Supplementary Information

Bulk viscosity of molecular fluids

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1 Full pressure tensor

The pressure tensor elements are given by [1,2]

\[ P_{\alpha\beta} = \frac{1}{V} \left( \sum_{i} \frac{p_{i\alpha} p_{i\beta}}{m_{i}} + \sum_{i>1} \sum_{j>i} f_{ij\alpha} f_{ij\beta} \right) - \langle P_{\alpha\beta} \rangle, \]

(1)

where the sum is over \( N_{a} \) atoms, and \( p_{i\alpha} \) and \( p_{i\beta} \) are the particle momenta in the respective direction. The second term is a sum over all forces between atoms \( i \) and \( j \). \( \langle P_{\alpha\beta} \rangle \) is expressed as:

\[ \langle P_{\alpha\beta} \rangle = \delta_{\alpha\beta} \left[ P + \frac{\partial P}{\partial N} (N - \langle N \rangle) + \frac{\partial P}{\partial E} (E - \langle E \rangle) \right]. \]

(2)

Thus it is 0 for \( \alpha \neq \beta \). In the microcanonical ensemble (constant number, volume, energy) we have \( N = \langle N \rangle \) and \( E = \langle E \rangle \), such that \( \langle P_{\alpha\beta} \rangle = \delta_{\alpha\beta} P \).

2 Influence of the Lennard-Jones cut-off on transport properties

The influence of the potential cut-off, \( r_{\text{cut}} \), is examined briefly in order to choose a cut-off value which does not affect results, as done in previous studies [3, 4]. A near-critical isotherm (\( T^{*} = 1.35 \)) was chosen to investigate the effects on the enhanced bulk viscosity near the critical point. As can be seen in Fig. S1a, the shear viscosity, \( \eta^{*} \), remains unaffected by the chosen cut-off. The bulk viscosity, however, is larger for larger cut-off values. The increase of the ratio, \( \kappa^{*} / \eta^{*} \), for larger cut-offs is significant (Fig. S2). Therefore we can conclude that the long-range interactions contribute to the bulk viscosity, \( \kappa^{*} \), but not the shear viscosity. For larger \( r_{\text{cut}} \) the effect of this becomes smaller.
Figure S1: Shear and bulk viscosity for an LJ fluid at $T^* = 1.35$ for several cut-off parameters, $r_{\text{cut}}$. The filled markers present data from this work with the crosses being obtained from Meier et al. (2004,2005) [3,5] for $r_{\text{cut}} = 5.5$. 

S-3
Figure S2: Viscosity ratio, $\kappa^*/\eta^*$, for an LJ fluid at $T^* = 1.35$ for several cut-off parameters, $r_{\text{cut}}$.

3 Simulation parameters for water, CO$_2$ and n-decane

Tables S1, S2, S3, S4 and S5 show the simulation parameters used in this work.

Table S1: Simulation parameters for two atomistic water models, SPC/E and TIP4P/2005.

| Model       | SPC/E  | TIP4P/2005 |
|-------------|--------|------------|
| $\sigma_{O-O}$ (Å) | 3.166  | 3.1589     |
| $\epsilon_{O-O}$ (eV) | 0.00673 | 0.0080     |
| $\sigma_{H-O} = \epsilon_{H-O} = \sigma_{H-H} = \epsilon_{H-H}$ | 0 | 0 |
| $q_O$ (e) | -0.8476 | -1.1128 |
| $q_M$ (e) | - | - |
| $b$ (Å) | 1.0 | 0.9572 |
| $\theta$ (°) | 109.47 | 104.520 |
| $r_{O-M}$ (Å) | - | 0.1546 - |
Table S2: A selection of parameters for the SAFT-$\gamma$ Mie CGW1-ift and CGW1-vle water models as parametrised by Lobanova et al. [6].

| T (K) | $\sigma_{\text{ift}}$ (Å) | $\epsilon_{\text{ift}}$ (K/k_B) | $\sigma_{\text{vle}}$ (Å) | $\epsilon_{\text{vle}}$ (K/k_B) |
|-------|----------------|----------------|----------------|----------------|
| 293   | 2.9055         | 304.28         | -               | -               |
| 298   | 2.9016         | 305.21         | -               | -               |
| 313   | 2.8938         | 309.01         | -               | -               |
| 343   | 2.8811         | 318.84         | 3.0015          | 481.87          |
| 393   | 2.8721         | 332.18         | 3.0039          | 466.97          |
| 493   | 2.8666         | 350.25         | 3.0251          | 438.55          |
| 533   | -              | -              | 3.0403          | 426.99          |
| 613   | -              | -              | 3.1028          | 400.21          |

Table S3: Simulation parameters for different CO$_2$ models.

| Model        | TrapPE | EPM2 | SAFT-$\gamma$ dimer | SAFT-$\gamma$ monomer |
|--------------|--------|------|----------------------|------------------------|
| $\sigma_{C-C}$ (Å) | 2.8    | 2.757 | -                     | -                      |
| $\epsilon_{C-C}$ (eV) | 0.00233 | 0.002424 | -                      | -                      |
| $\sigma_{O-O}$ (Å) | 3.05   | 3.033 | -                     | -                      |
| $\epsilon_{O-O}$ (eV) | 0.00681 | 0.006938 | -                      | -                      |
| $q_C$ (e)     | 0.7    | 0.6512 | -                     | -                      |
| b (Å)         | 1.16   | 1.149 | -                     | -                      |
| $\theta$ (°)  | 180.0  | 180.0 | -                     | -                      |
| $K_{\theta}$ (eV/rad$^2$) | -     | (6.405) | -                     | -                      |
| $\sigma_{\text{SAFT}}$ (Å) | -     | -     | 2.85                 | 3.74                   |
| $\epsilon_{\text{SAFT}}$ (eV) | -     | -     | 190.14               | 0.03118                |
| $\lambda_r$   | 12.0   | 12.0  | 13.77                | 23.0                   |
| $\lambda_g$   | 6.0    | 6.0   | 6.0                  | 6.66                   |
Table S4: Simulation parameters for two atomistic decane models, OPLS and L-OPLS.

| Model | OPLS | L-OPLS |
|-------|------|--------|
| \(\sigma_{\text{C-C}}\) (Å) | 0.35 | 0.35 |
| \(\epsilon_{\text{C-C}}\) (eV) | 0.00286 | 0.00286 |
| \(\sigma_{\text{H-H}}\) (Å) | 0.25 | 0.25 |
| \(\epsilon_{\text{H-H,CH}_2}\) (eV) | 0.00130 | 0.00114 |
| \(\epsilon_{\text{H-H,CH}_3}\) (eV) | 0.00130 | 0.00130 |
| \(q_{\text{C,CH}_2}\) (e) | -0.120 | -0.148 |
| \(q_{\text{C,CH}_3}\) (e) | -0.180 | -0.222 |
| \(q_H\) (e) | 0.06 | 0.074 |
| \(b_{\text{C-H}}\) (Å) | 1.09 | 1.09 |
| \(k_{b_{\text{C-H}}}\) (eV) | 14.743 | 14.743 |
| \(b_{\text{C-C}}\) (Å) | 1.529 | 1.529 |
| \(k_{b_{\text{C-C}}}\) (eV) | 11.621 | 11.621 |
| \(\theta_{\text{C-C-H}}\) (°) | 110.7 | 110.7 |
| \(k_{\theta_{\text{C-C-H}}}\) (eV) | 1.6262 | 1.6262 |
| \(\theta_{\text{H-C-C-H}}\) (°) | 107.8 | 107.8 |
| \(k_{\theta_{\text{H-C-C-H}}}\) (eV) | 1.43 | 1.43 |
| \(\theta_{\text{C-C-C}}\) (°) | 112.7 | 112.7 |
| \(k_{\theta_{\text{C-C-C}}}\) (eV) | 2.503 | 2.503 |

Table S5: Dihedral parameters for two atomistic decane models, OPLS and L-OPLS.

|         | K_1(eV) | K_2(eV) | K_3(eV) | K_4(eV) |
|---------|---------|---------|---------|---------|
| OPLS    |         |         |         |         |
| H-C-C-H | 0.0     | 0.0     | 0.01379 | 0.0     |
| C-C-C-H | 0.0     | 0.0     | 0.01588 | 0.0     |
| H-C-C-C | 0.07545 | -0.00681| 0.01209 | 0.0     |
| L-OPLS  |         |         |         |         |
| H-C-C-H | 0.0     | 0.0     | 0.01379 | 0.0     |
| C-C-C-H | 0.0     | 0.0     | 0.01588 | 0.0     |
| H-C-C-C | -0.00317| 0.02796 | -0.00929| 0.00772 |
Figure S3: $P - \rho$ relation for different water models at 393 and 613 K. For 393 K, the models presented are SPC/E, SAFT-ift and SAFT-vle. For 613 K, the models presented are SPC/E, TIP4P/2005 and SAFT-vle.

4 Additional data: Water at different temperatures

In addition to the data presented in the paper we also examine water using both atomistic (SPC/E, TIP4P/2005) and coarse-grained models (two versions of the SAFT-$\gamma$ Mie model) at 393K and the near critical 613K.

The SAFT-$\gamma$ Mie force field for water [6] not only has temperature dependent parameters $\sigma$ and $\epsilon$ but there are also two different parametrizations performed over different temperature ranges. SAFT-ift is applicable in the lower temperature range (293 – 493 K) and was parametrized to fit to interfacial tension data whilst SAFT-vle was parametrized for 343 – 613 K using vapour-liquid equilibrium data.

We examine the pressure, shear viscosity and bulk viscosity data for water at two different temperatures; 393 K (SPC/E, SAFT-ift, SAFT-vle) and 613 K (SPC/E, TIP4P/2005 and SAFT-vle). The former is to compare the two coarse-grained parametrizations whilst the latter is to obtain data near the critical point. As SPC/E performs best in the liquid regime [7, 8] this is further a crude test for the applicability of SPC/E in the vapour phase. As no rotational or vibrational relaxation time data is available for these temperatures, only the configurational contribution of $\kappa$ is presented.
4.1 393 K

Figure S3a and S4 show the results for $T = 393$ K. The short falls of the SPC/E model in achieving the correct thermodynamic state are immediately clear as it overestimates the pressures. The tendency of atomistic models to overestimate pressures is already present at $T = 300$ K. SAFT-ift performs best, in particular at low densities, whilst SAFT-vle only obtains similar accuracy at very low pressures/densities but diverges in the high pressure regime.

Despite the inaccurate pressure values SPC/E does well in the prediction of shear viscosities, where on average only a slight overestimation compared to NIST values is observed. Notably SAFT-ift, while underestimating the shear viscosity, shows much improvement in this temperature range compared to room temperature data. SAFT-vle, however, overestimates the viscosities greatly and is as such not suitable for transport calculations.

Once again both SAFT models perform poorly in predicting bulk viscosity values of the order of the shear viscosity, with $\kappa_{\text{conf}}/\eta$ well below 1. SPC/E, however, obtains values between 3-4, similar to those at room temperature. These values are higher than that of 2.32 observed at 300 K. The increase of $\kappa_{\text{conf}}/\eta$ is contrary to an observed decrease with increase temperature in experiment [9, 10].

In conclusion we can assert that, despite its shortcomings in thermodynamic quantities, transport properties are still well predicted by the SPC/E model. The SAFT-ift model is superior to SAFT-vle both in terms of the thermodynamic predictions and the shear viscosity. The performance of SAFT-ift is much improved over room temperature simulations. Bulk viscosities, however, are still not well predicted.

4.2 613 K

SPC/E, TIP4P/2005 and SAFT-vle perform well in predicting the $P - \rho$ curve (Fig. S3b), with TIP4P/2005 giving the best agreement with NIST data. The SAFT-vle model diverges near the phase transition, giving pressures too low and even below zero, and overestimates the pressure for higher densities. SPC/E shows a great improvement over the 393 K case, with pressures within 200 bar or less for the whole density range. The shear viscosity, $\eta$, and the viscosity ratio, $\kappa_{\text{conf}}/\eta$, are presented in Fig. S5. $\eta$ is well predicted by the atomistic models over the entire range, underestimating slightly
Figure S4: $\eta$ and $\kappa_{\text{conf}}/\eta$ for SPC/E, SAFT-ift, SAFT-vle at $T = 393$ K.
Figure S5: $\eta$ and $\kappa_{\text{conf}}/\eta$ for SPC/E and SAFT-vle at $T = 613$ K.
compared to NIST at low densities. The SAFT-vle model performs particularly well in the vapour regime. In the liquid regime shear viscosities are overestimated considerably.

The viscosity ratio for SAFT-vle resembles that of other simple (monomer) fluids near the critical point with a clear peak observed near the critical density. SPC/E shows inconsistent results pointing to the difficulty of obtaining statistically relevant results near the critical point. TIP4P/2005, whilst more consistent, nevertheless does not show a clear enhancement near the critical point. However notably the viscosity ratio is increased significantly (to up to 17.5 for the SPC/E model). As no literature data exists for this region the validity of this cannot be confirmed. More investigations in the high temperature range are necessary.

4.3 Conclusions

The SAFT-ift model outperforms the SAFT-vle model both in terms of thermodynamic and transport properties. Attention should be drawn to the relatively good prediction of the shear viscosity at higher temperatures (SAFT-ift at 393 K and SAFT-vle at 613 K) showing that coarse-grained models for water may be successfully used in transport applications in some physical situations. An increase in the viscosity ratio near the critical point is observed similar to other fluids. However, as this is not present in generally superior atomistic models, further investigations near the critical point should be performed.

5 Additional data: Supercritical CO$_2$

We study two atomistic (EPM2, TraPPE) and two coarse-grained models (SAFT dimer, SAFT monomer) for the supercritical isotherm at 323 K. All models studied show qualitatively the same behaviour for supercritical CO$_2$ as at 300 K (Figure S6). The SAFT monomer model once again overestimates the shear viscosity at high densities. EPM2 is the most accurate in predicting $\eta$ whilst TraPPE and the SAFT dimer model have comparable results. The viscosity ratio (where only the configurational term is considered for $\kappa$) is overestimated in the lower density range, in particular in the vicinity of the critical point. With the exception of the high density region, where there is a
larger amount of scatter in the data together with bigger error bars, EPM2, TraPPE and SAFT dimer agree well with each other for the viscosity ratio. In the high density region the SAFT dimer model gives the lowest values compared to the atomistic and the monomer models. The dilute gas contribution, \( \kappa_{\text{dilute}} \), is not considered here as it is the same for all models.

6 Additional data: CO\(_2\) in the two-phase region

In order to investigate the simulation box size dependence of transport properties in the two-phase regions, we have simulated CO\(_2\) at \( \rho = 0.6 \) g/cm\(^3\) using different numbers of molecules: 500, 1372, 2048 (as presented in the paper), 4000, and 6912. The results are presented in Fig. S7. A clear dependence of the quantities on the simulation box size/number of molecules is observed. Notably, for all but the smallest box, \( \kappa_{\text{conf}}/\eta \) agree relatively well (almost within their error). Therefore we conclude that the results presented in the paper may still serve as a guide to the general behaviour in a two-phase region.

7 Additional data: Influence of constraints on n-decane at 300K

The constraints placed on the bonds and angles in alkanes such as n-decane are not usually treated with much care. In the work presented in the paper we constrained both the bonds involving hydrogens and the H-C-H angle for the OPLS force field, whilst this force field is often employed without any constraints whatsoever. However, it has been stated in the literature that for certain properties these constraints can have a significant impact [11].

We present here data for n-decane, where either both the hydrogen bonds and the H-C-H angle is constrained (OPLS, as presented in the paper) or both are governed by a harmonic potential (OPLS (flexible)). Our work shows that for the shear viscosities, while there are differences, they are small (see Fig. S8a). It should be noted that the rigid model achieves slightly better accuracy with regard to NIST data than its flexible counter part. The difference is more notable in the viscosity ratio (Fig. S8b). The ratio
Figure S6: $\eta$ and $\kappa_{conf}/\eta$ for the rigid EPM2, TraPPE, SAFT dimer and SAFT monomer models at the supercritical temperature of $T = 323K$. 
Figure S7: $\eta$, $\kappa$ and $\kappa_{\text{conf}}/\eta$ for the rigid EPM2 model with different numbers of molecules simulated.
of the flexible model is about 20 % bigger. Notably, the rigid model is in agreement with the experimental value [12]. For L-OPLS a similar investigation was conducted with no notable difference observed. As only one experimental observation was found to date, this may in practice lie within the error of the measurement. However, in any case it highlights an issue with how force fields for alkanes are used and presented. Further analysis needs to be done to confirm which constraints produce the results most in line with the true physical molecule.

8 Compressibilities for different water models

In order to investigate the differences in predicting the bulk viscosity between the SAFT monomer and atomistic models (SPC/E and TIP4P/2005), we calculate the compressibilities, $\beta_c$, for all three models at 300 K. As a relation between the bulk viscosity and the bulk modulus, $K = 1/\beta_c$, is proposed in [13], a difference in the models could give an insight into the reasons for the varying performance. The isothermal bulk viscosity is defined through

$$\beta_{c,T} = -\frac{1}{V_0} \left( \frac{\partial V}{\partial P} \right)_T,$$

where $V$ is the volume, $P$ the pressure and $V_0$ the volume at the desired pressure. In practice, we calculate this by performing simulations at different pressures either side from the target pressure (1 bar) in steps of 50 bar, i.e. between -100 and 100 bar. The slope of the resulting curve is then used to calculate $\partial V/\partial P$. The simulations are performed in an NPT ensemble and individual simulation runs last 2 ns each. The resulting compressibilities are given in Tab. S6. Whilst the SAFT monomer model is furthest from the experimental data we can nevertheless not see a correlation between success in computing $\kappa$ and $\beta_c$. The values for TIP4P/2005 and SAFT monomer are very similar, yet the resulting bulk viscosities are not. Further, SPC/E shows the best agreement with experiment but predicts values of $\kappa$ which are lower than both experiment and TIP4P/2005. Therefore the conclusion of this preliminary analysis is that the ability of a force field to obtain the correct compressibility does not influence its success in calculating the bulk viscosity, $\kappa$. 

S-15
Figure S8: $\eta$ and $\kappa/\eta$ for n-decane at 300 K. Two versions of the atomistic OPLS model is presented; one with rigid hydrogen bonds and a rigid H-C-H angle, and one fully flexible one.
Table S6: Compressibilities of water at 300 K and $P = 1$ bar for the SPC/E, TIP4P/2005 and SAFT monomer (SAFT-ift) water models compared to the experimental value [14].

| Model           | $\beta_c \times 10^{-6}$ (bar$^{-1}$) |
|-----------------|----------------------------------------|
| SPC/E           | 47.8                                   |
| TIP4P/2005      | 51.7                                   |
| SAFT Monomer    | 52.3                                   |
| Exp.            | 45.2                                   |

9 Benchmark testing for $\kappa_{\text{dilute}}$

Benchmark testing of $\kappa_{\text{dilute}}$ was performed by the example of CO$_2$ (i.e. using values of $c_v$ for CO$_2$) in order to assess the importance of the relaxation times, $\tau_{\text{rot}}$ and $\tau_{\text{vib}}$, and the wave number, $k$, on the rotational and vibrational contributions to $\kappa$, $\kappa_{\text{rot}}$ and $\kappa_{\text{vib}}$. The main conclusions are presented in the paper. The dependence on the parameters is illustrated in Figs. S9 and S10.

10 Complete data for the viscosities at different state points

Below is a collection of the data for water ($T = (300, 393, 613)$ K, models = (SPC/E, TIP4P/2005, SAFT1-ift, SAFT1-vle), CO$_2$ ($T = (300, 323)$ K, models = (EPM2, TraPPE, SAFT dimer, SAFT monomer) and n-decane ($T = 300$ K, models = (OPLS, OPLS[flexible], L-OPLS, SAFT three-bead). The density, pressure, shear viscosity, bulk viscosity and viscosity ratio are given for each data point.

11 Files

Example input files and the numerical results presented in the figures and tables above are deposited in the following data repository:

https://doi.org/10.6084/m9.figshare.5799132
Figure S9: $\kappa_{\text{rot}}$ and $\kappa_{\text{vib}}$ as a function of the relaxation times, $\tau_{\text{rot}}$ and $\tau_{\text{vib}}$. Two different values for the wave number, $k$, were chosen for $\kappa_{\text{vib}}$. 

(a) $\kappa_{\text{rot}}$

(b) $\kappa_{\text{vib}}, \ k = 700 \ \text{cm}^{-1}$

(c) $\kappa_{\text{vib}}, \ k = 3000 \ \text{cm}^{-1}$
Figure S10: Dependence of $\kappa_{\text{vib}}$ on the wavenumber, $k$, at different temperatures. The relaxation time was set to $\tau_{\text{vib}} = 4\mu$s.
### Table S7: Shear and bulk viscosity data for the SPC/E water model at $T = 300$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa$ (mPas) | $\kappa/\eta$ |
|-------------------|-----------|----------------|-----------------|---------------|
| 0.997             | 105.0     | 0.69 ± 0.019   | 1.532 ± 0.051   | 2.018 ± 0.097 |
| 1.002             | 208.4     | 0.681 ± 0.005  | 1.521 ± 0.078   | 2.301 ± 0.076 |
| 1.006             | 309.2     | 0.672 ± 0.019  | 1.648 ± 0.074   | 2.485 ± 0.246 |
| 1.01              | 409.1     | 0.688 ± 0.033  | 1.316 ± 0.086   | 1.914 ± 0.11  |
| 1.014             | 506.6     | 0.658 ± 0.031  | 1.407 ± 0.093   | 2.061 ± 0.178 |
| 1.018             | 605.4     | 0.666 ± 0.003  | 1.754 ± 0.03    | 2.578 ± 0.062 |
| 1.023             | 705.1     | 0.69 ± 0.026   | 1.746 ± 0.205   | 2.548 ± 0.252 |
| 1.026             | 806.1     | 0.655 ± 0.011  | 1.481 ± 0.202   | 2.162 ± 0.236 |
| 1.03              | 905.5     | 0.709 ± 0.018  | 2.0 ± 0.304     | 2.787 ± 0.492 |

### Table S8: Shear and bulk viscosity data for the TIP4P/2005 water model at $T = 300$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa$ (mPas) | $\kappa/\eta$ |
|-------------------|-----------|----------------|-----------------|---------------|
| 0.997             | 134.1     | 0.832 ± 0.04   | 2.161 ± 0.079   | 2.565 ± 0.171 |
| 1.002             | 236.0     | 0.839 ± 0.039  | 1.866 ± 0.13    | 2.22 ± 0.08  |
| 1.006             | 327.6     | 0.836 ± 0.0    | 2.317 ± 0.0     | 2.836 ± 0.0  |
| 1.01              | 432.7     | 0.792 ± 0.016  | 2.175 ± 0.073   | 2.814 ± 0.077 |
| 1.014             | 529.4     | 0.856 ± 0.005  | 2.174 ± 0.049   | 2.407 ± 0.073 |
| 1.018             | 630.2     | 0.762 ± 0.035  | 2.502 ± 0.266   | 3.359 ± 0.258 |
| 1.023             | 728.1     | 0.769 ± 0.004  | 2.297 ± 0.166   | 3.076 ± 0.305 |
| 1.026             | 833.0     | 0.778 ± 0.017  | 1.935 ± 0.15    | 2.299 ± 0.217 |
| 1.03              | 925.9     | 0.787 ± 0.017  | 2.178 ± 0.02    | 2.669 ± 0.037 |

### Table S9: Shear and bulk viscosity data for the SAFT1-ift water model at $T = 300$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa$ (mPas) | $\kappa/\eta$ |
|-------------------|-----------|----------------|-----------------|---------------|
| 0.997             | 12.3      | 0.247 ± 0.012  | 0.099 ± 0.017   | 0.388 ± 0.06  |
| 1.002             | 99.8      | 0.275 ± 0.007  | 0.096 ± 0.024   | 0.329 ± 0.077 |
| 1.006             | 188.5     | 0.274 ± 0.006  | 0.168 ± 0.031   | 0.628 ± 0.136 |
| 1.01              | 278.3     | 0.287 ± 0.012  | 0.065 ± 0.002   | 0.232 ± 0.006 |
| 1.014             | 369.0     | 0.274 ± 0.014  | 0.071 ± 0.01    | 0.268 ± 0.053 |
| 1.018             | 461.9     | 0.286 ± 0.005  | 0.065 ± 0.003   | 0.239 ± 0.007 |
| 1.023             | 554.5     | 0.283 ± 0.016  | 0.081 ± 0.001   | 0.297 ± 0.027 |
| 1.026             | 645.4     | 0.317 ± 0.005  | 0.06 ± 0.002    | 0.189 ± 0.005 |
| 1.03              | 736.6     | 0.305 ± 0.009  | 0.154 ± 0.062   | 0.5 ± 0.205  |
Table S10: Shear and bulk viscosity data for the SPC/E water model at $T = 393$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}}/\eta$ |
|-----------------|----------|---------------|------------------|------------------|
| 0.948           | 482.0    | 0.235 ± 0.013 | 0.864 ± 0.036    | 3.717 ± 0.367    |
| 0.953           | 569.9    | 0.247 ± 0.015 | 0.827 ± 0.027    | 3.362 ± 0.1     |
| 0.958           | 665.6    | 0.251 ± 0.003 | 0.831 ± 0.021    | 3.314 ± 0.049    |
| 0.962           | 756.3    | 0.248 ± 0.007 | 0.701 ± 0.016    | 2.831 ± 0.018    |
| 0.966           | 849.9    | 0.238 ± 0.002 | 0.871 ± 0.028    | 3.656 ± 0.078    |
| 0.971           | 939.2    | 0.26 ± 0.0    | 0.819 ± 0.094    | 3.144 ± 0.356    |
| 0.975           | 1032.4   | 0.239 ± 0.01  | 0.97 ± 0.028     | 4.088 ± 0.29     |
| 0.979           | 1126.8   | 0.265 ± 0.002 | 0.861 ± 0.144    | 3.24 ± 0.516     |

Table S11: Shear and bulk viscosity data for the SAFT1-if$^t$ water model at $T = 393$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}}/\eta$ |
|-----------------|----------|---------------|------------------|------------------|
| 0.948           | 117.8    | 0.199 ± 0.009 | 0.077 ± 0.0       | 0.388 ± 0.015    |
| 0.953           | 189.1    | 0.184 ± 0.001 | 0.079 ± 0.014    | 0.432 ± 0.078    |
| 0.958           | 259.3    | 0.21 ± 0.007  | 0.132 ± 0.027    | 0.638 ± 0.15     |
| 0.962           | 337.7    | 0.202 ± 0.013 | 0.088 ± 0.006    | 0.444 ± 0.057    |
| 0.966           | 412.1    | 0.202 ± 0.01  | 0.076 ± 0.001    | 0.379 ± 0.013    |
| 0.971           | 486.3    | 0.205 ± 0.009 | 0.075 ± 0.002    | 0.367 ± 0.024    |
| 0.975           | 560.5    | 0.212 ± 0.008 | 0.09 ± 0.001     | 0.426 ± 0.018    |
| 0.979           | 634.0    | 0.215 ± 0.011 | 0.116 ± 0.04     | 0.522 ± 0.158    |

Table S12: Shear and bulk viscosity data for the SAFT1-vle water model at $T = 393$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}}/\eta$ |
|-----------------|----------|---------------|------------------|------------------|
| 0.948           | 131.4    | 0.445 ± 0.013 | 0.091 ± 0.004    | 0.206 ± 0.016    |
| 0.953           | 310.0    | 0.447 ± 0.015 | 0.16 ± 0.042     | 0.353 ± 0.082    |
| 0.958           | 493.0    | 0.465 ± 0.001 | 0.257 ± 0.13     | 0.554 ± 0.281    |
| 0.962           | 685.9    | 0.501 ± 0.005 | 0.109 ± 0.01     | 0.217 ± 0.022    |
| 0.966           | 867.2    | 0.491 ± 0.015 | 0.113 ± 0.029    | 0.226 ± 0.052    |
| 0.971           | 1060.1   | 0.503 ± 0.011 | 0.105 ± 0.014    | 0.208 ± 0.024    |
| 0.975           | 1240.3   | 0.465 ± 0.017 | 0.103 ± 0.013    | 0.224 ± 0.037    |
| 0.979           | 1429.3   | 0.465 ± 0.002 | 0.126 ± 0.034    | 0.269 ± 0.073    |
Table S13: Shear and bulk viscosity data for the SPC/E water model at \( T = 613 \) K.

| \( \rho \) (g/cm\(^3\)) | \( P \) (bar) | \( \eta \) (mPas) | \( \kappa_{\text{conf}} \) (mPas) | \( \kappa_{\text{conf}} / \eta \) |
|-------------------------|-------------|-----------------|-----------------|-----------------|
| 0.0004                  | 1.0         | 0.001 ± 0.0     | 0.001 ± 0.0     | 0.864 ± 0.0     |
| 0.0472                  | 85.3        | 0.018 ± 0.001   | 0.218 ± 0.096   | 11.834 ± 1.332  |
| 0.0544                  | 91.2        | 0.018 ± 0.0     | 0.119 ± 0.0     | 6.759 ± 0.0     |
| 0.0692                  | 103.3       | 0.021 ± 0.0     | 0.271 ± 0.0     | 12.588 ± 0.0    |
| 0.0898                  | 113.7       | 0.017 ± 0.0     | 0.305 ± 0.0     | 17.451 ± 0.0    |
| 0.1196                  | 121.5       | 0.022 ± 0.0     | 0.209 ± 0.0     | 9.528 ± 0.0     |
| 0.2335                  | 124.4       | 0.025 ± 0.0     | 0.078 ± 0.0     | 3.06 ± 0.0      |
| 0.3486                  | 119.7       | 0.036 ± 0.0     | 0.196 ± 0.0     | 5.425 ± 0.0     |
| 0.4354                  | 127.6       | 0.043 ± 0.0     | 0.506 ± 0.0     | 11.635 ± 0.0    |
| 0.5535                  | 201.0       | 0.059 ± 0.0     | 0.548 ± 0.0     | 9.351 ± 0.0     |
| 0.6381                  | 400.3       | 0.077 ± 0.003   | 0.446 ± 0.006   | 5.847 ± 0.127   |
| 0.6703                  | 537.4       | 0.077 ± 0.0     | 0.429 ± 0.0     | 5.585 ± 0.0     |
| 0.6936                  | 662.4       | 0.085 ± 0.001   | 0.594 ± 0.011   | 7.004 ± 0.271   |
| 0.7122                  | 782.9       | 0.083 ± 0.002   | 0.52 ± 0.082    | 6.228 ± 0.824   |
| 0.728                   | 904.5       | 0.093 ± 0.003   | 1.626 ± 0.117   | 17.497 ± 0.772  |

Table S14: Shear and bulk viscosity data for the TIP4P/2005 water model at \( T = 613 \) K.

| \( \rho \) (g/cm\(^3\)) | \( P \) (bar) | \( \eta \) (mPas) | \( \kappa_{\text{conf}} \) (mPas) | \( \kappa_{\text{conf}} / \eta \) |
|-------------------------|-------------|-----------------|-----------------|-----------------|
| 0.0004                  | 1.0         | 0.001 ± 0.0     | 0.0 ± 0.025     | 0.499 ± 1.206   |
| 0.0472                  | 85.3        | 0.019 ± 0.001   | 0.083 ± 0.007   | 4.491 ± 0.145   |
| 0.0544                  | 91.6        | 0.02 ± 0.0      | 0.058 ± 0.012   | 2.929 ± 0.575   |
| 0.0692                  | 101.8       | 0.017 ± 0.0     | 0.07 ± 0.013    | 4.104 ± 0.673   |
| 0.0898                  | 110.3       | 0.018 ± 0.0     | 0.045 ± 0.001   | 2.587 ± 0.045   |
| 0.1196                  | 115.4       | 0.02 ± 0.001    | 0.051 ± 0.008   | 2.485 ± 0.285   |
| 0.164                   | 113.4       | 0.022 ± 0.0     | 0.089 ± 0.0     | 4.145 ± 0.142   |
| 0.2335                  | 109.2       | 0.026 ± 0.0     | 0.088 ± 0.011   | 3.345 ± 0.387   |
| 0.3486                  | 91.5        | 0.038 ± 0.002   | 0.154 ± 0.019   | 4.088 ± 0.331   |
| 0.4354                  | 79.0        | 0.046 ± 0.001   | 0.256 ± 0.003   | 5.55 ± 0.075    |
| 0.5535                  | 104.1       | 0.063 ± 0.001   | 0.239 ± 0.015   | 3.829 ± 0.139   |
| 0.6381                  | 264.5       | 0.077 ± 0.002   | 0.26 ± 0.012    | 3.374 ± 0.254   |
| 0.6703                  | 378.7       | 0.077 ± 0.0     | 0.261 ± 0.017   | 3.423 ± 0.976   |
| 0.6936                  | 496.0       | 0.093 ± 0.003   | 0.202 ± 0.037   | 2.176 ± 0.737   |
| 0.7122                  | 605.2       | 0.087 ± 0.002   | 0.299 ± 0.042   | 3.416 ± 0.407   |
| 0.728                   | 722.6       | 0.086 ± 0.004   | 0.41 ± 0.073    | 4.697 ± 0.627   |
Table S15: Shear and bulk viscosity data for the SAFT1-vle water model at $T = 613$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}} / \eta$ |
|-------------------|-----------|----------------|-------------------------------|-------------------------------|
| 0.004             | 1.0       | 0.001 ± 0.0    | 0.0 ± 0.0                     | 0.0 ± 0.0                     |
| 0.0472            | 108.3     | 0.022 ± 0.0    | 0.001 ± 0.0                   | 0.023 ± 0.0                   |
| 0.0544            | 120.5     | 0.023 ± 0.001  | 0.001 ± 0.0                   | 0.043 ± 0.0                   |
| 0.0692            | 142.2     | 0.022 ± 0.0    | 0.002 ± 0.0                   | 0.071 ± 0.002                 |
| 0.0898            | 164.9     | 0.024 ± 0.0    | 0.004 ± 0.0                   | 0.165 ± 0.0                   |
| 0.1196            | 183.3     | 0.026 ± 0.001  | 0.02 ± 0.001                  | 0.781 ± 0.001                 |
| 0.164             | 166.1     | 0.028 ± 0.001  | 0.186 ± 0.002                 | 6.595 ± 0.052                 |
| 0.2335            | 146.0     | 0.034 ± 0.0    | 0.187 ± 0.002                 | 5.503 ± 0.016                 |
| 0.3486            | 94.5      | 0.048 ± 0.0    | 0.398 ± 0.028                 | 8.215 ± 0.537                 |
| 0.5535            | −48.3     | 0.092 ± 0.002  | 0.21 ± 0.006                  | 2.282 ± 0.044                 |
| 0.6143            | 219.9     | 0.092 ± 0.003  | 0.126 ± 0.009                 | 1.371 ± 0.049                 |
| 0.6381            | 407.4     | 0.107 ± 0.009  | 0.098 ± 0.002                 | 0.915 ± 0.058                 |
| 0.6703            | 761.4     | 0.134 ± 0.0    | 0.078 ± 0.0                   | 0.59 ± 0.0                     |
| 0.6936            | 1089.4    | 0.144 ± 0.001  | 0.067 ± 0.0                   | 0.464 ± 0.0                   |
| 0.7122            | 1404.1    | 0.161 ± 0.0    | 0.073 ± 0.0                   | 0.454 ± 0.0                   |
| 0.728             | 1715.4    | 0.161 ± 0.001  | 0.074 ± 0.001                 | 0.461 ± 0.004                 |

Table S16: Shear and bulk viscosity data for the EPM2 CO$_2$ model at $T = 300$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa$ (mPas) | $\kappa / \eta$ |
|-------------------|-----------|----------------|-----------------|-----------------|
| 0.066             | 31.4      | 0.014 ± 0.001  | 32.121 ± 0.007  | 2386.03 ± 28.806 |
| 0.095             | 41.7      | 0.015 ± 0.001  | 32.122 ± 0.002  | 2147.449 ± 1.542 |
| 0.133             | 53.4      | 0.017 ± 0.0    | 32.123 ± 0.003  | 1926.003 ± 13.531 |
| 0.269             | 66.4      | 0.021 ± 0.002  | 32.138 ± 0.028  | 1519.786 ± 8.01  |
| 0.3               | 64.9      | 0.023 ± 0.001  | 32.149 ± 0.0    | 1411.617 ± 188.643 |
| 0.35              | 67.9      | 0.024 ± 0.001  | 32.164 ± 0.006  | 1330.27 ± 66.091 |
| 0.4               | 68.4      | 0.028 ± 0.001  | 32.189 ± 0.008  | 1155.162 ± 6.114 |
| 0.45              | 60.8      | 0.031 ± 0.0    | 32.22 ± 0.0     | 1037.52 ± 84.955 |
| 0.5               | 59.9      | 0.035 ± 0.0    | 32.23 ± 0.003   | 890.25 ± 18.897  |
| 0.55              | 55.1      | 0.038 ± 0.0    | 32.282 ± 0.0    | 877.054 ± 45.316 |
| 0.6               | 53.6      | 0.044 ± 0.001  | 32.305 ± 0.001  | 755.436 ± 25.476 |
| 0.803             | 59.2      | 0.077 ± 0.002  | 32.337 ± 0.029  | 410.421 ± 19.096 |
| 0.906             | 171.5     | 0.098 ± 0.001  | 32.32 ± 0.022   | 338.235 ± 1.495  |
| 0.96              | 272.4     | 0.115 ± 0.001  | 32.302 ± 0.008  | 292.696 ± 9.419  |
| 0.999             | 375.7     | 0.127 ± 0.001  | 32.335 ± 0.024  | 254.717 ± 8.775  |
| 1.029             | 460.2     | 0.135 ± 0.002  | 32.322 ± 0.014  | 235.3 ± 5.51     |
| 1.055             | 572.5     | 0.157 ± 0.004  | 32.314 ± 0.013  | 210.468 ± 4.868  |
| 1.077             | 696.0     | 0.147 ± 0.001  | 32.352 ± 0.013  | 216.659 ± 19.399 |
Table S17: Shear and bulk viscosity data for the TraPPE CO\(_2\) model at \(T = 300\) K.

| \(\rho\) (g/cm\(^3\)) | \(P\) (bar) | \(\eta\) (mPas) | \(\kappa\) (mPas) | \(\kappa/\eta\) |
|------------------------|-------------|-----------------|------------------|---------------|
| 0.066                  | 32.1        | 0.014 ± 0.0     | 32.121 ± 0.012   | 2194.316 ± 3.405 |
| 0.095                  | 41.9        | 0.016 ± 0.0     | 32.122 ± 0.001   | 1971.881 ± 19.13 |
| 0.133                  | 54.1        | 0.017 ± 0.0     | 32.124 ± 0.0     | 1906.14 ± 21.243 |
| 0.269                  | 72.4        | 0.022 ± 0.008   | 32.135 ± 0.004   | 1476.152 ± 10.44 |
| 0.3                    | 71.5        | 0.022 ± 0.0     | 32.144 ± 0.0     | 1413.32 ± 36.043 |
| 0.35                   | 72.5        | 0.025 ± 0.001   | 32.161 ± 0.002   | 1310.152 ± 3.181 |
| 0.4                    | 60.5        | 0.029 ± 0.003   | 32.196 ± 0.004   | 1141.722 ± 46.731 |
| 0.45                   | 68.0        | 0.032 ± 0.0     | 32.216 ± 0.0     | 1024.029 ± 36.78 |
| 0.5                    | 66.2        | 0.036 ± 0.0     | 32.241 ± 0.001   | 887.772 ± 6.582 |
| 0.55                   | 59.2        | 0.042 ± 0.0     | 32.26 ± 0.0      | 794.979 ± 24.905 |
| 0.6                    | 60.4        | 0.047 ± 0.0     | 32.278 ± 0.001   | 708.104 ± 22.523 |
| 0.803                  | 112.4       | 0.075 ± 0.003   | 32.309 ± 0.017   | 413.358 ± 21.753 |
| 0.906                  | 247.0       | 0.099 ± 0.008   | 32.362 ± 0.004   | 322.543 ± 8.757 |
| 0.96                   | 341.9       | 0.121 ± 0.001   | 32.34 ± 0.003    | 266.753 ± 0.499 |
| 0.999                  | 463.8       | 0.14 ± 0.001    | 32.33 ± 0.015    | 236.539 ± 7.346 |
| 1.029                  | 586.4       | 0.145 ± 0.001   | 32.344 ± 0.023   | 222.997 ± 4.747 |
| 1.055                  | 688.6       | 0.16 ± 0.005    | 32.358 ± 0.003   | 205.195 ± 3.764 |
| 1.077                  | 844.8       | 0.183 ± 0.002   | 32.33 ± 0.009    | 168.699 ± 20.927 |

Table S18: Shear and bulk viscosity data for the SAFT dimer CO\(_2\) model at \(T = 300\) K.

| \(\rho\) (g/cm\(^3\)) | \(P\) (bar) | \(\eta\) (mPas) | \(\kappa\) (mPas) | \(\kappa/\eta\) |
|------------------------|-------------|-----------------|------------------|---------------|
| 0.066                  | 31.6        | 0.013 ± 0.001   | 32.121 ± 0.004   | 2529.409 ± 38.636 |
| 0.095                  | 42.1        | 0.013 ± 0.001   | 32.121 ± 0.012   | 2408.441 ± 14.458 |
| 0.133                  | 52.9        | 0.016 ± 0.002   | 32.123 ± 0.005   | 1981.374 ± 53.609 |
| 0.269                  | 72.4        | 0.02 ± 0.002    | 32.136 ± 0.012   | 1588.841 ± 16.112 |
| 0.3                    | 73.8        | 0.023 ± 0.001   | 32.147 ± 0.001   | 1486.108 ± 144.781 |
| 0.35                   | 74.7        | 0.027 ± 0.0     | 32.165 ± 0.005   | 1156.416 ± 34.852 |
| 0.4                    | 75.5        | 0.03 ± 0.001    | 32.182 ± 0.031   | 1098.898 ± 24.77 |
| 0.45                   | 74.7        | 0.033 ± 0.0     | 32.222 ± 0.0     | 971.442 ± 23.866 |
| 0.5                    | 73.9        | 0.037 ± 0.001   | 32.256 ± 0.0     | 848.071 ± 37.848 |
| 0.55                   | 73.6        | 0.043 ± 0.0     | 32.305 ± 0.0     | 737.218 ± 14.062 |
| 0.6                    | 73.8        | 0.048 ± 0.0     | 32.339 ± 0.004   | 670.959 ± 42.513 |
| 0.803                  | 127.3       | 0.082 ± 0.001   | 32.329 ± 0.008   | 388.577 ± 4.449 |
| 0.906                  | 255.8       | 0.106 ± 0.001   | 32.285 ± 0.015   | 310.034 ± 5.696 |
| 0.96                   | 379.8       | 0.132 ± 0.005   | 32.3 ± 0.007     | 245.36 ± 9.569 |
| 0.999                  | 502.4       | 0.135 ± 0.002   | 32.301 ± 0.003   | 237.918 ± 8.707 |
| 1.029                  | 624.9       | 0.155 ± 0.003   | 32.294 ± 0.003   | 225.936 ± 9.978 |
| 1.055                  | 746.5       | 0.168 ± 0.003   | 32.295 ± 0.001   | 199.51 ± 7.577 |
| 1.077                  | 864.3       | 0.157 ± 0.002   | 32.297 ± 0.009   | 210.798 ± 5.792 |
Table S19: Shear and bulk viscosity data for the SAFT monomer CO\textsubscript{2} model at \( T = 300 \) K.

| \( \rho \) (g/cm\textsuperscript{3}) | \( P \) (bar) | \( \eta \) (mPas) | \( \kappa \) (mPas) | \( \kappa/\eta \) |
|-----------------|------------|----------------|----------------|-----------------|
| 0.066 | 31.5 | 0.014 ± 0.004 | 32.12 ± 0.003 | 2234.981 ± 7.396 |
| 0.095 | 41.9 | 0.015 ± 0.0 | 32.121 ± 0.0 | 2252.646 ± 93.524 |
| 0.133 | 52.4 | 0.017 ± 0.001 | 32.123 ± 0.003 | 1914.995 ± 4.993 |
| 0.269 | 69.2 | 0.022 ± 0.009 | 32.163 ± 0.013 | 1551.071 ± 5.71 |
| 0.3 | 68.7 | 0.021 ± 0.005 | 32.179 ± 0.008 | 1484.123 ± 5.494 |
| 0.35 | 68.4 | 0.026 ± 0.006 | 32.209 ± 0.043 | 1245.36 ± 0.553 |
| 0.4 | 65.9 | 0.028 ± 0.0 | 32.293 ± 0.0 | 1120.942 ± 3.076 |
| 0.45 | 65.0 | 0.034 ± 0.002 | 32.313 ± 0.01 | 932.982 ± 38.642 |
| 0.5 | 62.1 | 0.042 ± 0.0 | 32.395 ± 0.02 | 791.611 ± 4.444 |
| 0.55 | 60.8 | 0.045 ± 0.0 | 32.42 ± 0.007 | 699.534 ± 47.632 |
| 0.6 | 57.5 | 0.055 ± 0.01 | 32.502 ± 0.009 | 564.052 ± 8.227 |
| 0.804 | 75.4 | 0.088 ± 0.001 | 32.519 ± 0.004 | 378.187 ± 15.769 |
| 0.906 | 167.2 | 0.138 ± 0.001 | 32.451 ± 0.0 | 242.452 ± 34.709 |
| 0.96 | 274.7 | 0.154 ± 0.0 | 32.461 ± 0.024 | 217.891 ± 0.384 |
| 0.999 | 386.3 | 0.171 ± 0.0 | 32.399 ± 0.003 | 192.492 ± 44.607 |
| 1.029 | 501.8 | 0.206 ± 0.006 | 32.41 ± 0.007 | 159.746 ± 6.518 |
| 1.055 | 622.6 | 0.225 ± 0.001 | 32.436 ± 0.016 | 148.877 ± 13.995 |
| 1.077 | 744.7 | 0.254 ± 0.001 | 32.429 ± 0.054 | 130.867 ± 2.912 |
Table S20: Shear and bulk viscosity data for the EPM2 CO$_2$ model at $T = 323$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}}/\eta$ |
|-------------------|-----------|----------------|------------------------------|-----------------------------|
| 0.269             | 87.6      | 0.023 ± 0.001  | 0.017 ± 0.002                | 0.732 ± 0.093               |
| 0.3               | 94.0      | 0.024 ± 0.001  | 0.023 ± 0.001                | 0.939 ± 0.031               |
| 0.35              | 93.4      | 0.024 ± 0.001  | 0.031 ± 0.001                | 1.288 ± 0.001               |
| 0.4               | 102.8     | 0.029 ± 0.002  | 0.037 ± 0.039                | 1.281 ± 0.278               |
| 0.45              | 94.4      | 0.031 ± 0.001  | 0.065 ± 0.004                | 2.089 ± 0.158               |
| 0.5               | 108.0     | 0.035 ± 0.002  | 0.071 ± 0.003                | 2.044 ± 0.016               |
| 0.55              | 115.4     | 0.04 ± 0.001   | 0.086 ± 0.002                | 2.148 ± 0.012               |
| 0.6               | 132.1     | 0.047 ± 0.0    | 0.127 ± 0.009                | 2.685 ± 0.206               |
| 0.803             | 190.8     | 0.075 ± 0.001  | 0.16 ± 0.002                 | 2.124 ± 0.082               |
| 0.906             | 360.5     | 0.103 ± 0.006  | 0.253 ± 0.011                | 2.473 ± 0.138               |
| 0.96              | 478.5     | 0.116 ± 0.005  | 0.22 ± 0.004                 | 1.895 ± 0.108               |
| 0.999             | 578.4     | 0.129 ± 0.008  | 0.284 ± 0.025                | 2.249 ± 0.339               |
| 1.029             | 738.5     | 0.128 ± 0.005  | 0.196 ± 0.021                | 1.522 ± 0.104               |
| 1.055             | 804.2     | 0.15 ± 0.001   | 0.276 ± 0.047                | 1.847 ± 0.473               |
| 1.077             | 955.4     | 0.152 ± 0.0    | 0.247 ± 0.013                | 1.642 ± 0.182               |

Table S21: Shear and bulk viscosity data for the TraPPE CO$_2$ model at $T = 323$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}}/\eta$ |
|-------------------|-----------|----------------|------------------------------|-----------------------------|
| 0.269             | 91.7      | 0.024 ± 0.0    | 0.014 ± 0.002                | 0.593 ± 0.081               |
| 0.3               | 97.0      | 0.023 ± 0.0    | 0.025 ± 0.001                | 1.074 ± 0.022               |
| 0.35              | 100.5     | 0.025 ± 0.0    | 0.033 ± 0.001                | 1.282 ± 0.065               |
| 0.4               | 102.8     | 0.028 ± 0.0    | 0.049 ± 0.006                | 1.737 ± 0.038               |
| 0.45              | 107.1     | 0.032 ± 0.001  | 0.063 ± 0.0                  | 1.95 ± 0.039                |
| 0.5               | 110.8     | 0.034 ± 0.0    | 0.083 ± 0.002                | 2.409 ± 0.077               |
| 0.55              | 108.8     | 0.04 ± 0.0     | 0.094 ± 0.004                | 2.353 ± 0.123               |
| 0.6               | 131.2     | 0.044 ± 0.001  | 0.114 ± 0.007                | 2.561 ± 0.121               |
| 0.803             | 217.8     | 0.077 ± 0.001  | 0.152 ± 0.007                | 1.973 ± 0.233               |
| 0.906             | 403.0     | 0.099 ± 0.008  | 0.221 ± 0.007                | 2.246 ± 0.026               |
| 0.96              | 531.7     | 0.118 ± 0.001  | 0.232 ± 0.041                | 1.952 ± 0.331               |
| 0.999             | 698.8     | 0.128 ± 0.002  | 0.281 ± 0.013                | 2.193 ± 0.132               |
| 1.029             | 909.0     | 0.147 ± 0.002  | 0.417 ± 0.015                | 2.837 ± 0.067               |
| 1.055             | 947.5     | 0.162 ± 0.007  | 0.244 ± 0.001                | 1.506 ± 0.145               |
| 1.077             | 1071.3    | 0.181 ± 0.002  | 0.274 ± 0.002                | 1.516 ± 0.081               |
Table S22: Shear and bulk viscosity data for the SAFT dimer CO$_2$ model at $T = 323$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}} / \eta$ |
|-------------------|-----------|---------------|-------------------------------|-----------------------------|
| 0.269             | 92.6      | 0.022 ± 0.0   | 0.014 ± 0.0                  | 0.614 ± 0.033               |
| 0.3               | 97.1      | 0.023 ± 0.001 | 0.019 ± 0.0                  | 0.823 ± 0.019               |
| 0.35              | 103.6     | 0.026 ± 0.001 | 0.027 ± 0.001                | 1.011 ± 0.077               |
| 0.4               | 108.9     | 0.031 ± 0.002 | 0.046 ± 0.006                | 1.495 ± 0.028               |
| 0.45              | 114.2     | 0.033 ± 0.001 | 0.061 ± 0.001                | 1.844 ± 0.091               |
| 0.5               | 120.3     | 0.037 ± 0.0   | 0.075 ± 0.002                | 2.02 ± 0.026                |
| 0.55              | 126.4     | 0.042 ± 0.001 | 0.094 ± 0.001                | 2.284 ± 0.059               |
| 0.6               | 135.9     | 0.048 ± 0.001 | 0.131 ± 0.005                | 2.712 ± 0.054               |
| 0.803             | 254.2     | 0.075 ± 0.001 | 0.154 ± 0.01                 | 2.052 ± 0.307               |
| 0.906             | 434.1     | 0.114 ± 0.001 | 0.164 ± 0.003                | 1.44 ± 0.023                |
| 0.96              | 591.3     | 0.115 ± 0.0   | 0.137 ± 0.0                  | 1.199 ± 0.0                 |
| 0.999             | 740.8     | 0.14 ± 0.004  | 0.137 ± 0.008                | 0.985 ± 0.091               |
| 1.029             | 886.5     | 0.158 ± 0.0   | 0.188 ± 0.0                  | 1.187 ± 0.0                 |
| 1.055             | 1027.0    | 0.151 ± 0.006 | 0.136 ± 0.001                | 0.899 ± 0.083               |
| 1.077             | 1165.7    | 0.178 ± 0.0   | 0.144 ± 0.003                | 0.808 ± 0.055               |

Table S23: Shear and bulk viscosity data for the SAFT monomer CO$_2$ model at $T = 323$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa_{\text{conf}}$ (mPas) | $\kappa_{\text{conf}} / \eta$ |
|-------------------|-----------|---------------|-------------------------------|-----------------------------|
| 0.269             | 88.9      | 0.022 ± 0.003 | 0.021 ± 0.02                  | 0.964 ± 0.073               |
| 0.3               | 92.7      | 0.026 ± 0.002 | 0.03 ± 0.0                    | 1.167 ± 0.075               |
| 0.35              | 97.5      | 0.025 ± 0.003 | 0.046 ± 0.007                 | 1.853 ± 0.071               |
| 0.4               | 98.9      | 0.031 ± 0.002 | 0.083 ± 0.001                 | 2.641 ± 0.033               |
| 0.45              | 102.7     | 0.037 ± 0.0   | 0.132 ± 0.0                   | 3.574 ± 0.0                 |
| 0.5               | 105.1     | 0.04 ± 0.0    | 0.137 ± 0.0                   | 3.45 ± 0.0                  |
| 0.55              | 108.8     | 0.048 ± 0.0   | 0.229 ± 0.0                   | 4.776 ± 0.0                 |
| 0.6               | 113.7     | 0.055 ± 0.009 | 0.211 ± 0.019                 | 3.818 ± 0.03                |
| 0.804             | 191.2     | 0.093 ± 0.0   | 0.277 ± 0.0                   | 2.988 ± 0.0                 |
| 0.906             | 338.4     | 0.123 ± 0.0   | 0.263 ± 0.003                 | 2.149 ± 0.16                |
| 0.96              | 478.6     | 0.176 ± 0.0   | 0.289 ± 0.0                   | 1.648 ± 0.0                 |
| 0.999             | 619.5     | 0.195 ± 0.0   | 0.283 ± 0.0                   | 1.445 ± 0.0                 |
| 1.029             | 760.3     | 0.204 ± 0.0   | 0.27 ± 0.003                  | 1.325 ± 0.014               |
| 1.055             | 902.6     | 0.22 ± 0.0    | 0.28 ± 0.0                    | 1.274 ± 0.0                 |
| 1.077             | 1046.2    | 0.249 ± 0.007 | 0.331 ± 0.048                 | 1.322 ± 0.156               |
Table S24: Shear and bulk viscosity data for the OPLS n-decane model at \( T = 300 \) K, where the hydrogen bonds and the H-C-H angle is constrained.

| \( \rho \) (g/cm\(^3\)) | \( P \) (bar) | \( \eta \) (mPas)   | \( \kappa \) (mPas) | \( \kappa / \eta \) |
|-------------------------|--------------|---------------------|---------------------|---------------------|
| 0.725                   | 15.6         | 0.715 ± 0.039       | 2.812 ± 0.174       | 3.889 ± 0.006 |
| 0.733                   | 122.1        | 0.82 ± 0.004        | 3.813 ± 0.142       | 4.769 ± 0.169 |
| 0.74                    | 224.4        | 0.902 ± 0.02        | 3.547 ± 0.236       | 3.854 ± 0.341 |
| 0.746                   | 329.5        | 1.058 ± 0.017       | 3.682 ± 0.001       | 3.378 ± 0.02  |
| 0.752                   | 435.3        | 1.111 ± 0.021       | 3.85 ± 0.121        | 3.424 ± 0.018 |
| 0.758                   | 536.8        | 1.133 ± 0.023       | 3.826 ± 0.318       | 3.389 ± 0.333 |
| 0.763                   | 645.3        | 1.338 ± 0.052       | 4.028 ± 0.332       | 3.009 ± 0.425 |
| 0.768                   | 742.5        | 1.271 ± 0.004       | 4.341 ± 0.105       | 3.428 ± 0.097 |

Table S25: Shear and bulk viscosity data for the fully flexible OPLS n-decane model at \( T = 300 \) K.

| \( \rho \) (g/cm\(^3\)) | \( P \) (bar) | \( \eta \) (mPas)   | \( \kappa \) (mPas) | \( \kappa / \eta \) |
|-------------------------|--------------|---------------------|---------------------|---------------------|
| 0.725                   | 28.8         | 0.712 ± 0.039       | 3.292 ± 0.087       | 4.751 ± 0.285 |
| 0.733                   | 139.9        | 0.781 ± 0.009       | 3.315 ± 0.128       | 4.318 ± 0.125 |
| 0.74                    | 243.3        | 0.886 ± 0.026       | 4.027 ± 0.165       | 4.596 ± 0.282 |
| 0.746                   | 350.2        | 0.978 ± 0.009       | 4.456 ± 0.205       | 4.556 ± 0.465 |
| 0.752                   | 460.7        | 1.071 ± 0.011       | 4.555 ± 0.047       | 4.347 ± 0.018 |
| 0.758                   | 563.3        | 1.099 ± 0.064       | 4.427 ± 0.19        | 4.023 ± 0.386 |
| 0.763                   | 670.2        | 1.31 ± 0.053        | 5.074 ± 0.08        | 3.892 ± 0.135 |
| 0.768                   | 774.2        | 1.322 ± 0.0         | 4.631 ± 0.182       | 3.532 ± 0.126 |

Table S26: Shear and bulk viscosity data for the L-OPLS n-decane model at \( T = 300 \) K.

| \( \rho \) (g/cm\(^3\)) | \( P \) (bar) | \( \eta \) (mPas)   | \( \kappa \) (mPas) | \( \kappa / \eta \) |
|-------------------------|--------------|---------------------|---------------------|---------------------|
| 0.725                   | 111.8        | 0.734 ± 0.058       | 2.501 ± 0.422       | 3.363 ± 0.233 |
| 0.733                   | 218.5        | 0.853 ± 0.058       | 2.762 ± 0.22        | 3.231 ± 0.02  |
| 0.74                    | 321.7        | 0.904 ± 0.021       | 2.958 ± 0.032       | 3.251 ± 0.119 |
| 0.746                   | 424.3        | 0.991 ± 0.027       | 3.366 ± 0.138       | 3.298 ± 0.126 |
| 0.752                   | 527.8        | 0.999 ± 0.031       | 3.761 ± 0.318       | 3.594 ± 0.221 |
| 0.758                   | 633.6        | 1.21 ± 0.021        | 3.573 ± 0.017       | 2.92 ± 0.06  |
| 0.763                   | 738.9        | 1.281 ± 0.016       | 3.402 ± 0.032       | 2.737 ± 0.052 |
| 0.768                   | 842.2        | 1.437 ± 0.022       | 3.813 ± 0.137       | 2.639 ± 0.108 |
Table S27: Shear and bulk viscosity data for the three-bead SAFT model at $T = 300$ K.

| $\rho$ (g/cm$^3$) | $P$ (bar) | $\eta$ (mPas) | $\kappa$ (mPas) | $\kappa/\eta$ |
|------------------|-----------|---------------|-----------------|---------------|
| 0.725            | -64.3     | 0.699 ± 0.017 | 0.66 ± 0.002    | 0.933 ± 0.021 |
| 0.733            | 23.4      | 0.747 ± 0.019 | 0.71 ± 0.018    | 0.937 ± 0.045 |
| 0.74             | 111.9     | 0.831 ± 0.027 | 0.657 ± 0.009   | 0.8 ± 0.044   |
| 0.746            | 198.4     | 0.928 ± 0.044 | 0.762 ± 0.042   | 0.78 ± 0.026  |
| 0.752            | 283.6     | 1.09 ± 0.048 | 0.879 ± 0.095   | 0.847 ± 0.141 |
| 0.758            | 371.1     | 1.129 ± 0.032 | 0.877 ± 0.069   | 0.798 ± 0.025 |
| 0.763            | 459.3     | 1.238 ± 0.03  | 1.133 ± 0.172   | 0.915 ± 0.176 |
| 0.768            | 546.9     | 1.367 ± 0.06  | 1.129 ± 0.09    | 0.858 ± 0.044 |
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