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# Graphical Abstract

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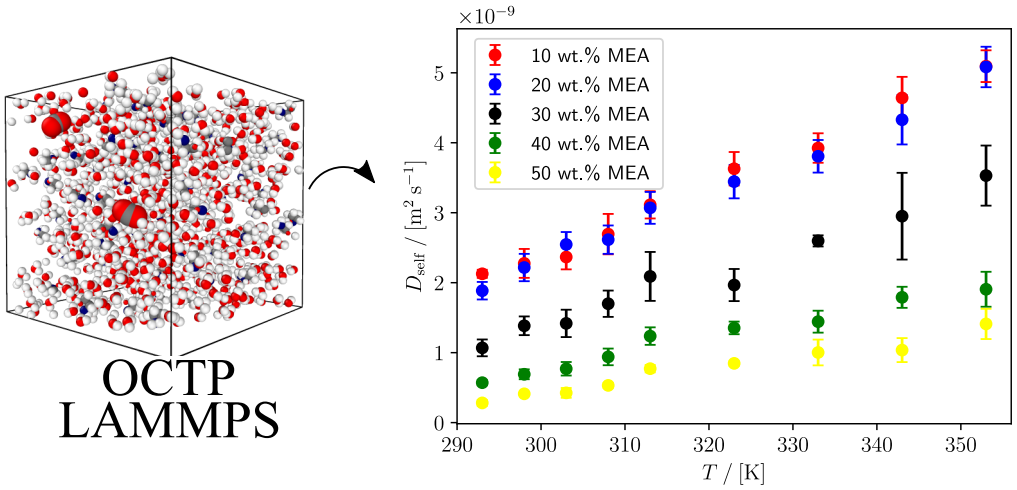

# Transport Properties of Mixtures of Acid Gases with Aqueous Monoethanolamine Solutions: A Molecular Dynamics Study

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**Abstract**

We investigated the effect of temperature and monoethanolamine (MEA) concentration on the self-diffusivity of acid gases, $CO_2$ , and $H_2S$ in aqueous MEA solutions. For this purpose, we computed densities of pure MEA and 30 wt.% MEA/water solutions while scaling the LJ energy ($\epsilon$) parameter and point charges of MEA. Results show that with a scaling factor of 0.80 applied to the point charges of MEA, computed densities agree well with the experimental ones from literature. This was tested by computing viscosities and the self-diffusivity of pure MEA and 30 wt.% MEA/water solutions and comparing these with experiments. We showed that the scaling factor of 0.80 also works well for predicting transport properties of MEA/water solutions. Finally, we computed self-diffusivities of infinitely diluted $CO_2$ and $H_2S$ for temperatures ranging from 293–353 K and MEA concentrations of 10–50 wt.%. Our results show that the self-diffusivity of both acid gases depends significantly on the temperature and MEA concentration in the solution. The results of this study will contribute to the development of more efficient acid gas treatment processes.

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*Keywords:* Molecular simulation, Carbon dioxide, Hydrogen sulfide, Transport properties, Diffusion

## 1. Introduction

Natural gas is the fossil fuel with the highest energy density per carbon atom [1]. $NO_x$ and particulate matter emissions from the process of natural gas burning are lower compared to other fossil fuels [2]. Natural gas will play a key role in hydrogen production [3]. These advantages make natural gas a promising candidate to replace liquid fossil fuels and coal, and to be a transition fuel until renewable energy sources are feasible on a large scale [4, 5]. It is well known that about 40% of the remaining natural gas sources have a $CO_2$ concentration higher than 2% and a $H_2S$ concentration higher than 100 ppm [6]. The acid gas concentration in natural gas needs to be reduced to <2% and <50 ppm of $CO_2$ for pipeline and liquefied natural gas (LNG) transport, respectively, and <4 ppm of $H_2S$ for both pipeline gas and LNG [6]. The removal of acid gases from natural gas streams can be achieved by several different processes such as adsorption-based separation [7], membrane separation [8], cryogenic distillation [9], direct conversion of $H_2S$ to elemental sulfur [10] and, absorption-based separation using liquid solvents. The latter option is widely preferred since it is a technically mature, and a reliable process, and it offers a low amount of absorbed hydrocarbons [11].

In absorption-based separation processes, $CO_2$ and $H_2S$ are removed by a liquid solvent, usually aqueous alkanolamines, by physical and/or chemical absorption [12, 13, 14, 15]. In this process, a natural gas stream flows through the absorption column at high pressure (20–100 bar) and mild temperature (313–353 K) and acid gases are absorbed into the liquid solvent [16]. After absorption, the liquid solvent is sent to the regeneration column where it is regenerated at high temperature (typically 363–383 K). This process can be optimized using process simulation software [17] in which the diffusion coefficients of the acid gases inside the liquid phase are used to simulate the reaction kinetics [11]. Since both $CO_2$ and $H_2S$ react with the solvent, it is experimentally not possible to directly measure their diffusion coefficients. Instead, the experimental studies measure the diffusion coefficient of non-reacting model molecules [18], such as $N_2O$ to replace $CO_2$, and calculate the diffusion coefficient of the required acid gas from the diffusion coefficient

of the model molecule [19, 20, 21]. Sada et al. [22] measured the diffusion coefficients of $N_2O$ in aqueous solutions of five different amines including monoethanolamine (MEA) at 298 K and calculated the diffusion coefficient of $CO_2$ using the diffusion coefficients of $N_2O$. Ko et al. [23] measured $N_2O$ absorption rates in aqueous solutions of various amines at 303, 308 and 313 K and calculated the diffusion coefficients using the absorption rates. Ying and Eimer [24] also measured the diffusion coefficients of $N_2O$ in aqueous MEA solutions for a temperature range between 298 K and 333 K and calculated the diffusion coefficients of $CO_2$ using the $CO_2/N_2O$ analogy [19].

Force field-based molecular dynamics (MD) simulations have been extensively used to predict diffusion coefficients of different solutes such as alkylbenzenes, ketones, and water in various solvents [25, 26]. This simulation method requires an accurate description of the interaction between the molecules of the solute and the solvent i.e., interaction potentials that describe the interactions between the molecules accurately. The advantage of MD simulations is that reactions in the system can be "switched off", eliminating the need for a model molecule in the experimental studies. Although MD simulations have been very promising and are widely used for this purpose [20, 15], we currently have limited knowledge of the diffusion coefficients of $CO_2$ and $H_2S$ and their temperature dependendence in solutions with different concentrations of alkanolamine in the solvent. The diffusivity of acid gases in pure water has been studied extensively [27, 28, 29]. Only two simulation studies in the literature report diffusion coefficients of acid gases in aqueous MEA solutions. To validate the $CO_2/N_2O$ analogy, Chen et al. [20] have computed the self-diffusivities of $CO_2$ and $N_2O$ in aqueous MEA solution at 303 K. Melnikov and Stein [30] have computed the diffusion coefficient of $CO_2$ in aqueous MEA solution as a function of $CO_2$ loading at 313 K. This study revealed that the diffusion coefficients of all the species in $CO_2$ loaded aqueous MEA solution decrease significantly with increasing $CO_2$ loading.

In this study, we compute self-diffusion coefficients ($D_{\text{self}}$) for $CO_2$ and $H_2S$ in aqueous MEA solutions for a wide range of temperatures and MEA concentrations in the solution. We studied aqueous MEA solutions because it is considered as an industry benchmark solvent [31] and it is also used for $CO_2$ capture from flue gas [32]. We first computed the density of pure MEA solution for the temperature range 293–353 K. It turns out that with the standard force fields from literature, the results did not agree with the experimental density values from literature. We then scaled the force field parameters of MEA molecules to find the optimum scaling factor that best

describes the experimental densities of the solvent. We validated this set of parameters by calculating the viscosities and $D_{\text{self}}$ of pure MEA and 30 wt.% MEA/water solution and compared these values to experimental values from literature. We used the validated force field for MEA to compute the self-diffusivities of $CO_2$ and $H_2S$ at infinite dilution for a temperature range of 293–353 K and MEA concentrations ranging from of 10–50 wt.% in the solvent. The results we provide will be useful for more accurate modelling in the process simulations, and will guide the design and development of acid gas removal process.

This article is organized as follows: the force field parameters and the simulation methods are discussed in the next section. In section 3, we discuss the results from the simulations and compare them with available literature data. In the final section, we provide conclusions regarding to the diffusivity of acid gases in aqueous MEA solutions.

## 2. Simulation Methods

Monte Carlo (MC) simulations to compute solvent densities were performed using the open source MC software, Brick-CFCMC [33, 34, 35]. For MEA molecules, the OPLS-AA [36, 37] force field was used for intermolecular Lennard-Jones (LJ) interactions because it was optimized for amines. Partial charges computed from quantum mechanical calculations were used for electrostatic interactions of MEA molecules. Quantum chemical calculations were performed using the Gaussian09 [38] software at second order Møller-Plesset perturbation theory (MP2) [39] level using the 6-311+G(2d,2p) basis set. We then multiply either the energy ($\epsilon$) parameters of LJ interactions of MEA molecule or the point charges of MEA molecule with a scaling factor $\chi$ to scale the interactions of this molecule. For water molecules, the SPC/E [40] force field was used. The SPC/E force field is known to predict the transport properties of water accurately [41]. For $CO_2$ molecules, the TraPPE [42] force field was used. The interactions between the TraPPE $CO_2$ molecules and the SPC/E water molecules were computed using the optimized intermolecular potential for $CO_2/H_2O$ developed by Orozco et al. [43]. For $H_2S$ molecules, the force field developed by Kristóf and Liszi [44] were used. All force field parameters for these molecules can be found in the Supporting Information (Tables S1-S3). LJ parameters of the interactions of different types of atoms except the interactions between $CO_2$ and water molecules [43] were computed using Lorentz-Berthelot mixing rules [45]. All molecules in

the molecular simulations were kept rigid. It was shown that the rigidity of small molecules (length of MEA molecule $\approx 3$ Å) does not significantly affect the dynamics in MD simulations [45]. Initial configurations were generated in a cubic simulation box with a length of 25.5 Å using Packmol [46]. For initialization, equilibration and production stages, $10^4$, $10^5$ and $10^5$ MC cycles were performed, respectively. In MC cycles, the number of trial moves is equal to the number of molecules in the simulation box. These moves were the translation of a randomly selected molecule (49.5%), the rotation of a randomly selected molecule (49.5%) and attempting to change the volume of the simulation box (1%). In these simulations, LJ interactions were truncated at 12 Å and analytic tail corrections [45] were applied. To compute the electrostatic interactions, the Ewald summation [47] was used with a precision of $10^{-6}$. Standard deviations for densities of pure MEA and 30 wt.% MEA/water solutions were computed using block averaging over the densities computed in the production stage of the MC simulations.

Initial configurations for the MD simulations were generated with a box length of 50 Å using Packmol [46]. The number of MEA and water molecules used for different concentrations of MEA in the solution are listed in Table 1. Two molecules of $CO_2$ or $H_2S$ were used to compute the self-diffusivity of these species. The MD simulations start with an equilibration period of 0.5 ns with a timestep of 1 fs in the $NPT$ ensemble using the Nosé-Hoover thermostat and barostat. After this equilibration, the temperature was equilibrated in the $NVT$ ensemble using the Nosé-Hoover thermostat for another 0.5 ns. In the production stage, the simulations were run for 100 ns in the $NVE$ ensemble with a timestep of 1 fs. In these simulations, LJ interactions were truncated at 12 Å. Analytic tail corrections [45] were applied to account for the long-range interactions. Electrostatic interactions were computed using the Particle-Particle Particle-Mesh (PPPM) method with a relative precision of $10^{-5}$. MD simulations to compute viscosities and self-diffusivities were performed using the LAMMPS [48] package (version 3 March 2020) with the OCTP [49] plugin. The computed self-diffusion coefficients were corrected for the finite-size effects [50, 51, 52]. It is important to note that the computed self-diffusion coefficients of the acid gases are practically equal to transport diffusion coefficients because the acid gases are at low loading [53]. The standard deviations of the self-diffusion coefficients and the viscosities were computed from ten independent simulations starting from different initial configurations. The radial distribution functions (RDFs) computed in this study are center-of-mass radial distribution functions.

Table 1: Number of MEA and water molecules in MD simulations for different concentrations of MEA in the MEA/water solutions.

| MEA concentration / [wt.%] | Number of MEA molecules | Number of water molecules | Average Box Size at 313 K / [Å] |
|---|---|---|---|
| 10 | 25 | 775 | 29.1 |
| 20 | 55 | 745 | 29.8 |
| 30 | 81 | 646 | 29.9 |
| 40 | 123 | 627 | 30.5 |
| 50 | 159 | 541 | 30.8 |

## 3. Results and Discussion

LJ interaction parameters for MEA were taken from the OPLS-AA force field [36, 37]. The point charges of MEA were computed using quantum chemical calculations as discussed in the previous section. Generic force fields such as OPLS-AA and point charges calculated using quantum chemical calculations may require scaling (with different methods) [54, 55, 56, 57, 58, 59]. The reason for this is that point charges calculated using quantum chemical calculations typically overestimate electrostatic interactions [54, 57, 60, 61, 62, 58]. To test the performance of the force field for MEA, we first calculated the density of a pure MEA solution and a 30 wt.% MEA/water solution for a temperature range of 293–353 K using MC simulations. Comparison between computed and experimental densities [63, 64, 65] are shown in Fig. 1. Results showed that computed densities using this force field do not agree well with experimental measurements [63, 64, 65]. This is because strong polarization and charge transfer in these solutions are not well produced by this force field [55]. We scaled the energy ($\epsilon$) parameter of the LJ potential and the point charges of the MEA molecule by multiplying either $\epsilon$ or the point charges with a scaling factor, $\chi$. Fig. 1 shows the densities of pure MEA solvent and 30 wt.% MEA/water solution as a function of temperature and $\chi$. Results show that changing the LJ potential does not affect the densities of both pure MEA and 30 wt.% MEA/water solution significantly, while scaling the point charges significantly affects the density of these solutions. Figure S1 of the Supplementary Material shows that scaling the LJ $\epsilon$ parameter of the MEA atoms by $\chi = 0.7$ changes the density of pure MEA solution (30 wt.%

MEA/water solution) by ca. 0.4% (1.1%) at 303 K. The scaling of the point charges of MEA by the same $\chi$ changes the density of pure MEA by ca 10% and the density of 30 wt.% MEA/water solution by ca. 4% (Fig. 1(b) and (d)). Overall, these results suggest that calculated densities of pure MEA and 30 wt.% MEA/water solutions agree well with the experimental values when the point charges of MEA are scaled by 0.8, with a maximum deviation of ca. 3% from experiments for both solutions (Fig. 1 (b) and (d)).

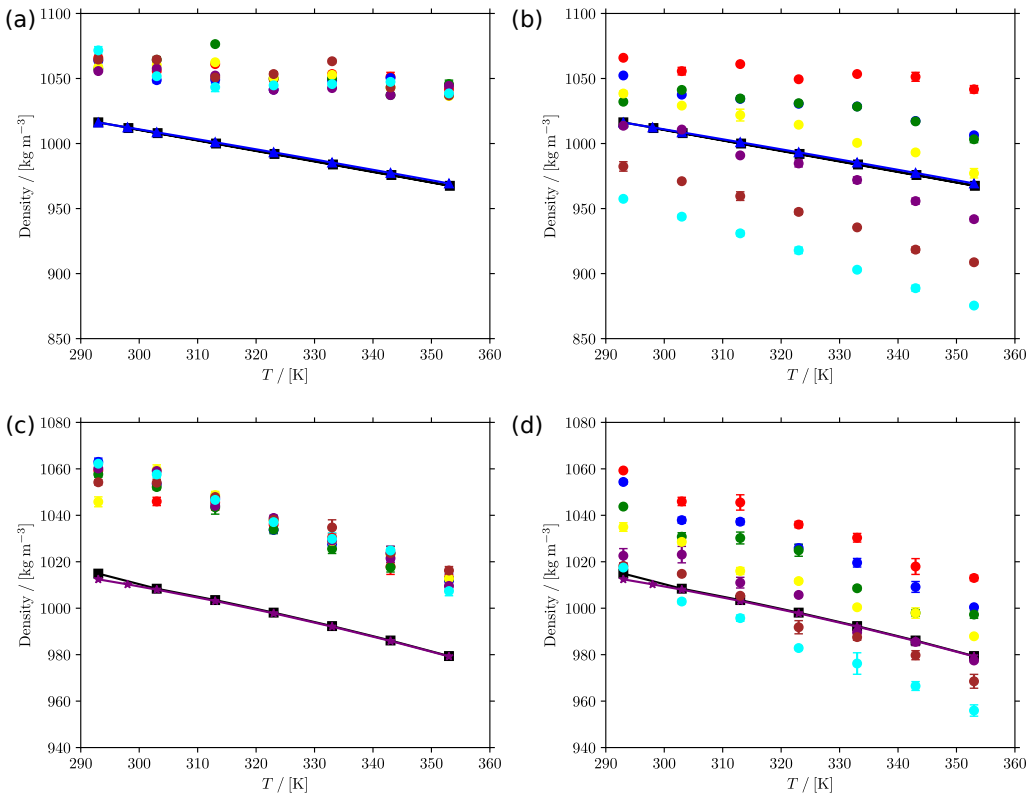


Figure 1: Comparison of simulated and experimental [63, 64, 65] densities of (a,b) pure MEA and (c,d) 30 wt.% MEA/water solutions as a function of temperature. Subfigures (a) and (c) show the scaling of LJ $\epsilon$ parameters of the MEA molecules while subfigures (b) and (d) show the scaling of the point charges of the MEA molecules. Red: $\chi = 1.00$; blue: $\chi = 0.95$; green: $\chi = 0.90$; yellow: $\chi = 0.85$; purple: $\chi = 0.80$; brown: $\chi = 0.75$; cyan: $\chi = 0.70$; black: experimental [63], blue: experimental correlation [64, 65]. The lines connecting the experimental data are to guide the eye.

Motivated by the good agreement between simulations and experiments on the densities of pure MEA and 30 wt.% MEA/water solutions, we validated the scaling factor for the point charges, i.e. chi = 0.80, of MEA by computing the viscosities and $D_{\text{self}}$ of these solutions using MD simulations for a temperature range of 293–353 K. We have used 30 wt.% MEA/water solution to validate our model for MEA because this is the most studied solution in literature and the industry standard for $CO_2$ capture [66]. It is important to note that we scaled the point charges of MEA with $\chi = 0.8$ in these simulations. Fig. 2 shows the comparison between the computed and experimental [63] viscosities and $D_{\text{self}}$ of pure MEA and 30 wt.% MEA/water solutions. Results show that the computed and experimental viscosities of pure MEA and 30 wt.% MEA/water solutions have coefficient of determination ($R^2$) [67] scores of 0.98 and 0.97, respectively. Both $R^2$ scores show that the simulations, and therefore this set of force field parameters for MEA, agree well with the experiments on viscosity in this temperature range. We also compare the simulation results with the experimental correlation obtained from Design Institute for Physical Properties (DIPPR) [64] (Fig. 2(a)). The simulations agree well also with the experimental correlation obtained from the DIPPR database. For example, the computed viscosities for pure MEA (30 wt.% MEA/water) were between 26.26–2.42 (2.69–0.91) mPa · s at 293–353 K. The experimental values for the same conditions vary between 24.09–2.92 and 2.91–0.77 mPa · s for pure MEA and 30 wt.% MEA/water solutions, respectively. The maximum (average) deviation between computed viscosities and experimental viscosities were computed as 17% (8.8%) and 15% (7.6%) for pure MEA and 30 wt.% MEA/water solutions, respectively. These results suggest that using the scaling factor ($\chi = 0.8$) for the point charges of MEA in these simulations can provide accurate predictions for the viscosity of MEA/water solutions. We also compared the computed $D_{\text{self}}$ (corrected for finite-size effects using computed viscosities [50, 51]) of MEA molecules in pure MEA solution with the experimental values from literature [68]. The experimental values are $4.2\times10^{-11}\,\text{m}^2\,\text{s}^{-1}$, $5.5\times10^{-11}\,\text{m}^2\,\text{s}^{-1}$, and $9.3\times10^{-11}\,\text{m}^2\,\text{s}^{-1}$ for 288, 298, and 308 K, while the computed $D_{\text{self}}$ are $4.5\times10^{-11}\,\text{m}^2\,\text{s}^{-1}$ (extrapolated slightly using an Arrhenius equation fit, $R^2$ for Arrhenius fit = 0.997), $5.6\times10^{-11}\,\text{m}^2\,\text{s}^{-1}$, and $1.1\times10^{-10}\,\text{m}^2\,\text{s}^{-1}$, respectively. To the best of our knowledge, there is no experimental data in literature to compare $D_{\text{self}}$ of water and MEA molecules in 30 wt.% MEA/water solutions. The value of $D_{\text{self}}$ of water molecules is 2.14-2.34 times larger than the MEA molecules in 30 wt.% MEA/water solutions. Also, the results show that the self diffusivity $D_{\text{self}}$ of MEA is an

order of magnitude higher in 30 wt.% MEA/water solution than that in a pure MEA solution. This indicates stronger MEA-MEA interactions than MEA-water interactions.

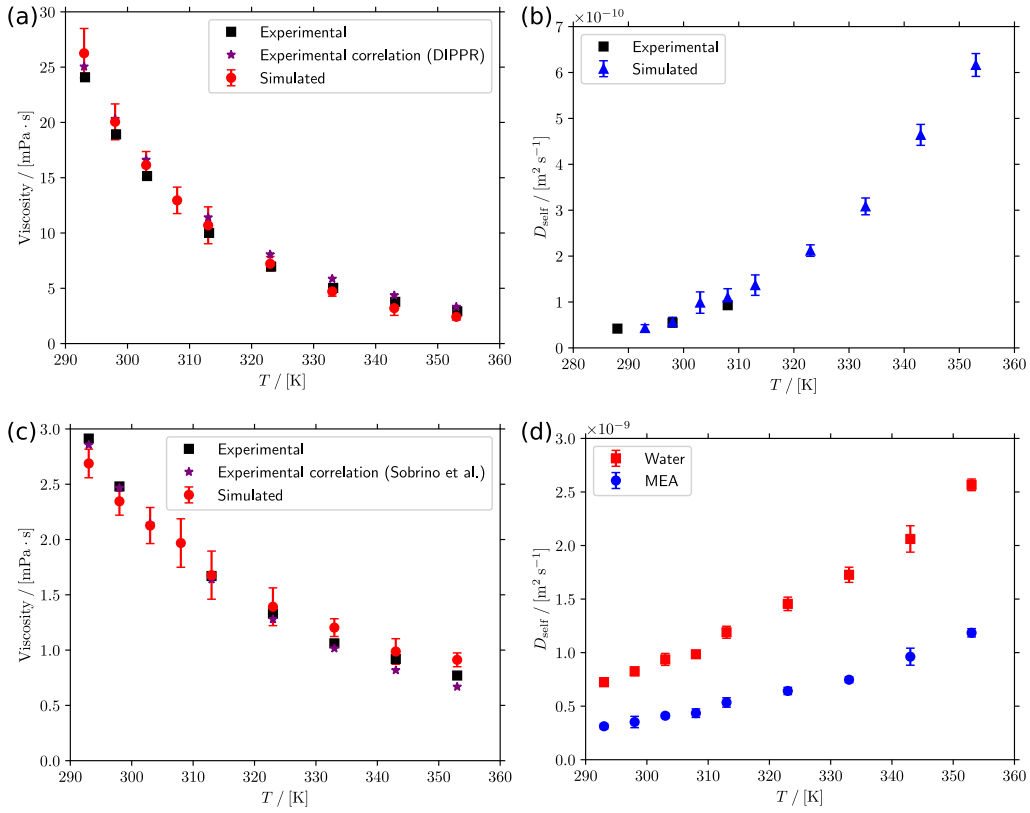


Figure 2: Comparison of simulated and experimental [63, 64, 65, 69] viscosities of (a) pure MEA and (c) 30 wt.% MEA/water solution as a function of temperature. $D_{\text{self}}$ of (b) MEA molecules in pure MEA and (d) MEA and water molecules in 30 wt.% MEA/water solution as a function of temperature.

To obtain a fundamental understanding of the transport mechanism of $CO_2$ and $H_2S$ in MEA/water solutions with different MEA concentrations, we computed $D_{self}$ of $CO_2$, $H_2S$, water, and MEA molecules in 10–50 wt.% MEA/water solutions at infinite dilution and 1 bar for a temperature range of 293–353 K using MD simulations. Fig. 3 shows $D_{self}$ of both acid gases in pure water [27, 28] and 10–50 wt.% MEA/water solutions as a function of temperature and MEA concentration. Fig S3. shows computed viscosities of aqueous MEA solutions as a function of temperature and MEA concentration. Fig. S4 shows $D_{self}$ of water and MEA molecules as a function of temperature and MEA concentration. We first compare computed values of $D_{self}$ of $CO_2$ with values of $D_{self}$ of $CO_2$ obtained using $CO_2/N_2O$ analogy [70]. Mandal et al. [70] estimated values of $D_{self}$ of $CO_2$ in 30 wt.% MEA/water solution as $1.61\times10^{-9}$ $m^2\,s^{-1}$, $1.74\times10^{-9}$ $m^2\,s^{-1}$, and $2.14\times10^{-9}$ $m^2\,s^{-1}$ at 293 K, 303 K, and 313 K, respectively. The values of $D_{self}$ of $CO_2$ we computed in 30 wt.% MEA/water solution are $1.1\times10^{-9}$ $m^2\,s^{-1}$, $1.4\times10^{-9}$ $m^2\,s^{-1}$, and $2.1\times10^{-9}$ $m^2\,s^{-1}$ at 293 K, 303 K, and 313 K, respectively. These results show that simulated values of $D_{self}$ of $CO_2$ are slightly underestimated for the temperatures 293 K and 303 K while at 313 K the computed value of $D_{self}$ of $CO_2$ agrees with the value obtained using $CO_2/N_2O$ analogy [70].

Our results show that $D_{self}$ of both acid gases increase with increasing temperature. Fig. 3 also shows that $D_{self}$ of $CO_2$ is larger than $D_{self}$ of $H_2S$ at the same conditions. Although $H_2S$ has a lower molar mass ($M_{H_2S}$ = 34.1 g mol$^{-1}$) than $CO_2$ ($M_{CO2}$ = 44.01 g mol$^{-1}$), its values of $D_{self}$ is lower because it can form hydrogen bonds with both water and MEA molecules, and the $H_2S$ molecule is more spherical than the linear $CO_2$ molecule [71]. Also, the results show that with the increasing concentration of MEA in the solution both $D_{self}$ of $CO_2$ and $H_2S$ in these solutions decrease. For $CO_2$ ($H_2S$), $D_{self}$ at 293 K decreases by a factor of 7.6 (6.8) times from 10 wt.% MEA to 50 wt.% MEA while at 353 K, $D_{self}$ decrease by a factor of 3.6 (3.4) times. The temperature dependency of the $D_{self}$ of both $CO_2$ and $H_2S$ decreases with increasing MEA concentration in the solution. The same temperature dependency can also be observed in $D_{self}$ of water and MEA molecules (Fig. S4). The slope of $D_{self}$ as a function of temperature in a 10 wt.% solution is 3.0 and 2.7 times higher than 50 wt.% solution for $CO_2$ and $H_2S$, respectively. Also, $D_{self}$ changes significantly for both acid gases from 40 wt.% solution to 30 wt.%, especially at low temperatures. However, the changes in $D_{self}$ of both acid gases are not as significant from 50 wt.% to 40 wt.%. For example, $D_{self}$ of $H_2S$ at 293 K increases by 2.2 times from 40 wt.%

solution to 30 wt.% solution while it only increases by a factor of 1.7 from 50 wt.% to 40 wt.%. This effect of MEA concentration on $D_{\text{self}}$ decreases with the increasing temperature as $D_{\text{self}}$ of $H_2S$ increases 1.5 times both from 40 wt.% to 30 wt.% and from 50 wt.% to 40 wt.% at 353 K. For $CO_2$, water and MEA, there is also a significant effect of concentration on $D_{\text{self}}$ from 30 wt.% MEA/water solution to 20 wt.% MEA/water solution (Fig. 3(a) and Fig. S4). We fit the value of $D_{\text{self}}$ of $CO_2$ and $H_2S$ to an Arrhenius equation using:

$$D_{\text{self}} = D_0 \exp\left[-\frac{E_A}{RT}\right] \tag{1}$$

where $D_0$ is the pre-exponential factor, $E_A$ is the activation energy for diffusion, $R$ is the ideal gas constant, and $T$ is the absolute temperature. Fig. 3(b,d) shows the Arrhenius fits for $D_{\text{self}}$ of $CO_2$ and $H_2S$. Tables 2 and 3 shows Arrhenius fit parameters for $D_{\text{self}}$ of $CO_2$ and $H_2S$. Tables 2 and 3 show that the activation energy for diffusion for both acid gases increase with increasing MEA concentration in the solution. This was also indicated by slower acid gas dynamics (Fig. 3) with increasing MEA concentration. We also fit the $D_{\text{self}}$ of $CO_2$ and $H_2S$ to the Speedy-Angell power equation [72] (Eq. S1) and the Vogel-Tamann-Fulcher (VTF) equation [73] (Eq. S2). Tables S8-11 of the Supplementary Material show the Speedy-Angell power equation and the VTF equation fit parameters for $CO_2$ and $H_2S$. Figure S2 of the Supplementary Material shows the Speedy-Angell and VTF fits for $D_{\text{self}}$ of $CO_2$ and $H_2S$ in aqueous MEA solutions. The pressure and temperature dependent form of the Speedy-Angell power equation has been shown to be able to predict the $CO_2$ diffusivity in water very accurately [29]. Our results show that the Speedy-Angell power equation has the highest coefficients of determination ($R^2$) for $D_{\text{self}}$ of $CO_2$ and $H_2S$ between the Arrhenius equation, the Speedy-Angell power equation and the VTF equation.

We performed two more sets of MD simulations to measure the sensitivity of values of $D_{\text{self}}$ of acid gases with respect to the point charge scaling factor $\chi$. We computed values of $D_{\text{self}}$ of $CO_2$ as $3.48_{0.3} \times 10^{-9}$ $m^2\,s^{-1}$, $3.53_{0.4} \times 10^{-9}$ $m^2\,s^{-1}$, and $2.66_{0.2} \times 10^{-9}$ $m^2\,s^{-1}$ for $\chi = 0.7$, $\chi = 0.8$, and $\chi = 1.0$, respectively, at 353 K and 1 bar in 30 wt.% MEA/water solution. This shows that the value of $D_{\text{self}}$ of $CO_2$ change significantly with the scaling from $\chi = 1.0$ to $\chi = 0.8$ while the change in the value of $D_{\text{self}}$ of $CO_2$ from $\chi = 0.8$ to $\chi = 0.7$ is not significant (within the error bars shown as subscripts in this paragraph).

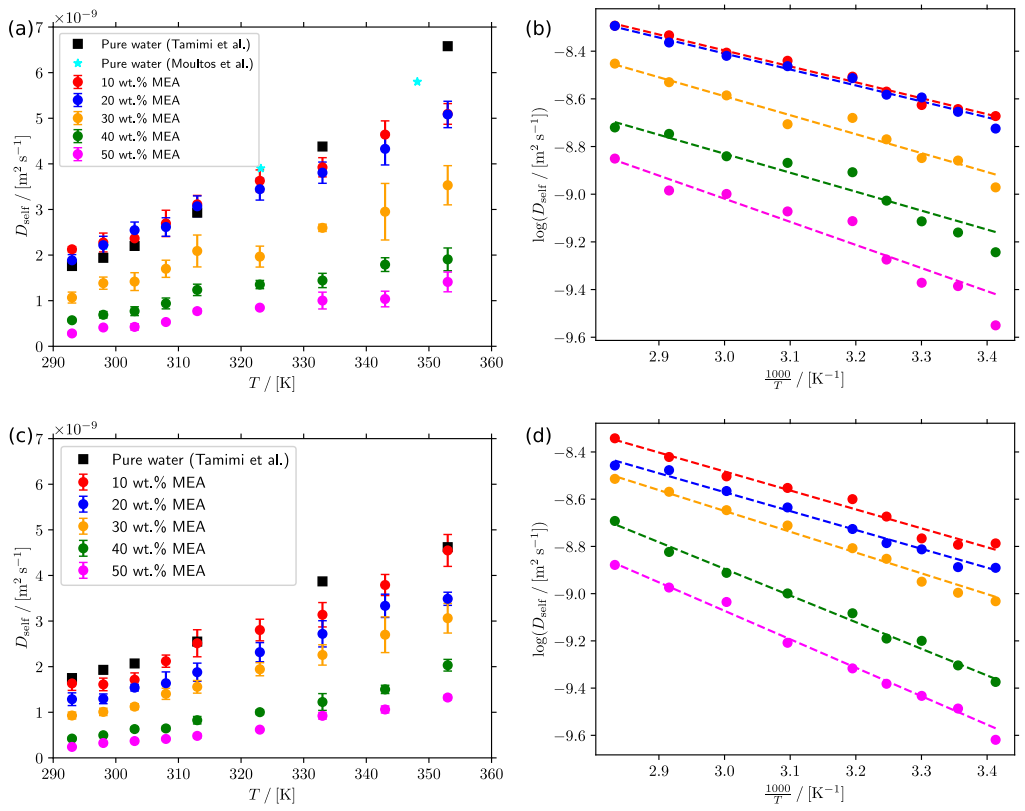


Figure 3: Self-diffusion coefficients of (a) $CO_2$ and (c) $H_2S$ in pure water [27, 28] and 10–50 wt.% MEA/water solutions as a function of temperature. Subfigures (b) and (d) show the Arrhenius plots of subfigures (a) and (c), respectively. In subfigures (b) and (d), color codes follow those in subfigures (a) and (c). Dashed lines represent the Arrhenius fits of the $D_{self}$. The Speedy-Angell [72] and the VTF [73] equation fits are shown in Figure S2 of the Supplementary Material.

Table 2: Arrhenius fit parameters (pre-exponential factor ($D_0$) and activation energy ($E_A$)) and coefficient of determinations ($R^2$) for $D_{self}$ of $CO_2$ in MEA/water solutions with different MEA concentrations. The values of $D_{self}$ of $CO_2$ were fitted for a temperature range of 293–353 K.

| MEA concentration / [wt.%] | $D_0$ / $[m^2\,s^{-1}]$ | $E_A$ / $[kJ\,mol^{-1}]$ | $R^2$ |
|---|---|---|---|
| 10 | $4.05 \times 10^{-7}$ | 12.79 | 0.989 |
| 20 | $3.98 \times 10^{-7}$ | 12.82 | 0.988 |
| 30 | $6.28 \times 10^{-7}$ | 15.23 | 0.970 |
| 40 | $3.59 \times 10^{-7}$ | 15.23 | 0.947 |
| 50 | $7.77 \times 10^{-7}$ | 18.57 | 0.944 |

Table 3: Arrhenius fit parameters (pre-exponential factor ($D_0$) and activation energy ($E_A$)) and coefficient of determinations ($R^2$) for $D_{self}$ of $H_2S$ in MEA/water solutions with different MEA concentrations. The values of $D_{self}$ of $H_2S$ were fitted for a temperature range of 293–353 K.

| MEA concentration / [wt.%] | $D_0$ / $[m^2\,s^{-1}]$ | $E_A$ / $[kJ\,mol^{-1}]$ | $R^2$ |
|---|---|---|---|
| 10 | $8.41 \times 10^{-7}$ | 15.36 | 0.985 |
| 20 | $6.76 \times 10^{-7}$ | 15.31 | 0.985 |
| 30 | $9.84 \times 10^{-7}$ | 16.86 | 0.991 |
| 40 | $3.10 \times 10^{-6}$ | 21.61 | 0.991 |
| 50 | $3.48 \times 10^{-6}$ | 23.06 | 0.992 |

Fig. 4 shows RDFs of $CO_2$ and $H_2S$ with water and MEA molecules as a function of the MEA concentration in MEA/water solutions. For the MEA concentrations, the peak positions of $CO_2$-MEA and $H_2S$-MEA RDFs are similar. However, the results show that the intensity of the first peaks in $CO_2$-MEA and $H_2S$-MEA RDFs increases with decreasing MEA concentration in the solution. These results indicate that acid gas-MEA interactions are stronger with respect to the decreasing MEA concentration in the solutions. In the $CO_2$-water RDF, it can be observed that the first peak gets widened and more intense with increasing MEA concentration in the solution. In the $H_2S$-water RDF, the first peak positions do not change while the intensities of the first peak show a trend of decreasing with increasing MEA concentration in the solution. These results mainly indicate a weaker interaction between $H_2S$ and water molecules with respect to the increase in the concentration of MEA in the solutions. The second peaks in $H_2S$-water RDFs slightly change position in the solutions with different MEA concentration. Intensities of the second peak in $H_2S$-water RDF also change with changing MEA concentration in the solution. The intensity decreases from 10 to 40 wt.% while it increases from 30 to 40 wt.%. Overall, our results show that the MEA concentration in aqueous MEA solutions significantly affects the acid gas-MEA and acid gas-water interactions. The RDFs we computed indicate that both acid gas-MEA interactions and acid gas-water interactions will become weaker with increasing MEA concentration in the solution. With weaker interactions with the surrounding molecules, we would expect that values of $D_{\text{self}}$ of both acid gases increase with increasing MEA concentration. However, Fig. 3 shows that values of $D_{\text{self}}$ decrease significantly with increasing MEA concentration in the solution. This is because of increased viscosity of the solution with increasing MEA concentration [65], i.e. values of $D_{\text{self}}$ of every molecule type in the solution decrease (Fig. 3 and Fig. S4) with increasing MEA concentration.

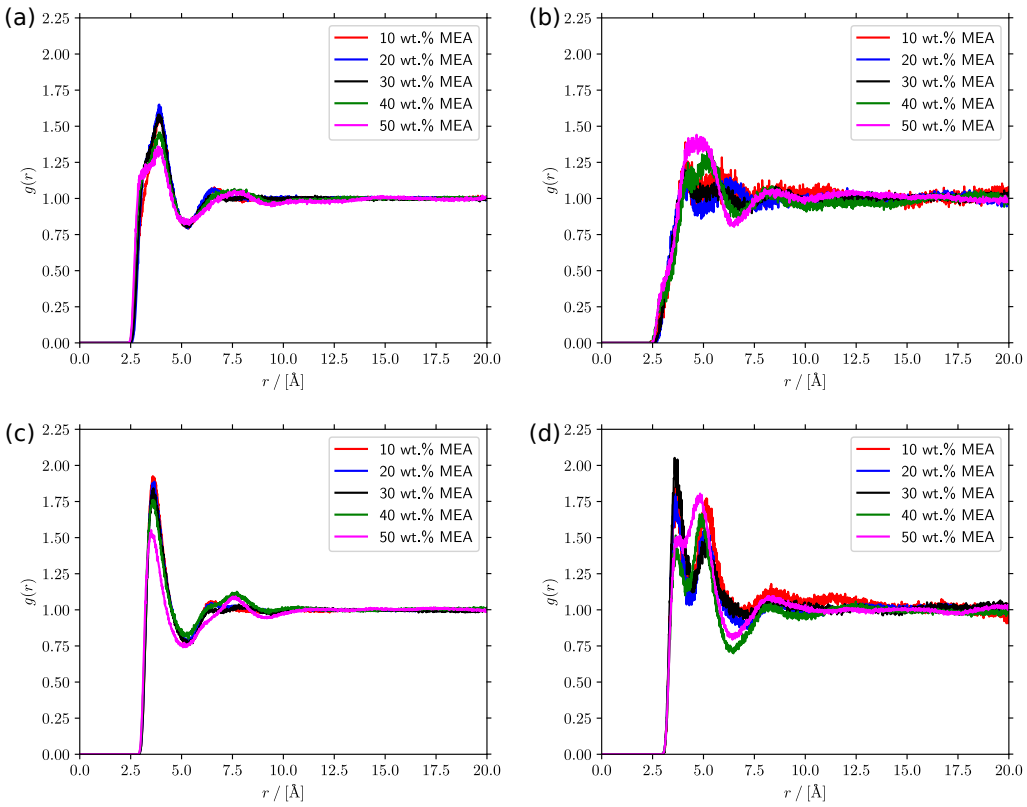


Figure 4: Radial distribution functions of (a) $CO_2$ -MEA, (b) $CO_2$ -water, (c) $H_2S$ -MEA, and (d) $H_2S$ -water for 10–50 wt.% MEA/water solutions at 293 K and 1 bar.

## 4. Conclusions

We investigated the effect of temperature and MEA concentration on the self-diffusivity of $CO_2$ and $H_2S$ in aqueous MEA solutions. For this purpose, we computed densities of pure MEA and 30 wt.% MEA/water solutions as a function of temperature and the scaling factor for point charges of MEA ($\chi$). We showed that scaling factor $\chi = 0.80$ can be used to obtain a good agreement between molecular simulations and experiments from literature. We validated this scaling factor by computing viscosities and self-diffusivity of pure MEA and 30 wt.% MEA/water solutions at 293–353 K. The scaling factor of $\chi = 0.80$ was validated by comparing the computed and experimental viscosities and the self-diffusivities of pure MEA and 30 wt.% MEA/water solutions. We computed the self-diffusivities of $CO_2$ and $H_2S$ at infinite dilution, at 293–353 K and 1 bar, for 10–50 wt.% MEA/water solutions. The results showed that $D_{\text{self}}$ of acid gases significantly depends on the MEA concentration in the solution. It is also shown that $D_{\text{self}}$ of $CO_2$ is larger than $D_{\text{self}}$ of $H_2S$ despite molecular weight of $CO_2$ ($44.01\,\text{g}\,\text{mol}^{-1}$) being higher than that of $H_2S$ ($34.1\,\text{g}\,\text{mol}^{-1}$).

## Acknowledgement

This work was supported by the Carbon Capture Utilization and Storage R&D program from TotalEnergies S.E. This work has been financially supported by ANRT, CIFRE Convention no. 2019/0860.We are grateful for the support by NWO Domain Science for the use of supercomputer facilities, with financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organisation for Scientific Research, NWO). T.J.H.V. acknowledges NWO-CW (Chemical Sciences) for a VICI grant.

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