SUPPLEMENTARY INFORMATION

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Optimisation results for dispersive and associative models



1. Results from model 1.0 using strategy a

Figure 1 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 2 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 3 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.



Figure 4 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. All isotherms follow the same line. The calculations were made at 1 bar.

2. Results from model 1.1 using strategy a.



Figure 5 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 1.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 6 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 1.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 7 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 1.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.



Figure 8 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 1.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. All isotherms follow the same line. The calculations were made at 1 bar.

3. Results from model 1.2 using strategy a.



Figure 9 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 1.2 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 10 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 1.2 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. All isotherms follow the same line. The calculations were made at 1.

4. Results from model 1.0 using strategy b.



Figure 11 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy b. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.



Figure 12 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy b. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 13 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 1.0 using optimisation strategy b. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.

5. Results from model 2.0 using strategy a.



Figure 14 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 2.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 15 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 2.0 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.

6. Results from model 2.1 using strategy a.



Figure 16 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 17 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 18 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.



Figure 19 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. All isotherms follow the same line. The calculations were made at 1 bar.

7. Results from model 2.2 using strategy a.



Figure 20 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 21 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 22 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. The calculations were made at 1 bar.



Figure 23 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 2.1 using optimisation strategy a. The symbols represent the experimental data and the lines represent the results obtained with the model. All isotherms follow the same line. The calculations were made at 1 bar.

8. Dispersive models



Figure 24 : Effect of the various terms on the logarithm of mean ionic activity coefficient for aqueous NaCl in function of the salt concentration at 298 K using model 1.0. Where hs=Hard sphere, disp=dispersion, asso=association, polar=polar, msa= MSA and born=Born terms, sum = sum of all terms and Exp= experimental data. The calculations were made at 1 bar.



Figure 25 : Effect of the various terms on the logarithm of mean ionic activity coefficient for aqueous NaCl in function of the salt concentration at 298 K using model 1.1. Where hs=Hard sphere, disp=dispersion, asso=association, polar=polar, msa= MSA and born=Born terms, sum = sum of all terms and Exp= experimental data. The calculations were made at 1 bar

9. Associative models



Figure 26 : Effect of the various terms on the logarithm of mean ionic activity coefficient for aqueous NaCl in function of the salt concentration at 298 K using model 2.1. Where hs=Hard sphere, disp=dispersion, asso=association, polar=polar, msa= MSA and born=Born terms, sum = sum of all terms and Exp= experimental data. The calculations were made at 1 bar.



Figure 27 : Effect of the various terms on the logarithm of mean ionic activity coefficient for aqueous NaCl in function of the salt concentration at 298 K using model 2.2. Where hs=Hard sphere, disp=dispersion, asso=association, polar=polar, msa= MSA and born=Born terms, sum = sum of all terms and Exp= experimental data. The calculations were made at 1 bar.

Dielectric constant as a function of salt concentration.

10. Schreckenberg model



Figure 28 : Dielectric constant in function of the salt concentration at various temperatures, using Schreckenberg model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.

11. Pottel model



Figure 29 : Dielectric constant in function of the salt concentration at various temperatures, using Pottel model. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.

12. Simonin model



Figure 30 : Dielectric constant in function of the salt concentration at various temperatures, using Simonin model with parameters from model 2.0. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.

Optimisations results from extension to 4 salts (NaCl, KCl, NaBr and KBr)

1,1 1,1 ▲298.15K -373.15K ◆273.15K × 323.15K ◆273.15K ▲288.15K ▲ 398.15K = 473.15K + 348.15K (b) (a) **×**291.15K + 298.15K 1 1 ×423.15K • 448.15K ▲ 323.15K - 308.15K 0,9 0,9 0,8 0,8 $^{\chi_{\pm}}_{\pi}$ 0,7 0,7 0,6 0,6 0,5 0,5 0,4 0,4 $\frac{2}{m/(\text{mol}\cdot\text{kg}^{-1})}$ 0 6 $\frac{2}{m/(\text{mol}\cdot\text{kg}^{-1})}$ 0 6 4 1,4 1,1 ▲ 278.15K ◆273.15K ◆298.15K 1,3 (d) ×283.15K +288.15K (c) 1 1,2 293.15K ▲ 298.15K × 303.15K • 313.15K 0,9 1,1 ${{\mathfrak{s}}_{+}\atop \overset{}{\scriptstyle \prec}}^{0,8}$ 1 ε₊₁ 0,9 0,7 0,8 0,7 0,6 0,6 0,5 0,5 0,4 0,4 4 0 2 4 6 0 2 6 m / (mol·kg-1) $m / (\text{mol} \cdot \text{kg}^{-1})$

13. Dispersive model (Model 1.0)

Figure 31 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 1.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 32 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 1.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 33 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 1.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1.



Figure 34 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 1.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.

14. Associative model (Model 2.0)



Figure 35 : Mean ionic activity coefficient in function of the salt concentration obtained from the optimisation of model 2.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 36 : Osmotic coefficient in function of the salt concentration obtained from the optimisation of model 2.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.



Figure 37 : Enthalpy of solution in function of the salt concentration obtained from the optimisation of model 2.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar.



Figure 38 : Apparent molar volume in function of the salt concentration obtained from the optimisation of model 2.0. The symbols represent the experimental data and the lines represent the results obtained with the model. (a) NaCl ; (b) KCl ; (c) NaBr ; (d) KBr. The calculations were made at 1 bar for temperatures up to 373.15 K, then the saturation pressure of the solvent was used for temperatures above 373.15 K.