Viscosity and Interfacial Tension of Binary Mixtures Consisting of an \( n \)-Alkane, Branched Alkane, Primary Alcohol, or Branched Alcohol and a Dissolved Gas Using Equilibrium Molecular Dynamics Simulations

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Abstract
This study aims to characterize binary mixtures consisting of a liquid with a dissolved gas by determining their dynamic viscosity and interfacial tension using equilibrium molecular dynamics (EMD) simulations in the temperature range between (298 and 573) K and for solute mole fractions up to 0.20. With the help of a systematic variation of solvent and solute molecules, the influence of their molecular characteristics, e.g., in form of size, shape, or polarity, on the thermophysical properties of the mixtures is discussed. For this, eight different alkanes and alcohols with a carbon number between 12 and 40 as solvents and seven solutes in form of hydrogen, helium, methane, water, nitrogen, carbon monoxide, or carbon dioxide are studied. Using EMD simulations, the liquid dynamic viscosity is determined in the slightly compressed liquid phase close to saturation conditions. Simulations at vapor–liquid-equilibrium (VLE) are performed to determine the interfacial tension and to calculate the solute molecules at the vapor–liquid interface. To check the applicability of the EMD simulations, data for the dynamic viscosity and interfacial tension from this work are compared to experimental data of binary mixtures with the same solutes and similar solvents. The results from this work show that the impact of the dissolved gas on the thermophysical properties is strongly depending on its molecular characteristics. For example, the properties of mixtures containing dissolved He are usually within combined uncertainties with the ones of the pure solvent. In contrast, dissolving \( \text{CO}_2 \) leads to a pronounced reduction in both properties at comparable solute mole fractions. For the molecular characteristics of the solvent, the carbon chain length is shown to influence mainly the interfacial tension and the polarity mainly influences the viscosity.

Keywords Binary mixtures · Gases · Hydrocarbons · Interfacial tension · Molecular dynamics simulations · Viscosity
1 Introduction

Mixtures consisting of liquids with dissolved gases are often in use as working fluids in many processes in chemical and energy engineering. Examples are the synthesis of base chemical components, such as methanol [1, 2] or dimethyl ether [2, 3], the conversion of carbon dioxide (CO2) and water to synthesis gas [4], or the esterification of biofuels [5, 6]. For the efficient design of processes and apparatuses, reliable data for the thermophysical properties of the working fluid is needed at process-relevant conditions in terms of temperature $T$, pressure $p$, and composition. Two important properties are the dynamic viscosity $\eta$ and interfacial tension $\sigma$, which are addressed within this work. For example, $\eta$ is related to the momentum transport and, therefore, required for the calculation of power requirements for mixers and pumps and the characterization of the flow field in pipes and reactors. $\sigma$ is important for describing the wetting behavior at catalysts or machineries and the mass transfer at gas–liquid interfaces.

Due to the infinite number of combinations of solvents, solutes, and thermodynamic states, the investigation of $\eta$ and $\sigma$ only by experiments is not a feasible task. For systems which cannot or only hardly be probed by experiments, also because of safety reasons, the use of computer simulations has proven to be a valuable tool for thermophysical property research.

Equilibrium molecular dynamics (EMD) simulations, which are used in this work for the determination of $\eta$ and $\sigma$, are based on the study of the molecular motion in a statistical fluid ensemble by solving Newton’s equations of motion using descriptions of the intra- and intermolecular interactions via so-called force fields (FF) [7–9]. In this way, multiple thermophysical properties can be calculated from the recorded molecule trajectories and energies. A further advantage of EMD simulations is the possibility to gain insight into the fluid structure on a molecular level which is valuable for the analysis of structure–property relationships and can be utilized for the development of prediction models [10–12].

The present work is part of a research project at the Institute of Advanced Optical Technologies-Thermophysical Properties (AOT-TP) at the Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU). The project aims at the characterization of pure linear, branched, or cyclic hydrocarbons and their mixtures without and with dissolved gases by the determination of $\eta$ and surface or interfacial tension $\sigma$ over a wide range of thermodynamic states via surface light scattering (SLS) experiments and EMD simulations. In a first step, a $T$-dependent modification of the L-OPLS FF [13, 14] served for improved predictions of the liquid density $\rho_L$, $\eta_L$, and $\sigma$ of 12 pure solvents up to $T=573$ K [15]. This modification has since been applied to binary liquid mixtures of $n$-hexadecane with $n$-octacosane, 2,2,4,4,6,8,8-heptamethylnonane (HMN), or 1-hexadecanol [16], and binary mixtures consisting of long linear, branched, or cyclic hydrocarbons and seven different gases between (303 and 573) K [17, 18]. For all investigations, the results of EMD simulations could be directly compared with the SLS experiments. Here again, EMD simulations are used to determine $\eta_L$ and $\sigma$ of 45 further binary mixtures consisting of a liquid with a dissolved gas up to $T=573$ K.
and mole fractions of the dissolved gas in the liquid phase up to 0.20. As solvents, linear and branched alkanes and alcohols of varying carbon number are investigated to cover a wide range of molecular size and weight as well as polarity. In detail, n-dodecane, 1-dodecanol, 2-butyl-1-octanol, HMN, 1-hexadecanol, n-octacosane, 2,6,10,15,19,23-hexamethyldodecasane (squalane), and n-tetracontane are selected as solvents. As solutes, hydrogen (H₂), helium (He), methane (CH₄), water (H₂O), nitrogen (N₂), carbon monoxide (CO), and CO₂ were chosen for their variety in size, weight, sphericity, and polarity.

In the following, the relevant information for the EMD simulations, including the applied FFs, simulation procedure, and data evaluation will be given. Afterwards, the influence of the molecular characteristics of the solvent and solute on the thermophysical properties of the mixtures is discussed. The influence of the solute is analyzed by comparing the results for binary mixtures consisting of different gases dissolved in either n-dodecane, 1-dodecanol, or n-tetracontane. The simulation results are compared to experimental data. Because of the scarce data situation in the literature, results for similar systems are used for comparison purpose mainly with results for binary mixtures based on similar solvents from our previous investigations [17, 18]. The influence of the solvent is analyzed by comparing the results for binary mixtures consisting of the solutes He, N₂, or CO₂ dissolved in various solvents. Finally, the enrichment of solute molecules at the vapor–liquid interface is calculated via the relative adsorption \( \Gamma_2 \) and related to the results for \( \sigma \) to deepen the understanding why different solute types have a different impact on \( \sigma \) of the binary mixtures.

2 EMD Simulations

EMD simulations predict the movement of atoms and molecules in a statistical ensemble by solving Newton’s classical equations of motions and describing the intra- and intermolecular interactions between two atoms. From the resulting trajectories, forces, and energies, a broad range of thermophysical properties, including the viscosity and interfacial tension which are of interest in this work, can be accessed. Further information on the underlining principals of MD simulations can be found in the respective literatures [7–9]. In the following, only the relevant information of the present study is summarized.

The accuracy of the predicted properties from EMD simulations strongly depends on the employed FFs. The L-OPLS FF was chosen for modeling of the solvent molecules, since it has shown the best agreement with experimental reference data for predicting equilibrium and transport properties of long alkanes and alcohols [19], including diffusivities in their mixtures with dissolved gases [10, 11, 20–22].

In our previous studies, by incorporating a \( T \) dependency in one parameter of the 6–12 Lennard–Jones (LJ) potential, which is used to describe repulsive and dispersive interactions between two atoms, the L-OPLS all-atom (AA) FF [13, 14] was modified for a better representation of the density \( \rho \), \( \eta \), and \( \sigma \) of 12 pure linear or branched alkanes and alcohols with carbon numbers between 12 and 40 at \( T \) between (298.15 and 573.15) K [15]. This modification led to improved predictions of \( \rho \), \( \eta \),
and $\sigma$ with average absolute relative deviations from SLS experiments, which were presented in the same work, of (1.1, 17, and 11) $\%$, respectively. The $T$-dependent modification [15], which is fully transferable to other substances, was then tested by comparing predicted $\eta$ and $\sigma$ of binary liquid mixtures of linear and branched alkanes and alcohols [16], cyclic liquid organic hydrogen carriers (LOHCs) [23] and their mixtures [24] as well as binary mixtures consisting of a hydrocarbon liquid with different dissolved gases [17, 18] to experimental values from SLS. In all cases, the $T$-dependent modification has shown to greatly improve the $T$-dependent drift of the simulated properties in comparison to the experimental ones. Therefore, the same FFs are employed within this study for the solvent molecules. The partial charges located on the center of the atoms were taken from the original publications and are summarized together with the FF parameters in the Supporting Information of our previous publication [15]. For the gas solutes, namely, H$_2$, He, CH$_4$, H$_2$O, CO, N$_2$ and CO$_2$, the FFs employed in our previous publications are used [17, 18]. All FFs used in this study are non-polarizable flexible all atom FFs, characterized by pair-additive potential energy functions describing intramolecular and intermolecular interactions.

2.1 Simulation Details

All EMD simulations were performed using the GROMACS software package, version 5.1.2 [25] at $T= (298.15, 323.15, 348.15, 373.15, 423.15, 473.15$ and 573.15) K or starting at elevated $T$ for solvents with a melting $T$ above 298.15 K. Equations of motion were integrated using the Leapfrog algorithm with an integration time step of 2 fs for all systems. The bonds between the lighter hydrogen atoms and the heavier carbon or oxygen atoms in the solvent molecules were constrained using the LINCS [26] algorithm. The LJ and electrostatic interactions are calculated from the 6–12 LJ potential and coulombic potential, within the cutoff radius of 1.6 nm. Long-range electrostatic interactions beyond the cutoff radius is modeled via the particle-mesh Ewald (PME) algorithm [27]. For simulations in the slightly compressed liquid phase, standard dispersion corrections for energy and pressure is considered for calculating long-range LJ interactions beyond the cutoff radius. Long-range LJ interaction in VLE simulations are calculated using LJ-PME algorithm [28].

The simulations were initialized by randomly inserting solvent molecules into a cubic simulation box with periodic boundary conditions in all directions. The number of solvent molecules corresponds to approximately 30,000 atoms in each case to maintain comparable computational effort for all mixtures. Three unique simulation boxes are created, which are the bases of three independent simulation runs for all following steps. Afterwards, solute molecules are added to match the calculated solubility data. Details on the solubility data used in this work are discussed in the Supporting Information. After an energy minimization step to approximate realistic atomic proximities following the random insertion of molecules into the simulation box, the desired $T$ and $p$ was attained in 1 ns simulation run in a canonical ensemble ($NVT$), followed by a 10 ns simulation run in an isothermal-isobaric ensemble ($NpT$). To avoid a possible phase separation, $p$ in the $NpT$ simulations was
set 0.2 MPa above the calculated saturation $p$. $\rho_L$ was determined from the $NpT$ simulation, neglecting first 0.5 ns for equilibration of $T$ and $p$, and the liquid dynamic viscosity $\eta_L$ was calculated from a subsequent simulation in an $NVT$ ensemble based on the Green–Kubo method [29, 30], which is further detailed in the “Data evaluation” section.

To create a simulation box in VLE, which is required for calculating $\sigma$, the cubic simulation boxes were extended in the z-direction to create two vapor–liquid interfaces. For typical simulation boxes with a box length of (6.0 to 7.0) nm, previous studies have shown that an extension by a factor of three is sufficient to ensure that the long solvent molecules in the vapor phase only interact with one of the two interfaces. Additional solute gas molecules were then added to the vapor phase to reach the vapor density of the solute at the required partial pressure. These values were obtained from the REFPROP data base [31]. Afterwards, VLE simulations of about (15 to 20) ns in an $NVT$ ensemble were performed to calculate $\sigma$, neglecting the first 5 ns for equilibration. From the same VLE simulations, the partial number densities of the solute and solvent as function of the $z$-coordinate of the simulation box were obtained to study a possible enrichment of the constituent species at the interface, which is further explained in Sect. 2.2.

For calculating the composition in the liquid phase and the related saturation $p$, solubility data are required. Experimental solubility data are only available for a few systems and had to be inter- and extrapolated with respect to the carbon number in the solvent molecules for all other systems. This is detailed in the Supporting Information.

### 2.2 Data Evaluation

The Green–Kubo formalism [29, 30] is used to calculate $\eta_L$ within this study. This formalism analyzes the pressure autocorrelation function (PACF) of the off-diagonal elements of the $p$ tensor. By averaging over the plateau of the running integral of the PACF $\eta_L(\tau)$ with respect to correlation time $\tau$, $\eta_L$ can be obtained. However, in practice, it is difficult to identify the plateau region due to an accumulation of statistical noise at long correlation times [32]. Alternatively, Zhang et al. [33] proposed a time-decomposition method for the reliable determination of $\eta_L$. Using this method, $\eta_L(\tau)$ of multiple short simulation runs are fitted to a double exponential function according to

$$\eta_L(\tau) = A \alpha \tau_1 (1 - e^{-\tau/\tau_1}) + A(1 - \alpha) \tau_2 (1 - e^{-\tau/\tau_2}).$$

In Eq. 1, $A$ and $\alpha$ are fitting parameters and $\tau_1$ and $\tau_2$ characterize the decay time of the two exponential functions. For more detail of this method, reader is referred to the respective publication [33]. In the following, the procedure adopted within this study is detailed.

To apply the method of Zhang et al. [33], the $NVT$ simulations are divided into multiple short simulations of 2 ns. In the upper part of Fig. 1, $\eta_L(\tau)$ for the resulting 28 short simulations for the binary mixture of 1-dodecanol and CO$_2$ with $x_{CO_2} = 0.20$ at 423.15 K is shown together with their running average indicated by
a black dashed line. The running average is then fitted with respect to $\tau$ according to Eq. 1. In a next step, the double standard deviation, $\sigma(\eta(\tau))$, between all individual runs is calculated as a function of $\tau$ and fitted to the functional form $\sigma(\eta(\tau)) = B \cdot \tau^{b}$ up to a cutoff time of $\tau_{\text{cut}} = 6 \cdot \max(\tau_1, \tau_2)$, where $B$ and $b$ are fitting parameters. In all the investigated systems within this study, $b$ values were between 0.6 and 0.8. In a final step, the running average is again fitted according to Eq. 1 up to $\tau_{\text{cut}}$ with a weighing function of $1/\tau^b$. $\eta_L$ follows directly from the plateau value of the theoretical equation according to Eq. 1. To compare the approach of Zhang et al. [33] with the results from one long simulation run, the running average of 28 simulations of 2 ns length is compared with $\eta_L(\tau)$ from a 60 ns long simulation run for the system of 1-dodecanol with dissolved CO$_2$ at $x_{\text{CO}_2} = 0.20$ is shown in the lower part of Fig. 1. While both methods yield similar $\eta_L$, the method proposed by Zhang et al. [33] has the advantage that the plateau range is calculated from the decay times of the exponential functions and is, therefore, not influenced by the subjectivity of the individual, as is the case for the classical approach.

$\sigma$ is calculated from the diagonal elements of the $p$ tensor from the simulations in VLE according to

$$\sigma(t) = \frac{L_z}{2} \left( p_{zz}(t) - \frac{p_{xx}(t) + p_{yy}(t)}{2} \right)$$

(2)

Here, $L_z$ is the box length in $z$-direction and $p_{xx}$, $p_{yy}$, and $p_{zz}$ are the diagonal elements of the pressure sensor. $\sigma$ is then determined by averaging $\sigma(t)$ over the simulation time excluding the first 5 ns for equilibration and formation of the vapor–liquid interface. For more details on the calculation of $\sigma$ from MD simulations, the reader
is referred to our previous publications [15, 34] as well as the general literature on MD simulations [7–9].

For a vapor–liquid interface parallel to the $x,y$-plane, the relative adsorption of the solute (component 2) with respect to the solvent (component 1) $\Gamma_2^{(1)}$ can be calculated using the component number density profiles along the $z$-axis $\rho_i(z)$, obtained from EMD simulations in VLE according to [35, 36]

$$\Gamma_2^{(1)} = - \left( \rho_{L,2} - \rho_{V,2} \right) \int_{-\infty}^{\infty} \left[ \frac{\rho_{L,1} - \rho_1(z)}{\rho_{L,1} - \rho_{V,2}} + \frac{\rho_{L,2} - \rho_2(z)}{\rho_{L,2} - \rho_{V,2}} \right] dz \quad (3)$$

Here, $\rho_{L,i}$ and $\rho_{V,i}$ represent the bulk liquid and vapor densities of component $i$ far away from the vapor–liquid interface. They can be calculated by averaging over $\rho_i(z)$ in the range where $\rho_i(z)$ is independent of $z$.

The final results for the thermophysical properties of the binary mixtures investigated in this work are summarized in Table 1 together with their statistical uncertainties and thermodynamic states. The final values for the properties are the averages of the three independent simulation runs. The expanded statistical uncertainties, based on a coverage factor $k=2$, are calculated from the double standard deviations of the values from the three simulations.

### 3 Results and Discussion

The results for $\rho_L, \eta_L, \sigma$, and $\Gamma_2^{(1)}$ for the 45 binary mixtures investigated in this work are summarized together with their expanded statistical uncertainties and thermodynamic states in Table 1. Systems based on solutes with a low solubility, i.e., $H_2$, He, and $N_2$, were investigated at constant $p$ of about 7.0 MPa to allow a comparison with previous publications containing binary mixtures with the same solutes [17, 18] which were conducted at a maximum $p=7.0$ MPa due to the limitations of the experimental setup. Systems with more soluble solutes, namely, $CH_4$, CO, CO$_2$, and $H_2O$, were investigated at $x_{\text{solute}}=(0.10$ and 0.20) in the case of $CH_4$ and CO$_2$, $x_{\text{solute}}=(0.05$ and 0.10) in the case of CO, and $x_{\text{solute}}=(0.045$ and 0.09) in the case of $H_2O$. The simulations were carried out at $T$ between (298.15 and 573.15) K, or starting from approximately 20 K above the melting $T$ of the solvent. Due to the low solubility of $H_2O$ in hydrocarbons at low $T$, the simulations with dissolved $H_2O$ were carried out between $T=(423.15$ and 573.15) K. In the following, first the results for $\eta_L$ and $\sigma$ are shown and discussed. To investigate the influence of the dissolved gas, the deviation of the reported mixture properties from the pure solvents investigated by EMD simulations [15] are calculated. For this, $\eta_L$ and $\sigma$ of the pure solvents were correlated with respect to $T$, as detailed in the Supporting Information. Here, it must be mentioned that the a general overprediction for $\eta_L$ and $\sigma$ of the pure solvents by up to 30 % with respect to the $T$-dependent correlations from SLS results could be observed in our previous publication [15]. In this work, the influence of the dissolved gas in form of the deviation between the mixtures and the pure solvents is compared to experimental and simulated data for similar systems.
Table 1. Liquid densities $\rho_L$ and dynamic viscosities $\eta_L$ in the compressed liquid phase close to saturation conditions as well as interfacial tensions $\sigma$ and relative adsorptions $\Gamma_{2}^{(1)}$ at VLE conditions of the investigated binary mixtures

| $T$ (K) | $p$ (MPa)\(^{a}\) | $x_{\text{solute}}$\(^{b}\) | $\rho_L$ (kg·m\(^{-3}\)) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m\(^{-1}\)) | $100 U_r(\sigma)$ | $\Gamma_{2}^{(1)}$ (nm\(^{-2}\)) | $U(\Gamma_{2}^{(1)})$ (nm\(^{-2}\)) |
|---------|------------------|--------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| n-Dodecane + H\(_2\) ($p \approx 7.0$ MPa) |
| 298.15  | 7.04 0.047 744.38 0.27 1.99 15 | 24.48 3.4 0.48 | 0.096 | 323.15 7.14 0.054 724.56 0.11 1.33 17 | 21.82 1.0 0.37 | 0.081 | 348.15 7.15 0.061 705.13 0.39 0.821 12 | 20.44 2.2 0.29 | 0.055 | 373.15 7.19 0.068 685.57 0.71 0.716 20 | 18.80 1.4 0.29 | 0.055 | 423.15 5.17 0.083 645.71 0.14 0.389 15 | 14.79 2.6 0.19 | 0.044 | 473.15 7.78 0.099 603.27 0.48 0.278 13 | 11.39 5.2 0.14 | 0.008 | 573.15 5.76 0.147 496.65 0.16 0.108 12 | 4.45 5.3 0.09 | 0.025 |
| n-Dodecane + He ($p \approx 7.0$ MPa) |
| 298.15  | 7.16 0.016 748.44 0.28 1.81 14 | 25.41 5.6 0.06 | 0.028 | 323.15 7.16 0.021 729.10 0.41 1.43 26 | 23.32 0.8 0.05 | 0.010 | 348.15 7.12 0.027 710.10 0.32 0.870 15 | 21.53 1.9 0.03 | 0.022 | 373.15 7.27 0.032 691.48 0.22 0.704 2.2 | 19.39 0.5 0.03 | 0.046 | 423.15 7.09 0.046 652.65 0.22 0.440 10 | 15.61 1.9 0.02 | 0.028 | 473.15 7.07 0.062 611.65 0.56 0.291 7.4 | 11.88 2.8 0.01 | 0.016 | 573.15 7.00 0.103 512.83 0.65 0.135 20 | 4.77 5.0 0.00 | 0.010 |
| n-Dodecane + CH\(_4\) ($x_{\text{solute}} \approx 0.10$) |
| 298.15  | 2.18 0.100 736.04 0.79 1.76 11 | 23.25 3.3 0.71 | 0.069 | 323.15 2.60 0.100 716.69 0.65 1.03 7.5 | 20.62 0.8 0.59 | 0.073 | 348.15 2.82 0.100 697.51 0.43 0.789 4.7 | 19.09 2.6 0.51 | 0.062 | 373.15 2.88 0.100 678.28 0.42 0.621 8.1 | 17.38 2.6 0.42 | 0.050 | 423.15 3.08 0.100 639.03 0.46 0.375 21 | 13.96 2.1 0.32 | 0.039 | 473.15 3.19 0.100 597.14 0.98 0.245 6.0 | 10.39 1.4 0.23 | 0.001 | 573.15 3.40 0.100 495.99 0.54 0.117 6.8 | 3.90 2.6 0.12 | 0.003 |
Table 1 (continued)

| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|---------|----------------|------------------|-----------------|-------------------|---------------|-----------------|-----------------|----------------|-----------------|----------------|
| 298.15  | 4.76           | 0.200            | 725.62          | 0.39              | 1.50          | 14              | 19.40           | 4.2             | 1.39            | 0.042           |
| 323.15  | 5.40           | 0.200            | 706.20          | 0.28              | 0.861         | 12              | 17.96           | 3.8             | 1.16            | 0.043           |
| 348.15  | 5.75           | 0.200            | 686.87          | 0.34              | 0.684         | 3.6             | 16.30           | 1.9             | 0.97            | 0.082           |
| 373.15  | 6.21           | 0.200            | 667.72          | 0.27              | 0.560         | 16              | 14.89           | 2.1             | 0.84            | 0.054           |
| 423.15  | 6.49           | 0.200            | 627.88          | 0.38              | 0.342         | 5.7             | 11.89           | 2.1             | 0.59            | 0.006           |
| 473.15  | 6.61           | 0.200            | 584.88          | 0.13              | 0.222         | 2.2             | 9.02            | 4.7             | 0.40            | 0.037           |
| 573.15  | 5.97           | 0.200            | 476.94          | 1.33              | 0.103         | 4.3             | 3.15            | 3.6             | 0.18            | 0.010           |
| 298.15  | 5.48           | 0.050            | 742.47          | 0.37              | 1.92          | 32              | 21.96           | 5.1             | 0.79            | 0.040           |
| 323.15  | 3.70           | 0.050            | 723.24          | 0.40              | 1.17          | 20              | 20.17           | 2.4             | 0.64            | 0.052           |
| 348.15  | 3.99           | 0.050            | 704.22          | 0.13              | 0.909         | 3.4             | 18.88           | 2.5             | 0.48            | 0.021           |
| 373.15  | 4.34           | 0.050            | 684.97          | 0.73              | 0.622         | 14              | 17.31           | 2.6             | 0.38            | 0.037           |
| 423.15  | 2.35           | 0.050            | 645.91          | 0.74              | 0.368         | 1.0             | 13.91           | 2.2             | 0.26            | 0.014           |
| 473.15  | 0.90           | 0.050            | 604.26          | 0.66              | 0.254         | 16              | 10.67           | 1.2             | 0.19            | 0.009           |
| 573.15  | 2.17           | 0.050            | 503.14          | 0.70              | 0.118         | 3.4             | 4.28            | 3.0             | 0.08            | 0.006           |
| 298.15  | 6.72           | 0.100            | 739.98          | 0.69              | 1.61          | 11              | 19.13           | 4.8             | 1.24            | 0.053           |
| 323.15  | 6.66           | 0.100            | 720.52          | 0.11              | 0.961         | 1.7             | 17.63           | 0.4             | 1.03            | 0.067           |
| 348.15  | 6.64           | 0.100            | 701.52          | 0.45              | 0.807         | 11              | 16.52           | 7.0             | 0.85            | 0.057           |
| 373.15  | 6.46           | 0.100            | 682.39          | 0.55              | 0.623         | 2.1             | 15.26           | 1.6             | 0.69            | 0.042           |
| 423.15  | 5.97           | 0.100            | 643.19          | 0.46              | 0.373         | 3.7             | 12.32           | 5.4             | 0.47            | 0.049           |
| 473.15  | 5.49           | 0.100            | 601.58          | 0.46              | 0.256         | 9.6             | 9.46            | 3.5             | 0.31            | 0.004           |
| 573.15  | 4.65           | 0.100            | 501.29          | 1.14              | 0.119         | 2.0             | 3.55            | 10.0            | 0.14            | 0.018           |
| T (K) | \( p \) (MPa)\(^a\) | \( x_{\text{solute}} \) | \( \rho_L \) (kg·m\(^{-3}\)) | \( 10^3 \cdot U_r(\rho_L) \) | \( \eta_L \) (mPa·s) | \( 100 \cdot U_r(\eta_L) \) | \( \sigma \) (mN·m\(^{-1}\)) | \( 100 \cdot U_r(\sigma) \) | \( \Gamma_{2}^{(1)} \) (nm\(^{-2}\)) | \( U(\Gamma_{2}^{(1)}) \) (nm\(^{-2}\)) |
|-------|----------------|-----------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| 298.15 | 3.74 | 0.089 | 743.12 | 0.36 | 1.79 | 11 | 20.43 | 6.8 | 1.03 | 0.035 |
| 323.15 | 8.09 | 0.090 | 726.91 | 0.34 | 1.39 | 14 | 19.08 | 1.3 | 0.87 | 0.049 |
| 348.15 | 7.66 | 0.091 | 707.64 | 0.16 | 0.850 | 2.5 | 17.60 | 1.8 | 0.71 | 0.059 |
| 373.15 | 7.34 | 0.094 | 688.28 | 0.11 | 0.636 | 6.8 | 15.90 | 1.9 | 0.58 | 0.103 |
| 423.15 | 7.21 | 0.104 | 648.93 | 0.57 | 0.394 | 10 | 12.86 | 3.8 | 0.41 | 0.017 |
| 473.15 | 7.28 | 0.120 | 606.64 | 1.09 | 0.272 | 18 | 9.54 | 1.7 | 0.31 | 0.020 |
| 573.15 | 7.27 | 0.168 | 500.77 | 1.76 | 0.120 | 11 | 3.42 | 5.8 | 0.15 | 0.022 |
| 298.15 | 1.06 | 0.100 | 749.26 | 0.47 | 1.46 | 13 | 23.89 | 2.2 | 0.68 | 0.084 |
| 323.15 | 1.08 | 0.100 | 729.26 | 0.26 | 1.21 | 19 | 21.85 | 1.9 | 0.64 | 0.080 |
| 348.15 | 1.52 | 0.100 | 709.93 | 0.43 | 0.859 | 7.7 | 19.83 | 4.1 | 0.57 | 0.138 |
| 373.15 | 1.70 | 0.100 | 690.36 | 0.47 | 0.612 | 2.4 | 17.76 | 1.2 | 0.51 | 0.055 |
| 423.15 | 1.87 | 0.100 | 650.24 | 0.77 | 0.373 | 1.9 | 13.95 | 2.7 | 0.38 | 0.016 |
| 473.15 | 2.29 | 0.100 | 607.96 | 1.13 | 0.251 | 2.3 | 10.49 | 2.9 | 0.33 | 0.009 |
| 573.15 | 3.08 | 0.100 | 506.99 | 2.09 | 0.121 | 3.3 | 3.72 | 7.2 | 0.19 | 0.029 |
| 298.15 | 1.77 | 0.100 | 754.39 | 0.04 | 1.34 | 22 | 22.15 | 1.4 | 0.94 | 0.051 |
| 323.15 | 2.40 | 0.100 | 734.11 | 0.20 | 0.990 | 10 | 19.74 | 3.1 | 0.94 | 0.099 |
| 348.15 | 2.93 | 0.100 | 714.05 | 0.12 | 0.733 | 15 | 17.70 | 5.4 | 0.81 | 0.089 |
| 373.15 | 3.25 | 0.100 | 693.81 | 0.16 | 0.571 | 3.8 | 15.94 | 1.4 | 0.72 | 0.078 |
| 423.15 | 4.06 | 0.100 | 652.77 | 1.04 | 0.342 | 6.9 | 12.20 | 3.6 | 0.57 | 0.063 |
| 473.15 | 4.56 | 0.100 | 608.79 | 0.12 | 0.238 | 7.2 | 8.89 | 4.0 | 0.44 | 0.024 |
| 573.15 | 5.05 | 0.100 | 500.91 | 1.68 | 0.109 | 4.1 | 2.79 | 11.4 | 0.20 | 0.019 |
Table 1 (continued)

| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|---------|---------------|-------------------|-------------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| $n$-Dodecane + H$_2$O ($x_{\text{solute}} \approx 0.045$) | | | | | | | | | | |
| 423.15  | $-0.12$       | $0.046$           | 646.85            | 0.47           | 0.395          | 10             | 15.60          | 2.2             | 0.02           | 0.019          |
| 473.15  | $0.23$        | $0.046$           | 602.82            | 1.16           | 0.272          | 21             | 11.79          | 0.5             | 0.01           | 0.006          |
| 573.15  | $0.98$        | $0.046$           | 498.12            | 1.89           | 0.116          | 4.0            | 4.72           | 4.2             | 0.01           | 0.014          |
| $n$-Dodecane + H$_2$O ($x_{\text{solute}} \approx 0.090$) | | | | | | | | | | |
| 423.15  | $-0.10$       | $0.091$           | 647.38            | 0.49           | 0.402          | 10             | 15.58          | 2.6             | 0.20           | 0.256          |
| 473.15  | $0.25$        | $0.091$           | 603.27            | 1.21           | 0.266          | 7.7            | 11.77          | 1.9             | 0.04           | 0.002          |
| 573.15  | $1.74$        | $0.091$           | 489.09            | 3.47           | 0.110          | 9.0            | 4.75           | 7.8             | 0.03           | 0.011          |
| 1-Dodecanol + H$_2$ ($\rho \approx 7.0$ MPa) | | | | | | | | | | |
| 323.15  | $6.72$        | $0.073$           | 805.39            | 1.25           | 6.35           | 29             | 29.94          | 4.2             | 0.41           | 0.030          |
| 348.15  | $7.07$        | $0.071$           | 785.20            | 0.21           | 3.32           | 17             | 24.98          | 11.1            | 0.33           | 0.056          |
| 373.15  | $7.06$        | $0.071$           | 765.04            | 0.18           | 1.93           | 5.7            | 23.04          | 2.7             | 0.26           | 0.061          |
| 423.15  | $7.17$        | $0.078$           | 723.61            | 0.53           | 0.866          | 23             | 19.85          | 5.2             | 0.20           | 0.032          |
| 473.15  | $5.94$        | $0.089$           | 680.48            | 0.35           | 0.452          | 4.6            | 15.31          | 1.5             | 0.15           | 0.013          |
| 573.15  | $7.11$        | $0.117$           | 584.44            | 0.70           | 0.191          | 3.5            | 8.27           | 2.3             | 0.10           | 0.040          |
| 1-Dodecanol + He ($\rho \approx 7.0$ MPa) | | | | | | | | | | |
| 323.15  | $7.00$        | $0.013$           | 813.49            | 0.57           | 9.09           | 17             | 31.62          | 4.8             | 0.06           | 0.042          |
| 348.15  | $7.38$        | $0.014$           | 793.03            | 1.03           | 3.61           | 29             | 26.72          | 3.3             | 0.04           | 0.027          |
| 373.15  | $7.15$        | $0.018$           | 772.67            | 0.45           | 2.10           | 18             | 24.20          | 2.8             | 0.03           | 0.024          |
| 423.15  | $7.22$        | $0.025$           | 732.17            | 0.47           | 0.921          | 11             | 20.77          | 5.2             | 0.02           | 0.029          |
| 473.15  | $7.03$        | $0.038$           | 690.15            | 0.44           | 0.479          | 3.6            | 16.44          | 2.7             | 0.01           | 0.006          |
| 573.15  | $6.04$        | $0.072$           | 597.55            | 1.11           | 0.203          | 17             | 8.70           | 1.2             | 0.00           | 0.026          |
| 1-Dodecanol + CH$_4$ ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 323.15  | $3.03$        | $0.100$           | 799.90            | 0.65           | 6.16           | 23             | 26.23          | 10              | 0.64           | 0.118          |
| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma(1)^{11}$ (nm$^{-2}$) | $U(\Gamma(1)^{11})$ (nm$^{-2}$) |
|--------|-------------|------------------|-----------------|----------------|---------------|----------------|----------------|---------------|----------------|----------------|
| 348.15 | 3.22        | 0.100            | 779.23          | 0.99           | 3.10          | 21             | 23.48          | 3.1           | 0.61            | 0.016          |
| 373.15 | 3.63        | 0.100            | 758.89          | 0.31           | 1.71          | 3.0            | 21.62          | 1.2           | 0.55            | 0.053          |
| 423.15 | 3.99        | 0.100            | 717.58          | 0.66           | 0.810         | 13             | 18.23          | 1.7           | 0.43            | 0.038          |
| 473.15 | 4.34        | 0.100            | 675.39          | 1.12           | 0.437         | 11             | 14.60          | 1.0           | 0.31            | 0.072          |
| 573.15 | 4.09        | 0.100            | 580.93          | 0.65           | 0.189         | 5.2            | 7.52           | 1.4           | 0.17            | 0.014          |
| 1-Dodecanol + CH$_4$ ($x_{\text{solute}} \approx 0.20$) |
| 323.15 | 5.79        | 0.200            | 787.40          | 0.29           | 4.50          | 17             | 22.05          | 7.6           | 1.28            | 0.105          |
| 348.15 | 6.45        | 0.200            | 766.98          | 0.20           | 2.36          | 16             | 20.29          | 1.2           | 1.13            | 0.105          |
| 373.15 | 7.07        | 0.200            | 746.58          | 0.31           | 1.40          | 3.6            | 18.77          | 7.8           | 0.97            | 0.063          |
| 423.15 | 7.56        | 0.200            | 704.90          | 0.86           | 0.635         | 10             | 15.86          | 1.5           | 0.72            | 0.032          |
| 473.15 | 8.15        | 0.200            | 662.42          | 0.39           | 0.390         | 3.4            | 12.60          | 2.9           | 0.55            | 0.044          |
| 573.15 | 8.37        | 0.200            | 569.04          | 0.32           | 0.175         | 2.7            | 6.12           | 4.4           | 0.30            | 0.022          |
| 1-Dodecanol + CO ($x_{\text{solute}} \approx 0.05$) |
| 323.15 | 3.26        | 0.051            | 806.51          | 0.54           | 5.94          | 31             | 26.48          | 9.3           | 0.65            | 0.068          |
| 348.15 | 3.54        | 0.051            | 785.99          | 0.31           | 3.40          | 13             | 22.98          | 2.3           | 0.57            | 0.039          |
| 373.15 | 3.70        | 0.051            | 765.67          | 0.38           | 1.83          | 11             | 22.20          | 5.4           | 0.46            | 0.024          |
| 423.15 | 3.82        | 0.051            | 724.65          | 0.65           | 0.810         | 1.9            | 18.38          | 2.6           | 0.35            | 0.024          |
| 473.15 | 3.68        | 0.051            | 682.45          | 0.47           | 0.451         | 5.0            | 14.69          | 1.6           | 0.24            | 0.005          |
| 573.15 | 3.44        | 0.051            | 589.91          | 0.64           | 0.204         | 11             | 7.64           | 4.7           | 0.13            | 0.016          |
| 1-Dodecanol + CO ($x_{\text{solute}} \approx 0.10$) |
| 323.15 | 7.44        | 0.100            | 803.40          | 0.33           | 5.73          | 23             | 23.98          | 3.8           | 1.00            | 0.100          |
| 348.15 | 7.57        | 0.100            | 782.77          | 0.23           | 3.19          | 19             | 20.69          | 4.9           | 0.90            | 0.016          |
| 373.15 | 7.32        | 0.100            | 762.35          | 0.28           | 1.80          | 24             | 19.80          | 4.2           | 0.80            | 0.014          |
| 423.15 | 7.49        | 0.100            | 721.57          | 0.64           | 0.786         | 7.1            | 16.60          | 2.3           | 0.58            | 0.053          |
Table 1 (continued)

| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|---------|-----------------|---------------------|--------------------------|---------------------|-----------------|---------------------|------------------|------------------|-----------------|-----------------|
| 473.15  | 7.25            | 0.100               | 679.71                   | 0.37                | 0.439           | 9.4                 | 13.04            | 0.4              | 0.41            | 0.050           |
| 573.15  | 8.68            | 0.100               | 588.98                   | 0.53                | 0.206           | 5.4                 | 6.56             | 1.1              | 0.22            | 0.039           |
| 1-Dodecanol + N$_2$ ($p \approx 7.0$ MPa) |
| 323.15  | 8.62            | 0.042               | 813.00                   | 1.40                | 7.31            | 28                  | 26.77            | 6.7              | 0.78            | 0.055           |
| 348.15  | 7.42            | 0.045               | 791.51                   | 0.38                | 2.85            | 7.2                 | 23.06            | 3.0              | 0.71            | 0.021           |
| 373.15  | 7.51            | 0.048               | 771.22                   | 0.12                | 1.92            | 24                  | 21.36            | 6.2              | 0.56            | 0.020           |
| 423.15  | 7.19            | 0.059               | 729.82                   | 0.17                | 0.896           | 11                  | 17.75            | 4.3              | 0.43            | 0.008           |
| 473.15  | 7.14            | 0.073               | 687.19                   | 0.15                | 0.469           | 10                  | 14.27            | 2.8              | 0.31            | 0.035           |
| 573.15  | 6.19            | 0.110               | 592.27                   | 0.76                | 0.197           | 9.7                 | 6.87             | 2.1              | 0.17            | 0.003           |
| 1-Dodecanol + CO$_2$ ($x_{\text{solute}} \approx 0.10$) |
| 323.15  | 1.60            | 0.100               | 813.48                   | 0.90                | 4.65            | 0.70                | 26.59            | 5.2              | 0.47            | 0.109           |
| 348.15  | 2.09            | 0.100               | 792.54                   | 0.28                | 2.66            | 16                  | 24.33            | 4.6              | 0.51            | 0.082           |
| 373.15  | 1.93            | 0.100               | 771.53                   | 0.09                | 1.75            | 18                  | 22.26            | 5.5              | 0.50            | 0.056           |
| 423.15  | 2.33            | 0.100               | 729.52                   | 0.20                | 0.838           | 8.2                 | 18.43            | 2.9              | 0.40            | 0.061           |
| 473.15  | 2.69            | 0.100               | 686.41                   | 0.67                | 0.450           | 14                  | 14.65            | 3.9              | 0.30            | 0.038           |
| 573.15  | 2.79            | 0.100               | 589.73                   | 0.30                | 0.195           | 12                  | 7.27             | 6.8              | 0.17            | 0.023           |
| 1-Dodecanol + CO$_2$ ($x_{\text{solute}} \approx 0.20$) |
| 323.15  | 2.72            | 0.200               | 817.05                   | 0.66                | 4.52            | 11                  | 23.37            | 4.1              | 1.19            | 0.227           |
| 348.15  | 3.44            | 0.200               | 795.92                   | 0.62                | 2.12            | 26                  | 21.28            | 1.2              | 1.07            | 0.206           |
| 373.15  | 3.86            | 0.200               | 774.67                   | 0.64                | 1.28            | 16                  | 19.70            | 2.4              | 1.00            | 0.107           |
| 423.15  | 4.79            | 0.200               | 731.89                   | 0.24                | 0.649           | 9.6                 | 16.10            | 0.7              | 0.75            | 0.105           |
| 473.15  | 5.14            | 0.200               | 687.43                   | 0.56                | 0.403           | 2.8                 | 12.60            | 3.5              | 0.56            | 0.020           |
| 573.15  | 4.73            | 0.200               | 588.19                   | 0.32                | 0.172           | 2.7                 | 5.87             | 5.3              | 0.31            | 0.011           |
Table 1 (continued)

| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^1$ (nm$^{-2}$) | $U(\Gamma_2^1)$ (nm$^{-2}$) |
|---------|----------------|----------------------|-------------------------|---------------------|------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 1-Dodecanol + H$_2$O ($x_{\text{solute}} \approx 0.045$) | | | | | | | | | | |
| 423.15  | 0.17           | 0.046               | 727.66                 | 0.48               | 0.841            | 4.6                 | 20.73               | 2.3                 | 0.02                | 0.015               |
| 473.15  | 0.25           | 0.046               | 684.64                 | 0.22               | 0.481            | 8.0                 | 16.59               | 0.8                 | 0.01                | 0.037               |
| 573.15  | 0.72           | 0.046               | 588.77                 | 1.02               | 0.194            | 4.8                 | 8.55                | 2.8                 | 0.02                | 0.006               |
| 1-Dodecanol + H$_2$O ($x_{\text{solute}} \approx 0.090$) | | | | | | | | | | |
| 423.15  | 0.07           | 0.091               | 727.96                 | 0.51               | 0.875            | 14                  | 20.67               | 1.4                 | 0.07                | 0.036               |
| 473.15  | 0.24           | 0.091               | 684.33                 | 0.97               | 0.486            | 6.6                 | 16.42               | 1.9                 | 0.03                | 0.010               |
| 573.15  | 1.02           | 0.091               | 586.37                 | 0.62               | 0.193            | 10                  | 8.27                | 2.7                 | 0.05                | 0.010               |
| n-Tetracontane + H$_2$ ($p \approx 7.0$ MPa) | | | | | | | | | | |
| 423.15  | 7.14           | 0.146               | 737.41                 | 0.49               | 3.13             | 20                  | 23.73               | 3.2                 | 0.21                | 0.040               |
| 473.15  | 7.07           | 0.169               | 704.79                 | 0.35               | 1.81             | 25                  | 20.53               | 1.9                 | 0.16                | 0.038               |
| 573.15  | 5.80           | 0.217               | 638.35                 | 0.73               | 0.982            | 11                  | 14.59               | 4.4                 | 0.09                | 0.036               |
| n-Tetracontane + He ($p \approx 7.0$ MPa) | | | | | | | | | | |
| 423.15  | 6.93           | 0.050               | 742.54                 | 0.34               | 3.21             | 24                  | 24.38               | 4.3                 | 0.00                | 0.010               |
| 473.15  | 7.06           | 0.065               | 711.34                 | 1.11               | 2.64             | 19                  | 21.66               | 2.3                 | 0.01                | 0.014               |
| 573.15  | 7.15           | 0.109               | 647.87                 | 0.68               | 0.954            | 12                  | 15.36               | 2.5                 | 0.01                | 0.006               |
| n-Tetracontane + CH$_4$ ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 423.15  | 1.29           | 0.103               | 734.97                 | 1.22               | 3.17             | 27                  | 23.68               | 4.7                 | 0.16                | 0.029               |
| 473.15  | 1.55           | 0.103               | 703.13                 | 0.92               | 1.85             | 10                  | 20.65               | 2.4                 | 0.12                | 0.026               |
| 573.15  | 1.64           | 0.103               | 638.10                 | 0.59               | 0.953            | 13                  | 15.18               | 2.4                 | 0.07                | 0.004               |
| n-Tetracontane + CH$_4$ ($x_{\text{solute}} \approx 0.20$) | | | | | | | | | | |
| 423.15  | 3.09           | 0.202               | 731.17                 | 0.23               | 2.48             | 25                  | 23.35               | 1.3                 | 0.31                | 0.009               |
| 473.15  | 3.08           | 0.202               | 698.88                 | 0.37               | 1.66             | 24                  | 20.18               | 4.4                 | 0.23                | 0.005               |
| 573.15  | 3.21           | 0.202               | 633.69                 | 0.21               | 0.949            | 17                  | 14.45               | 1.7                 | 0.14                | 0.018               |
| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|-------|----------------|-----------------|------------------|-----------------|----------------|----------------|-----------------|----------------|----------------|----------------|
| n-Tetracontane + CO ($x_{\text{solute}} \approx 0.05$) | | | | | | | | | | |
| 423.15 | 1.89 | 0.050 | 737.96 | 0.63 | 2.47 | 11 | 23.63 | 6.5 | 0.18 | 0.019 |
| 473.15 | 1.74 | 0.050 | 706.00 | 0.32 | 2.17 | 18 | 20.92 | 2.9 | 0.11 | 0.025 |
| 573.15 | 1.26 | 0.050 | 640.64 | 0.18 | 0.949 | 15 | 15.24 | 4.7 | 0.05 | 0.019 |
| n-Tetracontane + CO ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 423.15 | 3.68 | 0.103 | 737.20 | 0.43 | 2.67 | 25 | 23.21 | 4.2 | 0.33 | 0.033 |
| 473.15 | 3.34 | 0.103 | 705.11 | 0.68 | 1.95 | 17 | 20.04 | 5.0 | 0.23 | 0.015 |
| 573.15 | 2.67 | 0.103 | 639.83 | 0.24 | 0.892 | 13 | 14.82 | 1.3 | 0.10 | 0.016 |
| n-Tetracontane + N$_2$ ($p \approx 7.0$ MPa) | | | | | | | | | | |
| 423.15 | 7.19 | 0.225 | 737.63 | 1.21 | 3.15 | 1.2 | 62 | 1.45 | 6.7 | 0.44 | 0.037 |
| 473.15 | 7.11 | 0.246 | 704.85 | 0.16 | 2.10 | 32 | 18.85 | 2.8 | 0.32 | 0.014 |
| 573.15 | 7.95 | 0.277 | 638.69 | 0.31 | 0.922 | 19 | 13.91 | 3.9 | 0.19 | 0.040 |
| n-Tetracontane + H$_2$O ($x_{\text{solute}} \approx 0.045$) | | | | | | | | | | |
| 423.15 | 0.01 | 0.047 | 737.53 | 0.96 | 2.68 | 25 | 25.38 | 4.4 | 0.01 | 0.009 |
| 473.15 | 0.06 | 0.047 | 705.03 | 0.39 | 1.83 | 11 | 22.08 | 4.0 | 0.01 | 0.015 |
| 573.15 | 0.64 | 0.047 | 640.54 | 1.09 | 0.949 | 13 | 14.40 | 5.6 | 0.01 | 0.005 |
| n-Tetracontane + H$_2$O ($x_{\text{solute}} \approx 0.090$) | | | | | | | | | | |
| 423.15 | 0.21 | 0.093 | 737.62 | 0.52 | 2.66 | 16 | 25.18 | 3.5 | 0.01 | 0.042 |
| 473.15 | 0.08 | 0.093 | 704.21 | 1.47 | 1.65 | 3.4 | 21.35 | 7.5 | 0.02 | 0.009 |
| 573.15 | 0.87 | 0.093 | 639.11 | 0.47 | 0.954 | 17 | 15.27 | 3.2 | 0.03 | 0.007 |
| Squalane + CO$_2$ ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 298.15 | 0.50 | 0.102 | 803.12 | 1.10 | 18.7 | 24 | 28.64 | 14.3 | 0.22 | 0.031 |
| 323.15 | 0.26 | 0.102 | 785.63 | 0.22 | 7.87 | 14 | 25.60 | 11.2 | 0.21 | 0.088 |
| 348.15 | 0.67 | 0.102 | 768.81 | 0.18 | 4.16 | 8.0 | 24.56 | 3.0 | 0.21 | 0.039 |
| 373.15 | 0.86 | 0.102 | 752.05 | 0.65 | 2.94 | 24 | 21.85 | 4.0 | 0.19 | 0.059 |
### Table 1 (continued)

| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ $^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|---------|----------------|------------------|------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 423.15  | 1.00           | 0.102            | 718.49           | 0.61            | 1.56            | 6.8             | 18.79           | 2.2             | 0.15            | 0.018           |
| 473.15  | 1.29           | 0.102            | 684.72           | 0.24            | 0.930           | 25              | 15.94           | 6.0             | 0.13            | 0.024           |
| 573.15  | 1.60           | 0.102            | 612.46           | 0.84            | 0.412           | 16              | 10.21           | 3.7             | 0.09            | 0.011           |
| Squalane + CO$_2$ ($x_{\text{solute}} \approx 0.20$) |
| 298.15  | 1.00           | 0.201            | 805.10           | 0.16            | 14.4            | 3.9             | 26.47           | 10.7            | 0.48            | 0.065           |
| 323.15  | 1.21           | 0.201            | 787.66           | 0.08            | 6.54            | 18              | 24.46           | 12.3            | 0.47            | 0.055           |
| 348.15  | 1.54           | 0.201            | 770.53           | 0.29            | 3.90            | 15              | 22.35           | 5.2             | 0.43            | 0.060           |
| 373.15  | 1.57           | 0.201            | 753.26           | 0.47            | 2.76            | 31              | 21.10           | 4.2             | 0.40            | 0.071           |
| n-Octacosane + He ($p \approx 7.0$ MPa) |
| 423.15  | 7.03           | 0.049            | 725.73           | 0.69            | 2.07            | 28              | 22.90           | 2.9             | 0.02            | 0.006           |
| 473.15  | 7.03           | 0.065            | 693.07           | 0.30            | 1.47            | 17              | 19.77           | 5.4             | 0.01            | 0.012           |
| 573.15  | 7.04           | 0.108            | 624.82           | 0.82            | 0.555           | 16              | 12.88           | 3.7             | 0.00            | 0.011           |
| n-Octacosane + N$_2$ ($p \approx 7.0$ MPa) |
| 423.15  | 7.44           | 0.161            | 722.07           | 0.77            | 1.76            | 30              | 19.67           | 2.0             | 0.43            | 0.031           |
| 473.15  | 7.21           | 0.185            | 687.83           | 0.59            | 1.21            | 14              | 17.12           | 4.9             | 0.32            | 0.039           |
| 573.15  | 7.17           | 0.237            | 615.30           | 0.74            | 0.524           | 16              | 11.63           | 5.3             | 0.19            | 0.014           |
| 1-Hexadecanol + He ($p \approx 7.0$ MPa) |
| 348.15  | 7.12           | 0.010            | 794.22           | 0.38            | 4.74            | 28              | 25.76           | 5.0             | 0.03            | 0.026           |
| 373.15  | 7.08           | 0.010            | 775.44           | 0.30            | 2.42            | 20              | 23.51           | 4.4             | 0.03            | 0.024           |
| 423.15  | 7.25           | 0.034            | 736.31           | 0.22            | 1.03            | 4.2             | 19.77           | 3.4             | 0.01            | 0.009           |
| 473.15  | 4.63           | 0.081            | 690.31           | 0.35            | 0.530           | 12              | 16.46           | 1.9             | 0.01            | 0.010           |
| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$$^b$ | $\rho_{\text{L}}$ (kg·m$^{-3}$) | $10^3 U (\rho_{\text{L}})$ | $\eta_{\text{L}}$ (mPa·s) | $100 U (\eta_{\text{L}})$ | $\sigma$ (mN·m$^{-1}$) | $100 U (\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|--------|--------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 573.15 | 5.75         | 0.213           | 582.21          | 0.40            | 0.201           | 13              | 9.34            | 1.3             | 0.01            | 0.011           |
| 1-Hexadecanol + N$_2$ ($p \approx 7.0$ MPa) |
| 348.15 | 7.54         | 0.056           | 792.53          | 0.82            | 3.89            | 15              | 21.46           | 5.2             | 0.69            | 0.047           |
| 373.15 | 7.25         | 0.056           | 773.30          | 0.46            | 2.14            | 25              | 19.92           | 0.5             | 0.57            | 0.044           |
| 423.15 | 7.13         | 0.071           | 734.30          | 0.54            | 0.974           | 15              | 17.19           | 4.9             | 0.42            | 0.067           |
| 473.15 | 4.78         | 0.095           | 690.69          | 0.60            | 0.511           | 4.4             | 14.03           | 3.1             | 0.29            | 0.011           |
| 573.15 | 5.55         | 0.162           | 597.90          | 1.72            | 0.217           | 5.6             | 7.70            | 2.9             | 0.18            | 0.018           |
| 1-Hexadecanol + CO$_2$ ($x_{\text{solute}} \approx 0.10$) |
| 348.15 | 1.11         | 0.100           | 792.18          | 0.49            | 3.74            | 25              | 23.75           | 3.9             | 0.40            | 0.063           |
| 373.15 | 1.52         | 0.100           | 772.58          | 0.52            | 1.82            | 12              | 21.93           | 1.9             | 0.35            | 0.060           |
| 423.15 | 1.70         | 0.100           | 733.01          | 0.18            | 0.884           | 5.1             | 18.56           | 3.4             | 0.25            | 0.070           |
| 473.15 | 2.21         | 0.100           | 693.51          | 0.43            | 0.531           | 13              | 14.98           | 3.7             | 0.21            | 0.047           |
| 573.15 | 2.62         | 0.100           | 607.42          | 0.28            | 0.230           | 4.6             | 8.30            | 2.7             | 0.15            | 0.019           |
| 1-Hexadecanol + CO$_2$ ($x_{\text{solute}} \approx 0.20$) |
| 348.15 | 2.86         | 0.201           | 795.22          | 0.38            | 2.90            | 13              | 21.12           | 7.5             | 0.79            | 0.193           |
| 373.15 | 2.95         | 0.201           | 774.97          | 0.71            | 1.76            | 27              | 20.35           | 3.8             | 0.71            | 0.216           |
| 423.15 | 3.73         | 0.201           | 734.91          | 0.25            | 0.838           | 14              | 16.66           | 5.6             | 0.56            | 0.084           |
| 473.15 | 4.26         | 0.201           | 694.34          | 0.41            | 0.477           | 13              | 13.17           | 0.5             | 0.45            | 0.043           |
| 573.15 | 5.10         | 0.201           | 606.68          | 0.59            | 0.211           | 3.8             | 6.94            | 2.5             | 0.28            | 0.052           |
| HMN + N$_2$ ($p \approx 7.0$ MPa) |
| 298.15 | 4.38         | 0.091           | 786.32          | 0.64            | 3.45            | 27              | 20.01           | 0.8             | 1.06            | 0.086           |
| 323.15 | 4.64         | 0.095           | 769.45          | 0.98            | 2.07            | 23              | 18.93           | 2.7             | 0.83            | 0.013           |
| 348.15 | 4.51         | 0.095           | 752.58          | 0.27            | 1.39            | 21              | 18.09           | 1.9             | 0.67            | 0.011           |
| 373.15 | 8.45         | 0.095           | 739.90          | 2.49            | 1.16            | 1.3             | 16.79           | 2.5             | 0.56            | 0.025           |
Table 1 (continued)

| T (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|-------|----------------|------------------------|-------------------------|---------------------|------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 423.15 | 7.54 | 0.095 | 705.87 | 0.24 | 0.657 | 9.3 | 14.00 | 4.8 | 0.40 | 0.023 |
| 473.15 | 7.40 | 0.125 | 668.28 | 0.22 | 0.411 | 3.4 | 11.17 | 5.1 | 0.30 | 0.018 |
| 573.15 | 7.23 | 0.137 | 589.28 | 0.57 | 0.210 | 9.8 | 6.03 | 4.7 | 0.17 | 0.033 |

HMN + CO$_2$ ($x_{\text{solute}} \approx 0.10$)

| T (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|-------|----------------|------------------------|-------------------------|---------------------|------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 298.15 | 0.85 | 0.100 | 790.12 | 0.32 | 3.31 | 2.4 | 24.05 | 2.8 | 0.33 | 0.106 |
| 323.15 | 1.05 | 0.100 | 773.01 | 0.35 | 1.93 | 8.6 | 21.89 | 3.2 | 0.33 | 0.014 |
| 348.15 | 1.32 | 0.100 | 756.15 | 0.59 | 1.36 | 8.0 | 20.21 | 5.4 | 0.32 | 0.059 |
| 373.15 | 1.42 | 0.100 | 739.05 | 0.63 | 1.05 | 9.4 | 18.57 | 4.9 | 0.33 | 0.089 |
| 423.15 | 1.74 | 0.100 | 704.43 | 0.31 | 0.629 | 4.1 | 15.66 | 3.3 | 0.24 | 0.083 |
| 473.15 | 2.01 | 0.100 | 668.40 | 0.46 | 0.402 | 4.0 | 12.53 | 4.1 | 0.20 | 0.024 |
| 573.15 | 2.45 | 0.100 | 586.11 | 0.94 | 0.203 | 2.5 | 6.68 | 4.1 | 0.13 | 0.035 |

HMN + CO$_2$ ($x_{\text{solute}} \approx 0.20$)

| T (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|-------|----------------|------------------------|-------------------------|---------------------|------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 298.15 | 1.69 | 0.201 | 793.55 | 0.48 | 2.50 | 20 | 23.17 | 3.5 | 0.77 | 0.055 |
| 323.15 | 2.21 | 0.201 | 775.94 | 0.43 | 1.77 | 21 | 20.52 | 2.2 | 0.72 | 0.165 |
| 348.15 | 2.58 | 0.201 | 758.49 | 0.35 | 1.15 | 6.4 | 18.98 | 5.9 | 0.72 | 0.155 |
| 373.15 | 2.90 | 0.201 | 740.95 | 0.18 | 0.877 | 2.7 | 16.78 | 0.8 | 0.62 | 0.099 |
| 423.15 | 3.45 | 0.201 | 705.12 | 0.36 | 0.550 | 9.2 | 14.07 | 5.3 | 0.50 | 0.024 |
| 473.15 | 3.94 | 0.201 | 667.71 | 0.26 | 0.379 | 4.5 | 11.04 | 4.7 | 0.39 | 0.009 |
| 573.15 | 4.60 | 0.201 | 581.80 | 1.06 | 0.180 | 1.3 | 5.47 | 0.2 | 0.21 | 0.031 |

2-Butyl-1-octanol + N$_2$ ($p \approx 7.0$ MPa)

| T (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}^b$ | $\rho_L$ (kg·m$^{-3}$) | $10^3 U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100 U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100 U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|-------|----------------|------------------------|-------------------------|---------------------|------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 298.15 | 3.07 | 0.073 | 820.13 | 0.71 | 14.0 | 5.2 | 21.72 | 13 | 1.08 | 0.069 |
| 323.15 | 8.80 | 0.061 | 805.10 | 1.41 | 5.75 | 7.8 | 21.99 | 10 | 0.84 | 0.123 |
| 348.15 | 7.65 | 0.055 | 784.72 | 0.21 | 2.46 | 9.0 | 20.35 | 6.1 | 0.68 | 0.058 |
| 373.15 | 7.42 | 0.056 | 764.20 | 0.67 | 1.31 | 7.0 | 18.19 | 5.7 | 0.57 | 0.055 |
| $T$ (K) | $p$ (MPa)$^a$ | $x_{\text{solute}}$ | $\rho_L$ (kg·m$^{-3}$) | $10^3U_r(\rho_L)$ | $\eta_L$ (mPa·s) | $100U_r(\eta_L)$ | $\sigma$ (mN·m$^{-1}$) | $100U_r(\sigma)$ | $\Gamma_2^{(1)}$ (nm$^{-2}$) | $U(\Gamma_2^{(1)})$ (nm$^{-2}$) |
|--------|-------------|-----------------|-----------------|----------------|-------------|----------------|-----------------|----------------|----------------|----------------|
| 423.15 | 7.11        | 0.070           | 721.54          | 0.22           | 0.631       | 3.8           | 15.21           | 3.3            | 0.41           | 0.066          |
| 473.15 | 7.29        | 0.095           | 676.67          | 0.48           | 0.363       | 6.8           | 11.97           | 1.5            | 0.30           | 0.010          |
| 573.15 | 7.20        | 0.162           | 566.70          | 1.79           | 0.144       | 4.2           | 5.17            | 4.3            | 0.17           | 0.010          |
| 2-Butyl-1-octanol + CO$_2$ ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 298.15 | 0.74        | 0.101           | 826.26          | 0.43           | 12.1        | 29            | 25.79           | 6.9            | 0.52           | 0.058          |
| 323.15 | 1.37        | 0.101           | 806.26          | 0.24           | 4.42        | 30            | 24.08           | 1.1            | 0.44           | 0.162          |
| 348.15 | 1.46        | 0.101           | 785.51          | 0.20           | 1.97        | 7.1           | 22.40           | 2.7            | 0.37           | 0.021          |
| 373.15 | 1.71        | 0.101           | 764.35          | 0.68           | 1.18        | 4.4           | 20.01           | 1.1            | 0.41           | 0.038          |
| 423.15 | 1.94        | 0.101           | 721.23          | 0.69           | 0.532       | 3.9           | 16.57           | 3.7            | 0.30           | 0.032          |
| 473.15 | 2.18        | 0.101           | 676.63          | 0.11           | 0.340       | 4.1           | 12.61           | 6.2            | 0.23           | 0.058          |
| 573.15 | 2.61        | 0.101           | 574.30          | 0.88           | 0.155       | 9.2           | 5.77            | 6.7            | 0.13           | 0.006          |
| 2-Butyl-1-octanol + CO$_2$ ($x_{\text{solute}} \approx 0.10$) | | | | | | | | | | |
| 298.15 | 1.64        | 0.201           | 830.76          | 1.04           | 9.10        | 25            | 24.34           | 2.7            | 1.04           | 0.128          |
| 323.15 | 2.35        | 0.201           | 809.98          | 0.46           | 3.45        | 30            | 22.32           | 3.0            | 0.93           | 0.070          |
| 348.15 | 2.84        | 0.201           | 788.70          | 0.41           | 1.71        | 12            | 20.48           | 4.7            | 0.83           | 0.158          |
| 373.15 | 2.97        | 0.201           | 766.77          | 0.66           | 1.08        | 9.8           | 18.07           | 2.4            | 0.73           | 0.077          |
| 423.15 | 3.66        | 0.201           | 722.46          | 0.22           | 0.506       | 1.5           | 14.77           | 4.1            | 0.59           | 0.093          |
| 473.15 | 4.07        | 0.201           | 676.24          | 0.79           | 0.306       | 4.5           | 11.19           | 1.3            | 0.44           | 0.051          |
| 573.15 | 5.09        | 0.201           | 571.08          | 0.34           | 0.142       | 6.5           | 4.32            | 3.4            | 0.25           | 0.029          |

Expanded statistical relative or absolute uncertainties ($k = 2$) are included for all simulated thermophysical properties

$^a$The specified pressures are obtained from the simulation in the compressed liquid phase in a canonical ensemble

$^b$x$_{\text{solute}}$ refers to the gas mole fraction in simulations in the compressed liquid phase. In the simulations in VLE x$_{\text{solute}}$ can vary slightly due to surface enrichment
investigated within the same project [15–18] and further experimental results from the literature, when available. For each property, first, the influence of the solute is discussed using the results for binary mixtures consisting of the gases H₂, He, CH₄, H₂O, CO, N₂, or CO₂ dissolved in n-dodecane, n-tetracontane, or 1-dodecanol. Here, the different molecular characteristics of the solutes, e.g., molecule size, weight, or polarity, allow a systematic investigation of the influence of the solute on the properties of the mixture. To study the influence of the molecular characteristics of the solvents on the properties of the mixtures, binary mixtures based on the solutes He, N₂, or CO₂ dissolved in solvents with a variation in the chain length, hydroxylation, or branching are discussed. Finally, results for \( \Gamma_2^{(1)} \) are shown for some of the investigated binary mixtures and the influence of the surface enrichment on \( \sigma \) is discussed.

### 3.1 Liquid Viscosity

The results for \( \eta_L \) of binary mixtures consisting of H₂, He, N₂, CH₄, H₂O, CO, or CO₂ dissolved in n-dodecane (left panel), 1-dodecanol (middle panel), or n-tetracontane (right panel) are shown in Fig. 2 as a function of \( T \). Open and closed symbols are used to indicate different \( x_{\text{solute}} \) for systems with dissolved CH₄, H₂O, CO,

![Fig. 2](image-url)  

**Fig. 2** (Top) Liquid dynamic viscosity \( \eta_L \) of the binary mixtures of n-dodecane (left), 1-dodecanol (middle), or n-tetracontane (right) with the dissolved gases H₂, He, N₂, CH₄, H₂O, CO, or CO₂ by EMD simulations as a function of \( T \) and solute mole fraction (open and closed symbols). For comparison, the correlations for \( \eta_L \) of the solvents from EMD simulations [15] are shown (solid lines). (Bottom) Relative deviations between \( \eta_L \) of the binary mixtures and \( \eta_L \) of the pure solvents. The dotted lines mark the average expanded uncertainty (\( k = 2 \)) of the results for the pure solvents. Error bars are shown only exemplarily for the mixtures containing He for clarity. The mole fractions of the dissolved gas given in the legend are approximate values. The real composition at each \( T \) can be taken from Table 1.
and CO₂. For comparison, ηₜ of the pure solvents investigated by EMD simulations [15] are shown as solid lines. To study the influence of the dissolved gas on ηₜ, the deviations of ηₜ for the binary mixtures from the one of the pure solvents is given in the lower part of Fig. 2. Exemplary error bars are shown only for the He-based mixtures for legibility. For all the systems investigated within this section, the average expanded statistical uncertainty (k=2) of ηₜ is 12 %.

Due to the relatively large uncertainties for ηₜ in the range of (10 to 20) % (k=2), ηₜ of the binary mixtures shown in Fig. 2 are mostly within combined uncertainty with ηₜ of the pure solvents. However, even within combined uncertainties, the T-dependent trends of the deviations between the binary mixtures and the pure solvents, which represents the influence of the dissolved gas, can be compared to previous studies for ηₜ of binary mixtures based on the same solutes and similar solvents as shown here. For mixtures based on n-dodecane, which is shown in the left panel of Fig. 2, the results can be compared to the work of Klein et al. [18], who investigated binary mixtures of the same solutes dissolved in n-hexadecane over the same T range and similar compositions. For the solutes He and H₂ the EMD simulations in this work predict ηₜ of the mixtures to be larger than that of pure n-dodecane by up to (13 and 10) % for He and H₂. This agrees with the results from SLS for the binary mixtures based on n-hexadecane, which also show ηₜ of the mixtures to be within combined uncertainties at T<423 K and larger in comparison to pure n-hexadecane by up to (13 and 7) % for He and H₂ [18]. The results from this work also agree with the findings for the n-hexadecane-based mixtures from EMD simulations by Klein et al. [18], who also found ηₜ of the He- or H₂-based mixtures to be, on average, larger by approximately 10 % in comparison to pure n-hexadecane. The increase in ηₜ for mixtures containing He or H₂ with respect to the pure solvents can be explained by the effect of the increased p necessary to dissolve the solutes, while the pure solvents are investigated under saturation conditions or at p=0.1 MPa. The increase of p from saturation conditions to 7.0 MPa, for example, would lead to an increase in ηₜ of (9.1 or 30) % for T=(298.15 or 573.15) K [31]. For the smallest gases He and H₂, this p effect seems to be larger than the effect of the dissolved gases, which usually leads to a decrease in ηₜ, so that ηₜ for these mixtures is larger than that of pure n-dodecane.

For N₂, the EMD simulations from this work also predict ηₜ to be, on average, larger than that of pure n-dodecane. This agrees with the findings from SLS experiments for the binary mixture of n-hexadecane+N₂ of Klein et al. [18] only for T>473 K. At smaller T, the results from SLS show ηₜ of the mixtures to be smaller than that of pure n-hexadecane. The failure of the EMD simulations to predict a smaller ηₜ for the N₂-based mixtures in comparison to the pure solvent was already seen by Klein et al. [18] for the n-hexadecane-based mixture. This suggest that this failure can be explained by the inability of the applied L-OPLS FF [13, 14] to accurately predict the properties of the pure solvents at low T. This was already seen and discussed in our previous publications as well as by other authors [15, 16, 18, 19]. The L-OPLS FF seems to overpredict the interstitial space between the solvent molecules, which is supported by a too small ρₜ [15, 19], and overpredict the attractive intermolecular interactions, which is expressed by a too large ηₜ [15, 19]. With respect to the binary mixtures with dissolved N₂, this means that in the EMD
simulations, the small N₂ molecules can occupy the overmucly interstitial space, which leads to a larger \( \eta_L \) in comparison to the pure solvent.

For the binary mixtures of \( n \)-dodecane and H₂O, the EMD simulations from this work predict \( \eta_L \) of the mixture to be within combined uncertainties with the pure solvent with an average absolute deviation of about 4 %. Based on the two investigated \( x_{\text{solute}} = (0.045 \) and 0.090), no clear influence of \( x_{\text{solute}} \) on \( \eta_L \) can be found. This agrees with the finding from SLS and EMD simulations for the binary mixtures of \( n \)-hexadecane and H₂O of Klein et al. [18], which also found no clear influence of dissolved H₂O on \( \eta_L \) of the solvent over the entire range of \( T \) and \( x_{\text{solute}} \).

For the three solutes CH₄, CO, and CO₂, which were investigated over a wider range of \( x_{\text{solute}} \) due to their better solubility in the solvents in this work, also the strongest influence on \( \eta_L \) could be observed. For all three solutes, a smaller \( \eta_L \) of the mixtures in comparison to pure \( n \)-dodecane can be observed. This reduction by dissolving the solutes is most pronounced at the two lowest investigated \( T \) with deviations of about −25 % for CH₄ and CO₂ and −18 % for CO with respect to pure \( n \)-dodecane for the largest investigated \( x_{\text{solute}} \). With increasing \( T \) this reduction decreases so that the deviations at \( T = 573.15 \) K are −(14, 9.4, and 0.30) % for CH₄, CO₂, and CO, respectively. Both the \( T \)-dependent trend and the absolute deviations of the mixtures from the pure solvent agree well with the findings for \( n \)-hexadecane-based mixtures from SLS and EMD simulations from Klein et al. [18].

The investigation of binary mixtures consisting of the same solutes dissolved in 1-dodecanol allows an analysis of the influence of the solvent characteristics on \( \eta_L \) of the mixtures. Also here, the influence of the dissolved gases is investigated by calculating the deviation between \( \eta_L \) of the mixtures and that of pure 1-dodecanol was previously investigated by EMD simulations [15]. The absolute values for \( \eta_L \) for the eleven binary mixtures based on 1-dodecanol as well as the deviations from the pure solvent are shown in the middle panel in Fig. 2. For the three low-solubility gases H₂, He, and N₂, \( \eta_L \) of the binary mixtures agree within combined uncertainties with \( \eta_L \) of pure 1-dodecanol. This in in agreement with the findings for the mixtures
based on n-dodecane which suggests that the polar character of the alcohol molecule does not influence the interactions between solute and solvent molecules. The results for the mixture containing H$_2$ can further be compared to the results from Klein et al. [17], which investigated the binary mixture consisting of 1-hexadecanol and H$_2$ using SLS and EMD simulations. They also found $\eta_L$ of these mixtures to be within combined uncertainties of the pure solvent. The $\eta_L$ of the two binary mixtures containing H$_2$O are within 5% of pure 1-dodecanol. This is, however, contradictory to the findings for mixtures consisting of H$_2$O dissolved in 1-hexadecanol of Klein et al. [17], where the results from SLS have shown an approximately 40% larger $\eta_L$ for the mixtures in comparison to pure 1-hexadecanol at $T=573.15$ K. The EMD simulations presented together with the SLS results, however, were not able to predict larger $\eta_L$ for the mixtures [15], which agrees with the present work. Here it seems, that the combination of the modified L-OPLS FF [13, 14] and the SPC/E FF for water [38], which does not consider a $T$-dependent change of the FF parameters, fails to accurately predict the influence of the strong hydrogen bonds on $\eta_L$ at high $T$.

For the three gases with a larger solubility, namely CH$_4$, CO, and CO$_2$, the $\eta_L$ reduction in comparison to the pure solvent was found to be more pronounced in the 1-dodecanol than in n-dodecane at low $T$. The maximum deviation from the pure solvent is $- (44, 28, \text{and } 44)$% in 1-dodecanol and $- (29, 21, \text{and } 42)$% in n-dodecane for dissolved CH$_4$, CO, and CO$_2$, respectively. Since also the reduction of $\eta_L$ for the binary mixtures containing the non-polar CH$_4$ is more pronounced in the alcohol than the alkane, the difference is most likely not related to the polarity of the solvent but rather to the much larger $\eta_L$ of pure 1-dodecanol in comparison to n-dodecane. This means that dissolving a low-viscous solute leads to a stronger reduction in the mixture $\eta_L$ when the solvent $\eta_L$ is much larger at comparable compositions.

Finally, binary mixtures consisting of the same solutes, with the exception of CO$_2$, dissolved in n-tetracontane are studied, which allows the investigation of the influence of the solvent carbon chain length on $\eta_L$ of the mixtures. Due to the large melting point of n-tetracontane [15, 39], simulations could be performed only at $T>423.15$ K. Results for $\eta_L$ of the binary mixtures and pure n-tetracontane [15] as well as the deviations between $\eta_L$ of the binary mixture and the pure solvent are shown in the right panel of Fig. 2. Since large reductions in $\eta_L$ due to a dissolved gas are usually more pronounced at $T$ between (298.15 and 373.15) K and because of the larger statistical uncertainties in determining $\eta_L$ for large molecules, all $\eta_L$ results for the binary mixture are within combined uncertainties of the pure solvents. This suggests that the influence of the carbon chain length of the solvent is rather weak. This is supported also by experimental SLS investigations of the same solutes as investigated in this work dissolved in either n-hexadecane [18] or n-octacosane [17], which showed almost identical deviations between $\eta_L$ of the binary mixtures and the pure solvents in both cases.

To further study the influence of the molecular characteristics of the solvent on the thermophysical properties of the mixtures, binary systems consisting of either He, N$_2$, or CO$_2$ dissolved in various solvents were investigated by EMD simulations. The results for $\eta_L$ for the binary mixtures as well as the relative deviation from $\eta_L$ of the pure solvents [15] are shown in Fig. 3 for the solutes He (left panel), N$_2$ (middle panel), and CO$_2$ (right panel).
In the case of He as the solute, $\eta_L$ of the mixtures scatter around that of the pure solvents over the entire $T$ range for most solvents. The only exception is the mixture based on 1-hexadecanol, where a (16 to 33) % smaller $\eta_L$ of the mixture in comparison to pure 1-hexadecanol can be found over the entire $T$ range. Here, it seems that the He molecules are interfering with the intermolecular interactions of 1-hexadecanol more strongly than with other solvents. This is also observed when comparing mixtures of $N_2$ or $CO_2$ dissolved in different solvents. The largest reduction in $\eta_L$ can be observed for the 1-hexadecanol-based mixtures. Therefore, it seems that a failure of the solute FF can be ruled out.

Results for $\eta_L$ of further binary mixtures based on $N_2$ as the solute show no systematic deviations from the pure solvents, which agrees with the results from EMD simulations for the binary mixture of $n$-hexadecane and $N_2$ from our previous investigation [18]. Here, one should keep in mind, however, that SLS results from the same publication [18] have shown that at $T<373.15$ K a reduction in $\eta_L$ in comparison to pure $n$-hexadecane could be observed, as discussed in the previous section.

Finally, investigations of binary mixtures with $CO_2$ as the solute and $x_{\text{solute}}=0.20$, which are shown in the right panel of Fig. 3, show a typical $T$-dependent trend. There is first a strong reduction of $\eta_L$ caused by adding $CO_2$ at low $T$, followed by a convergence to $\eta_L$ to that of the pure solvent with increasing $T$. This behavior could already be seen for $CO_2$ dissolved in various solvents by SLS and EMD simulations [17, 18]. While the $T$-dependent trend is very similar for the different solvents, the magnitudes of the relative deviations change significantly with the molecular
characteristics. In general, dissolved CO$_2$ has a stronger impact on $\eta_L$ of alcohol-based mixtures. This can be explained by the presence of strong hydrogen bond, which are partly replaced by interactions with the polar CO$_2$ molecules. The dispersive van-der-Waals interactions, which are present in both alkanes and alcohols, are weaker in comparison, which explains the smaller impact of dissolved CO$_2$ on $\eta_L$ of alkane-based mixtures. The comparison between the linear 1-dodecanol and the branched 2-butyl-1-octanol as the solvent shows a stronger reduction for the linear alcohol. An explanation for this behavior can be found in the steric effect of the side branch in 2-butyl-1-octanol, which allows CO$_2$ molecules to occupy the free volume created by the steric effect without interfering with the intermolecular interactions between the solvent molecules.

### 3.2 Interfacial Tension

The interfacial tension $\sigma$ was calculated using EMD simulations for the same binary mixtures and the same thermodynamic states as for $\eta_L$. In the following, the influence of the molecular characteristics of the solvent and solute on $\sigma$ is investigated by comparing the results for the binary mixture with $\sigma$ of the pure solvents also investigated using EMD simulations and published in a previous publication [15].

The results for $\sigma$ of binary mixtures consisting of H$_2$, He, N$_2$, CH$_4$, H$_2$O, CO, or CO$_2$ dissolved in $n$-dodecane (left panel), 1-dodecanol (middle panel), or $n$-tetracontane (right panel) are shown in Fig. 4 as function of $T$. Open and closed symbols are used to indicate different $x_{\text{solute}}$ for systems with dissolved CH$_4$, H$_2$O, CO, and CO$_2$. For comparison, $\sigma$ of the pure solvents investigated by EMD simulations [15] are shown as solid lines. To study the influence of the dissolved gases on $\sigma$, the deviations of $\sigma$ for the binary mixtures from that of the pure solvents is given in the lower part of Fig. 4. Exemplary error bars are shown only for the He-based mixtures for legibility. The results in Fig. 4 show that due to the much smaller statistical uncertainty ($k=2$) of approximately 2.5 $\%$, the influence of the solute on $\sigma$ of the binary mixtures can be better resolved than was possible for $\eta_L$.

In the case of the solvent $n$-dodecane, the results from this work can be compared to the binary mixtures based on the same solutes and the solvent $n$-hexadecane investigated by SLS and EMD simulations [18]. For the two solutes He and H$_2$O, $\sigma$ of the binary mixtures are within combined uncertainty of pure $n$-dodecane over the entire $T$ range. This agrees well with the findings for the same solutes dissolved in $n$-hexadecane by SLS and EMD simulations [18]. For the solute H$_2$, a smaller $\sigma$ for the mixtures in comparison to pure $n$-dodecane with deviations up to 10 $\%$ were found in this work. Since both SLS and EMD simulations have shown agreement between $\sigma$ for binary mixtures of H$_2$ dissolved in $n$-hexadecane and that of pure $n$-hexadecane, it seems that H$_2$ has a stronger influence on $\sigma$ for shorter $n$-alkanes. A possible explanation for this, is the smaller difference between the liquid and vapor densities for shorter alkanes. Since the density difference is often used to correlate the interfacial tension [40], this can explain why dissolved H$_2$ seems to have a stronger influence on $\sigma$ in mixtures based on shorter alkanes.
For the more soluble gases, a clear reduction in $\sigma$ is observed from the addition of the dissolved gas. This reduction grows stronger with increasing $x_{\text{solute}}$. The magnitude and $T$-dependent trend of the deviations of $\sigma$ for the mixtures from pure $n$-dodecane are similar to those observed in $n$-hexadecane [18]. At $T<473$ K, the deviation between $\sigma$ of the binary mixtures and pure $n$-hexadecane are within 27 % and show only a small $T$-dependent trend. At $T=573.15$ K, this deviation increases for most mixtures and reaches a maximum of approximately 44 % in the case of the binary mixture with CO$_2$ and $x_{\text{solute}}=0.20$. An explanation for the larger deviations at the highest investigated $T$ is the small absolute value for $\sigma$ at this $T$, which leads to an increasing relative deviation in comparison to mixtures based on $n$-hexadecane, which has a larger $\sigma$ [15, 41].

The relative deviations between the binary mixtures based on 1-dodecanol with the same dissolved gases as investigated in $n$-dodecane are depicted in the middle panel of Fig. 4 and show a very similar $T$-dependent trend as the $n$-dodecane-based mixtures. The largest difference can be found at $T=573.15$ K, where the deviations in the 1-dodecanol-based mixtures are in general smaller in comparison to the $n$-dodecane-based mixtures. This can again be explained with the larger absolute value for $\sigma$ of 1-dodecanol than $n$-dodecane. The comparison
between \(n\)-dodecane and 1-dodecanol as the solvents shows that the terminal hydroxyl group seems to have only a very small influence on \(\sigma\) in binary mixtures with dissolved gases. This can be explained by the tendency of long primary alcohols to arrange themselves in such a way at the interface that the hydroxyl group is oriented into the liquid bulk phase and the alkane tail is oriented towards the interface [15, 16, 42].

The results for \(\sigma\) of binary mixtures consisting of \(n\)-tetracontane and the same solutes, with the exception of \(\text{CO}_2\), are shown in the right panel of Fig. 4. For the solutes \(\text{He, H}_2\), and \(\text{H}_2\text{O}\), agreement within combined uncertainties with \(\sigma\) of pure \(n\)-tetracontane can be found, which agrees with the observations done for the same solutes dissolved in \(n\)-dodecane, \(n\)-hexadecane [18], or \(n\)-octacosane [17]. For the mixtures containing \(\text{CH}_4\), \(\text{N}_2\), or \(\text{CO}\), however, the reduction in \(\sigma\) is, in comparison to the pure solvent, much less pronounced in the \(n\)-tetracontane-based mixtures than in other solvents. A possible explanation for this is the molecule size or shape of \(n\)-tetracontane. The linear character with a very long carbon chain containing 40 carbon atoms can lead to bending and entanglement between the solvent molecules, which causes free volume within the fluid structure. This free volume can be occupied by the solute molecules and leads to a depletion of solute molecules at the vapor–liquid interface. Therefore, only a small influence of the solute on \(\sigma\) of the binary mixture can be observed. The larger influence of \(\text{N}_2\) in comparison to \(\text{CO}\) or \(\text{CH}_4\) can again be explained by the larger vapor density in the case of \(\text{N}_2\) due to the larger \(p\).

As it was done for \(\eta_L\), the influence of the molecular characteristics of the solvent on the \(\sigma\) of the mixtures is addressed with the binary systems consisting of either \(\text{He, N}_2\), or \(\text{CO}_2\) dissolved in various solvents. The results for \(\sigma\) of the binary mixtures as well as the relative deviation from \(\sigma\) of the pure solvents are shown in Fig. 5 for the solutes \(\text{He}\) (left panel), \(\text{N}_2\) (middle panel), and \(\text{CO}_2\) (right panel). In the case of \(\text{He}\) as the solute, no clear influence of the dissolved \(\text{He}\) on \(\sigma\) of the binary mixtures can be found, with the exception of the mixture containing 1-hexadecanol as the solvent. Similar to what was shown for \(\eta_L\), \(\sigma\) of the binary mixture consisting of \(\text{He}\) dissolved in 1-hexadecanol is between (9 and 17) \% smaller than that of pure 1-hexadecanol. Since there are no experimental data available for mixtures consisting of \(\text{He}\) dissolved in a primary alcohol, this behavior cannot be verified.

For the binary mixtures containing \(\text{N}_2\) as the solvent, which are shown in the middle panel of Fig. 5, we can find a reduction in \(\sigma\) in comparison to the pure solvent between (10 and 31) \%. Up to \(T=473\ \text{K}\), no clear \(T\)-dependent trend for the relative deviation between \(\sigma\) of the binary mixtures and that of the pure solvent can be observed. At \(T=573.15\ \text{K}\), the relative deviation is in general larger for mixtures where \(\sigma\) of the solvent is small. The fact that the relative deviation between \(\sigma\) of the mixtures and that of the pure solvents are very similar for the different investigated solvents shows that the molecular characteristics of the solvent, e.g., chain length, branching, or hydroxylation, have only a small influence on \(\sigma\) in mixtures with dissolved \(\text{N}_2\). For mixtures based on the solute \(\text{CO}_2\), which are shown in the right panel of Fig. 5, however, a clear influence of the solvent characteristics on \(\sigma\) of the mixtures can be found. While the relative deviation between \(\sigma\) of the mixtures and that of the pure solvent are clearly visible for all investigated solvents, the
magnitude and $T$-dependent trend varies greatly. The influence of the dissolved CO$_2$ is less pronounced in the branched alkanes or alcohols, i.e., squalane, HMN, and 2-butyl-1-octanol, than in the linear ones. For the mixture based on squalane, for example, the deviation between $\sigma$ of the mixture and that of pure squalane are within 11%, while mixtures based on the linear isomers $n$-triacontane deviate by up to 20% from the pure solvent. The dissolved CO$_2$ also seems to have a smaller influence on the branched 2-butyl-1-octanol than on the linear 1-dodecanol at $T< 423.15$ K. The smaller influence in branched molecules can be explained by the free volume in the fluid structure caused by the steric effects of the side branches. As previously mentioned, this free volume can be occupied by the solute molecules, which leads to a smaller impact of the dissolved gases on $\sigma$ of the mixtures.

3.3 Structure–Property Relationship—Influence of Surface Enrichment on the Interfacial Tension

The relative adsorption of the solute (component 2) relative to the solvent (component 1) $\Gamma^{(1)}$ as a measurement for the enrichment of solutes molecules at the vapor–liquid interface can be used to investigate how the fluid structure close to the interface influences $\sigma$ [35, 43, 44]. In this work, $\Gamma^{(1)}$ is used to study why different solutes with comparable $x_{\text{solute}}$ in the bulk liquid phase have a different impact on $\sigma$. For this, the results for $\Gamma^{(1)}$ of binary mixtures consisting of H$_2$, n-dodecane, n-octacosane, n-triacontane, HMN, squalane, 1-dodecanol, 1-hexadecanol, and 2-butyl-1-octanol.
He, N₂, CH₄, H₂O, CO, or CO₂ dissolved in n-dodecane (left panel), 1-dodecanol (middle panel), or n-tetracontane (right panel) are shown in Fig. 6 as function of T. Open and closed symbols are used to indicate different solutes for systems with dissolved CH₄, H₂O, CO, and CO₂.

The results in Fig. 6 for all three solvents show a general decrease of \( \Gamma(1)_2 \) with increasing T. This can be explained by the increasing kinetic energy and dynamics of the molecules, which counteracts the enrichment of the solute molecules at the vapor–liquid interface by the mixing process. For all three investigated solvents, \( \Gamma(1)_2 \) for the mixtures containing H₂O and He are close to zero, indicating that there is no or only a very small enrichment of the solute molecules at the interface. This is not surprising, since also \( \sigma \) for these systems are mostly within combined uncertainties with that of the pure solvents. Also for the solute H₂, only small \( \Gamma(1)_2 \) within 0.5 nm⁻² can be found in all three solvents. This agrees well with the value found for the binary mixtures consisting of n-hexadecane with dissolved H₂, \( \Gamma(1)_2 = 0.54 \) nm⁻² was calculated for \( T = 323.15 \) K [18]. A large difference in \( \Gamma(1)_2 \) can be found, for example, when comparing the solutes CH₄, CO, and CO₂ dissolved in either n-dodecane or 1-dodecanol at \( x_{\text{solute}} = 0.10 \). Here, \( \Gamma(1)_2 \) of the mixtures containing CO is approximately 50 % larger than that of the other two solutes at \( T = 298.15 \) K, indicating a larger surface enrichment of CO. This observation can explain why \( \sigma \) of the binary mixture containing CO deviate more from \( \sigma \) of the pure solvent, as shown in Fig. 4. In the case of n-tetracontane, the largest \( \Gamma(1)_2 \) can be found for the mixture containing N₂, which agrees with the observations from the investigation of \( \sigma \), where the largest deviation between \( \sigma \) of the mixtures and that of pure n-tetracontane was found for the mixture containing N₂. Additionally, the smaller influence of dissolved CH₄ on \( \sigma \) in mixtures based on n-tetracontane in comparison to other linear alkanes can directly be linked to the smaller \( \Gamma(1)_2 \) in n-tetracontane-based mixtures.

**Fig. 6** Relative Adsorption of the solute (component 2) relative to the solvent (component 1) \( \Gamma(1)_2 \) of the binary mixtures of n-dodecane (left), 1-dodecanol (middle), or n-tetracontane (right) with the dissolved gases H₂, He, N₂, CH₄, H₂O, CO, or CO₂ by EMD simulations as a function of T and solute mole fraction (open and closed symbols). Error bars are shown only exemplarily for the mixtures containing CH₄ and \( x_{\text{solute}} = 0.20 \) for clarity. The mole fractions of the dissolved gas given in the legend are approximate values. The real composition at each T can be taken from Table 1.
In general, the results for $\Gamma^{(1)}_2$ shown in Fig. 6 and given in Table 1 show that the deviation between $\sigma$ of the binary mixtures from that of the pure solvents are inversely proportional to $\Gamma^{(1)}_2$, which shows that the enrichment of solute molecules at the vapor–liquid interface is an important property for understanding, correlating, and predicting $\sigma$ of mixtures consisting of liquids with dissolved gases [35, 43, 44].

4 Conclusions

In the present study, results for $\eta_L$, $\sigma$, and $\Gamma^{(1)}_2$ of 45 binary mixtures consisting of a linear, branched, or oxygenated hydrocarbon and a dissolved gas obtained by EMD simulations are presented. The binary mixtures consist of the solvents $n$-dodecane, $n$-octacosane, $n$-tetracontane, HNN, squalane, 1-dodecanol, 1-hexadecanol, or 2-butyl-1-octanol and the solutes H$_2$, He, CH$_4$, H$_2$O, N$_2$, CO, or CO$_2$ and were investigated in the $T$ range between (298.15 and 573.15) K and $x_{\text{solute}}$ up to 0.20. Using EMD simulations, $\eta_L$, $\sigma$, and $\Gamma^{(1)}_2$ of the binary mixtures could be accessed with average expanded statistical uncertainties ($k=2$) of (12, 3.7, and 12) %, respectively. By comparing the thermophysical properties of the binary mixtures with those of the pure solvents, the influence of the dissolved gas on $\eta_L$ and $\sigma$ could be analyzed. Due to the relatively large statistical uncertainties for $\eta_L$, such an influence could not be resolved within combined uncertainties of the binary mixture and the pure solvent data. For the more soluble gases CH$_4$, CO, or CO$_2$, however, an influence of the dissolved gas could be pointed out and the results were compared to those obtained by SLS and EMD simulations for binary mixtures with similar solvents and the same dissolved gases. Here, deviations of up to $-50$ % with respect to $\eta_L$ of the pure solvent could be observed for binary mixtures containing CH$_4$ or CO$_2$ at $T$ between (298.15 and 323.15) K. With increasing $T$, the difference between $\eta_L$ of the mixtures and that of the pure solvents reduced for all binary mixtures based on these solutes and was mostly within combined uncertainties at $T=573.15$ K. For the solute CO, a similar $T$-dependent trend for the difference between $\eta_L$ of the mixtures and that of the pure solvents was observed. However, the relative deviation was always within 27 % and, therefore, always smaller in comparison to the mixtures containing CH$_4$ or CO$_2$. An investigation of the solutes He, N$_2$, or CO$_2$ in different solvents has shown that the relative reduction of $\eta_L$ in comparison to the pure solvents is larger in mixtures containing linear or branched alcohols than in linear or branched alkanes. The observations with respect to the influence of the gas on $\eta_L$ agree well with experimental and simulated data for binary mixtures based on similar solvents from the literature.

In the case of $\sigma$, the relatively small statistical uncertainties allowed a more thorough analysis of the influence of the dissolved gases for all binary mixtures. While $\sigma$ of mixtures containing the solutes He or H$_2$O are mostly within uncertainties of the pure solvents, large deviations outside combined uncertainties were found for CH$_4$, CO, or CO$_2$ over the entire $T$ range. Here, the influence of the dissolved gas was found to strongly depend on the solute type. The binary mixture of CO dissolved in $n$-dodecane with $x_{\text{solute}}=0.10$, for example, deviates by $-27$ % from $\sigma$ of
pure \( n \)-dodecane at \( T = 298.15 \) K, while the mixtures with dissolved CO\(_2\) at the same \( T \) and \( x_{\text{solute}} \) deviates only by approximately \( –8\% \). With the help of \( \Gamma_2^{(1)} \), which could also be calculated from EMD simulations in VLE, the stronger impact of CO on \( \sigma \) could be related to its stronger enrichment at the vapor–liquid interface. Although absolute values for \( \eta_L \) and \( \sigma \) can differ by up to 30\%, the agreement between experimental and simulated results shows that EMD simulations are a valuable tool for the prediction of how a dissolved gas may influence mixture properties.

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**Declarations**

**Conflict of interest** The authors have no competing interests to declare that are relevant to the content of this article.

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