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1 Systematic evaluation of parameterization approaches for the ePPC- 2 SAFT model for aqueous alkali halide solutions

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10 **Abstract:** This work presents an analysis of various electrolyte SAFT model approaches
11 through a rigorous benchmarking on extensively collected and critically evaluated databases.
12 The primitive mean spherical approximation (MSA) and the Born equation are used
13 respectively for the long-range ion-ion and ion-solvent interactions. For the short range
14 interactions either dispersion, or association, or both (full) are investigated. Doing so, state-of-
15 the-art parameter sets are obtained for the ePPC-SAFT model. Physical consistency is enforced
16 for the parameters in the regression. Efforts are made to reduce the number of adjustable
17 parameters with minimum loss of accuracy. This is done by analyzing the physical indication
18 of the parameters, parameter sensitivity analysis, parameter trends, and trial-and-error. The
19 model and parameter sets accurately represent the mean ionic activity coefficient (MIAC),
20 vapor-liquid equilibria, and density, and accurately predict the osmotic coefficient extrapolated
21 to temperature and salt composition ranges beyond the range of the MIAC data used in the
22 regression. The ion-specific association strategies are found to be approximately as accurate as
23 the salt-specific strategies, and are more accurate than the ion-specific dispersion and full
24 strategies. Contributions of the model terms to the MIAC are analyzed. Temperature-
25 dependence of the MIAC is discussed. The ion-specific association strategies successfully
26 predicts the opposite relative magnitudes of the cation and anion individual ion activity
27 coefficient of the aqueous NaCl and KCl solutions. The information is not included in model
28 parameterization, while all the salt-specific strategies and ion-specific dispersion and full
29 strategies fail. We recommend including the Wertheim association for the short-range ion-ion

30 and ion-solvent interactions, and parameterizing SAFT models in an ion-specific manner using
31 physically consistent parameters.

32 **Keywords:** equation of state, aqueous electrolyte solutions, activity coefficient, vapor-liquid
33 equilibria, density.

34 **1 Introduction**

35 Thermodynamics of electrolyte solutions is important in many industrial and chemical
36 processes, e.g., carbon capture and sequestration [1], flue gas treatment [2], geochemical
37 processes [3], desalination [4], and batteries [5,6]. When ions are present in mixtures, long-
38 range ion-ion and ion-solvent interactions occur; thermodynamic properties of the electrolyte
39 solutions are significantly different from those of molecular systems even at low salt
40 concentration. Modeling these properties over ranges of temperature, pressure, and
41 composition is important for the design and improvements of the relevant processes. Over the
42 years, experimental data have been obtained for many electrolyte solutions; activity coefficient
43 models and equations of state have been extended to mixtures involving electrolytes. In
44 previous works [7–9], we have extensively collected and critically evaluated experimental
45 thermodynamic property data of alkali halides in water and mixed-solvents. The databases
46 cover various properties, including the mean-ionic activity coefficient (MIAC), vapor pressure,
47 osmotic coefficient (OC), density, etc. Benchmarking the thermodynamic property models to
48 these verified databases can help identify the contribution terms that are to be included in the
49 models, the parameters that are to be regressed, and the properties that are to be included in the
50 objective functions. The information will be useful when the models are applied on other
51 electrolyte systems, for which experimental data are not as extensively available as for systems
52 that have been covered by the reference databases.

53 Thermodynamics of electrolyte solutions has been reviewed in the literature, e.g., by
54 Loehe and Donohue [10], Kontogeorgis et al. [11], Ahmed et al. [12], Held [13], etc. Activity
55 coefficient models and equations of state (EoS) have been combined with electrolyte terms to
56 account for the long-range ion-ion and ion-solvent interactions. There are a few works in which
57 a non-primitive mean spherical approximation (MSA) model has been implemented [14–17],
58 i.e., long-range ion-ion, ion-dipole, and dipole-dipole interactions are accounted for explicitly.
59 The primitive MSA [18] or Debye-Hückel (DH) [19] theories are usually implemented in the
60 EoS, in which the solvent is treated as a dielectric continuum. The ion-dipole and dipole-dipole
61 interactions are accounted for using extra contribution terms in the EoS, e.g., the dispersion,

62 association, Born [20], and multipolar [21–23] terms. Maribo-Mogensen et al. [24] compared
63 the MSA and DH theories, and concluded that they are similar within the EoS framework. In
64 the following brief summary, a special focus would be on how the short-range interactions have
65 been treated differently in the SAFT models, because benchmarking the short-range
66 approaches is one of the goals of this work. The short-range interactions are modeled very
67 differently in the literature:

- 68 - Some models use dispersion approach; some use the association approach; some use both;
69 some use neither.
- 70 - In the dispersion approach, some models include like-ion dispersion; some do not include
71 like-ion dispersion; some do not include ion-ion dispersion at all, but only include ion-
72 water dispersion.
- 73 - In the association approach, some models include both cation-anion association and ion-
74 water association; some only include ion-water association; some include cation-water
75 association, but not anion-water association.

76 Galindo et al. [25] proposed the SAFT-VRE model, combining the SAFT-VR [26] with
77 the MSA term. Ion-water dispersion was included, while ion-ion dispersion was not. Gil-
78 Villegas et al. [27] compared the dispersion and association approaches for the short-range ion-
79 water interaction. The Born term was added to the model as the model was applied on mixed-
80 solvent electrolyte solutions [28]. Eriksen et al. [29] replaced the square-well dispersion term
81 with the Mie potential. They included the dispersion interaction for the ion-ion and ion-water
82 pairs, and the association interaction for the ion-water pairs. Liu et al. [30] extended a SAFT
83 model to electrolyte solutions using a primitive MSA term. Later, cation-water association was
84 included in the model, improving the results [31]; the MSA term was replaced using an
85 expansion of the non-primitive MSA theory [32]. However, Das et al. [16] commented that the
86 ion-solvent term in the low-density expansion reduced to the Born expression in the primitive
87 MSA model. Cameretti et al. [33] combined the PC-SAFT model [34,35] with the DH term.
88 Ion-solvent dispersion was included. Cation-anion dispersion was shown to improve results,
89 while no like-ion dispersion was included [36]. Later on, the model was revised, adding
90 composition dependent relative permittivity and the Born term [37]. Rozmus et al. [38]
91 combined the PPC-SAFT model (a PC-SAFT revision with a multipolar term) with the MSA
92 and Born terms. Association was included for the ion-ion and ion-solvent pairs. The model was
93 revised with a temperature-dependent water diameter parameter, significantly improving the
94 accuracy for liquid density, and was applied on mixed-solvent electrolyte solutions [12,39].

95 Roa Pinto et al. [40] investigated the temperature-dependence, short-range interactions, and
96 relative permittivity models in the ePPC-SAFT framework. Association and dispersion
97 approaches were compared for the short-range ion-ion and ion-solvent interactions. Like-ion
98 dispersion was firstly included in the dispersion approach, and was removed in an attempt to
99 reduce the number of adjustable parameters. The increasing mean ionic activity coefficient
100 (MIAC) with temperature of the aqueous NaCl solution at low temperature was captured in the
101 association approach. Maribo-Mogensen et al. [41] combined the CPA equation of state [42]
102 with the DH theory. Walker et al. [43] compared the SAFT-VR Mie + DH and + MSA, and
103 investigated the impact of the temperature-, density-, and composition-dependence of the
104 relative permittivity model. Short-range ion-ion and ion-solvent interactions were modeled in
105 the dispersion approach. Selam et al. [44] combined the SAFT-VR Mie model with the DH and
106 Born terms. Both like- and unlike-ion dispersions were included. An attempt was made to
107 capture the temperature-dependence of aqueous NaCl MIAC at low temperature using a
108 temperature-dependent ion-water dispersion energy parameter. The model was applied on
109 modeling gas solubility in aqueous electrolyte solutions [45].

110 The use of the readily available Wertheim association term for short-range ion-ion and
111 ion-solvent interactions has been successful [28,31,40]. Wu and Prausnitz [46] stated that the
112 Born term “gives only part of the change in Helmholtz energy due to charging neutral particles
113 because the interaction between water and the charged particles also depends on the
114 electrostatic states”, and introduced “additional association interactions between ions and
115 solvent (water) molecules to represent interactions between charged ions and water”. However,
116 when it comes to ion-ion interaction, the association decreases with temperature, contrary to
117 the increasing trend of ion-pairing. As discussed in the last paragraph, the short-range
118 interactions have been considered using very different assumptions, giving rise to the following
119 questions:

- 120 - It has been assumed in some works that like-ion dispersion could be neglected. One could
121 expect the like-ions to be far from each other, reducing this contribution. However, is the
122 model “smart” enough to exclude dispersion between like-ions?
- 123 - Should dispersion, association, or both short-range interactions be used for ion-ion and
124 ion-solvent interactions in the models? A full model would include both dispersion and
125 association for the short-range ion-ion and ion-solvent interaction. After all, one could not
126 assume associating ions to be free of dispersion interaction. However, the approach was
127 rarely considered. With advanced algorithms, the increased number of parameters would

128 not pose difficulty in the regression. Would there be any improvements if the complete
129 approach was considered?

130 - Can the experimental data be represented accurately without introducing physically
131 inconsistent parameters?

132 - Is there any drawback in using the ion-specific parameters instead of salt-specific ones?

133 Following Ref [40], the ePPC-SAFT model is parameterized in this work in an ion-
134 specific approach; physical consistency of parameters is enforced in the regression. State-of-
135 the-art parameter sets are obtained to facilitate the benchmarking of the modeling approaches,
136 based on extensively collected and critically evaluated databases. Efforts are made to reduce
137 the number of parameters with minimum loss of accuracy. The final parameter set is
138 determined based on analyzing the physical indication of the parameters, parameter sensitivity
139 analysis, parameter trends, and trial-and-error. Ion-specific parameters are obtained within a
140 series of alkali chloride aqueous solutions. Properties that are useful in practical applications
141 are included in the objective function, i.e., MIAC, vapor-liquid equilibria (VLE), and density.
142 In future works, the model will be extended to mixed-solvent electrolyte solutions. The osmotic
143 coefficient is not included in the regression, but is used as a validation of the model and
144 parameter set. Efforts are made to improve density correlation, using our previously evaluated
145 reference database for aqueous alkali halide solutions [9]. The association, dispersion, and full
146 approaches are compared. In the dispersion and full approaches, the effect of the like-ion
147 dispersion is investigated. The obtained ion-specific parameter sets are tested on the qualitative
148 behavior of cation and anion individual ion activity coefficients (IIAC) of aqueous NaCl and
149 KCl solutions. It is observed that the respective magnitudes of the cation and anion IIAC is
150 opposite for the 2 aqueous solutions. This phenomenon has never been successfully predicted
151 in the literature. The manuscript is organized as follows. In Section 2, the model, database, and
152 parameterization strategies for the short-range ion-ion and ion-solvent interactions are
153 introduced. In Section 3, comparisons are presented for the dispersion, association, and full
154 strategies, and for the salt- and ion-specific parameterizations. In Section 4, contributions of
155 the terms are investigated; of the MIAC temperature dependence represented using the model
156 and parameter sets is shown; the model is validated using the osmotic coefficient and individual
157 ion activity coefficient, which have not been used in the parameterization, In Section 5,
158 concluding remarks are presented.

159 **2 Method**

160 In this section, the model, database, salt- and ion-specific parameterization strategies, and
 161 the different parameterization strategies for the short-range ion-ion and ion-solvent interactions
 162 are introduced.

163 **2.1 ePPC-SAFT**

164 In the ePPC-SAFT model [12,38–40], the residual Helmholtz energy is given as,

$$A^{\text{res}} = A^{\text{hc}} + A^{\text{disp}} + A^{\text{assoc}} + A^{\text{polar}} + A^{\text{MSA}} + A^{\text{Born}} \quad (1)$$

165 where the superscript hc denotes hard-chain, disp denotes dispersion, assoc denotes association,
 166 polar denotes multipolar as presented by Ref [21–23]. For details about the equations for each
 167 term and the adjustable parameters (unary and binary), please refer to Ref [12,40]. Here the
 168 electrolyte terms of the model are briefly introduced.

169 The primitive MSA is used for describing the long-range interaction between cation and
 170 anion.

$$A^{\text{MSA}} = -\frac{N_A q_e^2}{4\pi\epsilon_0\epsilon_r} \sum_{\text{ions}} \frac{n_i Z_i^2 \Gamma}{1 + \Gamma \sigma_i^{\text{MSA}}} + \frac{\Gamma^3 RT}{3\pi\rho N_A} \quad (2)$$

$$\Gamma^2 = \frac{q_e^2 N_A^2}{4\epsilon_0\epsilon_r RT} \sum_{\text{ions}} n_i \rho \left[\frac{Z_i}{1 + \Gamma \sigma_i^{\text{MSA}}} \right]^2 \quad (3)$$

171 where N_A is the Avogadro constant, q_e is the elementary charge, ϵ_0 is the dielectric constant in
 172 the vacuum, ϵ_r is the relative permittivity, n_i is the number of moles of ion i , Z_i is the ionic
 173 valence, σ_i^{MSA} is the MSA diameter, R is the universal gas constant, T is temperature, and ρ is
 174 density.

175 The Born term is given in the usual form:

$$A^{\text{Born}} = \frac{N_A q_e^2}{4\pi\epsilon_0 RT} \left(1 - \frac{1}{\epsilon_r}\right) \sum_{\text{ions}} \frac{n_i Z_i^2}{\sigma_i^{\text{Born}}} \quad (4)$$

176 where σ_i^{Born} is the Born diameter. The Born diameter is calculated using the Born term and the
 177 experimental Gibbs energy of solvation obtained by Ref [47]. According to Ref [40], obtaining

178 the Born diameter in this way ensures accurate representation of the Gibbs energy of solvation
 179 of the ePPC-SAFT model.

180 The relative permittivity models shown in Ref [28] and [48] are used in this work, denoted
 181 as RPM-1 and RPM-2, respectively. In RPM-1, the salt-composition dependence is accounted
 182 for only via the density:

$$\varepsilon_r = 1 + \frac{n_{\text{solv}}}{V} d_v \left(\frac{d_T}{T} - 1 \right) \quad (5)$$

183 where n_{solv} is the number of moles of the solvent of volume V , $d_v = 0.3777 \text{ dm}^3/\text{mol}$ and
 184 $d_T = 1403 \text{ K}$ for water [28].

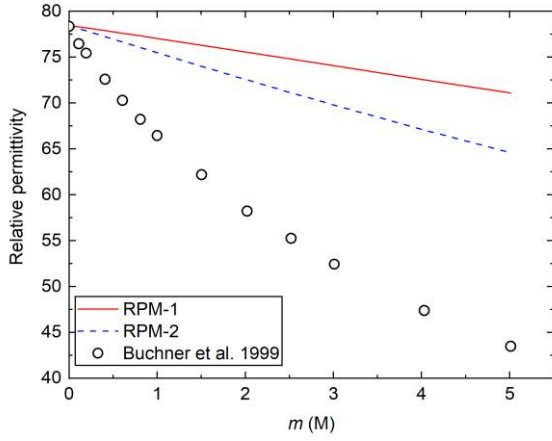
185 In RPM-2, an additional correction factor is applied for the salt composition dependence:

$$\varepsilon_r = 1 + (\varepsilon_{r0} - 1) \frac{1 - \xi}{1 + \frac{\xi}{2}} \quad (6a)$$

$$\xi = \frac{N_A \pi}{6} \sum_{\text{ions}} \frac{n_i \sigma_i^{\text{HS}3}}{V} \quad (6b)$$

186 where ε_{r0} is calculated using RPM-1, and σ_i^{HS} is the ionic hard sphere diameter.

187 One should note that neither of these models represents accurately the experimental salt
 188 composition dependence of the relative permittivity, as shown in Figure 1. The needed
 189 solutions density and ionic diameters are from a preliminary parameter set. In Roa Pinto et al.'s
 190 work [40], Simonin's empirical model [49] was also investigated, with the adjustable
 191 parameters regressed along with the ionic parameters of the ePPC-SAFT model. However, the
 192 resulting relative permittivity approached the RPM-1 rather than the experimental data,
 193 indicating that the RPM that is needed in the EoS should present a much less drastic decrease
 194 compared to the experimental data. In addition, Maribo-Mogensen et al. [50] suggested that
 195 the kinetic depolarization part of the relative permittivity, which was more than half of the
 196 observed decrease (Figure 1 in Ref [50]), was a dynamic property, and should not be included
 197 in the thermodynamic modeling of electrolytes. Therefore, the observed significant
 198 overestimations via RPM-1 and -2 compared to the experimental RP data maybe partially
 199 justified.



200

201 Figure 1. Comparison of the RPMs with the experimental relative permittivity data of aqueous NaCl
 202 solution [51].

203 For water, the parameter set from Ref [39] is used. Unlike other implementations of the
 204 PC-SAFT models, the sphere softness was made adjustable for water rather than fixed at 0.12.
 205 Parameters were regressed over liquid densities and vapor pressure pure water from 273.15 to
 206 550 K and on the LLE of (water + n-alkane) mixtures. The abnormal behavior of water density
 207 was captured using a temperature dependent segment diameter:

$$\sigma_{w,T} = \sigma_w + n_{T_1} e^{n_{T_2} T} + \frac{n_{T_3}}{T^2} \quad (7)$$

208 Ionic parameters are regressed to thermodynamic properties of aqueous solutions of the
 209 salts, as will be explained in detail in the following sections.

210 2.2 Objective function and reference database

211 In this work, aqueous solutions of 4 alkali chloride salts (NaCl, KCl, RbCl, and CsCl) are
 212 investigated. Mean-ionic activity coefficient (MIAC, γ^\pm), vapor-liquid equilibria (VLE), and
 213 density are included in the objective function:

$$\text{OF} = \frac{1}{2} \frac{\sum_{i=1}^{n_{\text{dp}}^{\gamma^\pm}} \left(\frac{\gamma_{\text{cal}}^\pm - \gamma_{\text{exp}}^\pm}{\gamma_{\text{exp}}^\pm} \right)^2}{n_{\text{dp}}^{\gamma^\pm}} + \frac{1}{2} \frac{\sum_{i=1}^{n_{\text{dp}}^p} \left(\frac{p_{\text{cal}} - p_{\text{exp}}}{p_{\text{exp}}} \right)^2}{n_{\text{dp}}^p} + \frac{1}{2} \frac{\sum_{i=1}^{n_{\text{dp}}^\rho} \left(\frac{\rho_{\text{cal}} - \rho_{\text{exp}}}{\rho_{\text{exp}}} \right)^2}{n_{\text{dp}}^\rho} \quad (8)$$

214 where n_{dp} is the number of data points of the property, p is the VLE pressure. The MIAC
 215 reference state is at infinite dilution in the aqueous solution in the molality unit. The involved

216 reference states and conventions, and equations for deriving them from the EoS have been
 217 explained in [7–9,40].

218 The data used in the regression are from the evaluated databases in our previous works
 219 [8,9]. In the data evaluation, datasets that contain outliers have been removed. Table 2 shows
 220 the information for each property of each aqueous electrolyte solution. The data used in the
 221 regression are: for MIAC from Ref [52–78]; for VLE from Ref [79–91]; for density from Ref
 222 [81,92–298]. The osmotic coefficient is not used in the objective function, but is used for
 223 validating the obtained model and parameter sets. The osmotic coefficient data have been
 224 evaluated in Ref [7].

225 Table 1. Number of datasets, number of data points, and ranges for each property of each aqueous
 226 electrolyte solution used in the parameter regression in this work.

Water +	Property	No. datasets	No. data points	T (K)	x_{ion}
NaCl	MIAC	20	344	273.15~333.15	~0.089
	VLE	3	166	294.34~368.07	~0.091
	Density	129	2578	253.00~473.15	~0.098
KCl	MIAC	4	43	273.15~298.15	~0.056
	VLE	8	192	298.15~374.15	~0.095
	Density	104	1615	273.15~473.15	~0.11
RbCl	MIAC	1	21	298.15	~0.076
	VLE	3	65	303.15~368.15	~0.10
	Density	14	168	288.15~328.15	~0.12
CsCl	MIAC	4	51	298.15	~0.049
	VLE	2	75	303.15~368.15	~0.12
	Density	30	351	273.85~473.15	~0.19

227 **2.3 Salt-specific parameterization**

228 Both salt- and ion-specific parameterizations are investigated in this work. Here, the
 229 adjustable parameters of the ions for the salt-specific strategies are explained. The number of
 230 parameters are reduced to facilitate ion-specific parameterization in the next step, as will be
 231 explained in Section 2.4.

232 Efforts are made to improve density accuracy by including the suitable volumetric
 233 parameters in the regression in this work. In a comparison of an electrolyte SAFT-VR model
 234 with Monte Carlo simulations, Zhao et al. [14] showed that, when the primitive MSA model
 235 was used, using the Pauling diameters as the ionic diameters resulted in systematic deviations

236 of density results. Therefore, the ionic hard sphere (HS) diameters are adjusted in this work.
 237 To ensure the “correct order” of the ionic HS diameters, it is assumed that they are proportional
 238 to the Pauling diameters [299]; the ratio (r^{HS}) of the HS diameter over the Pauling diameter,
 239 Eq. (9), is regressed instead of the diameters themselves. The same value is taken for the cation
 240 and anion in the salt-specific parameterization. To conveniently enforce the constraint that the
 241 MSA diameter has to be larger than the HS diameter, the ratio (r^{MSA}) of the MSA diameter
 242 over the HS diameter, Eq. (10), is regressed rather than the diameters themselves. Roa Pinto et
 243 al. [40] showed that, the ePPC-SAFT model accurately reproduced the Gibbs energy of
 244 solvation when the Born diameters were calculated based on the Born term alone. In this work,
 245 the same approach is taken for the Born diameters. Ionic Pauling diameters and Born diameters
 246 are provided in the Supplementary Material.

$$r^{\text{HS}} = \frac{\sigma^{\text{HS}}}{\sigma^{\text{Pauling}}} \quad (9)$$

$$r^{\text{MSA}} = \frac{\sigma^{\text{MSA}}}{\sigma^{\text{HS}}} \quad (10)$$

247 As discussed in Section 1, in addition to the MSA or DH term for the long-range ion-ion
 248 interaction and the Born term for the long-range ion-solvent interaction, different approaches
 249 have been investigated for the short-range ion-ion and ion-solvent interactions. In this work,
 250 10 parameterization strategies are compared for the aqueous NaCl solution, as shown in Table
 251 2. In the following text, the abbreviations (Disp-1, Assoc-1, Full-1, etc.) are used for denoting
 252 the parameterization strategies. r^{HS} and r^{MSA} are regressed for all strategies. For the dispersion
 253 strategies, the dispersion term is used for describing the short-range ion-ion and ion-solvent
 254 interactions. Hence, the dispersion energy of the cation and anion (ε_c and ε_a), and the binary
 255 interaction parameter between the cation and anion (k_{c-a}) are also regressed. The dispersion
 256 energy between the cation and water (ε_{c-w}), between the anion and water (ε_{a-w}), and between
 257 the cation and anion (ε_{c-a}) are obtained using Eq. (11). No binary interaction parameter is used
 258 for the ion-water dispersion interaction. When like-ion dispersion is not included, ε_{c-c} and
 259 ε_{a-a} are artificially set at 0. Naturally, when like-ion dispersion is included, $\varepsilon_{c-c} = \varepsilon_c$, and
 260 $\varepsilon_{a-a} = \varepsilon_a$.

$$\varepsilon_{i-w} = \sqrt{\varepsilon_i \varepsilon_w} \quad (11a)$$

$$\varepsilon_{c-a} = \sqrt{\varepsilon_c \varepsilon_a} (1 - k_{c-a}) \quad (11b)$$

261 For the association strategies, the Wertheim association term is used for describing the
 262 short-range ion-ion and ion-solvent interactions. The number of association sites is set to 7 for
 263 the cation, and 6 for the anion, same as in previous works [12,40]. Hence, the association
 264 energy parameters of the cation and anion (ε_c^{AB} and ε_a^{AB}), and the binary interaction parameter
 265 between the anion and cation (w_{c-a}) are regressed. The association interaction is included
 266 between water and water, ion and water, and cation and anion. The cross-association energies
 267 (ε_{c-w}^{AB} , ε_{a-w}^{AB} , and ε_{c-a}^{AB}) are obtained using Eq. (12). The association volumes of the cation (β_c^{AB})
 268 and anion (β_a^{AB}) are also regressed. The cross-association volume is obtained using Eq. (13).
 269 No binary interaction parameter is included for the unlike-ion association volume.

$$\varepsilon_{i-w}^{AB} = (\varepsilon_i^{AB} + \varepsilon_w^{AB})/2 \quad (12a)$$

$$\varepsilon_{c-a}^{AB} = (\varepsilon_c^{AB} + \varepsilon_a^{AB})(1 - w_{c-a})/2 \quad (12b)$$

$$\beta_{ij}^{AB} = \sqrt{\beta_i^{AB} \beta_j^{AB}} \quad (13)$$

270 For the full strategies, both the dispersion and Wertheim association terms are used for
 271 describing the short-range ion-ion and ion-solvent interactions. Hence, the parameter set also
 272 includes ε_c , ε_a , ε_c^{AB} , ε_a^{AB} , w_{c-a} , β_c^{AB} , and β_a^{AB} . In order to keep the number of adjustable
 273 parameters reasonable, no binary interaction parameter is included for the unlike-ion dispersion
 274 energy and association volume. We notice that there is a different sensitivity of the cation and
 275 anion energy parameters (dispersion or association). Furthermore, the solvation effect of the
 276 cation and anion are very different as reported in nuclear magnetic resonance measurements
 277 [300] and molecular simulations [301]. Liu et al. [30] included cation-solvent association, but
 278 excluded anion-solvent association in their electrolyte SAFT model. However, from an
 279 engineering perspective, we find this sensitivity to be case-specific in the parameter sensitivity
 280 analysis. Furthermore, removing one of the ionic interaction parameters would place a strong
 281 impact on the individual ion activity coefficient results. Therefore, both the cation-water and
 282 anion-water interactions are included. Regression boundaries of the parameters are provided in
 283 the Supplementary Material.

284 Table 2. Salt-specific parameterization strategies.

	Solvent-ion	Ion-ion	RPM-	Like-ion dispersion	Parameters
Disp-1	Dispersion, Born	Dispersion, MSA	1	No	5: $\epsilon_c, \epsilon_a, k_{c-a}, r^{HS}, r^{MSA}$
Disp-2			2		
Disp-3			1	Yes	
Disp-4			2		
Assoc-1	Association, Born	Association, MSA	1	-	7: $\epsilon_c^{AB}, \epsilon_a^{AB}, \beta_c^{AB}, \beta_a^{AB}, w_{c-a}, r^{HS}, r^{MSA}$
Assoc-2			2		
Full-1	Dispersion, association, Born	Dispersion, association, MSA	1	No	9: $\epsilon_c, \epsilon_a, \epsilon_c^{AB}, \epsilon_a^{AB}, \beta_c^{AB}, \beta_a^{AB}, w_{c-a}, r^{HS}, r^{MSA}$
Full-2			2		
Full-3			1	Yes	
Full-4			2		

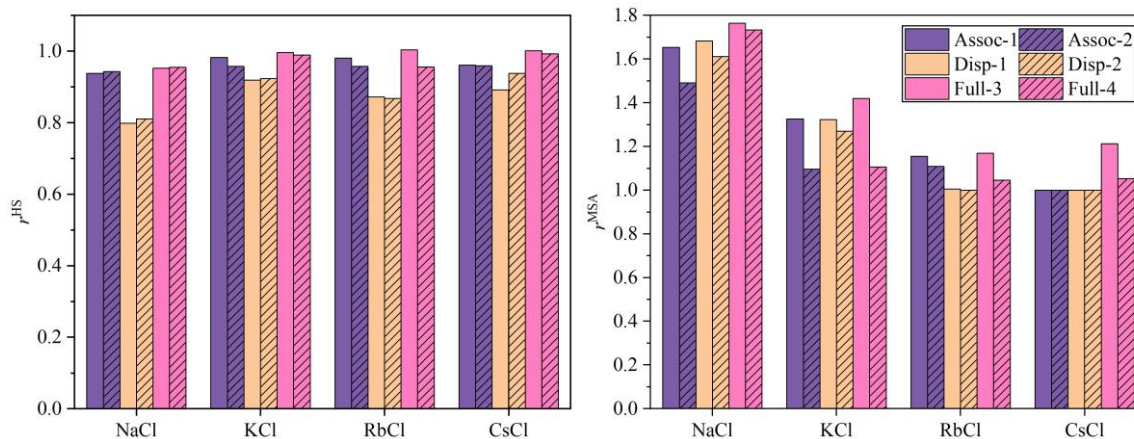
285 One might argue that comparisons are only fair with the same number of adjustable
286 parameters. However, the larger number of adjustable parameters and the resulting advantage
287 of being more flexible are intrinsic in the association and full strategies. Therefore, no effort
288 has been made to compare the strategies based on the same number of adjustable parameters.
289 Furthermore, as will be shown in Section 3, the Full strategies do not provide better results
290 despite using a larger number of adjustable parameters. In addition, as will be shown in Section
291 2.4, the ion-specific strategies will be compared using the same number of adjustable
292 parameters.

293 Parameters are regressed using an evolutionary algorithm [302], which is helpful for
294 avoiding local minima. In the salt-specific parameterization, different Cl^- parameters are taken
295 for each salt. The regressed salt-specific parameters for the 4 salts are provided in the
296 Supplementary Material. The resulting deviations will be discussed in Section 3.

297 For the aqueous NaCl solution, all 10 strategies are used. For Disp-3 and -4, the dispersion
298 energy parameters are too large (ϵ/k is approximately 1000 K for Na^+ compared to 201.747 K
299 for water), the binary interaction parameter, k_{c-a} , is close to 1 (the upper boundary), while the
300 accuracy is lower compared to the other strategies. Therefore, these two dispersion strategies
301 are not further pursued for the other aqueous electrolyte solutions, while the remaining 8
302 strategies are investigated for the other 3 salts. At this point, we have partly addressed the
303 question posted earlier “Is the model “smart” enough to exclude dispersion between like-ions?”
304 For the dispersion strategies, yes: $k_{c-a} = 1$ indicates zero dispersion between unlike-ions,
305 which is not reasonable with a non-zero dispersion between like-ions. However, the full

306 strategies with (Full-3 and -4) and without (Full-1 and -2) like-ion dispersion do not present
307 such distinction, with Full-3 and -4 presenting slightly better trend in parameters and higher
308 accuracy than Full-1 and -2.

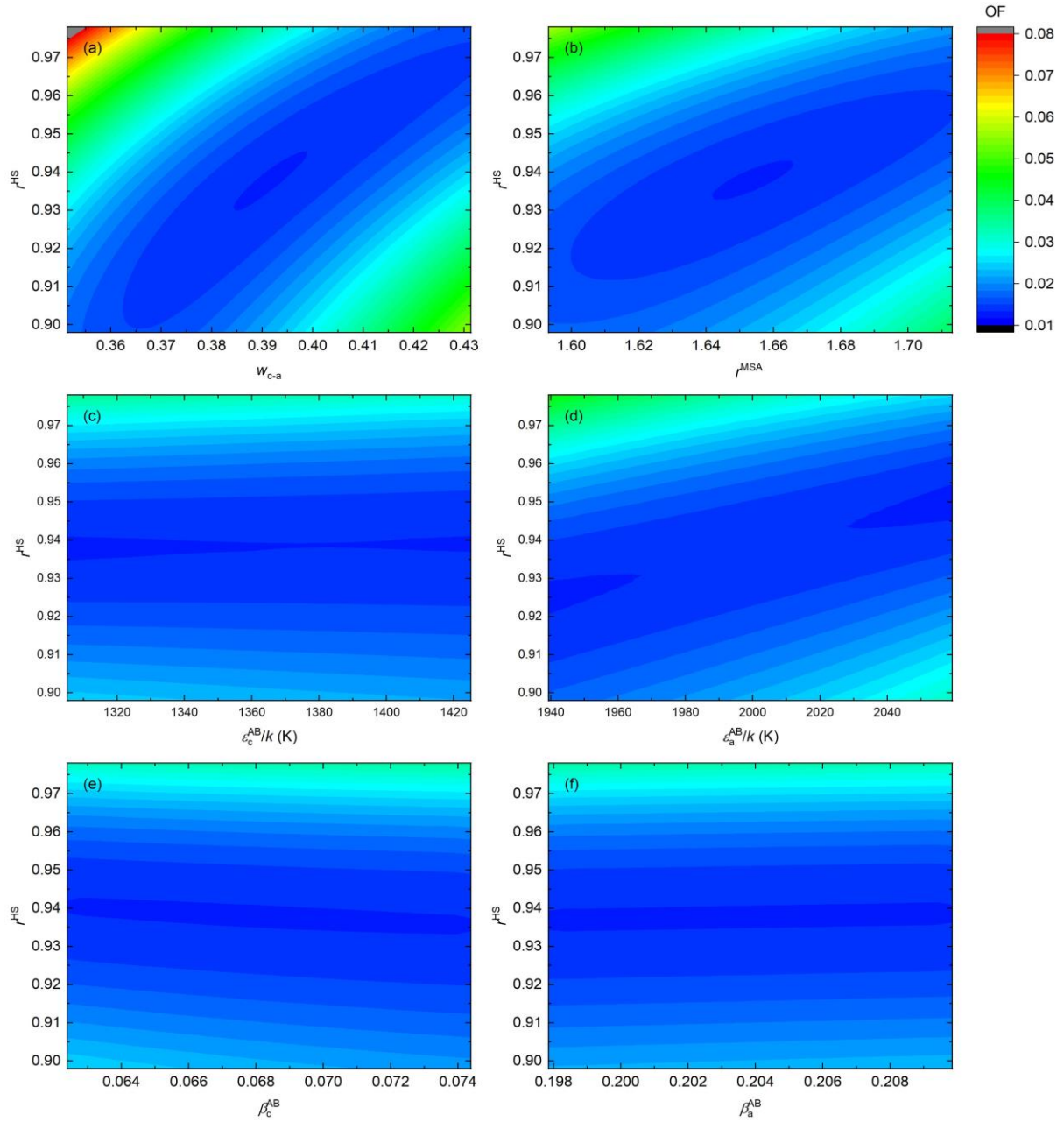
309 The parameters r^{HS} and r^{MSA} are the only ones that present trends with the cations in the
310 salt-specific regressions. Figure 2 shows the regressed r^{HS} and r^{MSA} in the salt-specific
311 strategies, Assoc-1, Assoc-2, Disp-1, Disp-2, Full-3, Full-4. r^{HS} is approximately the same for
312 the 4 alkali chlorides in each of the parameterization strategies. In most of the cases, r^{HS} is
313 smaller than 1, i.e., the HS diameter is smaller than the Pauling diameter, and is very close to
314 1 in a few cases of the full strategies. r^{HS} is approximately 0.8 in the dispersion strategies, and
315 is in general between 0.93 and 1 in the association and full strategies. In Ref [40], r^{MSA} was
316 set at 1.5 for the aqueous NaCl solution in order to reduce the number of adjustable parameters.
317 The value agrees with the regressed r^{MSA} in this work. For the heavier cations, however, the
318 regressed r^{MSA} is smaller. For CsCl, r^{MSA} reaches 1 (regression lower boundary) in some of
319 the cases; deviations are larger compared to the other salts (as will be shown in Sections 3 and
320 4), and can be reduced given a r^{MSA} that is smaller than 1. For RbCl, r^{MSA} is also 1 or very
321 close to 1 in the dispersion strategies. The parameter behavior might have resulted from certain
322 assumptions in the SAFT, MSA, and Born terms. In addition, the contributions except for that
323 of the MSA term are all close to linear, and compensate for each other, as will be shown in
324 Section 4.1. As will be shown in Section 3, deviations in VLE are also larger compared to the
325 association strategies, presenting another reason for preferring the association strategies over
326 the dispersion strategies. However, the boundary is set at 1 regardless of this potential slight
327 accuracy improvement, to avoid the inconsistency of smaller MSA diameters than the HS
328 diameters. The value of r^{HS} is approximately the same for the 4 salts in each of the strategies,
329 while r^{MSA} is smaller for the heavier salts. Therefore, in the ion-specific parameterization, the
330 same r^{HS} is used for all 5 ions, while different r^{MSA} are used for each ion, as will be discussed
331 in the next section.



332

333 Figure 2. Regressed r^{HS} (left) and r^{MSA} (right) in the salt-specific strategies Assoc-1, Assoc-2, Disp-
 334 1, Disp-2, Full-3, and Full-4.

335 Furthermore, the response surface is analyzed so that the number of adjustable parameters
 336 could be reduced with minimum loss in accuracy. Figure 3 shows the response surface of the
 337 objective function, Eq. (8), over the parameters in the salt-specific strategy, Assoc-1, for the
 338 aqueous NaCl solution. Contours are plotted on the same levels over r^{HS} and each of the other
 339 parameters. Obviously, there is parameter degeneracy. r^{HS} , w_{c-a} and r^{MSA} are the most
 340 pronounced parameters, while the association energy and volume parameters are less
 341 pronounced. The “valleys” are almost parallel to the association volume axis, indicating that
 342 the objective function is not sensitive to these parameters. The behavior is similar for the other
 343 aqueous electrolyte solutions. Therefore, β_c^{AB} and β_a^{AB} are fixed to that of water in the ion-
 344 specific parameterization. Although the “valley” is also parallel to the ϵ_c^{AB}/k axis, the situation
 345 is case-specific. Therefore, the association energy parameters are kept adjustable in the ion-
 346 specific parameterization.



347

348 Figure 3. Response surface of the objective function, Eq. (8), as a function of the parameters in the
 349 salt-specific strategy , Assoc-1, for the aqueous NaCl solution: (a) $r^{\text{HS}} - w_{\text{c-a}}$, (b) $r^{\text{HS}} - r^{\text{MSA}}$, (c) r^{HS}
 350 $- \epsilon_c^{\text{AB}}/k$, (d) $r^{\text{HS}} - \epsilon_a^{\text{AB}}/k$, (e) $r^{\text{HS}} - \beta_c^{\text{AB}}$, (f) $r^{\text{HS}} - \beta_a^{\text{AB}}$. In all the sub-graphs, all the other parameters
 351 are fixed at their optimal values.

352 2.4 Ion-specific parameterization

353 Ion-specific parameterization is desirable because it facilitates simple extension to mixed-
 354 salt systems, as no mixing rule is needed for the parameters of the same ion from different salts.
 355 Furthermore, it could potentially bring in information about the contributions of the cation and
 356 anion to the mean properties, even though individual ion information is not sufficiently
 357 included in the parameterization. However, obtaining these parameters requires regressing the
 358 parameters of a series of electrolyte solutions simultaneously [11]. Therefore, in this work,

359 efforts have been made to reduce the number of adjustable parameters with minimum loss in
 360 accuracy compared to the salt-specific parameterization, based on the findings in the salt-
 361 specific parameterization.

362 Table 3 shows the ion-specific parameterization strategies, abbreviated as Assoc-I-X,
 363 Disp-I-X, and Full-I-X, where the symbol “X” indicates the number of adjustable parameters.
 364 At first, the 15-parameter scenarios are investigated. Thus, the resulting ion-specific
 365 association strategy is Assoc-I-15, including the parameters, r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $4 w_{\text{c-a}}$
 366 for the 4 salts, and $5 \varepsilon_i^{\text{AB}}$ for the 5 ions. Similarly, for Disp-I-15, the $4 k_{\text{c-a}}$ and $5 \varepsilon_i$ parameters
 367 are optimized instead of their association counterparts; for Full-I-15, the parameter set is the
 368 same as Assoc-I-15, the ε_i parameters are set at the water value, while $k_{\text{c-a}}$ is set at 0. Like-
 369 ion dispersion is excluded in the dispersion strategies, but is included in the full strategies,
 370 according to the discussions in Section 2.3. Regression boundaries of the parameters are
 371 provided in the Supplementary Material. Rather than obtaining the parameters incrementally,
 372 i.e., starting from one of the salts and extending to the others, the 15 parameters are optimized
 373 simultaneously to the thermodynamic property data of the 4 aqueous alkali halide solutions.
 374 This facilitates a genuine ion-specific parameterization, as the contributions of the cation and
 375 anion are introduced into the regression by the difference of the mean thermodynamic property
 376 data of the 4 aqueous electrolyte solutions. In addition, compared to using salt-specific
 377 parameters of a reference system and extending to other salts that share a common ion (for
 378 instance, using the Cl^- parameters from NaCl and extending to other Cl^- salts), this approach
 379 avoids preferring over the accuracy of the reference system in the incremental approach while
 380 compromising that of the other systems.

381 Table 3. Parameter sets for the ion-specific strategies, including 4 cations (Na^+ , K^+ , Rb^+ , and Rb^+) and
 382 1 anion (Cl^-).

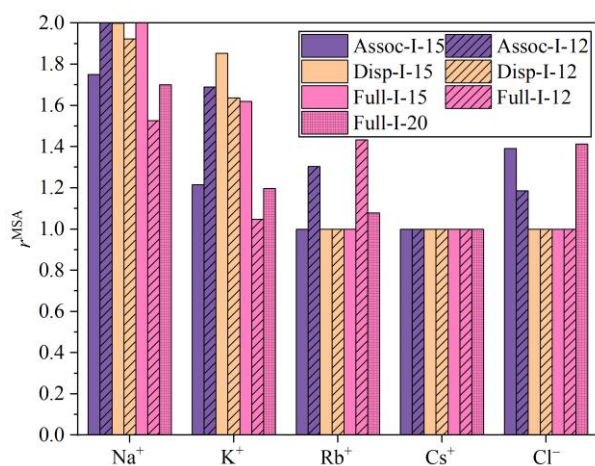
	Parameter set
Assoc-I-15	15: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $4 w_{\text{c-a}}$ for the 4 salts, and $5 \varepsilon_i^{\text{AB}}$ for the 5 ions
Assoc-I-12	12: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $w_{\text{c-a}}$, and $5 \varepsilon_i^{\text{AB}}$ for the 5 ions
Disp-I-15	15: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $4 k_{\text{c-a}}$ for the 4 salts, and $5 \varepsilon_i$ for the 5 ions
Disp-I-12	12: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $k_{\text{c-a}}$, and $5 \varepsilon_i$ for the 5 ions
Full-I-15	15: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $4 w_{\text{c-a}}$ for the 4 salts, and $5 \varepsilon_i^{\text{AB}}$ for the 5 ions
Full-I-12	12: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $w_{\text{c-a}}$, and $5 \varepsilon_i^{\text{AB}}$ for the 5 ions
Full-I-20	20: r^{HS} , $5 r^{\text{MSA}}$ for the 5 ions, $4 w_{\text{c-a}}$ for the 4 salts, $5 \varepsilon_i^{\text{AB}}$ for the 5 ions, and $5 \varepsilon_i$ for the 5 ions

383 In a next step, efforts are made to reduce the number of adjustable parameters. First, the
384 same ε_c^{AB} is regressed using a single parameter for all cations, reducing the number of
385 adjustable parameters from 15 to 12. However, even though the accuracy is as good as the 15-
386 parameter strategies, the individual ion activity coefficient (IIAC) is predicted to be smaller for
387 cation than for anion for both the aqueous NaCl and KCl solutions, i.e., the model and
388 parameter set fail in predicting the opposite relative magnitudes of cation and anion IIAC for
389 aqueous NaCl and KCl solutions. To obtain the qualitatively correct behavior, different
390 association energy parameters have to be used for Na⁺ and K⁺. Therefore, it is attempted to use
391 the same w_{c-a} for the 4 aqueous solutions, reducing the number of adjustable parameters from
392 15 to 12. The 12-parameter ion-specific association strategy is denoted as Assoc-I-12. Similarly,
393 the numbers of parameters for the dispersion and full ion-specific strategies are reduced from
394 15 to 12; the 12-parameter strategies are denoted as Disp-I-12 and Full-I-12.

395 Unlike the salt-specific cases, results of Full-I-15 and Full-I-12 are much worse than
396 Assoc-I-15 and Assoc-I-12. It is suspected that the increased deviations result from the fixed
397 dispersion energy parameter. Therefore, the dispersion energy parameters of the 5 ions are
398 included in the ion-specific full strategy, i.e., Full-I-20.

399 In the salt-specific cases, using RPM-1 or -2 results in only marginally different results.
400 Therefore, RPM-1 is used in all the ion-specific cases; the impact of the relative permittivity
401 model is not further pursued in this work.

402 The regressed parameters of the ion-specific strategies are provided in the Supplementary
403 Material. Like in the salt-specific cases, r^{HS} is smaller for the dispersion strategies than for the
404 association and full strategies. Figure 4 shows the regressed r^{MSA} in the ion-specific strategies,
405 Assoc-I-15, Assoc-I-12, Disp-I-15, Disp-I-12, Full-I-15, Full-I-12, and Full-I-20. For all the
406 strategies except for Full-I-12, r^{MSA} decreases with ionic size within the cations. In all ion-
407 specific strategies, r^{MSA} is 1 for Cs⁺. Accuracy could be improved if the constraint that the
408 MSA diameter must be larger than the HS diameter is relaxed. However, it is decided not to do
409 so. For Disp-I-15, Disp-I-12, Full-I-15, and Full-I-12, r^{MSA} is also 1 for Cl⁻. These strategies
410 also result in larger deviations compared to the others, as will be discussed in Section 3.2.



411

412 Figure 4. Regressed r^{MSA} in the ion-specific strategies, Assoc-I-15, Assoc-I-12, Disp-I-15, Disp-I-12,
 413 Full-I-15, Full-I-12, and Full-I-20.

414 3 Results

415 In this section, we compare the salt-specific association, dispersion, and full strategies as
 416 well as the ion- and salt-specific strategies approaches.

417 3.1 Comparison of the dispersion, association, full strategies for the short-range ion-ion 418 and ion-solvent interactions

419 The percentage average absolute deviations (AADs) and percentage maximum absolute
 420 deviations (MADs) of the salt-specific strategies for the 4 aqueous electrolyte solutions are
 421 shown in Table 4. For all 4 salts, the association and full strategies are more accurate than the
 422 dispersion strategies. Therefore, it appears necessary to include the association term for the
 423 short-range ion-ion and ion-solvent interactions. For the aqueous NaCl solution, Disp-3 and -
 424 4, in which like-ion dispersion is included, present slightly larger MADs compared to Disp-1
 425 and Disp-2, in which like-ion dispersion is excluded. Unlike the dispersion strategies, whether
 426 the like-ion dispersion is accounted for does not significantly affect the result for the full
 427 strategies. Considering the unreasonable parameters of Disp-3 and -4, as discussed in Section
 428 2.3, we conclude here that like-ion dispersion should not be included. Considering the slightly
 429 more accurate results and better trends in parameter of Full-3 and -4 compared to Full-1 and -
 430 2 (r^{MSA} of the salts, as shown in the Supplementary Material), we recommend including like-
 431 ion dispersion in the full strategies.

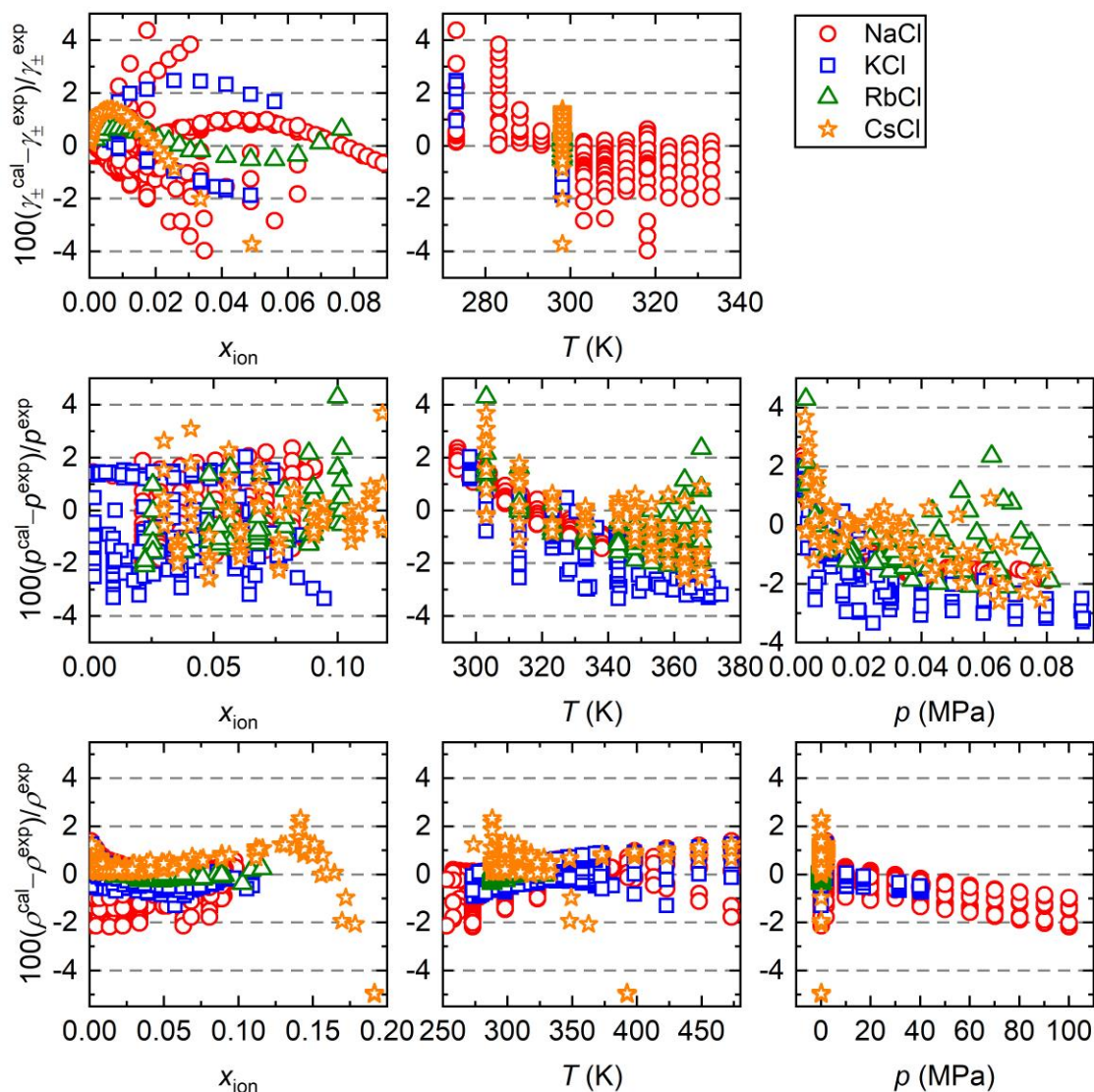
432 Table 4. Percentage average absolute deviations (AADs) and percentage maximum absolute
 433 deviations (MADs) of the salt-specific strategies for the 4 aqueous electrolyte solutions. Deviations
 434 larger than 10% are marked as bold and italic.

	MIAC		VLE p		Density	
	AAD (%)	MAD (%)	AAD (%)	MAD (%)	AAD (%)	MAD (%)
NaCl						
Disp-1	1.3	6.6	1.1	2.9	0.36	2.1
Disp-2	1.3	6.3	1.1	2.9	0.34	2.1
Disp-3	1.3	7.7	1.2	3.4	0.80	3.9
Disp-4	1.2	7.0	1.2	3.2	0.46	2.7
Assoc-1	0.62	4.4	1.1	2.4	0.32	2.2
Assoc-2	0.76	4.3	1.2	2.5	0.33	2.2
Full-1	0.81	5.4	1.1	2.5	0.69	4.2
Full-2	1.2	5.7	1.1	2.8	0.31	2.1
Full-3	0.57	4.3	1.1	2.2	0.37	2.4
Full-4	0.5	3.6	1.2	2.3	0.37	2.3
KCl						
Disp-1	1.2	4.4	1.5	3.3	0.26	1.3
Disp-2	1.1	3.8	1.5	3.3	0.27	1.3
Assoc-1	0.96	2.5	1.6	3.3	0.30	1.3
Assoc-2	0.42	2.0	1.6	3.3	0.23	1.6
Full-1	0.29	0.58	1.6	3.3	0.27	1.3
Full-2	0.24	0.84	1.6	3.3	0.30	1.5
Full-3	0.88	1.8	1.6	4.2	0.30	1.4
Full-4	0.39	1.6	1.7	3.5	0.29	1.4
RbCl						
Disp-1	0.59	2.6	1.5	6.9	0.27	1.4
Disp-2	0.73	2.0	1.4	7.0	0.44	2.3
Assoc-1	0.42	0.66	1.1	4.3	0.16	0.54
Assoc-2	0.37	0.93	1.1	4.2	0.29	1.0
Full-1	0.56	0.89	1.3	5.4	0.39	2.1
Full-2	1.5	4.2	1.1	2.3	2.4	<i>11</i>
Full-3	0.37	1.1	1.1	4.7	0.21	0.79
Full-4	0.51	0.95	1.2	5.8	0.62	2.9
CsCl						
Disp-1	1.8	3.2	2.7	<i>14</i>	0.71	3.5
Disp-2	2.1	3.3	2.6	<i>15</i>	0.24	1.3
Assoc-1	0.86	3.7	0.94	3.7	0.47	5.0

	MIAC		VLE p		Density	
	AAD (%)	MAD (%)	AAD (%)	MAD (%)	AAD (%)	MAD (%)
Assoc-2	1.1	4.7	1.1	6.0	0.61	4.1
Full-1	0.50	0.95	1.4	7.2	0.29	2.2
Full-2	0.73	2.3	1.5	6.9	0.32	3.2
Full-3	1.5	5.4	1.2	6.7	0.35	2.6
Full-4	1.1	5.4	1.6	5.7	0.40	2.2

435 The strategies using RPM-1 and -2 present only marginal difference in accuracy.

436 Figure 4 shows the deviations of the MIAC, VLE and density calculated with Assoc-1
437 from the experimental data. For references of the experimental data, please see Section 2.2.
438 Results for the other association and full strategies are similar. For MIAC, the data are all at
439 0.1 MPa. Thus, the deviation of MIAC plotted against p is not shown. In general, the model
440 agrees very well with experimental data over the entire temperature, pressure, and composition
441 range. For the 3 properties, the model is most accurate near 300 K, in which region the data are
442 more extensive. As temperature decreases, deviations increase slightly. Considering that the
443 deviation is at very small x_{ion} at low temperatures, it is much more pronounced compared to
444 that of approximately the same magnitude at much larger x_{ion} at high temperatures. For NaCl,
445 MIAC deviation clearly present a temperature-dependent behavior. Details will be discussed
446 in Section 4.2. Density data are available for the aqueous NaCl and KCl solutions at high
447 temperature; deviations increase very slightly as pressure increases, a large part of which is
448 attributed to the increased deviation of the model for pure water density at higher pressure.
449 Density data are available in the databases up to very high salt composition (close to $x_{\text{ion}} =$
450 0.2) for the aqueous CsCl solution; deviations increase to approximately 5% at high salt
451 composition, in which range MIAC and VLE data are not available in the database.



452

453 Figure 5. Deviations of the MIAC, VLE and density calculated with Assoc-1 from the experimental
 454 data. For references of the experimental data, please see Section 2.2.

455 3.2 Salt-specific vs. ion-specific parameterization

456 The AADs and MADs of the ion-specific parameter sets for MIAC, VLE, and density are
 457 shown in Table 5. Deviations larger than 10% are marked as bold and italic. Comparing the
 458 results in Tables 4 and 5, we can see that the Assoc-I-15 and Assoc-I-12 strategies are as
 459 accurate as the salt-specific parameter sets. This is achieved with a much smaller parameter set:
 460 15 or 12 parameters for the 4 salts as compared to 7 for each one of the 4 salts. However, unlike
 461 the association strategies, the dispersion and full strategies are not as accurate as the
 462 corresponding salt-specific strategies. Large deviations in the VLE of the aqueous CsCl
 463 solution are observed for Disp-I-15, Disp-I-12, Full-I-15, and Full-I-12. The only full strategy
 464 that achieved comparable accuracy as the association strategies includes 20 parameters (Full-
 465 I-20). In the following text, Assoc-I-15 is discussed as an example. Results calculated using

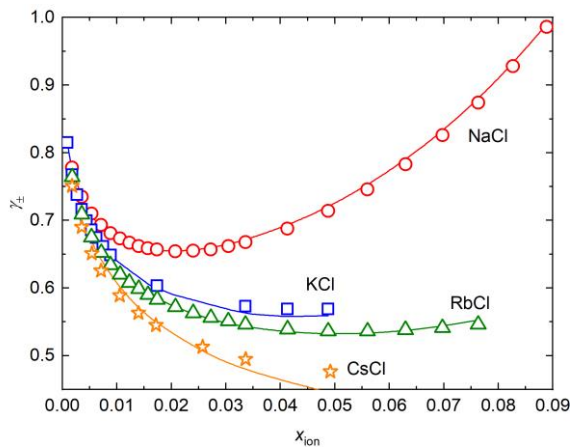
466 Assoc-I-12 are as accurate for all the properties, except for the density of the aqueous CsCl
467 solution, which has an MAD of 4.2% (still quite good) compared to the MIAC of 2.4% of
468 Assoc-I-15.

469 Table 5. AADs and MADs of MIAC, VLE, and density of the 4 aqueous alkali halide solutions
 470 calculated using the ion-specific strategies. Deviations larger than 10% are marked as bold and italic.

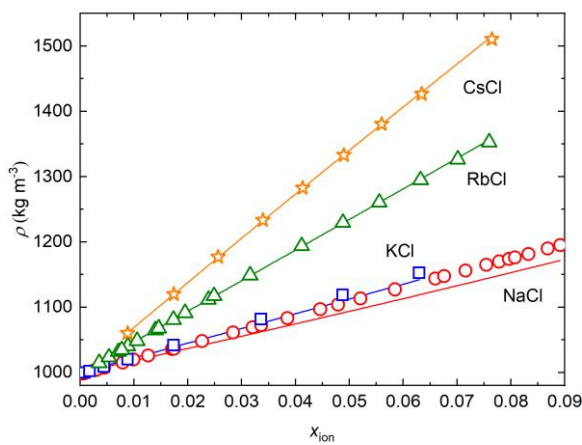
	MIAC		VLE p		Density	
	AAD (%)	MAD (%)	AAD (%)	MAD (%)	AAD (%)	MAD (%)
NaCl						
Disp-I-15	1.6	6.4	1.1	2.8	0.62	3.3
Disp-I-12	1.6	6.6	1.1	2.9	0.79	4.0
Assoc-I-15	0.84	5.4	1.1	2.1	0.64	3.4
Assoc-I-12	1.0	5.5	1.1	2.1	0.57	3.2
Full-I-15	1.4	5.8	1.1	2.4	0.72	3.8
Full-I-12	2.2	<i>10</i>	1.2	4.0	0.38	2.6
Full-I-20	0.76	4.6	1.1	2.3	0.74	4.2
KCl						
Disp-I-15	1.3	4.6	1.5	3.3	0.25	1.3
Disp-I-12	1.4	4.5	1.5	3.3	0.26	1.3
Assoc-I-15	1.2	4.0	1.6	3.5	0.39	2.0
Assoc-I-12	1.3	3.0	1.5	3.3	0.32	1.6
Full-I-15	1.3	4.4	1.5	3.3	0.32	1.3
Full-I-12	2.8	7.7	1.4	3.3	0.43	2.6
Full-I-20	1.1	3.6	1.5	3.3	0.32	1.4
RbCl						
Disp-I-15	0.64	1.7	1.4	6.8	0.20	1.0
Disp-I-12	0.68	1.9	1.4	6.9	0.20	0.96
Assoc-I-15	0.54	1.2	1.1	4.1	0.20	1.3
Assoc-I-12	0.64	1.1	1.1	3.8	0.25	1.6
Full-I-15	0.51	1.3	1.3	6.4	0.18	0.68
Full-I-12	1.8	4.4	1.4	7.5	0.26	1.6
Full-I-20	0.75	1.2	1.2	3.9	0.20	0.94
CsCl						
Disp-I-15	1.8	3.1	2.7	<i>14</i>	0.89	4.1
Disp-I-12	1.8	3.1	2.6	<i>14</i>	0.82	3.8
Assoc-I-15	1.5	6.5	1.2	5.5	0.42	2.4
Assoc-I-12	1.1	5.4	1.3	5.2	0.65	4.2
Full-I-15	1.8	2.8	2.4	<i>14</i>	0.36	2.4
Full-I-12	3.0	6.0	1.9	<i>11</i>	0.70	4.4
Full-I-20	1.3	4.5	1.1	5.1	0.58	3.4

471 Figures 6 and 7 show the comparison of the MIAC and density of the aqueous NaCl, KCl,
 472 RbCl, and CsCl solutions calculated using Assoc-I-15 and experimental data at 298.15 and 0.1

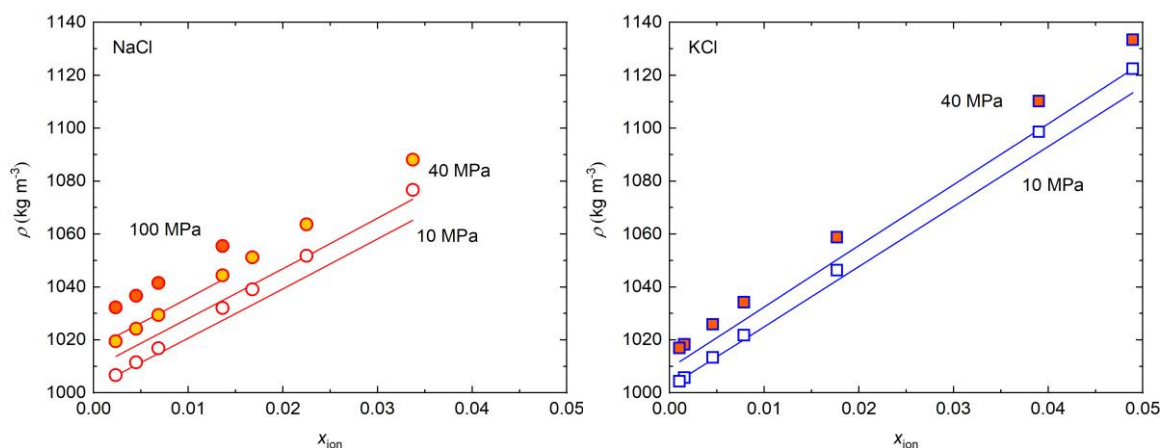
473 MPa. The experimental data are from Ref [52,57,70,107,161,242]. The model is very accurate
 474 at atmospheric conditions. Figure 8 shows the comparison of density of aqueous NaCl and KCl
 475 solutions calculated using Assoc-I-15 and experimental data at 298.15 K and elevated pressure,
 476 up to 100 MPa for NaCl and 40 MPa for KCl. The experimental data are from Ref [112,145].
 477 The deviations are slightly larger than at 0.1 MPa, and increase with increasing pressure and
 478 salt composition, but are within 2%. At higher pressure, the deviations for pure water of the
 479 PPC-SAFT model is also slightly larger, as shown in the left side of the graphs (at 0 salt
 480 composition). For the aqueous RbCl and CsCl solutions, data are not available up to such high
 481 pressure. Figure 9 shows the vapor pressure of aqueous KCl solution calculated using Assoc-
 482 I-15 and experimental data at 313.15 to 343.15 K. The experimental data are from Ref [88].
 483 Results are similar for the other aqueous electrolyte solutions. Overall, the properties are
 484 correlated very accurately.



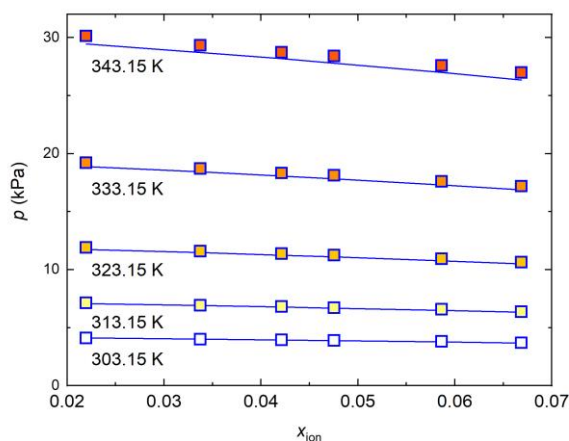
485
 486 Figure 6. Comparison of the MIAC of the aqueous NaCl, KCl, RbCl, and CsCl solutions calculated
 487 using Assoc-I-15 and experimental data at 298.15 K and 0.1 MPa. The experimental data are from Ref
 488 [52,57,70].



489
 490 Figure 7. Comparison of the density of the aqueous NaCl, KCl, RbCl, and CsCl solutions calculated
 491 using Assoc-I-15 and experimental data at 298.15 K and 0.1 MPa. The experimental data are from Ref
 492 [107,161,242].



493
 494 Figure 8. Comparisons of the density of the aqueous NaCl and KCl solutions calculated using Assoc-
 495 I-15 and experimental data at 298.15 K and elevated pressures. The experimental data are from Ref
 496 [112,145].



497
 498 Figure 9. Comparison of the vapor pressure of the aqueous KCl solution calculated using Assoc-I-15
 499 and experimental data at 313.15 to 343.15 K. The experimental data are from Ref [88].

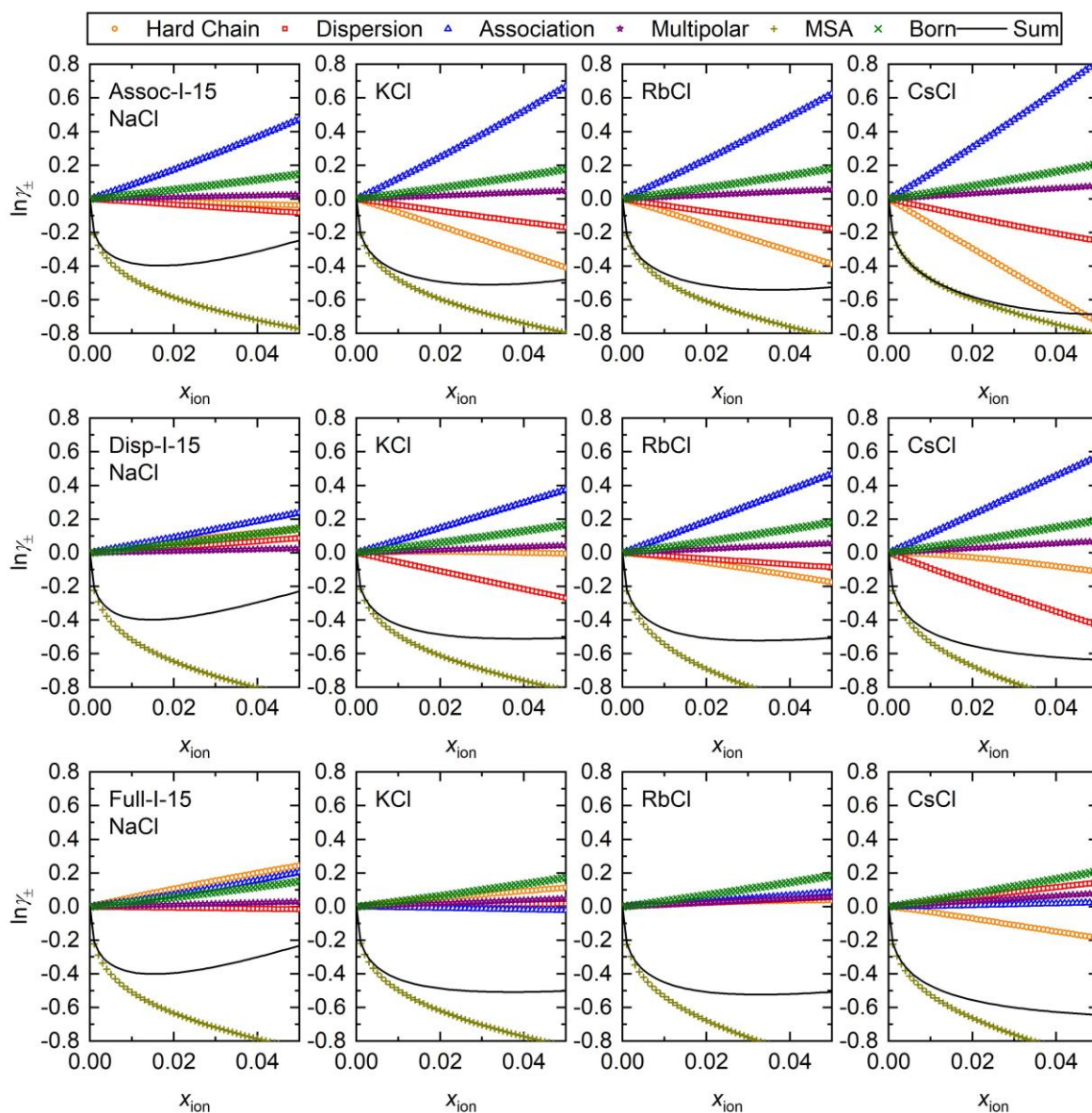
500 4 Discussion

501 In this section, the contributions of the terms are analyzed; the model is tested on the
 502 MIAC over large temperature ranges beyond the range of the data used in the regression. The
 503 model is also tested on the osmotic coefficient and individual ion activity coefficient, which
 504 have not been included in the parameter estimation.

505 4.1 Contributions of the terms

506 Figure 10 shows the contributions to $\ln \gamma_{\pm}$ of the terms (hard-chain, dispersion,
 507 association, multipolar, MSA, and Born) in the ion-specific strategies, Assoc-I-15, Disp-I-15,
 508 and Full-I-15, at 298.15 K and 0.1 MPa. The reference state of the sum of $\ln \gamma_{\pm}$ is naturally
 509 based on the molar fraction unit. The contributions of Assoc-I-12, Disp-I-12, Full-I-12, and
 510 Full-I-20 are similar to the corresponding 15-parameter cases, and are not shown here. At low
 511 salt composition, $\ln \gamma_{\pm}$ is dominated by the MSA term. The MSA term presents a curvature,

512 while all the other terms are close to linear in the entire ranges, counter-balancing the negative
513 MSA term. Thus, $\ln \gamma_{\pm}$ presents an upward curvature as salt composition increases. The Born
514 term is approximately the same in the different strategies for each aqueous electrolyte solution,
515 because it is determined by the relative permittivity and hard sphere diameters. The multipolar
516 term presents a very small contribution, indicating that our conclusions would also apply for a
517 SAFT model that does not include a multipolar term. The contributions of the salt-specific
518 parameterization strategies are provided in the Supplementary Material. The behavior is very
519 similar for the ion- and salt-specific association strategies, and is different for the dispersion
520 and full strategies. This could be linked to the observation that reducing the number of
521 adjustable parameters does not have a considerable impact on the accuracy of the model for
522 the association strategies.



523

524 Figure 10. Contributions to $\ln \gamma_{\pm}$ of the terms in the ion-specific strategies, Assoc-I-15, Disp-I-15, and
 525 Full-I-15, at 298.15 K and 0.1 MPa.

526 For Assoc-I-15, the association term presents the largest positive contribution for the 4
 527 aqueous electrolyte solutions, while the dispersion and hard chain terms are negative and quite
 528 pronounced. Note that, in the association strategies, the dispersion contribution is a
 529 consequence of only the water-water dispersion interaction. The segment number is set at 1 for
 530 the ions. The regressed segment number for water is also close to 1. Therefore, the hard chain
 531 contribution is approximately a hard sphere contribution.

532 For Disp-I-15, the association term also presents the largest positive contribution, but is
 533 smaller compared to the Assoc-I-15 cases. Note that, in the dispersion strategies, the
 534 association contribution is a consequence of only the water-water association interaction. The

535 hard chain and dispersion terms are positive for the aqueous NaCl solution, and are smaller (in
536 magnitude, negative) for the other aqueous solutions compared to the Assoc-I-15 cases.

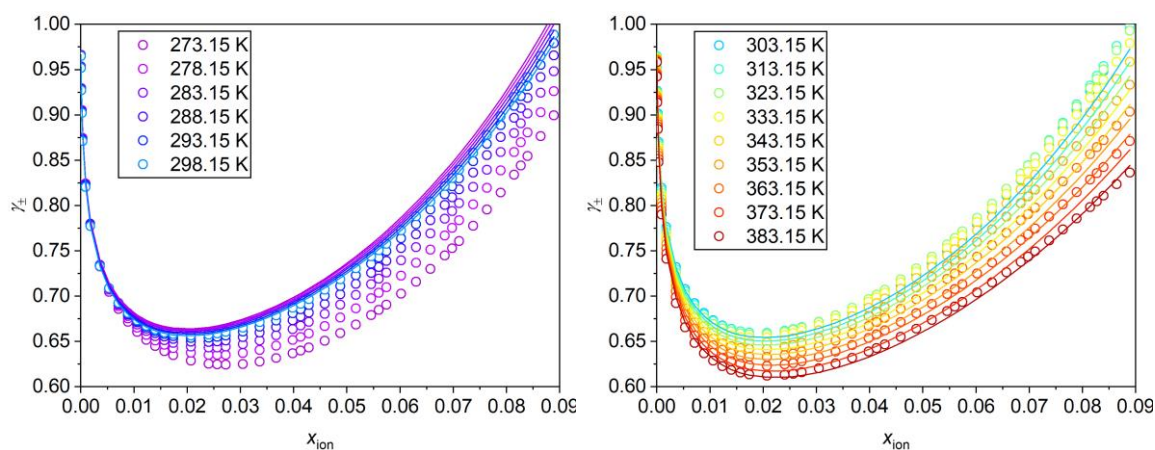
537 For Full-I-15, the association contribution is even smaller compared to the dispersion
538 cases. The association, hard chain, and dispersion terms present no trend. Considering that
539 these strategies resulted in no improvement in accuracy compared to the association strategies,
540 the counter-balancing behavior of the close-to-linear terms shows that there is no need to
541 account for both the association and dispersion contributions for the short-range ion-ion and
542 ion-solvent interactions.

543 Naturally, the total $\ln \gamma_{\pm}$ is of course approximately the same for the different strategies
544 for each salt. However, it is also interesting to notice that the magnitudes of the separate
545 contributions are larger for salts with heavier cations for the association and dispersion
546 strategies.

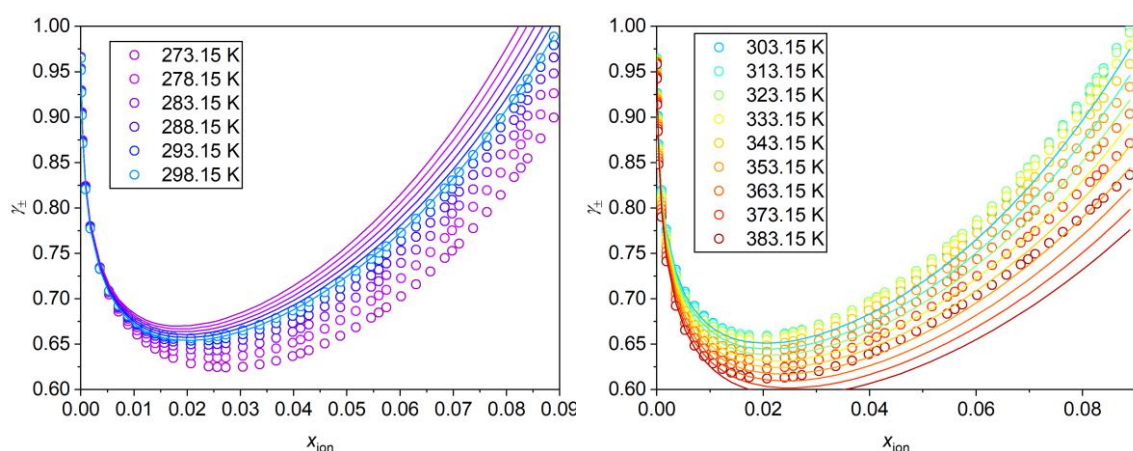
547 In the ePPC-SAFT model, the electrolyte contribution (MSA and Born) and the “physical”
548 contributions (that are all approximately linear) balance each other out.; Yet, the distribution
549 among the physical terms is sometimes very different, with, for example, sometimes positive,
550 sometimes negative contributions from the dispersion and hard sphere. The association
551 strategies show a logical trend when increasing the cation size in contrast to the other strategies.
552 At this point, we don’t have a sufficiently clear vision of the true physics to comment on the
553 meaning of these contrasting contributions, but the trend observed with the association strategy
554 provides better ground for further work.

555 **4.2 Temperature dependence of MIAC**

556 Figure 11 shows the comparison of the MIAC of the aqueous NaCl solution calculated
557 using Assoc-1 and experimental data from 273.15 to 383.15 K. The data used for comparison
558 are from Ref [303], and are not used in the regression. It covers larger temperature range than
559 the MIAC data used in the regression, which are only up to 333.15 K, and also covers larger
560 salt composition range at low temperatures. MIAC presents an anomaly: at low temperatures,
561 MIAC increases with increasing temperature. Figure 12 shows results calculated using the ion-
562 specific strategy, Assoc-I-15. Neither parameter set captures the temperature-dependence
563 anomaly. As temperature increases above the range of the data used in the regression, the salt-
564 specific parameter set, Assoc-1, agrees with experimental data very well, while the ion-specific
565 parameter set, Assoc-I-15, presents slightly larger deviations.



566
 567 Figure 11. Comparison of the MIAC of the aqueous NaCl solution calculated using Assoc-1 and
 568 experimental data from 273.15 to 383.15 K.



569
 570 Figure 12. Comparison of the MIAC of the aqueous NaCl solution calculated using Assoc-I-15 and
 571 experimental data from 273.15 to 383.15 K.

572 Efforts have been made in the literature to capture the temperature-anomaly of MIAC.
 573 Selam et al. [44] captured the temperature-anomaly using temperature-dependent dispersion
 574 energy parameters. Roa Pinto et al. [40] captured the temperature-dependence anomaly without
 575 introducing temperature-dependent interaction energy parameters. Specifically, the only
 576 parameter set that managed to capture the temperature-dependence is the 10-parameter
 577 association strategy, “model 2.0”. However, the regressed MSA diameter was smaller than the
 578 HS diameter for Cl⁻. Difference between the parameterization strategies of this work and Ref
 579 [40] are:

- 580 - The parameter sets are different. In Ref [40], there are more adjustable parameters, e.g.,
- 581 an extra binary interaction parameter was included for association volume.
- 582 - Ionic diameters were not forced to meet the physical consistency constraints in Ref [40].
- 583 The regressed anion MSA diameter was smaller than the HS diameter. In this work, the
- 584 ionic diameters consistency is enforced in the regression.

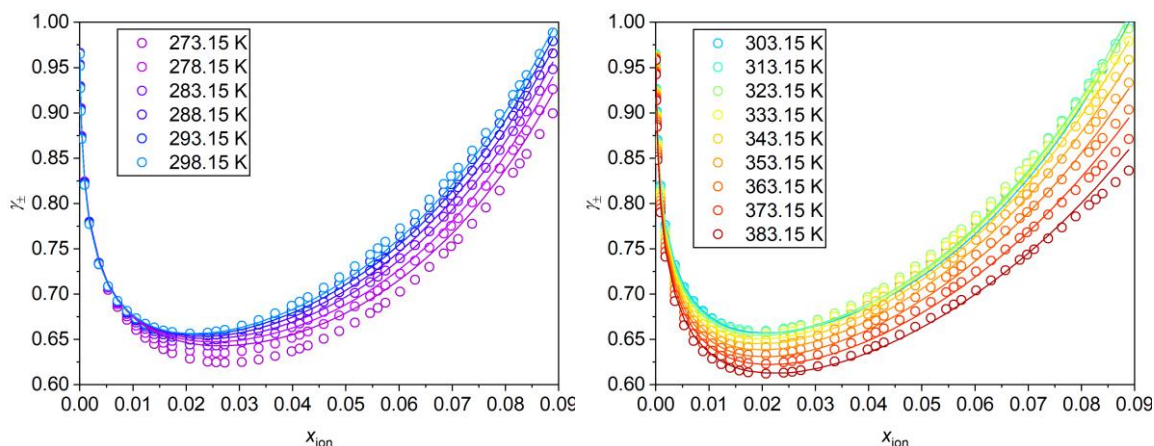
- 585 - The included data are different. In Ref [40], representative datasets were used, covering
586 larger ranges quite evenly. On the other hand, in this work, experimental data from
587 extensively collected and evaluated databases are used. The availability of such data
588 inevitably favors the ranges near room temperature (298.15 K).
- 589 - More importantly, enthalpy data was included in the objective function of Ref [40]. The
590 property, although only qualitatively represented, could be significantly important for the
591 temperature dependence of the MIAC.

592 Here we refrain from using enthalpy data for the following reasons:

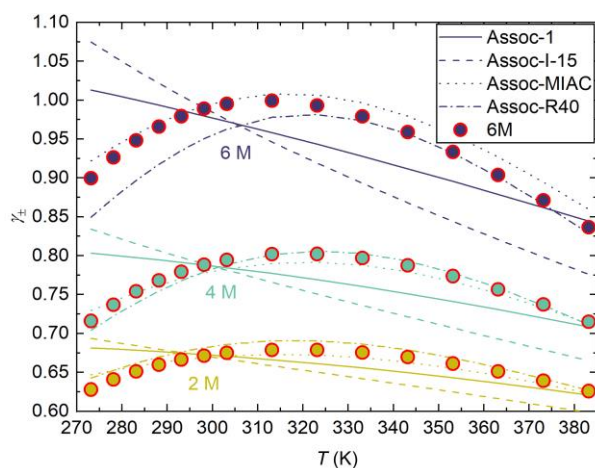
- 593 - In future works, the model will be extended to mixed-solvent electrolyte solutions, for
594 which enthalpy data are very scarce.
- 595 - The model has been shown to present large deviations from the experimental enthalpy
596 data in Ref [40]. The best that could be achieved was a qualitative agreement. Including
597 the data would jeopardize the highly accurate representation of the MIAC, VLE, and
598 density data in this work.

599 Enforcing the ionic diameter consistency in the regression, efforts are made to obtain the
600 same temperature dependence as in Ref [40]. When regressed to the evenly distributed pseudo-
601 experimental MIAC dataset from Ref [303], the obtained parameter set captures a maximum,
602 as shown in Figure 13. Only MIAC is included in this regression. This parameterization
603 strategy is here noted as Assoc-MIAC. When VLE and density data are also included in the
604 objective function along with the evenly distributed MIAC dataset, a maximum in MIAC is
605 also captured, however, the isotherms are rather close to each other; the temperature-
606 dependence is not captured as well as when using Asso-MIAC. In principle, the joint effect of
607 the decreasing Wertheim association term and increasing other terms can capture the maximum
608 of MIAC with temperature. However, whether it is captured is a matter of parameter regression.

609 The use of an extra binary interaction parameter for the association volume has been found to
610 result in only marginal difference. The parameters are provided in the Supplementary Material,
611 along with the comparisons of this parameter set and the Ref [40] with the data summarized in
612 Table 1 for the aqueous NaCl solution. Figure 14 shows a comparison of the MIAC
613 temperature-dependence of the modeling strategies Assoc-1, Assoc-I-15, Assoc-MIAC, and
614 Assoc-R40 (the parameter set from Ref [40]) at 2, 4, and 6 M from 273.15 to 383.15 K. Assoc-
615 MIAC and Assoc-R40 correctly represent the temperature dependence of MIAC, while Assoc-
616 1 and Assoc-15 present monotonically decreasing trends with temperature.



617
618 Figure 13. Comparison of the MIAC of the aqueous NaCl solution calculated using Assoc-MIAC and
619 experimental data from 273.15 to 383.15 K.



620
621 Figure 14. Comparison of the MIAC temperature-dependence of the modeling strategies Assoc-1,
622 Assoc-I-15, Assoc-MIAC, and Assoc-R40 at 2, 4, and 6 M from 273.15 to 383.15 K.

623 An advantage of the salt- and ion-specific parameter sets in this work (Asso-1 and Asso-
624 I-15), compared to Asso-MIAC and Asso-R40, is that they are more accurate close to 298.15
625 K, which is the region where most data are located, especially considering that the model is to
626 be extended to mixed-solvent electrolyte solutions. Therefore, the temperature-anomaly at low
627 temperature has not been further pursued in this work.

628 4.3 Osmotic coefficient

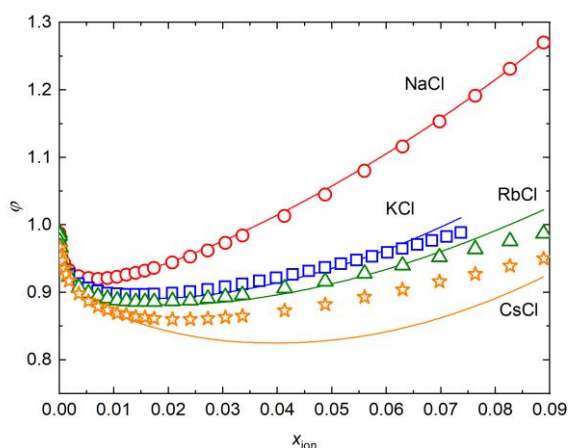
629 The model is tested on the osmotic coefficient, which is not included in the parameter
630 estimation. The osmotic coefficient is related to the MIAC according to the Gibbs-Duhem
631 equation. Therefore, if the MIAC is accurately represented with the model, the osmotic
632 coefficient is also expected to be accurately predicted. Therefore, the ThereforExperimental
633 osmotic coefficient data have been evaluated in Ref [7]. Because MIAC and VLE data are up
634 to 400 K in the regression, osmotic coefficient data above 400 K are removed in our study.
635 Furthermore, the osmotic coefficient data of the aqueous RbCl and CsCl solutions from Ref [7]

636 cover a much larger salt composition range compared to the MIAC data used in the regression.
 637 Therefore, the salt composition is limited to 6 M in this section (approximately $x_{\text{ion}} = 0.089$,
 638 close to the solubility of NaCl in water at 298.15 K). For the aqueous KCl, RbCl, and CsCl
 639 solutions, the range is larger than the MIAC data range (as shown in Table 1).

640 Table 6 shows the osmotic coefficient data and prediction deviations of the ion-specific
 641 parameter set, Assoc-I-15. Figure 15 shows the comparison of the osmotic coefficient of
 642 aqueous NaCl, KCl, RbCl, and CsCl solutions calculated using Assoc-I-15 and experimental
 643 data at 298.15 K and 0.1 MPa. Please note the very small scale in the graph. Overall, the model
 644 predicts the osmotic coefficient very well. For the aqueous NaCl solution, the calculated curve
 645 is in good agreement with the experimental data. For the aqueous KCl and RbCl solutions,
 646 deviations increase very slightly as salt composition exceeds the range of the MIAC data that
 647 are used in the regression; maximum deviations are within 5%. For the aqueous CsCl solution,
 648 deviation increases to more than 6% at approximately $x_{\text{ion}} = 0.05$. As shown in Table 1, CsCl
 649 MIAC data are available only up to $x_{\text{ion}} = 0.049$ for the mixture. Furthermore, the MIAC
 650 deviation is more than 4% at $x_{\text{ion}} = 0.049$ (as shown in Figure 6), in which range the largest
 651 osmotic coefficient deviation is located. Above the MIAC data range and up to $x_{\text{ion}} = 0.089$,
 652 the osmotic coefficient deviation decreases. However, outside of the range of the comparisons
 653 here, the osmotic coefficient data are available up to very high composition. The more drastic
 654 increase of the model compared to the experimental data retains as composition increases above
 655 what is shown in Figure 16, with the osmotic coefficient being 1.26 at $x_{\text{ion}} = 0.15$ (larger than
 656 11 M, far beyond the MIAC data range), where the experimental osmotic coefficient is 1.02.

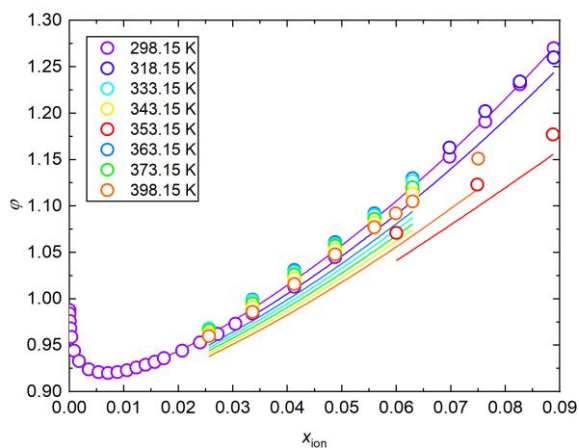
657 Table 6. Osmotic coefficient data and prediction deviations of the ion-specific parameter set, Assoc-I-
 658 15.

Wate +	T (K)	p (MPa)	x_{ion}	N_{dp}	References	AAD (%)	MAD (%)
NaCl	298.15 ~ 398.15	0.1 ~ 0.4	~ 0.089	73	[304–306]	1.6	3.5
KCl	273.15 ~ 372.75	0.1 ~ 0.2	~ 0.079	95	[74,307–310]	1.4	4.7
RbCl	298.15	0.1	~ 0.089	32	[304,311]	0.82	4.2
CsCl	298.15 ~ 383.15	0.1	~ 0.089	35	[60,312]	2.8	6.6



659
 660 Figure 15. Comparison of the osmotic coefficient of the aqueous NaCl, KCl, RbCl, and CsCl solutions
 661 calculated using Assoc-I-15 and experimental data at 298.15 K and 0.1 MPa.

662 Figure 16 shows the comparison of the osmotic coefficient of the aqueous NaCl solutions
 663 calculated using Assoc-I-15 and experimental data from 298.15 to 398.15 K. Unlike for the
 664 MIAC, experimental data are not available for the osmotic coefficient at lower temperatures.
 665 An overlap is observed in the experimental data at 298.15, 318.15, and 333.15 K, indicating
 666 that there is an osmotic coefficient maximum in the temperature range. The ion-specific
 667 parameter set predicts a monotonic decrease with temperature, like for the MIAC, and fails to
 668 capture the temperature maximum. Furthermore, the deviation is slightly larger at elevated
 669 temperatures. The salt-specific parameter sets, Assoc-1, Assoc-MIAC, and Assoc-R40, capture
 670 the high temperature behavior better than Assoc-I-15.



671
 672 Figure 16. Comparison of the osmotic coefficient of the aqueous NaCl solutions calculated using
 673 Assoc-I-15 and experimental data from 298.15 to 398.15 K.

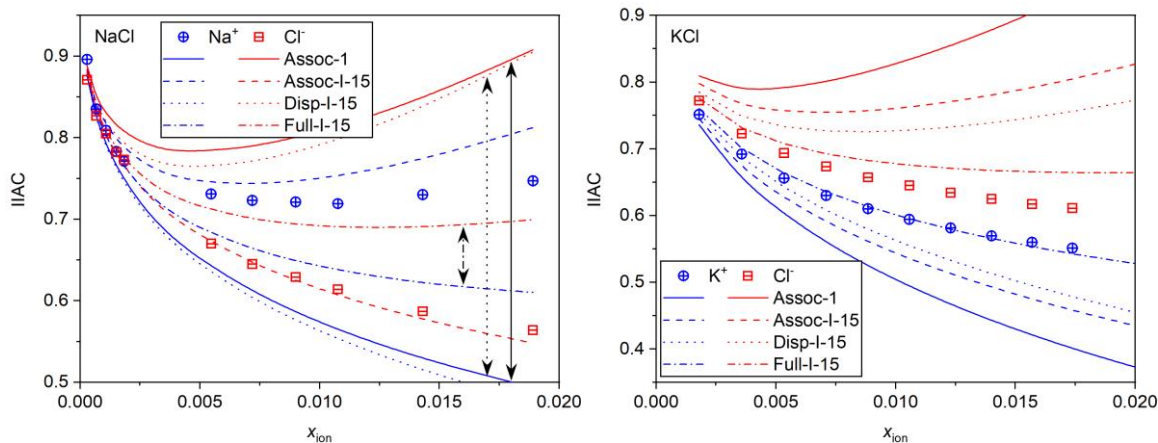
674 4.4 Individual ion activity coefficient (IIAC)

675 In addition to the mean properties, IIAC is an important property that can be used for
 676 further testing the model. IIAC is important in that it appears in the Nernst equation for the
 677 electrode potential in a half-cell [313]. In the ion-specific parameterizations, the 4 aqueous

678 alkali chloride solutions are regressed together, introducing more information about the
679 individual ions in the regression by the different cations and the common anion, Cl^- . Although
680 there is some debate about the validity of the experimental measurement of the activity
681 coefficient of individual ions [314–320], recent molecular simulations [321] qualitatively
682 agrees with the experimental data. Therefore, IIAC is used as a qualitative test here.

683 Experimental [322,323] and molecular simulation [321] investigations presented that the
684 relative magnitudes of the cation and anion IIAC for the aqueous NaCl and KCl solutions are
685 opposite. In the salt-specific parameterization in this work, the only individual ion information
686 that are introduced are: the HS diameters, which are proportional to the Pauling diameters, and
687 the Born diameters, which are determined based on the Gibbs energy of solvation. All 10 salt-
688 specific strategies fail to correctly predict the larger IIAC for Na^+ . All the properties used in
689 the objective function are mean properties, i.e., individual ion information is not introduced
690 from the data. In the ion-specific parameterizations, the 4 aqueous alkali chloride solutions are
691 regressed together, introducing more information about the individual ions in the regression by
692 the different cations and the common anion, Cl^- .

693 Figure 17 compares the IIAC of the aqueous NaCl and KCl solutions calculated using the
694 salt-specific strategy, Assoc-1, ion-specific strategies, Assoc-I-15, Disp-I-15, and Full-I-15,
695 and experimental data. The experimental data are from Ref [322,323]. This property is not
696 included in the regression. Thus, these results are pure predictions. Assoc-I-15 correctly
697 predicts the opposite relative magnitudes of the cation and anion IIAC of the aqueous NaCl
698 and KCl solutions, while the other strategies predict smaller IIAC for the cation in both cases.
699 Results are similar for the 12-parameter ion-specific strategies: Assoc-I-12 correctly predicts
700 the opposite relative magnitudes, while Disp-I-12 and Full-I-12 fail. Table 7 summarizes
701 whether the opposite relative magnitudes of the cation and anion IIAC of the aqueous NaCl
702 and KCl solutions is correctly predicted by the ion-specific strategies. In a nutshell, the ion-
703 specific association strategies present an advantage for predicting the qualitative behavior of
704 IIAC over the ion-specific dispersion and full strategies and the salt-specific strategies.



705

706 Figure 17. Comparison of the IIAC of the aqueous NaCl and KCl solutions calculated using the salt-
 707 specific strategy, Assoc-1, ion-specific strategies, Assoc-I-15, Disp-I-15, and Full-I-15, and
 708 experimental data.

709 Table 7. Whether the opposite relative magnitudes of the cation and anion IIAC of the aqueous NaCl
 710 and KCl solutions is correctly predicted by the ion-specific strategies.

Parameterization strategies	Opposite relative magnitudes
Asso-I-15	😊
Asso-I-12	😊
Disp-I-15	😞
Disp-I-12	😞
Full-I-15	😞
Full-I-12	😞
Full-I-20	😞

711

5 Conclusions

712 In this work, the ePPC-SAFT model is parameterized in an ion-specific approach
 713 enforcing physical consistency of the ionic parameters. Properties that are useful in practical
 714 applications are included in the objective function, i.e., MIAC, VLE, and density. Reference
 715 data are taken from extensively collected and critically evaluated databases. The association,
 716 dispersion, and full (association + dispersion) approaches are compared for the short-range ion-
 717 ion and ion-solvent interactions. Efforts are made to parameterize the model in an ion-specific
 718 manner with minimum loss of accuracy. The ion-specific parameter sets are compared with the
 719 salt-specific sets. The model and parameter sets are further tested on the MIAC beyond the data
 720 range of the regression, and on properties that have not been included in the regression. The
 721 major conclusions are:

- 722 - For the salt-specific cases, all strategies are quite accurate in the entire temperature,
 723 pressure, and salt composition range of the experimental database; the association and full
 724 strategies are more accurate than the dispersion strategies. Like-ion dispersion should not

725 be included in the dispersion strategies, but is recommended to be included in the full
726 strategies. Despite using a larger number of adjustable parameters and including more
727 completely the physical interactions, the full strategies are not more accurate than the
728 association strategies. Similar results are obtained both the investigated RPMs.

729 - For the ion-specific cases, the association strategies are approximately as accurate as the
730 salt-specific cases in terms of the average and maximum deviations, while the dispersion
731 and full strategies are less accurate. To obtain the same accuracy as the association
732 strategies, the full strategy has to use a larger number of adjustable parameters. This may
733 indicate a reason not to include both dispersion and association for the short-range ion-
734 ion and ion-solvent interactions. The 12-parameter ion-specific association strategy with
735 the same binary interaction parameter for the 4 salts is only slightly less accurate than the
736 15-parameter case.

737 - However, at higher temperature, the ion-specific association strategy is less accurate than
738 the salt-association association strategy. Neither salt- or ion-specific strategy represent the
739 MIAC temperature maximum of the aqueous NaCl solution, which was successfully
740 represented in the literature, using temperature-dependent energy parameters or a
741 physically inconsistent parameter set. The behavior is successfully represented here using
742 the association approach by regressing only to an evenly distributed pseudo-experimental
743 MIAC dataset. However, the attempt is dropped because the ion-specific association
744 parameter set is more extendable and more accurate overall speaking for all investigated
745 properties, with the temperature-anomaly in MIAC resulting in only slightly larger
746 deviations.

747 - An analysis of the contributions of the terms shows that the dispersion, association, and
748 hard chain terms are all close to linear with salt composition, and counter-balance each
749 other in all cases. The various terms present no trend within the alkali chloride series in
750 the full strategies. Therefore, the association strategies are possibly more favorable
751 compared to the dispersion and full strategies.

752 - The model and parameter set is tested on the osmotic coefficient and individual ion
753 activity coefficient, which are not included in the regression. The ion-specific association
754 parameter set accurately represents the osmotic coefficient over wide temperature and salt
755 composition ranges, beyond those of the MIAC data used in the regression. The ion-
756 specific association strategies correctly predict the opposite relative magnitudes of the
757 cation and anion individual ion activity coefficient of the aqueous NaCl and KCl solutions,
758 while the ion-specific dispersion and full strategies, and all the salt-specific strategies fail.

759 To sum up, state-of-the-art parameter sets have been obtained for the ePPC-SAFT model,
760 facilitating benchmarking of the short-range interaction approaches, and salt- and ion-specific
761 parameterizations. We recommend including the Wertheim association for the short-range ion-
762 ion and ion-solvent interactions, and parameterizing SAFT models in an ion-specific manner
763 using physically consistent parameters. In the future, the model will be extended to other
764 aqueous electrolyte solutions, and mixed-solvent and mixed-salt electrolyte solutions.

765 **CRedit authorship contribution statement**

766 Fufang Yang: Conceptualization, Methodology, Software, Validation, Formal Analysis,
767 Investigation, Data Curation, Writing – Original Draft, Visualization. Tri-Dat Ngo: Software.
768 Juan Sebastian Roa Pinto: Methodology. Georgios M. Kontogeorgis: Conceptualization,
769 Supervision, Writing – Review & Editing, Funding Acquisition. Jean-Charles de Hemptinne:
770 Conceptualization, Resources, Supervision, Software, Writing – Review & Editing, Project
771 Administration, Funding Acquisition.

772 **Declaration of competing interest**

773 The authors declare no competing financial interest.

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778 Thermodynamics”.

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