A SAFT Equation of State for the H₂S-CO₂-H₂O-NaCl system and applications for CO₂ - H₂S transportation and geological storage

Xiaoyan Jiᵃ⁻*, Chen Zhuᵇ

ᵃEnergy Engineering, Division of Energy Science, Luleå University of Technology, 97187 Luleå, Sweden
ᵇDepartment of Geological Science, Indiana University, Bloomington, IN 47405, USA

Abstract

When H₂S is co-injected with CO₂, we need to know thermodynamic properties and phase equilibria for the CO₂-H₂S-H₂O-NaCl system in order to evaluate the storage capacity, optimal transportation and injection conditions, potential for pipeline corrosion, and increased risk of storage and leakage. Here, we summarize the results of the phase equilibrium and densities for CO₂-sequestration related systems from a thermodynamic model that is based on statistical associating fluid theory equation of state. The proposed thermodynamic model can be used to represent equilibrium compositions in both phases (H₂O-rich/aqueous phase and gas-rich/non-aqueous phase) and the density at equilibrium or in the one phase region. The effect of H₂S impurities on the properties of CO₂ streams and on the CO₂ solubility and the corresponding aqueous solution density can be predicted to provide a knowledge base for both transportation and geological storage. In addition, as other ions such as Ca, Mg, K and SO₄ have been taken into account in our model, the properties for brine in geological reservoirs in addition to simplified aqueous NaCl solutions can also be predicted. The equation of state from this study can be incorporated into reservoir simulators to model the separation of phases in multi-phase flow and fluid reactions with reservoir rocks.

© 2013 The Authors. Published by Elsevier Ltd.
Selection and/or peer-review under responsibility of GHGT

Keywords: CO₂ geological storage; H₂S co-injection; SAFT; thermodynamic properties; phase equilibria

1. Introduction

* Corresponding author. Tel.: +46-920-492837; fax: +46-920-491074.
E-mail address: xiaoyan.ji@ltu.se.
CO₂ storage in deep saline aquifers has been proposed as a promising option to mitigate CO₂ emission. Large-scale injection of CO₂ into saline aquifers will induce a variety of coupled physical and chemical processes including multiphase fluid flow, solute transport as well as chemical reactions between fluids and formation minerals. The presence of impurities in CO₂ streams makes these processes even more complicated. H₂S is a main impurity of CO₂ streams from flue gases and natural gas fields, and co-injection of CO₂ and H₂S is proposed as a means of substantially reducing the capture and sequestration costs. Therefore, it is crucial to know the thermodynamic properties and phase equilibrium for H₂S-CO₂-brine systems in order to understand the fate of the co-injected H₂S and CO₂, the risk of leakage, the environmental impacts as well as the effects of H₂S on all these processes. In general, the dominant dissolved salts in brines in saline aquifers are Na and Cl, and thus brine chemistry can be simplified to be aqueous NaCl solution, and thus the system of interest in the current work is H₂S-CO₂-H₂O-NaCl.

Experimental measurements have been carried out for both gas solubility and aqueous solution density, in related binary and ternary subsystems and the summary has been described in detail in our previous work [1]. In brief, CO₂ or H₂S solubility in water and aqueous NaCl solution has been measured in a wide temperature and pressure range [2-6]. However, there are almost no experimental data on the solubility of H₂S-CO₂ mixtures. The density was measured [7-13], but they are scarcer than those of the solubility data and no data are available for the CO₂-H₂S-H₂O-NaCl.

To simultaneously represent phase equilibrium and density at equilibrium or in the one phase region, a model based on equation of state (EoS) is preferred. Previous modeling work in the literature has been summarized in our previous work [1, 14], which reveals that for CO₂-H₂S-H₂O-NaCl, the research is limited due to the lack of the experimental data. It should be mentioned here that a model based on Peng-Robinson (PR) EoS, TMGAS, was used to represent thermophysical properties and phase equilibria of CO₂-H₂S-H₂O-NaCl [15]. However, in this model, the density of aqueous solution is independent from EoS, which we considered incorrect here. The detailed discussion on TMGAS was presented in our previous work [14].

Based on statistical associating fluid theory (SAFT) EoS, a model was developed to represent thermodynamic properties and phase equilibria for CO₂-H₂S-H₂O-NaCl [1, 14] and its subsystems [12, 16-22]. The modelling results were verified with available experimental data from their subsystems, and the phase equilibrium and aqueous solution density were investigated extensively. To systematically review the model results, our previous work was summarized in the current work, and then discussions as well as perspectives were provided.

**Nomenclature**

- \( a \) activity
- \( m \) molality of electrolyte
- \( P \) pressure
- \( T \) temperature
- \( x/y \) mole fraction in aqueous / non-aqueous phase
- \( \gamma \) mean activity coefficient of electrolyte
- \( \rho \) density
2. Modeling

SAFT EoS was proposed 20 years ago. It has been widely used and accepted that it is superior over the simple cubic EoS. The key of this model is their firm statistical-mechanics basis, which allows for a rigorous physical interpretation of the contributions due to the various intermolecular interactions, such as hard-sphere, dispersion, chain, association, polar and ionic interactions. This provides a framework from which the effects on the thermodynamic properties of the various molecular features can be separated and quantified. It has been used successfully to describe the thermodynamic properties and phase behavior of fluids including associating molecular, electrolytes as well as polymers up to high pressures for which the ubiquitous and popular cubic equations of state fail to provide an adequate description [23].

For the system of interest in the current work, CO₂ is a molecule with quadrupolar interactions, H₂S is a polar molecule, H₂O is a molecule with hydrogen bond, and NaCl is a charged molecule, which makes SAFT EoS a best option to choose. While SAFT model itself has several versions, such as SAFT-VR, soft-SAFT, Perturbation chain (PC)-SAFT, SAFT1-RPM/SAFT2 [23]. Up to now, only SAFT2, the extension of SAFT1-RPM, has been developed to represent properties of aqueous electrolyte solutions up to high temperatures and pressures [20, 21], and the extension of SAFT2 to the system of interest was feasible. Thus, SAFT2 was chosen in our work. Following SAFT2, the dimensionless residual Helmholtz energy is defined as the summation of terms accounting for the segment, association, chain, and ionic interactions, respectively. The detailed description was in reference [14].

SAFT2 models a neutral substance with four parameters: segment number \( m \), segment volume \( \nu^{oo} \), segment energy \( u/k \), and the reduced range of the potential well \( \lambda \). For ionic (charged) substances, there is one more parameter, effective diameter \( d \). For the substance with association interactions, two additional parameters are needed: the well depth of the association site-site potential \( \kappa \) and the parameter related to the volume available for bonding \( \kappa \). Generally, these parameters are determined by fitting to liquid-density and/or vapor-pressure data of a pure component.

Mixing rules are required for mixtures. To improve the model performance, two adjustable parameters can be used to adjust the cross association to account for special interactions such as hydrogen bond and, in general, one parameter is also needed to adjust the dispersion energy. These parameters (if required) are usually determined by fitting to binary or ternary data, e.g., to the gas solubility in solutions [14].

3. Results

To represent properties of CO₂-H₂S-H₂O-NaCl up to high pressures, research has been carried out for several years for systems of NaCl-H₂O/brine [16-21], CO₂-H₂O-NaCl/brine [12, 14], H₂S-H₂O-NaCl [14, 22], and CO₂-H₂S-H₂O-NaCl [1, 14].

3.1. NaCl-H₂O / brine
In the field of geological carbon storage, it is common that the brine is simplified as or refers to aqueous NaCl solution as NaCl is usually the main component. This is feasible when the gas solubility and aqueous solution density are the main research focus. However, in the process that the dissolved CO₂reacts with rock, it can happen that an existent rock is dissolved and new minerals precipitate. In this case, it is also necessary to know the properties of other ions, such as their activities. This is because that the pressure is high, which makes the activity considerably different from concentration. Thus, to cover the whole interests in geochemistry, it is necessary to study the properties of “true” brine. Generally, the components in brines include Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, and SO₄²⁻.

The properties of aqueous solutions with electrolytes including Li⁺, Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, Br⁻, HCO₃⁻, SO₄²⁻, and CO₃²⁻ at temperatures from 298.15 to 473.15 K, at pressures up to 1000 bar with 0 to 6 mol/kgH₂O electrolytes in ionic strength were studied in our work [16-21]. The properties of aqueous NaCl solution, such as density, can be predicted up to 1000 bar reliably as shown in Figure 1a in which the symbols represent the results from the well-known Pitzer model [20]. As all the main components (ions) in brine have been studied, the properties of brine can be predicted. The density of brine at different temperatures was predicted with the typical composition taken from literature [24] and compared with available experimental data from 298.15 to 423.15 K. The model prediction agreed well with experimental data [16] as shown in Figure 1b where the chlorinity is the mass of chlorine in 1 kg solution.

As mentioned in the foregoing text that the properties of brine are assumed to be those for aqueous NaCl solution. To study the error caused by this assumption, the properties for both brine of typical composition and aqueous NaCl solutions were predicted, respectively, and then compared. As shown in Figure 2a, the activity of water in aqueous NaCl solution is similar to that for brine at different salinity (= total mass of salts in 1 kg solution), which proves that the simplification of brine to aqueous NaCl solution is reasonable when phase equilibrium is the research focus. The density of brine is slightly higher than that for aqueous NaCl solution at high chlorinity as shown in Figure 2b, and the largest error goes up to 3.5% under the conditions studied, which is still acceptable. Therefore, the comparison confirms that it is reasonable to assume brine as aqueous NaCl solutions.

The activity coefficient is used to adjust concentration when the non-ideal behavior is considerable. When the activity coefficient is almost a constant in the concentration range of interest, the adjustment can be merged to other parameters those are independent of concentration. In general, compared to NaCl, the concentration of Ca (CaCl₂) or Mg (MgCl₂) in brine is low, but it is still unclear that the activity
Coefficient is a constant or not. Using the developed model, the mean activity coefficients of both CaCl₂ and MgCl₂ in brine with the typical composition taken from literature [24] were predicted and illustrated in Figure 3a. For comparison, the mean activity coefficients of NaCl in brine are also illustrated in Figure 3a. When the ratio of Ca²⁺/Mg²⁺ to Na⁺ is fixed, with increasing salinity, the mean activity coefficients of both CaCl₂ and MgCl₂ decrease significantly and then increase. We also predicted the mean activity coefficients of CaCl₂ and MgCl₂ in water at 400 bar and 323.15 K, respectively. The prediction was compared with experimental data [20] which is illustrated in Figure 3b. Again, the activity coefficients vary from 1 to 0.45 when the concentration goes from zero to 0.5 mol/kgH₂O for both of them. All these observations imply that it is necessary to study the properties of other main components besides NaCl in water.

![Figure 2. Properties for aqueous NaCl solutions and brine/seawater at 298.15 K. Symbols: experimental data [16]; Curves: model prediction; salinity: total mass of salts in 1 kg solution; Chlorinity: the mass of chlorine in 1 kg solution.](image)

![Figure 3. Mean activity coefficients. (a) NaCl, CaCl₂, MgCl₂ in brine (Na⁺-K⁺-Ca²⁺-Mg²⁺-Cl⁻-SO₄²⁻-H₂O solutions with typical compositions taken from literature [24]) at 298.15 K. (b) CaCl₂ and MgCl₂ in aqueous CaCl₂ and aqueous MgCl₂ solutions, respectively, at 323.15 K.](image)

3.2. CO₂-H₂O-NaCl (CO₂-brine)
As NaCl is the main component of brine, the phase equilibrium and properties of CO₂-H₂O-NaCl play an important role in CO₂ geological storage. Models based on equation of state have been proposed, but predictive capacity is quite limited for most of them. One of the main reasons is the difficulty in description of systems with charged species under high pressures. For cubic equation of state, the reference state is the ideal gas state, which makes the prediction at high pressures not promising. In addition, the involvement of NaCl, the charged species, increases the challenge of the research work. Pitzer equation is a successful method for the system with charged species, and the extension to high pressures has been carried out, however, the intrinsic characterization of the G² model limits the description of other thermodynamic properties, for example, the density.

As shown in the foregoing section, the SAFT-based models have been proposed to represent the properties of aqueous NaCl solutions up to high pressure. Meanwhile, it is worthy to mention that the hard-sphere is used as reference state in SAFT-based model, which ensures reliable prediction at high pressures as the higher the pressure, the closer the system goes to hard sphere. The SAFT-based model has been extended to CO₂-H₂O-NaCl systems, and the verification of the model has been described in our previous work [12, 14].

Using the extended model, the phase equilibria of CO₂-H₂O at 298.15, 323.15, 348.15 and 373.15 K and at pressures up to 400 bar were predicted in the current work. The corresponding equilibrium compositions in H₂O-rich (aqueous) and CO₂-rich (non-aqueous) phases are illustrated in Figure 4. The solubility of H₂O in CO₂ decreases and then increases with increasing pressure, and it increases with increasing temperature. The equilibrium H₂O content in CO₂ is a limiting factor for CO₂ transportation. To avoid the risk of corrosion and hydrate formation, it is better to choose the pressure range in which the H₂O content increases with increasing pressure. The CO₂ solubility in H₂O increases rapidly and then slowly with increasing pressure at low temperatures. With increasing temperature, the CO₂ solubility in water decreases in the low pressure range, and it decreases first and then increases in the high pressure range. At low temperature, the phase equilibrium is changed from vapor-liquid equilibrium to liquid-liquid equilibrium with increasing pressures.

![Figure 4. Phase equilibrium for CO₂-H₂O.](image)

In the foregoing text, it has been shown the activity of water in brine and the density of brine are almost the same values as those for aqueous NaCl solution, which means that it is reasonable to simplify brine as aqueous NaCl solutions when the thermodynamic properties and phase equilibrium are the research focus. To further verify this statement, CO₂ solubility and aqueous solution density with dissolved CO₂ calculated for CO₂-H₂O-NaCl were compared with the experimental data of CO₂ solubility in brine and solution density of brine with dissolved CO₂ measured by Yang and Gu [5]. The comparison is shown in
Figure 5 with good agreement. This observation proves again that it is reasonable to simplify brine as aqueous NaCl solutions when the thermodynamic properties and phase equilibrium are the research focus.

The CO₂ solubility in aqueous NaCl solutions and the solution density with dissolved CO₂ were predicted at temperatures from 298.15 to 373.15 K and at pressures up to 400 bar with 2 mol/kg H₂O of NaCl. As shown in Figure 6a, similar to the CO₂ solubility in water, the CO₂ solubility in aqueous NaCl solutions increases rapidly and then slowly with increasing pressure at low temperatures. With increasing temperature, the CO₂ solubility in aqueous NaCl solutions decreases at low pressures, and it decreases first and then increases at high pressures. Compared to CO₂ solubility in water, the CO₂ solubility in aqueous NaCl solutions decreases with increasing concentration of NaCl.

![Figure 5. CO₂ solubility in brine and the aqueous solution density with dissolved CO₂. Symbols: (a) experimental CO₂ solubility in brine and (b) experimental brine density with dissolved gas [5]. Curves: model prediction for CO₂-H₂O-NaCl (n_{NaCl} = 0.07414 mol/kg H₂O)](image)

![Figure 6. CO₂ solubility in aqueous NaCl solutions and the aqueous solution density with dissolved CO₂.](image)
The density of solution at equilibrium is depicted in Figure 6b. The density at equilibrium increases with increasing pressure and decreasing temperature. At a fixed temperature, with increasing pressure, the CO₂ solubility increases. From the results shown in Figure 6b, it is not clear that this increasing dissolved CO₂ will increase or decreases the solution density. To study specifically the effect of dissolved CO₂ on the solution density, a special case was chosen at 348.15 K, 135 bar and with 2 mol/kgH₂O NaCl. The model results are shown in Figure 7a. It is clear that the dissolved CO₂ will increase solution density. This is very important for CO₂ dissolution process. With CO₂ dissolution, the aqueous solution density becomes denser, which can induce natural convection and hence enhance CO₂ dissolution (solubility trapping) in the reservoir.

![Figure 7. Density of (a) aqueous solution with dissolved gas at 135 bar, 348.15 K and m_{NaCl} = 2 mol/kgH₂O and (b) mixture of H₂S+CO₂ at 323.15 K.](image)

3.3. CO₂-H₂S-H₂O-NaCl

H₂S is one of the most common components in natural gas and products derived from oil processing and production. For example, the natural gas industry in the Alberta basin, Canada produces a significant amount of sour gases (H₂S+CO₂). Since surface desulphurization and surface storage is not economical and carries significant liability, acid gas (mixture of CO₂ and H₂S, also referred to as “sour gas”) disposal is operated through injection into depleted oil and gas reservoirs and deep saline formations.

In addition, in general, CO₂ streams from power stations and other CO₂ intensive industries are mixtures, and the separation of CO₂ from gas mixtures is the main cost for CO₂ capture. In fact, it is estimated that ¼ of the cost is used for CO₂ separation from gas streams. H₂S is one of the main impurities of flue gas streams from thermal power plants. The injection of H₂S-CO₂ mixtures can lead to potentials for great capital and energy savings in capture, but it may also seriously increase the costs and risks of CO₂ transportation and storage. For example, the inclusion of H₂S will increase the risk of pipeline corrosion and the dissolution of H₂S into the brine in the reservoirs seriously alters the geochemistry.

In order to understand the effects of H₂S on CO₂ transportation and geological storage, the SAFT-based model has been developed for H₂S-H₂O, H₂S-H₂O-NaCl, and H₂S-CO₂ systems. The model results have been verified with available experimental data to ensure the accuracy. Based on this, the model has
been extended to CO₂-H₂S-H₂O-NaCl, and the effect of H₂S on both CO₂ transportation and geological storage were discussed in our recent publication [1], and the main observations are summarized in the following.

To study the effect of H₂S on CO₂ transportation, using the developed model, the phase state and density for CO₂-H₂S streams were predicted. The model prediction shows that with inclusion of H₂S, phase split may occur, and the density increases and then decreases with increasing pressure. Figure 7b shows the model prediction at 323.15 K in which the dashed lines represent two-phase flow and the solid lines represent density in the single-phase region. In CO₂ transportation, two-phase flow in the pipeline could cause cavitation and pressure peaks and would damage the pipeline. Two-phase flow is also problematic in the operation of pumps, as well as compressors and injection wells. Therefore, the inclusion of H₂S increases the difficulty for CO₂ transportation.

The density of the (supercritical) gas phase reflects the amount of the fluid that can be stored at a fixed volume in reservoirs, as well as the transportation capacity. As shown in Figure 7b, due to the inclusion of H₂S, the mass density increases at low pressures, which means that the volume of the mixture decreases, leading to an increase in the amount of the gases that can be stored in a fixed volume in the reservoirs. Thus, the inclusion of H₂S in this low pressure range is favorable for CO₂ geological storage from the point of view of gas volume.

The inclusion of H₂S will also affect the gas solubility as well as the aqueous solution density with dissolved gas. Figure 7a shows the density of aqueous solution with dissolved H₂S+CO₂ at 348.15 K, 135 bar and with 2 mol/kgH₂O NaCl. With inclusion of H₂S, the aqueous solution density with dissolved gas decreases and may even less than that without dissolved gas. This is very important for gas dissolution process. If dissolution of gas into brine causes an increase of brine density and this density difference may also be large enough to trigger Rayleigh instability, which can strongly enhance dissolution processes due to mixing. However, if the dissolution of gas into the brine decreases the brine mass density, the brine can be subject to buoyancy-driven migration (and potential escape from formation) associated with separate gas phase. Thus, the inclusion of H₂S is not good for dissolution process.

---

**Figure 8.** Gas solubility (a) at 323.15 K, \( m_{NaCl} = 0 \); (b) at 348.15 K, 135 bar, \( m_{NaCl} = 2 \) mol/kgH₂O.
H$_2$S is more soluble in H$_2$O or brine than CO$_2$, and CO$_2$ can also be dissolved in liquid H$_2$S. With the inclusion of H$_2$S, the solubility of gas mixture of (H$_2$S+CO$_2$) is higher than that for pure CO$_2$. With increasing inclusion of H$_2$S, the H$_2$S solubility increases while the CO$_2$ solubility decreases.

4. Conclusion

Based on statistical associating fluid theory (SAFT) EoS, a model was developed to represent thermodynamic properties and phase equilibrium for CO$_2$-H$_2$S-H$_2$O-NaCl and its subsystems. To systematically review the model results, in the current work, we summarize the results of the phase equilibrium and densities for CO$_2$-sequestration related systems. The proposed thermodynamic model can be used to represent equilibrium compositions in both phases (H$_2$O-rich/aqueous phase and gas-rich/non-aqueous phase) and the density at equilibrium or in the one phase region. The effect of H$_2$S on the properties of CO$_2$ streams and on the CO$_2$ solubility and the corresponding aqueous solution density with dissolved gas can be predicted to provide knowledge for both transportation and geological storage. In addition, as other ions such as Ca, Mg, K and SO$_4$ have been taken into account in the model, the properties for brine instead of aqueous NaCl solutions can be predicted. The equation of state can be incorporated into reservoir simulators to simulate the separation of phases in multi-phase flow and fluid reactions with reservoir rocks.

Acknowledgements

X. Ji thanks the Swedish Research Council for the financial support, and CZ acknowledges the support of the U.S. Department of Energy grant DE-FE0004381, the Faculty Research Support Program from Indiana University, the support from the Norwegian Center of Excellence Subsurface CO$_2$ storage – Critical Elements and Superior Strategy (SUCCESS), and a Fulbright scholarship to Norway.
References

[1] Ji X, Zhu C. Predicting possible effects of H₂S impurity on CO₂ transportation and geological storage. *Environ. Sci Technol.* 2012, in press.

[2] Ji Y, Ji X, Feng X, Liu C, Lu L, Lu X. Progress in the study on the phase equilibria of the CO₂-H₂O and CO₂-H₂O-NaCl systems. *Chinese J. Chem. Eng.* 2007, 15(3):439-448.

[3] Duan Z, Sun R, Liu R, Zhu C. Accurate thermodynamic model for the calculation of H₂S solubility in pure water and brines. *Energy & Fuels* 2007, 21(4):2056-2065

[4] Yan W, Huang S, Stenby E. Measurement and modeling of CO₂ solubility in NaCl brine and CO₂-saturated NaCl brine density. *Int. J. Greenhouse Gas Control* 2011, 5(6):1460-1477

[5] Yang C, Gu Y. Accelerated mass transfer of CO₂ in reservoir brine due to density-driven natural convection at high pressures and elevated temperatures. *Ind. & Eng. Chem. Res.* 2006, 45(8):2430-2436.

[6] El-Maghraby R, Pentland C, Iglauer S, Blunt M. A fast method to equilibrate carbon dioxide with brine at high pressure and elevated temperature including solubility measurements. *J. Supercrit. Fluids* 2012, 62:55-59.

[7] Bachu S, Watson T. Review of failures for wells used for CO₂ and acid gas injection in Alberta. *Canada. Energy Procedia* 2009, 1(1):3531-3537

[8] Hidalgo J, Carrera J. Effect of dispersion on the onset of convection during CO₂ sequestration. *J. Fluid Mech.* 2009, 640:441-452

[9] Riaz A, Hesse H, Tchelepi F. Onset of convection in a gravitationally unstable, diffusive boundary layer in porous media. *J. Fluid Mech.* 2006, 548:87-111

[10] Vilarasa V, Bolster D, Dentz M, Olivella S, Carrera J. Effects of CO₂ Compressibility on CO₂ Storage in Deep Saline Aquifers. *Transp. Porous Media* 2010, 85:619-639

[11] Adams J, Bachu S. Equations of state for basin geofluids: algorithm review and intercomparison for brines. *GeoFluids* 2002, 2:257-271

[12] Ji X, Tan S, Adidharma H, Radosz M. SAFT1-RPM approximation extended to phase equilibria and densities of CO₂-H₂O and CO₂-H₂O-NaCl systems. *Ind. & Eng. Chem. Res.* 2005, 44(22):8419-8427

[13] Zhang Y, Chang F, Song Y, Zha J, Zhan Y, Jian, W. Density of Carbon Dioxide plus Brine Solution from Tianjin Reservoir under Sequestration Conditions. *J. Chem. Eng. Data* 2011, 56(3):565-573

[14] Ji X, Zhu C. A SAFT equation of state for the quaternary H₂S-CO₂-H₂O-NaCl system. *Geochim. Cosmochim. Acta* 2012, 91:40-59

[15] Battistelli A, Marcolini, M. TMGAS: A new TOUGH2 EOS module for the numerical simulation of gas mixtures injection in geological structures. *Int. J. Greenhouse Gas Control* 2009, 3:481-493

[16] Ji X, Tan S, Adidharma H, Radosz M. Statistical associating fluid theory coupled with restricted primitive model to represent aqueous strong electrolytes: Multiple-salt solutions. *Ind. & Eng. Chem. Res.* 2005, 44(19):7584-7590

[17] Ji X, Tan S, Adidharma H, Radosz M. Statistical associating fluid theory coupled with restricted primitive model extended to bivalent ions. SAFT2: 2. Brine/seawater properties predicted. *J. Phys. Chem. B* 2006, 110(33):16700-16706

[18] Tan S, Ji X, Adidharma H, Radosz M. Statistical associating fluid theory coupled with restricted primitive model extended to bivalent ions. SAFT2: 1. Single salt plus water solutions. *J. Phys. Chem. B* 2006, 110(33):16694-16699

[19] Ji X, Adidharma H. Ion-based SAFT2 to represent aqueous single- and multiple-salt solutions at 298.15 K. *Ind. & Eng. Chem. Res.* 2006, 45(22):7719-7728

[20] Ji X, Adidharma H. Ion-based statistical associating fluid theory (SAFT2) to represent aqueous single-salt solutions at temperatures and pressures up to 473.15 K and 1000 bar. *Ind. & Eng. Chem. Res.* 2007, 46(13):4667-4677

[21] Ji X, Adidharma H. Ion-based SAFT2 to represent aqueous multiple-salt solutions at ambient and elevated temperatures and pressures. *Chem. Eng. Sci.* 2008, 63(1):131-140
[22] Ji X, Zhu C. Modelling of phase equilibria in the H$_2$S-H$_2$O system with statistical associating fluid theory. 
*Energy & Fuels* 2010, **24**:6208-6213

[23] Tan S, Adidharma H, Radosz M. Recent Advances and Applications of Statistical Associating Fluid Theory, 
*Ind. & Eng. Chem. Res.* 2008, **47**(21):8063-8082

[24] Platford R. The activity coefficient of sodium chloride in seawater. *J. Mar. Res.* 1965, **23**(2):55-62