J Solution Chem*, *47*, 468–483. https://doi.org/10.1007/s10953-018

Bouarab, A. F., Martins, M. A. R., Stolarska, O., Smiglak, M., Harvey, J. P., Coutinho, J. A. P., & Robelin, C. (2020). Physical properties and solid-liquid equilibria for hexafluorophosphate-based ionic liquid ternary mixtures and their corresponding subsystems. *Journal of Molecular Liquids*, *316*. https://doi.org/10.1016/j.molliq.2020.113742

Calvar, N., Gómez, E., Macedo, E. A., & Domínguez, Á. (2013). Thermal analysis and heat capacities of pyridinium and imidazolium ionic liquids. *Thermochimica Acta*, *565*, 178–182. https://doi.org/10.1016/j.tca.2013.05.007

Cao, Y., & Mu, T. (2014). Comprehensive Investigation on the Thermal Stability of 66 Ionic Liquids by Thermogravimetric Analysis. *Industrial & Engineering Chemistry Research*, *53*(20), 8651–8664. https://doi.org/10.1021/ie5009597

chen et al. (2025). *ai4solvent.com*.

Dassault Systèmes. (2021). *BIOVIA COSMOtherm* (21). http://www.3ds.com

Diedenhofen, M., & Klamt, A. (2010). COSMO-RS as a tool for property prediction of IL mixtures—A review. *Fluid Phase Equilibria*, *294*(1–2), 31–38. https://doi.org/10.1016/j.fluid.2010.02.002

Domańska, U., Królikowski, M., Pobudkowska, A., & Bocheńska, P. (2012). Solubility of ionic liquids in water and octan-1-ol and octan-1-ol/water, or 2-phenylethanol/water partition coefficients. *Journal of Chemical Thermodynamics*, *55*, 225–233. https://doi.org/10.1016/j.jct.2012.06.003

Domańska, U., Okuniewska, P., & Markowska, A. (2015). Phase equilibrium in binary systems of ionic liquids, or deep eutectic solvents with 2-phenylethanol (PEA), or water. *Fluid Phase Equilibria*, *424*, 68–78. https://doi.org/10.1016/j.fluid.2015.10.016

Domańska, U., Roguszewska, M., Królikowski, M., Ramjugernath, D., & Naidoo, P. (2015). Phase equilibria study of binary systems comprising an (ionic liquid + hydrocarbon). *Journal of Chemical Thermodynamics*, *83*, 90–96. https://doi.org/10.1016/j.jct.2014.12.005

Dong, Q., Muzny, C. D., Kazakov, A., Diky, V., Magee, J. W., Widegren, J. A., Chirico, R. D., Marsh, K. N., & Frenkel, M. (2007). ILThermo: A Free-Access Web Database for Thermodynamic Properties of Ionic Liquids. *Journal of Chemical & Engineering Data*, *52*(4), 1151–1159. https://doi.org/10.1021/je700171f

Eckert, F., & Klamt, A. (2002). Fast solvent screening via quantum chemistry: COSMO-RS approach. *AIChE Journal*, *48*(2), 369–385. https://doi.org/10.1002/aic.690480220

Fadeeva, Y. A., Gruzdev, M. S., Kudryakova, N. O., Shmukler, L. E., & Safonova, L. P. (2020). Physico-chemical characterization of alkyl-imidazolium protic ionic liquids. *Journal of Molecular Liquids*, *297*. https://doi.org/10.1016/j.molliq.2019.111305

Fernandez, L., Silva, L. P., Martins, M. A. R., Ferreira, O., Ortega, J., Pinho, S. P., & Coutinho, J. A. P. (2017). Indirect assessment of the fusion properties of choline chloride from solid-liquid equilibria data. *Fluid Phase Equilibria*, *448*, 9–14. https://doi.org/10.1016/j.fluid.2017.03.015

Grimme, S., Antony, J., Ehrlich, S., & Krieg, H. (2010). A consistent and accurate *ab initio* parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu. *The Journal of Chemical Physics*, *132*(15). https://doi.org/10.1063/1.3382344

Huang, Y., Ke, T., Ke, Y., Ren, Q., Yang, Q., & Xing, H. (2018). Carboxylate Ionic Liquids with Large Free Volume and Strong Hydrogen Bonding Basicity for Efficient Separation of Butadiene and n -Butene. *Industrial and Engineering Chemistry Research*, *57*(40), 13519–13527. https://doi.org/10.1021/acs.iecr.8b03315

Khan, A. S., Man, Z., Bustam, M. A., Gonfa, G., Chong, F. K., Ullah, Z., Nasrullah, A., Sarwono, A., Ahmad, P., & Muhammad, N. (2017). Effect of Structural Variations on the Thermophysical Properties of Protic Ionic Liquids: Insights from Experimental and Computational Studies. *Journal of Chemical and Engineering Data*, *62*(10), 2993–3003. https://doi.org/10.1021/acs.jced.6b00450

Kiss, A. A., Pragt, J. J., Vos, H. J., Bargeman, G., & de Groot, M. T. (2016). Novel efficient process for methanol synthesis by CO 2 hydrogenation. *Chemical Engineering Journal*, *284*, 260–269. https://doi.org/10.1016/j.cej.2015.08.101

Klamt, A. (1995). Conductor-like screening model for real solvents: A new approach to the quantitative calculation of solvation phenomena. *Journal of Physical Chemistry*. https://doi.org/10.1021/j100007a062

Klamt, A., Eckert, F., & Arlt, W. (2010). COSMO-RS: An alternative to simulation for calculating thermodynamic properties of liquid mixtures. *Annual Review of Chemical and Biomolecular Engineering*, *1*, 101–122. https://doi.org/10.1146/annurev-chembioeng-073009-100903

Królikowska, M., Zawadzki, M., & Kuna, T. (2019). Physicochemical and thermodynamic properties of the {1-alkyl-1-methylpiperidinium bromide [C1Cn=2,4PIP][Br], or 1-butylpyridinium bromide, [C4Py][Br], or tri(ethyl)butylammonium bromide [N2,2,2,4] [Br] + water} binary systems. *Thermochimica Acta*, *671*, 220–231. https://doi.org/10.1016/j.tca.2018.11.018

Kurzin, A. V., Evdokimov, A. N., Antipina, V. B., & Feofanova, M. A. (2016). Isothermal Vapor-Liquid Equilibrium Data for the Methanol + Benzene + Tetraphenylphosphonium Chloride System. *Journal of Chemical and Engineering Data*, *61*(3), 1131–1135. https://doi.org/10.1021/acs.jced.5b00770

Lazzús, J. A. (2012). A group contribution method to predict the melting point of ionic liquids. *Fluid Phase Equilibria*, *313*, 1–6. https://doi.org/10.1016/j.fluid.2011.09.018

Li, J., Anderson, J. L., & Smith, E. A. (2022). Determination of Infinite Dilution Activity Coefficients of Molecular Solutes in Ionic Liquids and Deep Eutectic Solvents by Factorization-Machine-Based Neural Networks. *ACS Sustainable Chemistry & Engineering*, *10*(42), 13927–13935. https://doi.org/10.1021/acssuschemeng.2c02600

Merck. (2025). *Introduction to Ionic Liquids*. Https://Www.Sigmaaldrich.Com/PT/En/Technical-Documents/Technical-Article/Chemistry-and-Synthesis/Reaction-Design-and-Optimization/Introduction-to-Ionic-Liquids?Srsltid=AfmBOorVKfeajWIVgIA88yDTZgSGkvrx8EArQdtx6fVEc2e3G-5IX8SX.

Nakamoto, H., & Watanabe, M. (2007). Brønsted acid-base ionic liquids for fuel cell electrolytes. *Chemical Communications*, *24*, 2539–2541. https://doi.org/10.1039/b618953a

Neves, C. M. S. S., Rodrigues, A. R., Kurnia, K. A., Esperança, J. M. S. S., Freire, M. G., & Coutinho, J. A. P. (2013). Solubility of non-aromatic hexafluorophosphate-based salts and ionic liquids in water determined by electrical conductivity. *Fluid Phase Equilibria*, *358*, 50–55. https://doi.org/10.1016/j.fluid.2013.07.061

Paduszyński, K. (2018). Extensive Evaluation of the Conductor-like Screening Model for Real Solvents Method in Predicting Liquid-Liquid Equilibria in Ternary Systems of Ionic Liquids with Molecular Compounds. *Journal of Physical Chemistry B*, *122*(14), 4016–4028. https://doi.org/10.1021/acs.jpcb.7b12115

Parajó, J. J., Villanueva, M., Sánchez, P. B., & Salgado, J. (2018). Liquid window of some biologically-active ionic liquids. *Journal of Chemical Thermodynamics*, *126*, 1–10. https://doi.org/10.1016/j.jct.2018.06.014

Parsana, V. M., Parikh, S., Ziniya, K., Dave, H., Gadhiya, P., Joshi, K., Gandhi, D., Vlugt, T. J. H., & Ramdin, M. (2023). Isobaric Vapor-Liquid Equilibrium Data for Tetrahydrofuran + Acetic Acid and Tetrahydrofuran + Trichloroethylene Mixtures. *Journal of Chemical and Engineering Data*, *68*(2), 349–357. https://doi.org/10.1021/acs.jced.2c00593

Qiu, Y., Song, Z., Chen, G., Chen, W., Chen, L., Zhu, K., Qi, Z., Duan, X., & Chen, D. (2025). Large chemical language models for property prediction and high-throughput screening of ionic liquids. *Digital Discovery*, *4*(6), 1505–1517. https://doi.org/10.1039/D5DD00035A

Simulations, M. (n.d.). *Mahinder Ramdin CO 2 Capture with Ionic Liquids*.

Steffen, C., Thomas, K., Huniar, U., Hellweg, A., Rubner, O., & Schroer, A. (2010). TmoleX—A graphical user interface for TURBOMOLE. *Journal of Computational Chemistry*, *31*(16), 2967–2970. https://doi.org/10.1002/jcc.21576

TURBOMOLE. (2019). *TURBOMOLE V7.4 2019, a development of University of Karlsruhe and Forschungszentrum Karlsruhe GmbH, 1989-2007* (7.4 2019). University of Karlsruhe. www.turbomole.com

Venkatraman, V., Evjen, S., & Lethesh, K. C. (n.d.). *The Ionic Liquid Property Explorer: An Extensive Library of Task-Specific Solvents*. https://doi.org/10.5281/zenodo.3251643