Supplementary Materials for

A thermodynamic model for predicting carbon dioxide solubility in aqueous sodium chloride solutions using an association equation of state

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Section 1: CPA EoS

The CPA EoS is used for the vapor phase as follows:

$$P=\frac{RT}{v-b}-\frac{\alpha(T)}{v(v+b)}-\frac{1}{2}\frac{RT}{v}\left(1+\frac{1}{v}\frac{\partial \ln g}{\partial(1/v)}\right)\sum_{i=1}^{n}y_i\sum\nolimits_{A_i}\left(1-X_{A_i}\right) \quad \text{(S1)}$$

where v is molar volume, $cm^3 \cdot mol^{-1}$; $\alpha(T)$ and b are respectively the energy parameter and co-volume of CPA EoS; A and B stand for the bonding sites; y_i is the molar fraction of given component i; X_{A_i} is the molar fraction of i molecules not bonded at site A; g is the radial distribution function.

The energy parameter is given by:

$$\alpha(T)=a_0\left[1+c_1\left(1-\sqrt{T_r}\right)\right]^2 \quad \text{(S2)}$$

where a_0 and c_1 are parameters of CPA EoS; T_c is the critical temperature, K; T_r is equal to T/T_c.

The X_{A_i} in the association term can be calculated by

$$X_{A_i}=\left(1+\left(\frac{1}{v}\right)\sum_j y_j\sum_{B_j}X_{B_j}\Delta^{A_iB_j}\right)^{-1} \quad \text{(S3)}$$

$$\Delta^{A_iB_j}=g(v)\left[\exp\left(\frac{\varepsilon^{A_iB_j}}{RT}\right)-1\right]b_{ij}\beta^{A_iB_j} \quad \text{(S4)}$$

where $\varepsilon^{A_iB_j}$ is association energy; $\beta^{A_iB_j}$ is interaction volume; $\Delta^{A_iB_j}$ is association strength.

The radial distribution function is as follows:

$$g(v)=\frac{1}{(1-1.9\eta)} \quad \text{(S5)}$$

$$\eta=\frac{1}{4v}b \quad \text{(S6)}$$

where η is reduced fluid density.

Table S1 includes the parameters of the CPA EoS of pure CO_2 and H_2O.

Table S1 Parameters of CPA EoS of pure CO_2 and H_2O.

Fluid	Association scheme	a_0 ($cm^2 \cdot MPa \cdot mol^{-2}$)	b ($cm^3 \cdot mol^{-1}$)	c_1	ε ($cm \cdot MPa \cdot mol^{-1}$)	β
CO_2	Inert	350790	27.2	0.7602	-	-
H_2O	4C	122770	14.5	0.6736	16655	0.0692

Section 2: Derivatives of a_{CO2} and a_{H2O}

The derivatives of a_{CO_2} and a_{H_2O} with respect to m_{CO2} are Eqs. (S7) and (S8).

$$\frac{\partial a_{\mathrm{CO_2}}}{\partial m_{\mathrm{CO_2}}} = \gamma_{\mathrm{CO_2}} + \gamma_{\mathrm{CO_2}} m_{\mathrm{CO_2}} \left[2\lambda_{nn} + 6 m_{\mathrm{CO_2}} \mu_{nnn} + 6 m_{\mathrm{NaCl}} \left(\mu_{nnc} + \mu_{nna} \right) \right] \tag{S7}$$

$$\begin{aligned} \frac{\partial a_{\mathrm{H_2O}}}{\partial m_{\mathrm{CO_2}}} = -\frac{a_{\mathrm{H_2O}} M_{\mathrm{H_2O}}}{1000} \Big\{ 1 + 2 \Big[& m_{\mathrm{CO_2}} \lambda_{nn} + 3 m_{\mathrm{CO_2}}^2 \mu_{nnn} + m_{\mathrm{NaCl}} \left(\lambda_{nc} + \lambda_{na} \right) \\ & + 6 m_{\mathrm{CO_2}} m_{\mathrm{NaCl}} \left(\mu_{nnc} + \mu_{nna} \right) + m_{\mathrm{NaCl}}^2 \xi_{nca} \Big] \Big\} \end{aligned} \tag{S8}$$

The parameters of λ_{nn} and μ_{nnn} are adopted from Zhao et al. (2015[5]) as follows:

$$\lambda_{nn} = -0.16662 + 4.9482 \times 10^{-4} T - 5.1643 \times 10^{-7} T^2 \tag{S9}$$

$$\mu_{nnn} = -0.089196 - 2.8829 \times 10^{-4} T + 2.3197 \times 10^{-7} T^2 \tag{S10}$$

The parameters of λ_{nc}, λ_{na}, μ_{nna}, and μ_{nnc} are reported by Akinfiev and Diamond (2010)[82] as follows:

$$\begin{aligned} \left(\lambda_{nc} + \lambda_{na} \right) = {} & 0.057123 + \frac{2.6994}{T - 228} \\ & + \frac{0.045635}{2I} \left[1 - \left(1 + 2\sqrt{I} \right) \exp\left(-2\sqrt{I} \right) \right] \end{aligned} \tag{S11}$$

$$\left(\mu_{nnc} + \mu_{nna} \right) = -5.76 \times 10^{-4} \tag{S12}$$