# Supplementary Material:

## Activity Coefficients of Aqueous Electrolytes from Implicit-Water Molecular Dynamics Simulations

Sina Hassanjani Saravi and Athanassios Z. Panagiotopoulos*

Department of Chemical and Biological Engineering, Princeton University, Princeton, New Jersey 08544, USA

## I. Supplementary Figures

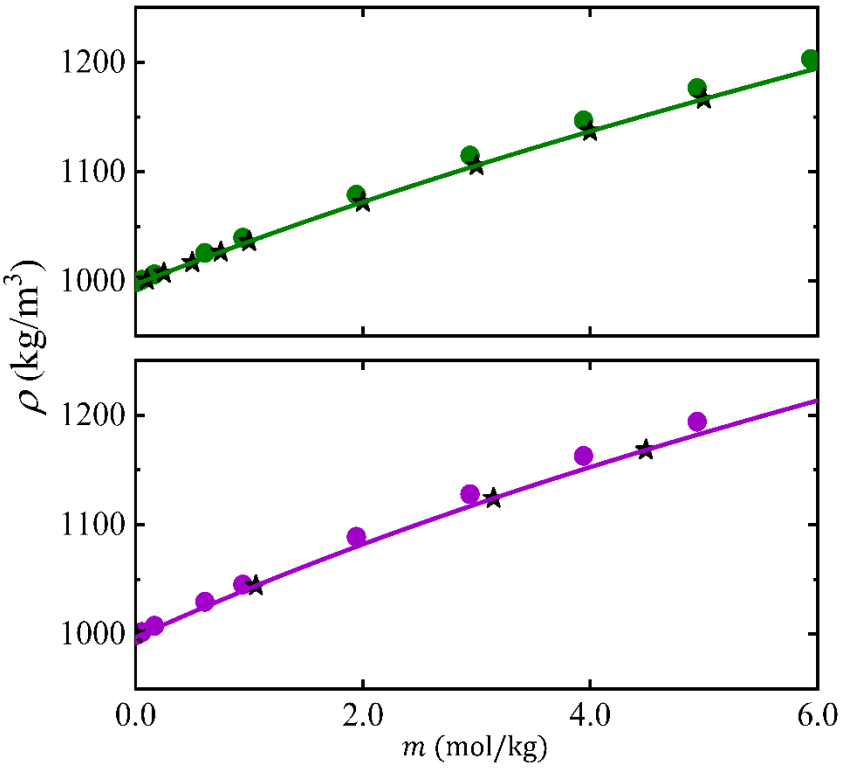


**Figure S1.** Solution density versus concentration in mol salt / kg $H_2O$ in aqueous NaCl and KCl solutions at 298.15 K and 1 bar. Green and purple circles denote simulation results for NaCl and KCl from explicit-water molecular dynamics simulations, respectively, with 5500 SPC/E[1] water molecules. Black stars in both panels show experimental data for NaCl[2] and KCl[3], fitted to cubic polynomials denoted by Green and purple solid curves, respectively. The polynomial parameters are shown in Table S49. Statistical uncertainties are smaller than symbol size.

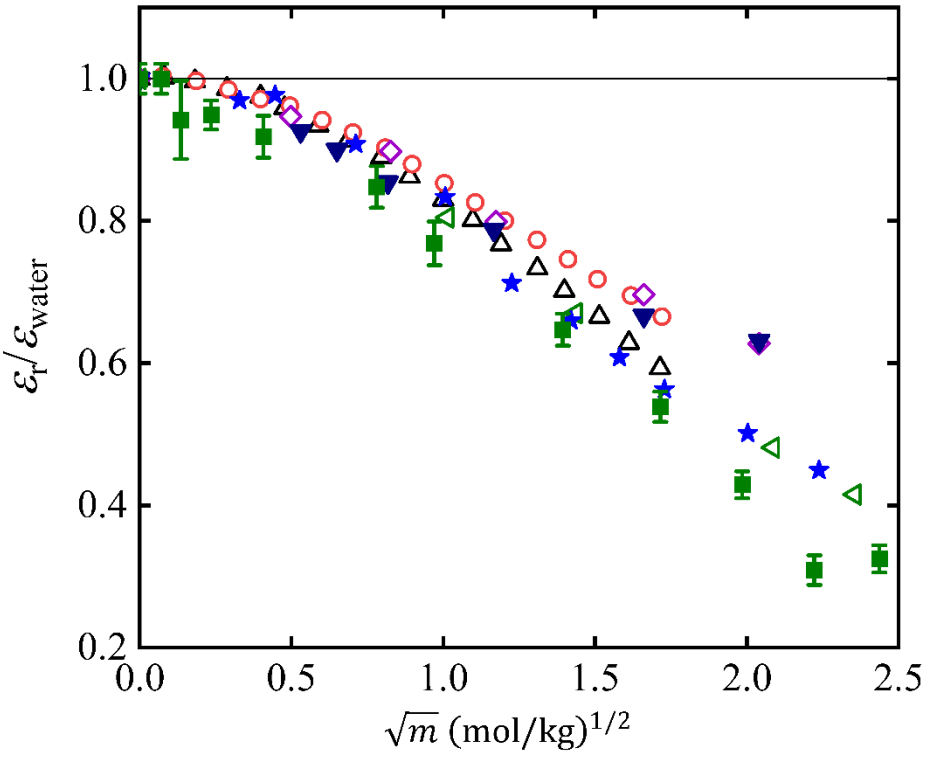


**Figure S2.** Relative permittivity ($\varepsilon_r$) in aqueous NaCl solutions versus $\sqrt{m}$, where $m$ is mol salt / kg $H_2O$, at 298.15 K and 1 bar, normalized with respect to the value of relative permittivity in pure water ($\varepsilon_{water}$). Specifically, the experimental data for $\varepsilon_r$ were normalized by 78.49, which is the experimental value for pure water[4], our simulation results were all divided by 73 ($\varepsilon_r$ for SPC/E water[1]). Filled symbols denote simulation results in explicit-water whereas open symbols represent experimental data: Squares for this study (JC[5]+SPC/E[1]), stars for Seal et al.[6] (SD[7]+SPC/E[1]), down-pointing triangles for Renou at al.[8] (polarizable force field with drude oscillator), up-pointing triangles for Barthel et al.[9], circles for Buchner et al.[10], left-pointing triangles for Christensen et al.[11], and diamonds for Smith and Dang[7]. The error bars indicate one standard deviation.

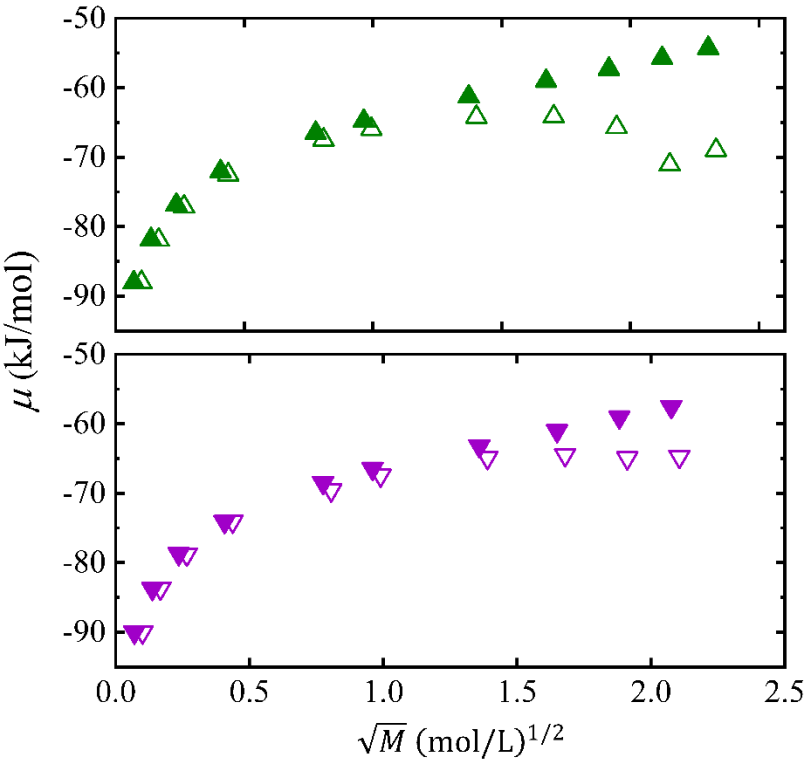


**Figure S3.** Simulation results for the chemical potential, $\mu$ (kJ/mol) in implicit-water NaCl and KCl solutions versus $\sqrt{M}$, where $M$ is mol salt / L, at 298.15 K and 1 bar. Filled green and purple triangles denote simulation results for NaCl and KCl, respectively, with $\varepsilon_r$ fixed at 73 at all concentrations. Open green and purple triangles denote results with concentration dependent $\varepsilon_r$ for NaCl and KCl, respectively. Symbol size is larger than the statistical uncertainties. Slight horizontal offset is applied to avoid overlapping symbols, where the smaller x values are the correct ones.

## II. Supplementary Tables

**Table S1.** Model parameters for SPC/E water[1] and Joung and Cheatham[5].

| Atom | $\epsilon$ (kJ/mol) | $\sigma$ (nm) | Charge |
|---|---|---|---|
| O | 0.6502 | 0.3165 | –0.8476 |
| H | - | - | 0.4238 |
| $Na^+$ | 1.4755 | 0.2159 | +1.0 |
| $K^+$ | 1.7978 | 0.2838 | +1.0 |
| $Cl^-$ | 0.0535 | 0.4830 | –1.0 |
| Geometry of SPC/E water[1] | | | |
| $R_{OH} = 0.1$ nm | | $\angle HOH = 109.47°$ | |

### Implicit-water NaCl solutions (constant relative permittivity):

**Table S2.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.005 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 998.989$ kg/m$^3$. $\mu^{IG}$ ($Na^+$) = $-43.09$ kJ/mol, $\mu^{IG}$ ($Cl^-$) = $-44.70$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -43.57 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.36 ± 0.01 | -0.36 ± 0.01 | -43.45 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.29 ± 0.03 | -0.29 ± 0.03 | -43.38 ± 0.03 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.28 ± 0.01 | -0.28 ± 0.01 | -43.37 ± 0.01 |
| | | $L \to \infty$ | | | -0.11 ± 0.02 | -43.20 ± 0.02 |
| $Cl^-$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.18 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.38 ± 0.01 | -0.38 ± 0.01 | -45.08 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.32 ± 0.02 | -0.32 ± 0.02 | -45.02 ± 0.02 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.27 ± 0.03 | -0.27 ± 0.03 | -44.97 ± 0.03 |
| | | $L \to \infty$ | | | -0.14 ± 0.02 | -44.84 ± 0.02 |

**Table S3.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.019 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 999.621\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -39.78\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -41.39\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.60 ± 0.02 | -0.60 ± 0.02 | -40.38 ± 0.02 |
| | 4.5 | 7.33 | 0.00 ± 0.00 | -0.46 ± 0.01 | -0.46 ± 0.01 | -40.24 ± 0.01 |
| | 11.5 | 10.02 | 0.00 ± 0.00 | -0.47 ± 0.03 | -0.47 ± 0.03 | -40.25 ± 0.03 |
| | 22.5 | 12.53 | 0.00 ± 0.00 | -0.41 ± 0.07 | -0.41 ± 0.07 | -40.19 ± 0.07 |
| | | $L \to \infty$ | | | -0.23 ± 0.05 | -40.01 ± 0.05 |
| $Cl^-$ | 1.5 | 5.08 | 0.01 ± 0.00 | -0.60 ± 0.02 | -0.59 ± 0.02 | -41.98 ± 0.02 |
| | 4.5 | 7.33 | 0.00 ± 0.00 | -0.55 ± 0.02 | -0.55 ± 0.02 | -41.94 ± 0.02 |
| | 11.5 | 10.02 | 0.01 ± 0.00 | -0.48 ± 0.03 | -0.47 ± 0.03 | -41.86 ± 0.03 |
| | 22.5 | 12.53 | 0.01 ± 0.00 | -0.51 ± 0.02 | -0.50 ± 0.02 | -41.89 ± 0.02 |
| | | $L \to \infty$ | | | -0.42 ± 0.03 | -41.82 ± 0.03 |

**Table S4.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.056 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1001.270\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -37.11\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -38.72\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 4.5 | 5.11 | 0.00 ± 0.00 | -0.71 ± 0.02 | -0.71 ± 0.02 | -37.82 ± 0.02 |
| | 14.5 | 7.55 | 0.00 ± 0.00 | -0.63 ± 0.04 | -0.63 ± 0.04 | -37.74 ± 0.04 |
| | 33.5 | 9.98 | 0.00 ± 0.00 | -0.61 ± 0.01 | -0.61 ± 0.01 | -37.72 ± 0.01 |
| | 65.5 | 12.49 | 0.00 ± 0.00 | -0.56 ± 0.05 | -0.56 ± 0.05 | -37.67 ± 0.05 |
| | | $L \to \infty$ | | | -0.50 ± 0.03 | -37.61 ± 0.03 |
| $Cl^-$ | 4.5 | 5.11 | 0.02 ± 0.00 | -0.72 ± 0.01 | -0.70 ± 0.01 | -39.42 ± 0.01 |
| | 14.5 | 7.55 | 0.02 ± 0.00 | -0.56 ± 0.03 | -0.54 ± 0.03 | -39.26 ± 0.03 |
| | 33.5 | 9.98 | 0.02 ± 0.00 | -0.64 ± 0.05 | -0.62 ± 0.05 | -39.34 ± 0.05 |
| | 65.5 | 12.49 | 0.03 ± 0.00 | -0.68 ± 0.03 | -0.65 ± 0.03 | -39.37 ± 0.03 |
| | | $L \to \infty$ | | | -0.53 ± 0.04 | -39.24 ± 0.04 |

**Table S5.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.166 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1006.340\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -34.40\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -36.01\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 12.5 | 5.00 | 0.01 ± 0.00 | -1.00 ± 0.04 | -0.99 ± 0.04 | -35.39 ± 0.04 |
| | 42.5 | 7.51 | 0.01 ± 0.00 | -0.87 ± 0.04 | -0.86 ± 0.04 | -35.26 ± 0.04 |
| | 100.5 | 10.01 | 0.01 ± 0.00 | -0.90 ± 0.02 | -0.89 ± 0.02 | -35.29 ± 0.02 |
| | 195.5 | 12.49 | 0.01 ± 0.00 | -0.88 ± 0.06 | -0.87 ± 0.06 | -35.27 ± 0.06 |
| | | $L \to \infty$ | | | -0.79 ± 0.05 | -35.19 ± 0.05 |
| $Cl^-$ | 12.5 | 5.00 | 0.08 ± 0.00 | -1.01 ± 0.03 | -0.93 ± 0.03 | -36.94 ± 0.03 |
| | 42.5 | 7.51 | 0.08 ± 0.00 | -1.05 ± 0.03 | -0.97 ± 0.03 | -36.98 ± 0.03 |
| | 100.5 | 10.01 | 0.07 ± 0.00 | -0.96 ± 0.03 | -0.89 ± 0.03 | -36.90 ± 0.03 |
| | 195.5 | 12.49 | 0.09 ± 0.01 | -0.94 ± 0.04 | -0.85 ± 0.04 | -36.86 ± 0.04 |
| | | $L \to \infty$ | | | -0.85 ± 0.05 | -36.86 ± 0.05 |

**Table S6.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.605 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1025.820\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -31.20\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -32.81\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 45.5 | 5.00 | 0.04 ± 0.01 | -1.39 ± 0.03 | -1.35 ± 0.03 | -32.55 ± 0.03 |
| | 153.5 | 7.50 | 0.04 ± 0.00 | -1.34 ± 0.02 | -1.30 ± 0.02 | -32.50 ± 0.02 |
| | 364.5 | 10.00 | 0.04 ± 0.01 | -1.41 ± 0.02 | -1.37 ± 0.02 | -32.57 ± 0.02 |
| | 711.5 | 12.50 | 0.04 ± 0.00 | -1.35 ± 0.06 | -1.31 ± 0.06 | -32.51 ± 0.06 |
| | | $L \to \infty$ | | | -1.35 ± 0.05 | -32.55 ± 0.05 |
| $Cl^-$ | 45.5 | 5.00 | 0.29 ± 0.01 | -1.61 ± 0.02 | -1.32 ± 0.02 | -34.13 ± 0.02 |
| | 153.5 | 7.50 | 0.31 ± 0.01 | -1.62 ± 0.02 | -1.31 ± 0.02 | -34.12 ± 0.02 |
| | 364.5 | 10.00 | 0.31 ± 0.01 | -1.47 ± 0.02 | -1.16 ± 0.02 | -33.97 ± 0.02 |
| | 711.5 | 12.50 | 0.30 ± 0.01 | -1.67 ± 0.04 | -1.37 ± 0.04 | -34.18 ± 0.04 |
| | | $L \to \infty$ | | | -1.15 ± 0.04 | -33.97 ± 0.04 |

**Table S7.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.930 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1039.760\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -30.13\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -31.75\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 70.5 | 5.01 | 0.07 ± 0.01 | -1.47 ± 0.03 | -1.40 ± 0.03 | -31.53 ± 0.03 |
| | 236.5 | 7.50 | 0.07 ± 0.00 | -1.51 ± 0.03 | -1.44 ± 0.03 | -31.57 ± 0.03 |
| | 560.5 | 10.00 | 0.06 ± 0.01 | -1.52 ± 0.04 | -1.46 ± 0.04 | -31.59 ± 0.04 |
| | 1094.5 | 12.50 | 0.06 ± 0.00 | -1.56 ± 0.04 | -1.50 ± 0.04 | -31.63 ± 0.04 |
| | | $L \to \infty$ | | | -1.55 ± 0.06 | -31.68 ± 0.06 |
| $Cl^-$ | 70.5 | 5.01 | 0.45 ± 0.01 | -1.79 ± 0.02 | -1.34 ± 0.02 | -33.09 ± 0.02 |
| | 236.5 | 7.50 | 0.48 ± 0.01 | -1.80 ± 0.03 | -1.32 ± 0.03 | -33.07 ± 0.03 |
| | 560.5 | 10.00 | 0.45 ± 0.01 | -1.79 ± 0.04 | -1.34 ± 0.04 | -33.09 ± 0.04 |
| | 1094.5 | 12.50 | 0.45 ± 0.02 | -1.84 ± 0.04 | -1.39 ± 0.04 | -33.14 ± 0.04 |
| | | $L \to \infty$ | | | -1.37 ± 0.05 | -33.11 ± 0.05 |

**Table S8.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 1.883 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1079.000\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -28.39\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -30.00\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 141.5 | 5.00 | 0.14 ± 0.01 | -1.72 ± 0.03 | -1.58 ± 0.03 | -29.97 ± 0.03 |
| | 478.5 | 7.50 | 0.14 ± 0.01 | -1.72 ± 0.03 | -1.58 ± 0.03 | -29.97 ± 0.03 |
| | 1133.5 | 10.00 | 0.13 ± 0.01 | -1.76 ± 0.02 | -1.63 ± 0.02 | -30.02 ± 0.02 |
| | 2214.5 | 12.50 | 0.14 ± 0.01 | -1.72 ± 0.04 | -1.58 ± 0.04 | -29.97 ± 0.04 |
| | | $L \to \infty$ | | | -1.64 ± 0.05 | -30.03 ± 0.05 |
| $Cl^-$ | 141.5 | 5.00 | 0.95 ± 0.02 | -2.26 ± 0.02 | -1.31 ± 0.03 | -31.31 ± 0.03 |
| | 478.5 | 7.50 | 0.95 ± 0.02 | -2.29 ± 0.04 | -1.34 ± 0.04 | -31.34 ± 0.04 |
| | 1133.5 | 10.00 | 0.96 ± 0.01 | -2.28 ± 0.02 | -1.32 ± 0.02 | -31.32 ± 0.02 |
| | 2214.5 | 12.50 | 0.96 ± 0.01 | -2.23 ± 0.01 | -1.27 ± 0.01 | -31.27 ± 0.01 |
| | | $L \to \infty$ | | | -1.25 ± 0.03 | -31.25 ± 0.03 |

**Table S9.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 2.798 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1114.710\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}\ (\mathrm{Na^+}) = -27.40\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}\ (\mathrm{Cl^-}) = -29.01\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 210.5 | 5.00 | 0.24 ± 0.01 | -1.83 ± 0.03 | -1.59 ± 0.03 | -28.99 ± 0.03 |
| | 710.5 | 7.50 | 0.23 ± 0.01 | -1.80 ± 0.01 | -1.57 ± 0.01 | -28.97 ± 0.01 |
| | 1685.5 | 10.00 | 0.22 ± 0.01 | -1.79 ± 0.03 | -1.57 ± 0.03 | -28.97 ± 0.03 |
| | 3291.5 | 12.50 | 0.21 ± 0.00 | -1.81 ± 0.02 | -1.60 ± 0.02 | -29.00 ± 0.02 |
| | | $L \to \infty$ | | | -1.60 ± 0.04 | -29.00 ± 0.04 |
| $Cl^-$ | 210.5 | 5.00 | 1.52 ± 0.01 | -2.58 ± 0.01 | -1.06 ± 0.01 | -30.07 ± 0.01 |
| | 710.5 | 7.50 | 1.55 ± 0.02 | -2.56 ± 0.02 | -1.01 ± 0.03 | -30.02 ± 0.03 |
| | 1685.5 | 10.00 | 1.55 ± 0.03 | -2.57 ± 0.03 | -1.02 ± 0.04 | -30.03 ± 0.04 |
| | 3291.5 | 12.50 | 1.52 ± 0.03 | -2.59 ± 0.02 | -1.07 ± 0.04 | -30.08 ± 0.04 |
| | | $L \to \infty$ | | | -1.01 ± 0.05 | -30.03 ± 0.05 |

**Table S10.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 3.674 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1147.060\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}\ (\mathrm{Na^+}) = -26.73\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}\ (\mathrm{Cl^-}) = -28.34\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 276.5 | 5.00 | 0.31 ± 0.01 | -1.80 ± 0.02 | -1.49 ± 0.03 | -28.22 ± 0.03 |
| | 933.5 | 7.50 | 0.32 ± 0.01 | -1.83 ± 0.04 | -1.51 ± 0.05 | -28.24 ± 0.05 |
| | 2212.5 | 10.00 | 0.32 ± 0.01 | -1.81 ± 0.01 | -1.49 ± 0.02 | -28.22 ± 0.02 |
| | 4321.5 | 12.50 | 0.33 ± 0.01 | -1.83 ± 0.01 | -1.50 ± 0.02 | -28.23 ± 0.02 |
| | | $L \to \infty$ | | | -1.50 ± 0.03 | -28.23 ± 0.03 |
| $Cl^-$ | 276.5 | 5.00 | 2.09 ± 0.02 | -2.81 ± 0.02 | -0.72 ± 0.03 | -29.06 ± 0.03 |
| | 933.5 | 7.50 | 2.08 ± 0.01 | -2.80 ± 0.01 | -0.72 ± 0.01 | -29.06 ± 0.01 |
| | 2212.5 | 10.00 | 2.11 ± 0.02 | -2.81 ± 0.00 | -0.70 ± 0.02 | -29.04 ± 0.02 |
| | 4321.5 | 12.50 | 2.07 ± 0.03 | -2.83 ± 0.02 | -0.76 ± 0.04 | -29.10 ± 0.04 |
| | | $L \to \infty$ | | | -0.72 ± 0.04 | -29.06 ± 0.04 |

**Table S11.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 4.510 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1176.440\ \mathrm{kg/m^3}$. $\mu^{IG}\ (Na^+) = -26.22\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -27.83\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| | 339.5 | 5.00 | 0.42 ± 0.01 | -1.84 ± 0.02 | -1.42 ± 0.03 | -27.64 ± 0.03 |
| | 1145.5 | 7.50 | 0.41 ± 0.01 | -1.78 ± 0.03 | -1.37 ± 0.04 | -27.59 ± 0.04 |
| $Na^+$ | 2715.5 | 10.00 | 0.42 ± 0.01 | -1.82 ± 0.01 | -1.40 ± 0.02 | -27.62 ± 0.02 |
| | 5304.5 | 12.50 | 0.41 ± 0.01 | -1.81 ± 0.02 | -1.40 ± 0.03 | -27.62 ± 0.03 |
| | | $L \to \infty$ | | | -1.38 ± 0.04 | -27.60 ± 0.04 |
| | 339.5 | 5.00 | 2.68 ± 0.02 | -2.98 ± 0.01 | -0.30 ± 0.02 | -28.13 ± 0.02 |
| | 1145.5 | 7.50 | 2.67 ± 0.02 | -2.98 ± 0.02 | -0.31 ± 0.03 | -28.14 ± 0.03 |
| $Cl^-$ | 2715.5 | 10.00 | 2.75 ± 0.03 | -2.98 ± 0.03 | -0.23 ± 0.04 | -28.06 ± 0.04 |
| | 5304.5 | 12.50 | 2.68 ± 0.01 | -2.99 ± 0.01 | -0.31 ± 0.01 | -28.14 ± 0.01 |
| | | $L \to \infty$ | | | -0.31 ± 0.03 | -28.14 ± 0.03 |

**Table S12.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 5.304 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1203.020\ \mathrm{kg/m^3}$. $\mu^{IG}\ (Na^+) = -25.82\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -27.43\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| | 399.5 | 5.00 | 0.55 ± 0.01 | -1.80 ± 0.01 | -1.25 ± 0.01 | -27.07 ± 0.01 |
| | 1347.5 | 7.50 | 0.55 ± 0.01 | -1.82 ± 0.03 | -1.27 ± 0.03 | -27.09 ± 0.03 |
| $Na^+$ | 3194.5 | 10.00 | 0.56 ± 0.01 | -1.83 ± 0.01 | -1.27 ± 0.01 | -27.09 ± 0.01 |
| | 6238.5 | 12.50 | 0.55 ± 0.01 | -1.80 ± 0.02 | -1.25 ± 0.02 | -27.07 ± 0.02 |
| | | $L \to \infty$ | | | -1.28 ± 0.03 | -27.09 ± 0.03 |
| | 399.5 | 5.00 | 3.32 ± 0.04 | -3.14 ± 0.01 | 0.18 ± 0.04 | -27.25 ± 0.04 |
| | 1347.5 | 7.50 | 3.34 ± 0.02 | -3.12 ± 0.02 | 0.22 ± 0.03 | -27.21 ± 0.03 |
| $Cl^-$ | 3194.5 | 10.00 | 3.35 ± 0.04 | -3.14 ± 0.01 | 0.21 ± 0.04 | -27.22 ± 0.04 |
| | 6238.5 | 12.50 | 3.33 ± 0.04 | -3.12 ± 0.01 | 0.21 ± 0.04 | -27.22 ± 0.04 |
| | | $L \to \infty$ | | | 0.24 ± 0.06 | -27.19 ± 0.06 |

**Implicit-water NaCl solutions (concentration-dependent relative permittivity):**

**Table S13.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.005 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 998.989\ kg/m^3$. $\mu^{IG}(Na^+) = -43.09\ kJ/mol$, $\mu^{IG}(Cl^-) = -44.70\ kJ/mol$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -43.57 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.37 ± 0.01 | -0.37 ± 0.01 | -43.46 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.31 ± 0.03 | -0.31 ± 0.03 | -43.40 ± 0.03 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.25 ± 0.02 | -0.25 ± 0.02 | -43.34 ± 0.02 |
| | | $L \to \infty$ | | | -0.10 ± 0.02 | -43.19 ± 0.02 |
| $Cl^-$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.18 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.33 ± 0.02 | -0.33 ± 0.02 | -45.03 ± 0.02 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.31 ± 0.02 | -0.31 ± 0.02 | -45.01 ± 0.02 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.30 ± 0.03 | -0.30 ± 0.03 | -45.00 ± 0.03 |
| | | $L \to \infty$ | | | -0.11 ± 0.03 | -44.81 ± 0.03 |

**Table S14.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.019 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 999.621\ kg/m^3$. $\mu^{IG}(Na^+) = -39.78\ kJ/mol$, $\mu^{IG}(Cl^-) = -41.39\ kJ/mol$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.59 ± 0.02 | -0.59 ± 0.02 | -40.37 ± 0.02 |
| | 4.5 | 7.33 | 0.00 ± 0.00 | -0.48 ± 0.03 | -0.48 ± 0.03 | -40.26 ± 0.03 |
| | 11.5 | 10.02 | 0.00 ± 0.00 | -0.55 ± 0.04 | -0.55 ± 0.04 | -40.33 ± 0.04 |
| | 22.5 | 12.53 | 0.00 ± 0.00 | -0.45 ± 0.02 | -0.45 ± 0.02 | -40.23 ± 0.02 |
| | | $L \to \infty$ | | | -0.37 ± 0.03 | -40.15 ± 0.03 |
| $Cl^-$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.60 ± 0.02 | -0.60 ± 0.02 | -41.99 ± 0.02 |
| | 4.5 | 7.33 | 0.01 ± 0.00 | -0.50 ± 0.03 | -0.49 ± 0.03 | -41.88 ± 0.03 |
| | 11.5 | 10.02 | 0.01 ± 0.00 | -0.49 ± 0.05 | -0.48 ± 0.05 | -41.87 ± 0.05 |
| | 22.5 | 12.53 | 0.01 ± 0.00 | -0.49 ± 0.05 | -0.48 ± 0.05 | -41.87 ± 0.05 |
| | | $L \to \infty$ | | | -0.35 ± 0.06 | -41.74 ± 0.06 |

**Table S15.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.056 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1001.270\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -37.11\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -38.72\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 4.5 | 5.11 | 0.00 ± 0.00 | -0.72 ± 0.03 | -0.72 ± 0.03 | -37.83 ± 0.03 |
| | 14.5 | 7.55 | 0.00 ± 0.00 | -0.72 ± 0.01 | -0.72 ± 0.01 | -37.83 ± 0.01 |
| | 33.5 | 9.98 | 0.00 ± 0.00 | -0.67 ± 0.03 | -0.67 ± 0.03 | -37.78 ± 0.03 |
| | 65.5 | 12.49 | 0.00 ± 0.00 | -0.68 ± 0.01 | -0.68 ± 0.01 | -37.79 ± 0.01 |
| | | $L \to \infty$ | | | -0.64 ± 0.02 | -37.74 ± 0.02 |
| $Cl^-$ | 4.5 | 5.11 | 0.03 ± 0.00 | -0.79 ± 0.03 | -0.76 ± 0.03 | -39.48 ± 0.03 |
| | 14.5 | 7.55 | 0.03 ± 0.00 | -0.79 ± 0.04 | -0.76 ± 0.04 | -39.48 ± 0.04 |
| | 33.5 | 9.98 | 0.03 ± 0.00 | -0.74 ± 0.06 | -0.71 ± 0.06 | -39.43 ± 0.06 |
| | 65.5 | 12.49 | 0.03 ± 0.00 | -0.71 ± 0.06 | -0.68 ± 0.06 | -39.40 ± 0.06 |
| | | $L \to \infty$ | | | -0.67 ± 0.07 | -39.38 ± 0.07 |

**Table S16.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.166 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1006.340\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -34.40\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -36.01\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 12.5 | 5.00 | 0.01 ± 0.00 | -1.12 ± 0.05 | -1.11 ± 0.05 | -35.51 ± 0.05 |
| | 42.5 | 7.51 | 0.01 ± 0.00 | -1.08 ± 0.02 | -1.07 ± 0.07 | -35.47 ± 0.07 |
| | 100.5 | 10.01 | 0.01 ± 0.00 | -1.13 ± 0.04 | -1.12 ± 0.04 | -35.52 ± 0.04 |
| | 195.5 | 12.49 | 0.01 ± 0.00 | -1.14 ± 0.05 | -1.13 ± 0.05 | -35.53 ± 0.05 |
| | | $L \to \infty$ | | | -1.13 ± 0.07 | -35.53 ± 0.07 |
| $Cl^-$ | 12.5 | 5.00 | 0.06 ± 0.00 | -1.18 ± 0.01 | -1.12 ± 0.01 | -37.13 ± 0.01 |
| | 42.5 | 7.51 | 0.08 ± 0.00 | -1.14 ± 0.05 | -1.06 ± 0.05 | -37.07 ± 0.05 |
| | 100.5 | 10.01 | 0.08 ± 0.01 | -1.13 ± 0.04 | -1.05 ± 0.04 | -37.06 ± 0.04 |
| | 195.5 | 12.49 | 0.07 ± 0.00 | -1.08 ± 0.04 | -1.01 ± 0.04 | -37.02 ± 0.04 |
| | | $L \to \infty$ | | | -0.95 ± 0.05 | -36.96 ± 0.05 |

**Table S17.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.605 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1025.820\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -31.20\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -32.81\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 45.5 | 5.00 | 0.04 ± 0.00 | -1.66 ± 0.03 | -1.62 ± 0.03 | -32.82 ± 0.03 |
| | 153.5 | 7.50 | 0.03 ± 0.00 | -1.74 ± 0.03 | -1.71 ± 0.03 | -32.91 ± 0.03 |
| | 364.5 | 10.00 | 0.04 ± 0.00 | -1.75 ± 0.02 | -1.71 ± 0.02 | -32.91 ± 0.02 |
| | 711.5 | 12.50 | 0.04 ± 0.00 | -1.72 ± 0.01 | -1.68 ± 0.01 | -32.88 ± 0.01 |
| | | $L \to \infty$ | | | -1.72 ± 0.03 | -32.92 ± 0.03 |
| $Cl^-$ | 45.5 | 5.00 | 0.30 ± 0.01 | -1.85 ± 0.02 | -1.55 ± 0.02 | -34.36 ± 0.02 |
| | 153.5 | 7.50 | 0.29 ± 0.02 | -1.92 ± 0.03 | -1.63 ± 0.04 | -34.44 ± 0.04 |
| | 364.5 | 10.00 | 0.28 ± 0.02 | -1.98 ± 0.04 | -1.70 ± 0.04 | -34.51 ± 0.04 |
| | 711.5 | 12.50 | 0.30 ± 0.01 | -1.97 ± 0.03 | -1.67 ± 0.03 | -34.48 ± 0.03 |
| | | $L \to \infty$ | | | -1.78 ± 0.05 | -34.59 ± 0.05 |

**Table S18.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 0.930 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1039.760\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -30.13\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -31.75\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 70.5 | 5.01 | 0.06 ± 0.00 | -2.15 ± 0.03 | -2.09 ± 0.03 | -32.22 ± 0.03 |
| | 236.5 | 7.50 | 0.06 ± 0.01 | -2.21 ± 0.03 | -2.15 ± 0.03 | -32.28 ± 0.03 |
| | 560.5 | 10.00 | 0.06 ± 0.01 | -2.18 ± 0.03 | -2.12 ± 0.03 | -32.25 ± 0.03 |
| | 1094.5 | 12.50 | 0.06 ± 0.01 | -2.21 ± 0.06 | -2.15 ± 0.06 | -32.28 ± 0.06 |
| | | $L \to \infty$ | | | -2.18 ± 0.06 | -32.31 ± 0.06 |
| $Cl^-$ | 70.5 | 5.01 | 0.45 ± 0.01 | -2.54 ± 0.05 | -2.09 ± 0.05 | -33.84 ± 0.05 |
| | 236.5 | 7.50 | 0.48 ± 0.02 | -2.51 ± 0.03 | -2.03 ± 0.04 | -33.78 ± 0.04 |
| | 560.5 | 10.00 | 0.45 ± 0.01 | -2.45 ± 0.03 | -2.00 ± 0.03 | -33.75 ± 0.03 |
| | 1094.5 | 12.50 | 0.48 ± 0.01 | -2.45 ± 0.04 | -1.97 ± 0.04 | -33.72 ± 0.04 |
| | | $L \to \infty$ | | | -1.90 ± 0.06 | -33.64 ± 0.06 |

**Table S19.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 1.883 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1079.000\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -28.39\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -30.00\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 141.5 | 5.00 | 0.15 ± 0.00 | -3.16 ± 0.02 | -3.01 ± 0.02 | -31.40 ± 0.02 |
| | 478.5 | 7.50 | 0.13 ± 0.01 | -3.19 ± 0.01 | -3.06 ± 0.01 | -31.45 ± 0.01 |
| | 1133.5 | 10.00 | 0.13 ± 0.01 | -3.17 ± 0.03 | -3.04 ± 0.03 | -31.43 ± 0.03 |
| | 2214.5 | 12.50 | 0.14 ± 0.01 | -3.13 ± 0.04 | -2.99 ± 0.04 | -31.38 ± 0.04 |
| | | $L \to \infty$ | | | -3.08 ± 0.04 | -31.46 ± 0.04 |
| $Cl^-$ | 141.5 | 5.00 | 0.95 ± 0.01 | -3.82 ± 0.04 | -2.87 ± 0.04 | -32.87 ± 0.04 |
| | 478.5 | 7.50 | 0.97 ± 0.01 | -3.78 ± 0.03 | -2.81 ± 0.03 | -32.81 ± 0.03 |
| | 1133.5 | 10.00 | 0.96 ± 0.03 | -3.75 ± 0.01 | -2.79 ± 0.03 | -32.79 ± 0.03 |
| | 2214.5 | 12.50 | 0.96 ± 0.02 | -3.77 ± 0.02 | -2.81 ± 0.03 | -32.81 ± 0.03 |
| | | $L \to \infty$ | | | -2.75 ± 0.05 | -32.75 ± 0.05 |

**Table S20.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 2.798 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1114.710\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -27.40\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -29.01\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 210.5 | 5.00 | 0.23 ± 0.00 | -4.22 ± 0.03 | -3.99 ± 0.03 | -31.39 ± 0.03 |
| | 710.5 | 7.50 | 0.22 ± 0.00 | -4.24 ± 0.02 | -4.02 ± 0.02 | -31.42 ± 0.02 |
| | 1685.5 | 10.00 | 0.23 ± 0.01 | -4.26 ± 0.02 | -4.03 ± 0.02 | -31.43 ± 0.02 |
| | 3291.5 | 12.50 | 0.22 ± 0.00 | -4.24 ± 0.02 | -4.02 ± 0.02 | -31.42 ± 0.02 |
| | | $L \to \infty$ | | | -4.05 ± 0.04 | -31.45 ± 0.04 |
| $Cl^-$ | 210.5 | 5.00 | 1.44 ± 0.03 | -5.16 ± 0.02 | -3.72 ± 0.04 | -32.73 ± 0.04 |
| | 710.5 | 7.50 | 1.48 ± 0.01 | -5.17 ± 0.03 | -3.69 ± 0.03 | -32.70 ± 0.03 |
| | 1685.5 | 10.00 | 1.40 ± 0.02 | -5.14 ± 0.03 | -3.74 ± 0.04 | -32.75 ± 0.04 |
| | 3291.5 | 12.50 | 1.45 ± 0.01 | -5.14 ± 0.02 | -3.69 ± 0.02 | -32.70 ± 0.02 |
| | | $L \to \infty$ | | | -3.69 ± 0.04 | -32.70 ± 0.04 |

**Table S21.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 3.674 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1147.060\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -26.73\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -28.34\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 276.5 | 5.00 | 0.32 ± 0.01 | -5.90 ± 0.02 | -5.58 ± 0.03 | -32.31 ± 0.03 |
| | 933.5 | 7.50 | 0.31 ± 0.01 | -5.89 ± 0.02 | -5.58 ± 0.03 | -32.31 ± 0.03 |
| | 2212.5 | 10.00 | 0.32 ± 0.01 | -5.86 ± 0.01 | -5.54 ± 0.02 | -32.27 ± 0.02 |
| | 4321.5 | 12.50 | 0.32 ± 0.01 | -5.93 ± 0.03 | -5.61 ± 0.04 | -32.34 ± 0.04 |
| | | $L \to \infty$ | | | -5.54 ± 0.04 | -32.27 ± 0.04 |
| $Cl^-$ | 276.5 | 5.00 | 1.97 ± 0.03 | -7.14 ± 0.03 | -5.17 ± 0.04 | -33.51 ± 0.04 |
| | 933.5 | 7.50 | 2.03 ± 0.01 | -7.08 ± 0.02 | -5.05 ± 0.02 | -33.39 ± 0.02 |
| | 2212.5 | 10.00 | 2.01 ± 0.02 | -7.13 ± 0.04 | -5.12 ± 0.04 | -33.46 ± 0.04 |
| | 4321.5 | 12.50 | 2.00 ± 0.02 | -7.14 ± 0.03 | -5.14 ± 0.04 | -33.48 ± 0.04 |
| | | $L \to \infty$ | | | -5.08 ± 0.06 | -33.42 ± 0.06 |

**Table S22.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 4.510 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1176.440\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{Na^+}) = -26.22\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -27.83\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 339.5 | 5.00 | 0.45 ± 0.01 | -9.18 ± 0.02 | -8.73 ± 0.03 | -34.95 ± 0.03 |
| | 1145.5 | 7.50 | 0.46 ± 0.02 | -9.25 ± 0.03 | -8.79 ± 0.05 | -35.01 ± 0.05 |
| | 2715.5 | 10.00 | 0.43 ± 0.01 | -9.22 ± 0.04 | -8.79 ± 0.05 | -35.01 ± 0.05 |
| | 5304.5 | 12.50 | 0.43 ± 0.01 | -9.18 ± 0.03 | -8.75 ± 0.04 | -34.97 ± 0.04 |
| | | $L \to \infty$ | | | -8.80 ± 0.06 | -35.02 ± 0.06 |
| $Cl^-$ | 339.5 | 5.00 | 2.44 ± 0.01 | -10.76 ± 0.01 | -8.32 ± 0.01 | -36.15 ± 0.01 |
| | 1145.5 | 7.50 | 2.46 ± 0.02 | -10.76 ± 0.02 | -8.30 ± 0.03 | -36.13 ± 0.03 |
| | 2715.5 | 10.00 | 2.46 ± 0.02 | -10.72 ± 0.03 | -8.26 ± 0.04 | -36.09 ± 0.04 |
| | 5304.5 | 12.50 | 2.47 ± 0.02 | -10.73 ± 0.02 | -8.26 ± 0.03 | -36.09 ± 0.03 |
| | | $L \to \infty$ | | | -8.22 ± 0.04 | -36.05 ± 0.04 |

**Table S23.** Chemical potentials and extrapolation to infinite system size for added $Na^+$ and $Cl^-$ in implicit-water NaCl solutions at a concentration of 5.304 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1203.020\ \mathrm{kg/m^3}$. $\mu^{IG}\ (Na^+) = -25.82\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -27.43\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $Na^+$ | 399.5 | 5.00 | 0.58 ± 0.02 | -8.86 ± 0.03 | -8.28 ± 0.04 | -34.10 ± 0.04 |
| | 1347.5 | 7.50 | 0.56 ± 0.01 | -8.75 ± 0.03 | -8.19 ± 0.03 | -34.01 ± 0.03 |
| | 3194.5 | 10.00 | 0.55 ± 0.01 | -8.74 ± 0.01 | -8.19 ± 0.01 | -34.01 ± 0.01 |
| | 6238.5 | 12.50 | 0.56 ± 0.01 | -8.75 ± 0.02 | -8.19 ± 0.02 | -34.01 ± 0.02 |
| | | $L \to \infty$ | | | -8.12 ± 0.04 | -33.94 ± 0.04 |
| $Cl^-$ | 399.5 | 5.00 | 3.08 ± 0.02 | -10.57 ± 0.02 | -7.49 ± 0.03 | -34.92 ± 0.03 |
| | 1347.5 | 7.50 | 3.02 ± 0.04 | -10.54 ± 0.03 | -7.52 ± 0.05 | -34.95 ± 0.05 |
| | 3194.5 | 10.00 | 3.07 ± 0.02 | -10.63 ± 0.03 | -7.56 ± 0.04 | -34.99 ± 0.04 |
| | 6238.5 | 12.50 | 3.05 ± 0.02 | -10.61 ± 0.02 | -7.56 ± 0.03 | -34.99 ± 0.03 |
| | | $L \to \infty$ | | | -7.61 ± 0.04 | -35.04 ± 0.04 |

## Implicit-water KCl solutions (constant relative permittivity):

**Table S24.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.005 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 998.989\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -45.07\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -44.70\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.55 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.37 ± 0.01 | -0.37 ± 0.01 | -45.44 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.33 ± 0.02 | -0.33 ± 0.02 | -45.40 ± 0.02 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.30 ± 0.03 | -0.30 ± 0.03 | -45.37 ± 0.03 |
| | | $L \to \infty$ | | | -0.14 ± 0.02 | -45.21 ± 0.02 |
| $Cl^-$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.18 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.35 ± 0.01 | -0.35 ± 0.01 | -45.05 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.33 ± 0.02 | -0.33 ± 0.02 | -45.03 ± 0.02 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.26 ± 0.02 | -0.26 ± 0.02 | -44.96 ± 0.02 |
| | | $L \to \infty$ | | | -0.09 ± 0.02 | -44.79 ± 0.02 |

**Table S25.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.019 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 999.791\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -41.76\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -41.39\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.58 ± 0.01 | -0.58 ± 0.01 | -42.34 ± 0.01 |
| | 4.5 | 7.33 | 0.00 ± 0.00 | -0.43 ± 0.02 | -0.43 ± 0.02 | -42.19 ± 0.02 |
| | 11.5 | 10.02 | 0.00 ± 0.00 | -0.45 ± 0.02 | -0.45 ± 0.02 | -42.21 ± 0.02 |
| | 22.5 | 12.54 | 0.01 ± 0.00 | -0.35 ± 0.03 | -0.34 ± 0.03 | -42.10 ± 0.03 |
| | | $L \to \infty$ | | | -0.23 ± 0.03 | -41.99 ± 0.03 |
| $Cl^-$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.59 ± 0.02 | -0.59 ± 0.02 | -41.98 ± 0.02 |
| | 4.5 | 7.33 | 0.01 ± 0.00 | -0.53 ± 0.02 | -0.52 ± 0.02 | -41.91 ± 0.02 |
| | 11.5 | 10.02 | 0.01 ± 0.00 | -0.48 ± 0.04 | -0.47 ± 0.04 | -41.86 ± 0.04 |
| | 22.5 | 12.54 | 0.01 ± 0.00 | -0.46 ± 0.02 | -0.45 ± 0.02 | -41.84 ± 0.02 |
| | | $L \to \infty$ | | | -0.35 ± 0.03 | -41.75 ± 0.03 |

**Table S26.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.056 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1001.820\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -39.08\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -38.72\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 4.5 | 5.11 | 0.00 ± 0.00 | -0.72 ± 0.04 | -0.72 ± 0.04 | -39.80 ± 0.04 |
| | 14.5 | 7.55 | 0.00 ± 0.00 | -0.68 ± 0.03 | -0.68 ± 0.03 | -39.76 ± 0.03 |
| | 33.5 | 9.99 | 0.00 ± 0.00 | -0.53 ± 0.04 | -0.53 ± 0.04 | -39.61 ± 0.04 |
| | 65.5 | 12.49 | 0.00 ± 0.00 | -0.58 ± 0.06 | -0.58 ± 0.06 | -39.66 ± 0.06 |
| | | $L \to \infty$ | | | -0.43 ± 0.07 | -39.51 ± 0.07 |
| $Cl^-$ | 4.5 | 5.11 | 0.03 ± 0.00 | -0.72 ± 0.02 | -0.69 ± 0.02 | -39.41 ± 0.02 |
| | 14.5 | 7.55 | 0.03 ± 0.00 | -0.68 ± 0.03 | -0.65 ± 0.03 | -39.37 ± 0.03 |
| | 33.5 | 9.99 | 0.03 ± 0.00 | -0.63 ± 0.04 | -0.60 ± 0.04 | -39.32 ± 0.04 |
| | 65.5 | 12.49 | 0.03 ± 0.00 | -0.55 ± 0.05 | -0.52 ± 0.05 | -39.24 ± 0.05 |
| | | $L \to \infty$ | | | -0.48 ± 0.06 | -39.20 ± 0.06 |

**Table S27.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.166 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1007.510\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -36.38\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -36.01\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 12.5 | 5.00 | 0.01 ± 0.00 | -0.90 ± 0.03 | -0.89 ± 0.03 | -37.27 ± 0.03 |
| | 42.5 | 7.52 | -0.01 ± 0.00 | -0.87 ± 0.03 | -0.88 ± 0.07 | -37.26 ± 0.07 |
| | 100.5 | 10.01 | -0.01 ± 0.00 | -0.89 ± 0.08 | -0.90 ± 0.08 | -37.28 ± 0.08 |
| | 195.5 | 12.50 | -0.01 ± 0.00 | -0.88 ± 0.03 | -0.89 ± 0.03 | -37.27 ± 0.03 |
| | | $L \to \infty$ | | | -0.89 ± 0.05 | -37.27 ± 0.05 |
| $Cl^-$ | 12.5 | 5.00 | 0.08 ± 0.01 | -1.04 ± 0.03 | -0.96 ± 0.03 | -36.97 ± 0.03 |
| | 42.5 | 7.52 | 0.08 ± 0.00 | -0.97 ± 0.02 | -0.89 ± 0.02 | -36.90 ± 0.02 |
| | 100.5 | 10.01 | 0.08 ± 0.00 | -0.99 ± 0.03 | -0.91 ± 0.03 | -36.92 ± 0.03 |
| | 195.5 | 12.50 | 0.08 ± 0.01 | -0.95 ± 0.01 | -0.87 ± 0.01 | -36.88 ± 0.01 |
| | | $L \to \infty$ | | | -0.82 ± 0.03 | -36.83 ± 0.03 |

**Table S28.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.602 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1029.510\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -33.19\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -32.83\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 45.5 | 5.01 | -0.02 ± 0.01 | -1.26 ± 0.02 | -1.28 ± 0.02 | -34.47 ± 0.02 |
| | 152.5 | 7.49 | -0.02 ± 0.01 | -1.25 ± 0.05 | -1.27 ± 0.05 | -34.46 ± 0.05 |
| | 362.5 | 10.00 | -0.04 ± 0.01 | -1.27 ± 0.04 | -1.31 ± 0.04 | -34.50 ± 0.04 |
| | 707.5 | 12.50 | -0.02 ± 0.01 | -1.27 ± 0.04 | -1.29 ± 0.04 | -34.48 ± 0.04 |
| | | $L \to \infty$ | | | -1.31 ± 0.05 | -34.50 ± 0.05 |
| $Cl^-$ | 45.5 | 5.01 | 0.31 ± 0.01 | -1.49 ± 0.05 | -1.18 ± 0.05 | -34.01 ± 0.05 |
| | 152.5 | 7.49 | 0.29 ± 0.01 | -1.55 ± 0.04 | -1.26 ± 0.04 | -34.09 ± 0.04 |
| | 362.5 | 10.00 | 0.31 ± 0.01 | -1.43 ± 0.07 | -1.12 ± 0.07 | -33.95 ± 0.07 |
| | 707.5 | 12.50 | 0.31 ± 0.01 | -1.48 ± 0.04 | -1.17 ± 0.04 | -34.00 ± 0.04 |
| | | $L \to \infty$ | | | -1.16 ± 0.07 | -33.99 ± 0.07 |

**Table S29.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.922 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1045.210\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -32.13\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -31.77\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 69.5 | 5.00 | -0.03 ± 0.00 | -1.40 ± 0.03 | -1.43 ± 0.03 | -33.56 ± 0.03 |
| | 234.5 | 7.50 | -0.03 ± 0.01 | -1.38 ± 0.02 | -1.41 ± 0.02 | -33.54 ± 0.02 |
| | 555.5 | 10.00 | -0.03 ± 0.01 | -1.36 ± 0.03 | -1.39 ± 0.03 | -33.52 ± 0.03 |
| | 1084.5 | 12.50 | -0.04 ± 0.01 | -1.31 ± 0.05 | -1.35 ± 0.05 | -33.48 ± 0.05 |
| | | $L \to \infty$ | | | -1.34 ± 0.05 | -33.47 ± 0.05 |
| $Cl^-$ | 69.5 | 5.00 | 0.47 ± 0.02 | -1.75 ± 0.02 | -1.28 ± 0.03 | -33.05 ± 0.03 |
| | 234.5 | 7.50 | 0.48 ± 0.01 | -1.76 ± 0.03 | -1.28 ± 0.03 | -33.05 ± 0.03 |
| | 555.5 | 10.00 | 0.50 ± 0.01 | -1.68 ± 0.03 | -1.18 ± 0.03 | -32.95 ± 0.03 |
| | 1084.5 | 12.50 | 0.49 ± 0.01 | -1.74 ± 0.01 | -1.25 ± 0.01 | -33.02 ± 0.01 |
| | | $L \to \infty$ | | | -1.22 ± 0.03 | -32.98 ± 0.03 |

**Table S30.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 1.848 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1088.870\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -30.41\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -30.04\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 139.5 | 5.00 | -0.03 ± 0.01 | -1.54 ± 0.04 | -1.57 ± 0.04 | -31.98 ± 0.04 |
| | 469.5 | 7.50 | -0.05 ± 0.02 | -1.57 ± 0.03 | -1.62 ± 0.04 | -32.03 ± 0.04 |
| | 1112.5 | 10.00 | -0.03 ± 0.02 | -1.54 ± 0.03 | -1.57 ± 0.04 | -31.98 ± 0.04 |
| | 2173.5 | 12.50 | -0.04 ± 0.01 | -1.58 ± 0.02 | -1.62 ± 0.02 | -32.03 ± 0.02 |
| | | $L \to \infty$ | | | -1.64 ± 0.04 | -32.05 ± 0.04 |
| $Cl^-$ | 139.5 | 5.00 | 1.04 ± 0.02 | -2.03 ± 0.03 | -0.99 ± 0.04 | -31.03 ± 0.04 |
| | 469.5 | 7.50 | 1.02 ± 0.02 | -2.09 ± 0.04 | -1.07 ± 0.04 | -31.11 ± 0.04 |
| | 1112.5 | 10.00 | 1.02 ± 0.02 | -2.07 ± 0.03 | -1.05 ± 0.04 | -31.09 ± 0.04 |
| | 2173.5 | 12.50 | 1.03 ± 0.01 | -2.11 ± 0.04 | -1.08 ± 0.04 | -31.12 ± 0.04 |
| | | $L \to \infty$ | | | -1.13 ± 0.06 | -31.18 ± 0.06 |

**Table S31.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 2.721 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1127.880\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -29.45\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -29.08\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 204.5 | 5.00 | 0.00 ± 0.01 | -1.55 ± 0.03 | -1.55 ± 0.03 | -31.00 ± 0.03 |
| | 691.5 | 7.50 | -0.02 ± 0.01 | -1.62 ± 0.04 | -1.64 ± 0.04 | -31.09 ± 0.04 |
| | 1638.5 | 10.00 | -0.02 ± 0.02 | -1.65 ± 0.03 | -1.67 ± 0.04 | -31.12 ± 0.04 |
| | 3200.5 | 12.50 | -0.01 ± 0.01 | -1.60 ± 0.02 | -1.61 ± 0.02 | -31.06 ± 0.02 |
| | | $L \to \infty$ | | | -1.68 ± 0.04 | -31.12 ± 0.04 |
| $Cl^-$ | 204.5 | 5.00 | 1.62 ± 0.03 | -2.29 ± 0.02 | -0.67 ± 0.04 | -29.75 ± 0.04 |
| | 691.5 | 7.50 | 1.61 ± 0.04 | -2.34 ± 0.03 | -0.73 ± 0.05 | -29.81 ± 0.05 |
| | 1638.5 | 10.00 | 1.62 ± 0.02 | -2.39 ± 0.03 | -0.77 ± 0.04 | -29.85 ± 0.04 |
| | 3200.5 | 12.50 | 1.64 ± 0.01 | -2.38 ± 0.02 | -0.74 ± 0.02 | -29.82 ± 0.02 |
| | | $L \to \infty$ | | | -0.80 ± 0.04 | -29.88 ± 0.04 |

**Table S32.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 3.542 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1162.840\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -28.79\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -28.43\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 266.5 | 5.00 | 0.03 ± 0.02 | -1.63 ± 0.03 | -1.60 ± 0.05 | -30.39 ± 0.05 |
| | 899.5 | 7.50 | 0.03 ± 0.02 | -1.59 ± 0.02 | -1.56 ± 0.04 | -30.35 ± 0.04 |
| | 2133.5 | 10.00 | 0.02 ± 0.01 | -1.59 ± 0.03 | -1.57 ± 0.04 | -30.36 ± 0.04 |
| | 4166.5 | 12.50 | 0.04 ± 0.01 | -1.59 ± 0.03 | -1.55 ± 0.04 | -30.34 ± 0.04 |
| | | $L \to \infty$ | | | -1.52 ± 0.06 | -30.32 ± 0.06 |
| $Cl^-$ | 266.5 | 5.00 | 2.20 ± 0.03 | -2.51 ± 0.02 | -0.31 ± 0.04 | -28.74 ± 0.04 |
| | 899.5 | 7.50 | 2.20 ± 0.02 | -2.58 ± 0.02 | -0.38 ± 0.03 | -28.81 ± 0.03 |
| | 2133.5 | 10.00 | 2.22 ± 0.02 | -2.51 ± 0.02 | -0.29 ± 0.03 | -28.72 ± 0.03 |
| | 4166.5 | 12.50 | 2.21 ± 0.02 | -2.51 ± 0.03 | -0.30 ± 0.04 | -28.73 ± 0.04 |
| | | $L \to \infty$ | | | -0.29 ± 0.05 | -28.72 ± 0.05 |

**Table S33.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 4.311 mol salt / L ($\varepsilon_r$ fixed at 73) – $\rho = 1194.180\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(\mathrm{K^+}) = -28.31\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(\mathrm{Cl^-}) = -27.94\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 324.5 | 5.00 | 0.12 ± 0.01 | -1.56 ± 0.02 | -1.44 ± 0.03 | -29.75 ± 0.03 |
| | 1095.5 | 7.50 | 0.11 ± 0.02 | -1.64 ± 0.02 | -1.53 ± 0.04 | -29.84 ± 0.04 |
| | 2596.5 | 10.00 | 0.11 ± 0.01 | -1.60 ± 0.03 | -1.49 ± 0.04 | -29.80 ± 0.04 |
| | 5071.5 | 12.50 | 0.12 ± 0.01 | -1.61 ± 0.01 | -1.49 ± 0.02 | -29.80 ± 0.02 |
| | | $L \to \infty$ | | | -1.53 ± 0.04 | -29.83 ± 0.04 |
| $Cl^-$ | 324.5 | 5.00 | 2.81 ± 0.01 | -2.61 ± 0.02 | 0.20 ± 0.02 | -27.74 ± 0.02 |
| | 1095.5 | 7.50 | 2.85 ± 0.03 | -2.60 ± 0.03 | 0.25 ± 0.04 | -27.69 ± 0.04 |
| | 2596.5 | 10.00 | 2.84 ± 0.03 | -2.63 ± 0.03 | 0.21 ± 0.04 | -27.73 ± 0.04 |
| | 5071.5 | 12.50 | 2.85 ± 0.01 | -2.64 ± 0.02 | 0.21 ± 0.02 | -27.73 ± 0.02 |
| | | $L \to \infty$ | | | 0.22 ± 0.04 | -27.72 ± 0.04 |

## Implicit-water NaCl solutions (concentration-dependent relative permittivity):

**Table S34.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.005 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 998.989\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(K^+) = -45.07\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(Cl^-) = -44.70\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.55 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.38 ± 0.01 | -0.38 ± 0.01 | -45.45 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.32 ± 0.03 | -0.32 ± 0.03 | -45.39 ± 0.03 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.22 ± 0.04 | -0.22 ± 0.04 | -45.29 ± 0.04 |
| | | $L \to \infty$ | | | -0.13 ± 0.03 | -45.20 ± 0.03 |
| $Cl^-$ | 0.5 | 5.50 | 0.00 ± 0.00 | -0.48 ± 0.00 | -0.48 ± 0.00 | -45.18 ± 0.00 |
| | 1.5 | 7.93 | 0.00 ± 0.00 | -0.37 ± 0.01 | -0.37 ± 0.01 | -45.07 ± 0.01 |
| | 3.5 | 10.52 | 0.00 ± 0.00 | -0.30 ± 0.03 | -0.30 ± 0.03 | -45.00 ± 0.03 |
| | 5.5 | 12.23 | 0.00 ± 0.00 | -0.24 ± 0.02 | -0.24 ± 0.02 | -44.94 ± 0.02 |
| | | $L \to \infty$ | | | -0.09 ± 0.02 | -44.79 ± 0.02 |

**Table S35.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.019 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 999.791\ \mathrm{kg/m^3}$. $\mu^{\mathrm{IG}}(K^+) = -41.76\ \mathrm{kJ/mol}$, $\mu^{\mathrm{IG}}(Cl^-) = -41.39\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{\mathrm{ex,LJ}}$ (kJ/mol) | $\mu^{\mathrm{ex,Coul}}$ (kJ/mol) | $\mu^{\mathrm{ex}}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.61 ± 0.02 | -0.61 ± 0.02 | -42.37 ± 0.02 |
| | 4.5 | 7.33 | 0.00 ± 0.00 | -0.49 ± 0.04 | -0.49 ± 0.04 | -42.25 ± 0.04 |
| | 11.5 | 10.02 | 0.00 ± 0.00 | -0.42 ± 0.05 | -0.42 ± 0.05 | -42.18 ± 0.05 |
| | 22.5 | 12.54 | 0.00 ± 0.00 | -0.48 ± 0.05 | -0.48 ± 0.05 | -42.24 ± 0.05 |
| | | $L \to \infty$ | | | -0.31 ± 0.06 | -42.07 ± 0.06 |
| $Cl^-$ | 1.5 | 5.08 | 0.00 ± 0.00 | -0.59 ± 0.02 | -0.59 ± 0.02 | -41.98 ± 0.02 |
| | 4.5 | 7.33 | 0.01 ± 0.00 | -0.52 ± 0.06 | -0.51 ± 0.06 | -41.90 ± 0.06 |
| | 11.5 | 10.02 | 0.01 ± 0.00 | -0.41 ± 0.03 | -0.40 ± 0.03 | -41.79 ± 0.03 |
| | 22.5 | 12.54 | 0.01 ± 0.00 | -0.39 ± 0.02 | -0.38 ± 0.02 | -41.77 ± 0.02 |
| | | $L \to \infty$ | | | -0.23 ± 0.03 | -41.63 ± 0.03 |

**Table S36.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.056 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1001.820$ kg/m$^3$. $\mu^{IG}(K^+) = -39.08$ kJ/mol, $\mu^{IG}(Cl^-) = -38.72$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 4.5 | 5.11 | 0.00 ± 0.00 | -0.73 ± 0.02 | -0.73 ± 0.02 | -39.81 ± 0.02 |
| | 14.5 | 7.55 | 0.00 ± 0.00 | -0.60 ± 0.03 | -0.60 ± 0.03 | -39.68 ± 0.03 |
| | 33.5 | 9.99 | 0.00 ± 0.00 | -0.56 ± 0.03 | -0.56 ± 0.03 | -39.64 ± 0.03 |
| | 65.5 | 12.49 | 0.00 ± 0.00 | -0.66 ± 0.05 | -0.66 ± 0.05 | -39.74 ± 0.05 |
| | | $L \to \infty$ | | | -0.44 ± 0.05 | -39.52 ± 0.05 |
| $Cl^-$ | 4.5 | 5.11 | 0.03 ± 0.00 | -0.72 ± 0.02 | -0.69 ± 0.02 | -39.41 ± 0.02 |
| | 14.5 | 7.55 | 0.03 ± 0.00 | -0.74 ± 0.05 | -0.71 ± 0.05 | -39.43 ± 0.05 |
| | 33.5 | 9.99 | 0.03 ± 0.00 | -0.71 ± 0.05 | -0.68 ± 0.05 | -39.40 ± 0.05 |
| | 65.5 | 12.49 | 0.03 ± 0.00 | -0.64 ± 0.02 | -0.61 ± 0.02 | -39.33 ± 0.02 |
| | | $L \to \infty$ | | | -0.57 ± 0.04 | -39.29 ± 0.04 |

**Table S37.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.166 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1007.510$ kg/m$^3$. $\mu^{IG}(K^+) = -36.38$ kJ/mol, $\mu^{IG}(Cl^-) = -36.01$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 12.5 | 5.00 | -0.01 ± 0.01 | -0.98 ± 0.05 | -0.99 ± 0.05 | -37.37 ± 0.05 |
| | 42.5 | 7.52 | -0.01 ± 0.00 | -0.98 ± 0.04 | -0.99 ± 0.07 | -37.37 ± 0.07 |
| | 100.5 | 10.01 | -0.01 ± 0.00 | -0.90 ± 0.03 | -0.91 ± 0.03 | -37.29 ± 0.03 |
| | 195.5 | 12.50 | -0.01 ± 0.00 | -0.88 ± 0.07 | -0.89 ± 0.07 | -37.27 ± 0.07 |
| | | $L \to \infty$ | | | -0.83 ± 0.07 | -37.21 ± 0.07 |
| $Cl^-$ | 12.5 | 5.00 | 0.07 ± 0.00 | -1.04 ± 0.05 | -0.97 ± 0.05 | -36.98 ± 0.05 |
| | 42.5 | 7.52 | 0.09 ± 0.01 | -1.06 ± 0.04 | -0.97 ± 0.04 | -36.98 ± 0.04 |
| | 100.5 | 10.01 | 0.09 ± 0.00 | -0.98 ± 0.04 | -0.89 ± 0.04 | -36.90 ± 0.04 |
| | 195.5 | 12.50 | 0.08 ± 0.00 | -0.95 ± 0.06 | -0.87 ± 0.06 | -36.88 ± 0.06 |
| | | $L \to \infty$ | | | -0.82 ± 0.08 | -36.83 ± 0.08 |

**Table S38.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.602 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1029.510\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -33.19\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -32.83\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 45.5 | 5.01 | -0.01 ± 0.01 | -1.70 ± 0.04 | -1.71 ± 0.04 | -34.90 ± 0.04 |
| | 152.5 | 7.49 | -0.02 ± 0.01 | -1.72 ± 0.05 | -1.74 ± 0.05 | -34.93 ± 0.05 |
| | 362.5 | 10.00 | -0.02 ± 0.01 | -1.78 ± 0.04 | -1.80 ± 0.04 | -34.99 ± 0.04 |
| | 707.5 | 12.50 | -0.04 ± 0.01 | -1.73 ± 0.03 | -1.77 ± 0.03 | -34.96 ± 0.03 |
| | | $L \to \infty$ | | | -1.83 ± 0.05 | -35.02 ± 0.05 |
| $Cl^-$ | 45.5 | 5.01 | 0.30 ± 0.01 | -1.98 ± 0.03 | -1.68 ± 0.03 | -34.51 ± 0.03 |
| | 152.5 | 7.49 | 0.30 ± 0.01 | -1.96 ± 0.05 | -1.66 ± 0.05 | -34.49 ± 0.05 |
| | 362.5 | 10.00 | 0.30 ± 0.02 | -2.01 ± 0.04 | -1.71 ± 0.04 | -34.54 ± 0.04 |
| | 707.5 | 12.50 | 0.30 ± 0.02 | -1.99 ± 0.03 | -1.69 ± 0.04 | -34.52 ± 0.04 |
| | | $L \to \infty$ | | | -1.70 ± 0.05 | -34.53 ± 0.05 |

**Table S39.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 0.922 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1045.210\ \mathrm{kg/m^3}$. $\mu^{IG}\ (K^+) = -32.13\ \mathrm{kJ/mol}$, $\mu^{IG}\ (Cl^-) = -31.77\ \mathrm{kJ/mol}$.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 69.5 | 5.00 | -0.03 ± 0.01 | -1.83 ± 0.07 | -1.86 ± 0.07 | -33.99 ± 0.07 |
| | 234.5 | 7.50 | -0.04 ± 0.01 | -1.86 ± 0.03 | -1.90 ± 0.03 | -34.03 ± 0.03 |
| | 555.5 | 10.00 | -0.03 ± 0.01 | -1.80 ± 0.04 | -1.83 ± 0.04 | -33.96 ± 0.04 |
| | 1084.5 | 12.50 | -0.04 ± 0.01 | -1.83 ± 0.04 | -1.87 ± 0.04 | -34.00 ± 0.04 |
| | | $L \to \infty$ | | | -1.84 ± 0.08 | -33.97 ± 0.08 |
| $Cl^-$ | 69.5 | 5.00 | 0.48 ± 0.01 | -2.17 ± 0.02 | -1.69 ± 0.02 | -33.46 ± 0.02 |
| | 234.5 | 7.50 | 0.48 ± 0.02 | -2.14 ± 0.05 | -1.66 ± 0.05 | -33.43 ± 0.05 |
| | 555.5 | 10.00 | 0.49 ± 0.02 | -2.25 ± 0.04 | -1.76 ± 0.04 | -33.53 ± 0.04 |
| | 1084.5 | 12.50 | 0.49 ± 0.01 | -2.09 ± 0.06 | -1.60 ± 0.06 | -33.37 ± 0.06 |
| | | $L \to \infty$ | | | -1.69 ± 0.07 | -33.45 ± 0.07 |

**Table S40.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 1.848 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1088.870$ kg/m$^3$. $\mu^{IG}(K^+) = -30.41$ kJ/mol, $\mu^{IG}(Cl^-) = -30.04$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 139.5 | 5.00 | -0.04 ± 0.02 | -2.43 ± 0.01 | -2.47 ± 0.02 | -32.88 ± 0.02 |
| | 469.5 | 7.50 | -0.04 ± 0.01 | -2.41 ± 0.03 | -2.45 ± 0.03 | -32.86 ± 0.03 |
| | 1112.5 | 10.00 | -0.06 ± 0.00 | -2.42 ± 0.02 | -2.48 ± 0.02 | -32.89 ± 0.02 |
| | 2173.5 | 12.50 | -0.04 ± 0.01 | -2.41 ± 0.01 | -2.45 ± 0.01 | -32.86 ± 0.01 |
| | | $L \to \infty$ | | | -2.45 ± 0.03 | -32.85 ± 0.03 |
| $Cl^-$ | 139.5 | 5.00 | 1.05 ± 0.02 | -3.04 ± 0.03 | -1.99 ± 0.04 | -32.03 ± 0.04 |
| | 469.5 | 7.50 | 0.99 ± 0.02 | -3.08 ± 0.04 | -2.09 ± 0.04 | -32.13 ± 0.04 |
| | 1112.5 | 10.00 | 1.04 ± 0.01 | -3.00 ± 0.05 | -1.96 ± 0.05 | -32.00 ± 0.05 |
| | 2173.5 | 12.50 | 1.03 ± 0.01 | -3.00 ± 0.03 | -1.97 ± 0.03 | -32.01 ± 0.03 |
| | | $L \to \infty$ | | | -1.97 ± 0.05 | -32.01 ± 0.05 |

**Table S41.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 2.721 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1127.880$ kg/m$^3$. $\mu^{IG}(K^+) = -29.45$ kJ/mol, $\mu^{IG}(Cl^-) = -29.08$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 204.5 | 5.00 | 0.01 ± 0.01 | -3.41 ± 0.03 | -3.40 ± 0.03 | -32.85 ± 0.03 |
| | 691.5 | 7.50 | 0.00 ± 0.01 | -3.34 ± 0.04 | -3.34 ± 0.04 | -32.79 ± 0.04 |
| | 1638.5 | 10.00 | 0.00 ± 0.02 | -3.33 ± 0.04 | -3.33 ± 0.04 | -32.78 ± 0.04 |
| | 3200.5 | 12.50 | 0.01 ± 0.01 | -3.34 ± 0.03 | -3.33 ± 0.03 | -32.78 ± 0.03 |
| | | $L \to \infty$ | | | -3.27 ± 0.05 | -32.72 ± 0.05 |
| $Cl^-$ | 204.5 | 5.00 | 1.58 ± 0.03 | -4.33 ± 0.04 | -2.75 ± 0.05 | -31.83 ± 0.05 |
| | 691.5 | 7.50 | 1.54 ± 0.03 | -4.28 ± 0.03 | -2.74 ± 0.04 | -31.82 ± 0.04 |
| | 1638.5 | 10.00 | 1.60 ± 0.02 | -4.33 ± 0.04 | -2.73 ± 0.04 | -31.81 ± 0.04 |
| | 3200.5 | 12.50 | 1.59 ± 0.01 | -4.27 ± 0.04 | -2.68 ± 0.04 | -31.76 ± 0.04 |
| | | $L \to \infty$ | | | -2.66 ± 0.07 | -31.74 ± 0.07 |

**Table S42.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 3.542 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1162.840$ kg/m$^3$. $\mu^{IG}(K^+) = -28.79$ kJ/mol, $\mu^{IG}(Cl^-) = -28.43$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 266.5 | 5.00 | 0.07 ± 0.02 | -4.31 ± 0.03 | -4.24 ± 0.05 | -33.03 ± 0.05 |
| | 899.5 | 7.50 | 0.07 ± 0.02 | -4.34 ± 0.04 | -4.27 ± 0.06 | -33.06 ± 0.06 |
| | 2133.5 | 10.00 | 0.04 ± 0.01 | -4.34 ± 0.03 | -4.30 ± 0.04 | -33.09 ± 0.04 |
| | 4166.5 | 12.50 | 0.06 ± 0.01 | -4.30 ± 0.04 | -4.24 ± 0.05 | -33.03 ± 0.05 |
| | | $L \to \infty$ | | | -4.30 ± 0.07 | -33.09 ± 0.07 |
| $Cl^-$ | 266.5 | 5.00 | 2.15 ± 0.03 | -5.54 ± 0.04 | -3.39 ± 0.05 | -31.82 ± 0.05 |
| | 899.5 | 7.50 | 2.11 ± 0.03 | -5.52 ± 0.05 | -3.41 ± 0.06 | -31.84 ± 0.06 |
| | 2133.5 | 10.00 | 2.08 ± 0.03 | -5.51 ± 0.03 | -3.43 ± 0.04 | -31.86 ± 0.04 |
| | 4166.5 | 12.50 | 2.16 ± 0.02 | -5.55 ± 0.02 | -3.39 ± 0.03 | -31.82 ± 0.03 |
| | | $L \to \infty$ | | | -3.40 ± 0.06 | -31.83 ± 0.06 |

**Table S43.** Chemical potentials and extrapolation to infinite system size for added $K^+$ and $Cl^-$ in implicit-water KCl solutions at a concentration of 4.311 mol salt / L (concentration-dependent $\varepsilon_r$) – $\rho = 1194.180$ kg/m$^3$. $\mu^{IG}(K^+) = -28.31$ kJ/mol, $\mu^{IG}(Cl^-) = -27.94$ kJ/mol.

| | $N$ | $L$ (nm) | $\mu^{ex,LJ}$ (kJ/mol) | $\mu^{ex,Coul}$ (kJ/mol) | $\mu^{ex}$ (kJ/mol) | $\mu$ (kJ/mol) |
|---|---|---|---|---|---|---|
| $K^+$ | 324.5 | 5.00 | 0.14 ± 0.02 | -4.98 ± 0.03 | -4.84 ± 0.05 | -33.15 ± 0.05 |
| | 1095.5 | 7.50 | 0.13 ± 0.02 | -4.91 ± 0.03 | -4.78 ± 0.05 | -33.09 ± 0.05 |
| | 2596.5 | 10.00 | 0.12 ± 0.01 | -4.98 ± 0.02 | -4.86 ± 0.03 | -33.17 ± 0.03 |
| | 5071.5 | 12.50 | 0.14 ± 0.01 | -4.97 ± 0.03 | -4.83 ± 0.04 | -33.14 ± 0.04 |
| | | $L \to \infty$ | | | -4.85 ± 0.06 | -33.16 ± 0.06 |
| $Cl^-$ | 324.5 | 5.00 | 2.75 ± 0.03 | -6.37 ± 0.03 | -3.62 ± 0.04 | -31.56 ± 0.04 |
| | 1095.5 | 7.50 | 2.77 ± 0.04 | -6.30 ± 0.03 | -3.53 ± 0.05 | -31.47 ± 0.05 |
| | 2596.5 | 10.00 | 2.79 ± 0.02 | -6.39 ± 0.02 | -3.60 ± 0.03 | -31.54 ± 0.03 |
| | 5071.5 | 12.50 | 2.77 ± 0.02 | -6.39 ± 0.02 | -3.62 ± 0.03 | -31.56 ± 0.03 |
| | | $L \to \infty$ | | | -3.61 ± 0.05 | -31.55 ± 0.05 |

**Table S44.** Simulation results for the relative permittivity of explicit-water NaCl and KCl solutions at 298.15 K and 1 bar. Each simulation data is the result of extrapolation to infinite system size. The uncertainties indicate one standard deviation for the extrapolation intercepts.

| $m$ | $\varepsilon_r$ (NaCl) | $\varepsilon_r$ (KCl) |
|---|---|---|
| 0 (pure water) | 73.0 ± 1.5 | 73.0 ± 1.5 |
| 0.056 | 69.3 ± 1.5 | 72.0 ± 2.1 |
| 0.167 | 67.1 ± 2.1 | 71.1 ± 1.4 |
| 0.611 | 61.9 ± 2.1 | 58.6 ± 1.4 |
| 0.944 | 56.1 ± 2.2 | 60.5 ± 1.7 |
| 1.943 | 47.2 ± 1.7 | 53.5 ± 1.4 |
| 2.942 | 39.3 ± 1.6 | 43.0 ± 0.6 |
| 3.941 | 31.3 ± 1.4 | 36.6 ± 0.9 |
| 4.940 | 26.2 ± 0.9 | 33.4 ± 0.8 |
| 5.939 | 22.0 ± 0.7 | – |

**Table S45.** Simulation results for individual and mean ionic activity coefficients in implicit-water NaCl solution at 298.15 K and 1 bar, with $\varepsilon_r$ fixed at 73.

| $M$ ($\frac{\text{mol}}{\text{L}}$) | $\ln \gamma_+$ | $\ln \gamma_-$ | $\ln \gamma_\pm$ |
|---|---|---|---|
| 0.005 | -0.09 ± 0.01 | -0.09 ± 0.01 | -0.09 ± 0.02 |
| 0.019 | -0.14 ± 0.02 | -0.21 ± 0.02 | -0.17 ± 0.03 |
| 0.056 | -0.25 ± 0.01 | -0.25 ± 0.02 | -0.25 ± 0.02 |
| 0.166 | -0.37 ± 0.02 | -0.38 ± 0.02 | -0.38 ± 0.03 |
| 0.605 | -0.60 ± 0.02 | -0.51 ± 0.02 | -0.56 ± 0.03 |
| 0.930 | -0.69 ± 0.02 | -0.60 ± 0.02 | -0.64 ± 0.03 |
| 1.883 | -0.74 ± 0.02 | -0.57 ± 0.02 | -0.66 ± 0.02 |
| 2.798 | -0.74 ± 0.02 | -0.49 ± 0.02 | -0.62 ± 0.03 |
| 3.674 | -0.72 ± 0.02 | -0.39 ± 0.02 | -0.56 ± 0.02 |
| 4.510 | -0.70 ± 0.02 | -0.25 ± 0.01 | -0.47 ± 0.02 |
| 5.304 | -0.68 ± 0.01 | -0.05 ± 0.03 | -0.36 ± 0.03 |

**Table S46.** Simulation results for individual and mean ionic activity coefficients in implicit-water NaCl solution at 298.15 K and 1 bar, with concentration-dependent $\varepsilon_r$.

| $M\ (\frac{\text{mol}}{\text{L}})$ | $\ln \gamma_+$ | $\ln \gamma_-$ | $\ln \gamma_\pm$ |
|---|---|---|---|
| 0.005 | -0.09 ± 0.01 | -0.09 ± 0.02 | -0.09 ± 0.02 |
| 0.019 | -0.20 ± 0.02 | -0.19 ± 0.03 | -0.20 ± 0.03 |
| 0.056 | -0.31 ± 0.01 | -0.32 ± 0.03 | -0.31 ± 0.04 |
| 0.166 | -0.51 ± 0.03 | -0.44 ± 0.02 | -0.47 ± 0.04 |
| 0.605 | -0.75 ± 0.01 | -0.77 ± 0.02 | -0.76 ± 0.03 |
| 0.930 | -0.95 ± 0.03 | -0.83 ± 0.03 | -0.89 ± 0.04 |
| 1.883 | -1.32 ± 0.02 | -1.19 ± 0.02 | -1.26 ± 0.03 |
| 2.798 | -1.73 ± 0.02 | -1.59 ± 0.02 | -1.66 ± 0.03 |
| 3.674 | -2.36 ± 0.02 | -2.17 ± 0.03 | -2.26 ± 0.03 |
| 4.510 | -3.69 ± 0.03 | -3.46 ± 0.02 | -3.57 ± 0.03 |
| 5.304 | -3.44 ± 0.02 | -3.23 ± 0.02 | -3.34 ± 0.03 |

**Table S47.** Simulation results for individual and mean ionic activity coefficients in implicit-water KCl solution at 298.15 K and 1 bar, with $\varepsilon_r$ fixed at 73.

| $M\ (\frac{\text{mol}}{\text{L}})$ | $\ln \gamma_+$ | $\ln \gamma_-$ | $\ln \gamma_\pm$ |
|---|---|---|---|
| 0.005 | -0.09 ± 0.01 | -0.09 ± 0.01 | -0.09 ± 0.02 |
| 0.019 | -0.13 ± 0.02 | -0.20 ± 0.02 | -0.16 ± 0.02 |
| 0.056 | -0.21 ± 0.03 | -0.25 ± 0.02 | -0.23 ± 0.04 |
| 0.166 | -0.40 ± 0.02 | -0.39 ± 0.02 | -0.39 ± 0.03 |
| 0.602 | -0.58 ± 0.02 | -0.54 ± 0.03 | -0.56 ± 0.04 |
| 0.922 | -0.60 ± 0.02 | -0.57 ± 0.01 | -0.58 ± 0.03 |
| 1.848 | -0.75 ± 0.02 | -0.56 ± 0.02 | -0.65 ± 0.03 |
| 2.721 | -0.79 ± 0.02 | -0.45 ± 0.02 | -0.62 ± 0.03 |
| 3.542 | -0.76 ± 0.03 | -0.28 ± 0.02 | -0.52 ± 0.04 |
| 4.311 | -0.79 ± 0.02 | -0.10 ± 0.02 | -0.44 ± 0.02 |

**Table S48.** Simulation results for individual and mean ionic activity coefficients in implicit-water KCl solution at 298.15 K and 1 bar, with concentration-dependent $\varepsilon_r$.

| $M\ (\frac{\text{mol}}{\text{L}})$ | $\ln \gamma_+$ | $\ln \gamma_-$ | $\ln \gamma_\pm$ |
|---|---|---|---|
| 0.005 | -0.09 ± 0.02 | -0.09 ± 0.01 | -0.09 ± 0.02 |
| 0.019 | -0.16 ± 0.03 | -0.15 ± 0.02 | -0.16 ± 0.03 |
| 0.056 | -0.22 ± 0.02 | -0.29 ± 0.02 | -0.25 ± 0.03 |
| 0.166 | -0.38 ± 0.03 | -0.39 ± 0.03 | -0.38 ± 0.04 |
| 0.602 | -0.79 ± 0.02 | -0.76 ± 0.02 | -0.77 ± 0.03 |
| 0.922 | -0.80 ± 0.03 | -0.76 ± 0.03 | -0.78 ± 0.04 |
| 1.848 | -1.08 ± 0.02 | -0.90 ± 0.02 | -0.99 ± 0.03 |
| 2.721 | -1.44 ± 0.02 | -1.21 ± 0.03 | -1.32 ± 0.04 |
| 3.542 | -1.88 ± 0.03 | -1.53 ± 0.02 | -1.71 ± 0.04 |
| 4.311 | -2.13 ± 0.03 | -1.65 ± 0.02 | -1.89 ± 0.03 |

**Table S49.** Fitting parameters of the cubic polynomial function for the experimental solution densities (illustrated in Figure S1) — $\rho(m) = B_0 + B_1 m + B_2 m^2 + B_3 m^3$

| Fitting Parameters | $B_0$ | $B_1$ | $B_2$ | $B_3$ |
|---|---|---|---|---|
| NaCl | 997 | 40.97 | -1.80 | 0.08 |
| KCl | 997 | 47.00 | -2.44 | 0.10 |

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