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Calculation of the free energy of solvation of the Li^+ and Na^+ ions in water and chloroform

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Abstract

The free energies of solvation of the lithium and sodium cations in water and in chloroform were calculated using Monte Carlo computer simulations. The free energies were obtained using thermodynamic integration either on a nearly linear path or on a polynomial path and the integral was evaluated with a Gaussian quadrature. The precision of the results was confirmed through comparison with additional calculations that obtained the solvation free energy differences by mutating a lithium to a sodium cation. Three sources for the discrepancy between the calculated and experimental solvation free energies were considered: the *multibody terms* were estimated using a polarization model; the *long-range contributions* were estimated with the Born correction and with a calculation using larger system size; and the extent of *charge transfer* was examined by ab initio calculations based on complete solvation shells extracted from the simulation.

Keywords: Charge transfer; Chloroform; Free energy; Lithium; Monte Carlo computer simulation; Sodium; Thermodynamic integration

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1. Introduction

Efficient calculation of the change in the free energy of association upon changing a ligand requires a thermocycle which involves the free energies of mutating the ligand both in the associated solvated state and in the free solvated state [1]. Since a ligand can take part in many different associations, it is of interest to develop a library of the calculated free energies of mutations and creations in the free state so that in subsequent studies of associations only the mutation and creation in the associated form has to be carried out. It is in this spirit that we undertook the calculation of the solvation free energies of the Na^+ and Li^+ ions, both in water and in chloroform, the former being the prototype associated liquid, the latter the prototype nonpolar liquid.

While such calculations have already been carried out in water [2], we decided to repeat them for three reasons. First, the discrepancy between the calculated and experimental values is rather large. The present work obtained quantitative estimates of several different contributing effects that were neglected or partially incorporated in these calculations, providing guidance to

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improving the representation of the interactions. Second, as our calculations were performed over a complete thermocycle involving creation of both ions independently, they provided further demonstration of the robustness of the methodology chosen for the calculation of solvation free energies. Third, comparison with the previous calculations also demonstrates the efficiency of the free energy methodology used.

2. Methods

The calculation of solvation free energy differences from computer simulation is generally based on a path that connects the two systems in the configuration space and on some quantity whose Boltzmann average is related to the free energy. The range of options has been reviewed in several publications [3–6]. Among these, thermodynamic integration (see below) on a nearly linear path was shown to be particularly well suited to changes involving the creation of new atoms [5,7–9]. The nearly linear path is defined by a coupling parameter

$$E(\lambda, \boldsymbol{X}^N) = \lambda^k \times E_1(\boldsymbol{X}^N) + (1-\lambda)^k \times E_0(\boldsymbol{X}^N) \tag{1}$$

where E_0 and E_1 are the energy functions for the two systems between which the free energy difference is to be computed and λ varies from 0 to 1. For the calculation of the solvation excess free energies (denoted by A') the reference system is the ideal gas at the same temperature and density, thus E_0 is simply taken as zero everywhere. Thermodynamic integration (TI), based on ideas of Kirkwood [10], relates the free energy difference between two systems to the integral

$$\Delta A = A_1 - A_0 = \int_0^1 \langle \partial E(\lambda)/\partial\lambda \rangle_\lambda \mathrm{d}\lambda \tag{2}$$

Substituting Eq. (1) into Eq. (2) gives

$$\Delta A = \int_0^1 k[-(1-\lambda)^{k-1}\langle E_0\rangle_\lambda + \lambda^{k-1}\langle E_1\rangle_\lambda]\mathrm{d}\lambda \tag{3}$$

For transitions involving the creation and/or annihilation of atoms a potential of the form $1/r^e$ contributes to the integrand an asymptotic behavior of $\propto \lambda^{(kd/e)-1}$ where d is the dimensionality of the space [3]. Thus k should be selected high enough to keep the asymptotic behavior finite [3,11,12]. Clearly, in three dimensions $k \geqslant e/3$ insures a convergent integrand.

It has been argued before [5] that TI over the path defined by Eq. (1) used in conjunction with a numerical (preferably Gaussian) quadrature is generally a better choice than TI with slow growth [13] or the perturbation method [14]. This conclusion has also been reached recently by Bayly and Kollman for the Li^+ and Cs^+ mutation [15].

Recently it has been demonstrated that a generalization of the nearly linear path that uses different λ exponents for the different types of potential energy terms for the definition of the path ('Polynomial TI') can increase the precision of the numerical quadrature [16,17]. In this case

$$\Delta A = \int_0^1 \sum_{e=12,6,1} k_e[-(1-\lambda)^{k_e-1}\langle EI_0(e)\rangle_\lambda + \lambda^{k_e-1}\langle EI_1(e)\rangle_\lambda]\mathrm{d}\lambda \tag{4}$$

where $EI(e)$ refers to terms containing $1/r^e$. One of our calculations used this technique with the exponent set $\{4,3,2\}$ for the repulsion, dispersion and electrostatic terms, respectively.

All simulations used the Metropolis algorithm [18] with the force-bias sampling technique of Rao et al. [19]. As proposed earlier [20], the force-biasing was scaled down near the solute. A bitmap was used to keep track of water molecules in each other's vicinity [21].

The perturbation of the water–water multibody contributions in the presence of the strong ionic field, not included in the parameterization of the ion–water interactions [22], was estimated using the Campbell–Mezei polarization model [23] that represents the multibody contributions with the energy of the cooperatively and self-consistently induced dipoles. In this model the waters are represented by a tenth-order multipole expansion. This model has been derived from ab initio calculations providing the monomer charge distribution and the interaction energies of a large number of dimers; it also uses the experimental dipole polarizability of water. Apart from inverse distance powers representing dispersion and exchange repulsion, it calculates the interaction of each water's charge distribution with the field of the rest. Introduction of the ion into the system would only add an additional term to the field. The ion charge distribution was represented by a point charge.

The estimate of the polarization contribution was based on the calculation of the linear dipole polarization energy both with the ion present and absent. The difference between the polarization energies of these two calculations represent the polarization energy due to the presence of the ion. To obtain the cooperative contribution of the ion, we subtracted from this polarization energy the sum of the polarization energies calculated between the ion and each water (since this contribution is included in a potential parameterized to ion–water pairwise energies). The induced energies were calculated with the program package MAXWELL [24] based on configurations extracted from the simulation.

The long-range contributions were estimated using the estimate for the energy of a unit charge inside a spherical cavity embedded in a dielectric continuum given by Born [25] as

$$A^0 = \frac{1-\epsilon}{2\epsilon R} \tag{5}$$

The application of this formula to the simulation in a polygonal simulation cell is not straightforward since the minimum image convention was used for the ion–solvent interactions. To obtain explicit contributions from a sphere only around the ion at each quadrature point the contribution to the solute–solvent binding energy from waters that lie outside the largest inscribed sphere in the simulation cell first have to be subtracted from the ion–solvent energy. However, these corrected energies have to be averaged with weights involving the subtracted contributions:

$$\langle E \rangle_{\mathrm{in}} = \frac{\langle E_{\mathrm{in}} \exp(E_{\mathrm{out}}/kT) \rangle_{\mathrm{in+out}}}{\langle \exp(E_{\mathrm{out}}/kT) \rangle_{\mathrm{in+out}}} \tag{6}$$

where E_{in} and E_{out} denote contributions coming from the inside sphere and outside the inside sphere, respectively, and $\langle\,\rangle_{\mathrm{in}}$ and $\langle\,\rangle_{\mathrm{in+out}}$ refer to Boltzmann averages based on E_{in} and $E_{\mathrm{in}} + E_{\mathrm{out}}$, respectively. The Born correction should then be applied to the free energy calculated with these contributions corrected to the inscribed sphere.

The extent of charge transfer from the ions was estimated based on the first solvation shell of the first ten configurations extracted from the simulations of the whole ion (i.e., at $\lambda = 1$). Ab initio calculations were next performed on these clusters using the Gaussian-92 program with 6-31G* basis set and the amount of charge transfer was estimated from Mulliken population analysis.

3. Calculations

The calculations reported in the paper were run at 298 K and at experimental partial molar volumes of the solute and solvent. Each system contained 215 solvent molecules and one ion under face-centered cubic periodic boundary conditions. The solute–solvent interactions used the minimum image convention while the solvent–solvent interactions were calculated with a spherical cutoff, 7.75 Å for water and 12.0 Å for chloroform. The minimum image convention was used for the ion–solvent interaction since a spherical cutoff would effectively partition the simulation cell into two regions with significant energy difference and that would have resulted in an artifactual density jump at the sphere boundary. In order to test the extent of applicability of the Born correction (see below) the aqueous sodium calculation was repeated in a larger system containing 599 waters. The λ exponent k in Eq. (1) was chosen to be 4 in the free energy calculations involving 215 solvent molecules and the polynomial TI exponent set $\{4, 3, 2\}$ was used in the larger sodium calculation. The integration over the coupling parameter λ was performed using 5-point Gaussian quadratures.

The intermolecular interactions were described with the OPLS family of potentials for the sodium and lithium [22] and for the chloroform [26]. For water, the TIP4P [27] model was used. A united atom representation was used for the CH group of the chloroform. The error estimates for the calculated free energies were derived from the error estimates on the integrands calculated at each quadrature point using the method of batch

Table 1
Solvation excess free energies[a] of the Li^+ and Na^+ cations in water

Run length[b]	$A'(\mathrm{Li}^+)$	$A'(\mathrm{Na}^+)$	$A'(\mathrm{Li}^+ - \mathrm{Na}^+)$	$A'(\mathrm{Li}^+) - A'(\mathrm{Na}^+)$	Error
1000 K	−144.3 ± 6.3	−116.5 ± 5.5	−24.9 ± 5.9	−27.8 ± 8.4	2.8
2000 K	−144.7 ± 3.9	−117.2 ± 3.7	−26.0 ± 4.6	−27.5 ± 5.4	1.5
3000 K	−144.3 ± 3.0	−117.7 ± 3.4	−25.4 ± 3.8	−26.6 ± 5.2	1.2
4000 K	−144.8 ± 2.6	−118.3 ± 3.3	−25.5 ± 3.2	−26.5 ± 5.1	1.0
5000 K	−144.5 ± 2.9	−117.9 ± 2.9	−	−26.6 ± 4.1	1.1
Exp.	−115	−89.6	−25.4	−25.4	

[a] In kcal mol^{-1}

[b] Run length in 1000 attempted MC steps.

[c] Experimental data from Ref. [32].

means [28,29] based on 10^5 Monte Carlo (MC) step blocks. The estimates given represent two standard deviations. To obtain an independent estimate of the precision of the calculated free energies, the free energy of mutating a Li^+ to a Na^+ was also computed with the same method providing a thermodynamic cycle over which the free energy changes should sum to zero.

The free energy simulations were supplemented with simulations using the full ion (i.e., $\lambda = 1$). The configurations used in estimating the multibody contributions and the charge transfer were extracted from these simulations. The simulations in chloroform (5×10^6 MC steps long each) were also used to calculate the ion–chloroform radial distribution functions and coordination number distributions.

4. Results and discussion

The calculated free energies are collected in Table 1 for the aqueous systems and in Table 2 for the solvation in chloroform. As the data in Table 2 show, the chloroform calculations converge significantly faster. It is gratifying to see that not only is the directly estimated error decreasing as the length of the run is increased but that the closure error is consistently well within the 95% confidence interval calculated from the block averages, providing an independent confirmation that the error estimates calculated directly from the thermodynamic integration runs do not underestimate the true error. In fact, the error estimates based on the block averages are rather conservative, similar to the case of the alanine dipeptide studied recently [17]. The TI integrand for the Na^+ system is shown in Fig. 1 using both the nearly linear TI (Eq. (3)) with $k = 4$ and polynomial TI with the exponent set $\{4, 3, 2\}$. It gives an additional demonstration of the ability of the polynomial TI to bring the TI integrand closer to linearity thereby reducing the quadrature error.

Table 2
Solvation excess free energies[a] of the Li^+ and Na^+ cations in chloroform

Run length[b]	$A'(\mathrm{Li}^+)$	$A'(\mathrm{Na}^+)$	$A'(\mathrm{Li}^+ - \mathrm{Na}^+)$	$A'(\mathrm{Li}^+) - A'(\mathrm{Na}^+)$	Error
500 K	−30.6 ± 4.7	−25.6 ± 4.4	−1.6 ± 4.0	−4.0 ± 6.4	2.4
1000 K	−29.3 ± 3.0	−26.3 ± 3.0	−4.3 ± 3.8	−3.0 ± 4.2	1.3
1500 K	−29.8 ± 2.3	−26.0 ± 2.2	−4.3 ± 2.9	−3.8 ± 3.2	0.5
2000 K	−30.0 ± 2.0	−25.7 ± 2.0	−4.1 ± 2.4	−4.3 ± 2.8	0.2

[a] In kcal mol^{-1}.

[b] Run length in 1000 attempted MC steps.

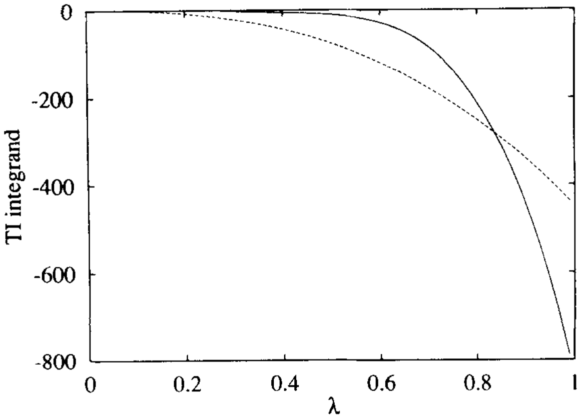

Fig. 1. Comparison of the TI integrand (in kcal mol^{-1}) with the exponent $k = 4$ on all terms (solid line) with polynomial TI using the exponent set $\{4, 3, 2\}$ (long dashed line).

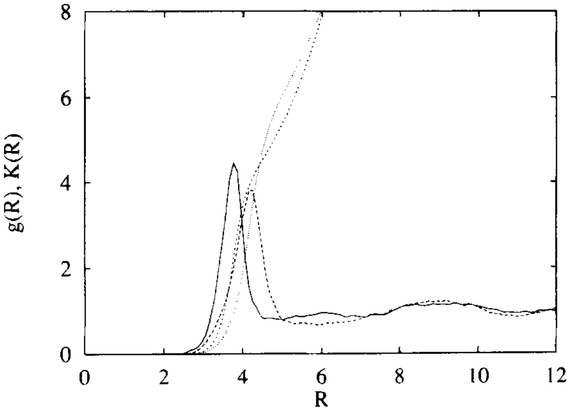

Fig. 2. Li^+–chloroform (full line) and Na^+–chloroform (long dashed line) radial distribution functions $g(R)$ and the corresponding running coordination number functions $K(R)$ (Li^+, short dashed line; Na^+, dotted line).

The present aqueous calculations can also be compared with the study of Jayaram et al. [2] where the free energy of changing an uncharged particle of the size of the sodium ion to a fully charged ion was calculated with the perturbation method in 5 simulation steps. The free energy of charging the sodium atom was obtained as -120 ± 1.7 kcal mol^{-1}. This is 2 kcal mol^{-1} more negative than the value obtained here for the complete hydration of the sodium ion. As the free energy of solvation of methane in water was found to be 2.5 kcal mol^{-1} by Jorgensen et al. [30] and the van der Waals radius of methane is 30% larger than that of a sodium ion, our calculations are in excellent agreement with their work. To appreciate the efficiency of the thermodynamic integration technique employed here it is to be noted that perturbation method calculations have to separate the creation of the van der Waals particle and the electrostatic charging to avoid numerical instabilities [31] and the computational expense of our calculations turned out to be comparable with the electrostatic charging process only (which is the computationally easier part).

The calculated radial distribution functions in chloroform are shown on Fig. 2 for the Li^+ and Na^+ ions. In sharp contrast to the aqueous system where the first peak is followed by a region of near zero density [22], even the first minima are rather shallow, indicating a much less sharply defined solvation shell than in water. The coordination number distributions, shown in Fig. 3 for the Li^+ and Na^+ ions, show similar trends. The coordination numbers were defined with a first shell radius of 4.85 Å and 5.2 Å for the Li^+ and Na^+ ions, respectively. These radii resulted in a mean coordination number of 4.8 and 6.2 for the Li^+ and Na^+ ions, respectively.

The calculated excess free energies of hydration are about 30 kcal mol^{-1} too negative in comparison with the experimental result for both ions [32]. This is similar to the discrepancy found by Chandrashekar et al. [22] for the enthalpy of solvation of these ions, where the calculated enthalpies

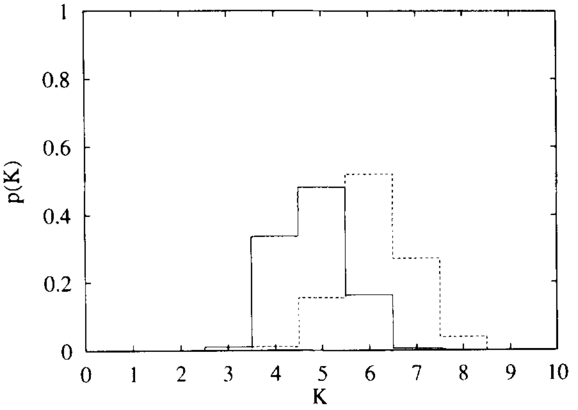

Fig. 3. Coordination number distribution $p(K)$ for the Li^+ (full line) and the Na^+ (long dashed line) ions in chloroform.

were 35 and 24 kcal mol^{-1} above the respective experimental values. No experimental data were found for the solvation free energies in chloroform.

Chanrashekar et al. [22] pointed out that since their potential was parameterized on ion–water dimer data it lacks multibody contributions and this might be responsible for this shortfall. To estimate the magnitude of the multibody terms, we extracted 20 configurations each (10^5 MC steps apart) from simulations of the Li^+ and Na^+ ions in water and obtained the estimate of the cooperative contribution of the ions based on the induced dipole energies, as described above. The average contribution of Li^+ to the induced energy was $+8.0 \pm 22.8$ kcal mol^{-1} and that of Na^+ was $+21.8 \pm 14.4$ kcal mol^{-1}. As these values are positive (i.e., repulsive), they are an upper bound to the multibody contributions since if the simulations were run using the multibody terms then the waters would rearrange themselves to reduce this repulsion.

Another neglected effect is the contribution of waters beyond the simulation cell (i.e., long-range contributions). It was estimated with the Born correction, as described in the previous section, based on a 10.4 Å sphere. With such a radius, the Born correction is -15.7 kcal mol^{-1}. Using Eq. (6) to transform our calculated free energies to a system with a cutoff sphere around the ions results in a reduction of -5.8 kcal mol^{-1} and -4.5 kcal mol^{-1} for the lithium and sodium ion free energies, respectively, giving the long-range correction estimates as -9.9 kcal mol^{-1} and -11.2 kcal mol^{-1} for the lithium and sodium ions, respectively.

A larger sodium system (containing 599 waters) was also studied to find out how close the waters outside the 10.4 Å sphere are to the limiting behavior (i.e., how applicable Eq. (5) is). The inscribed sphere of this system was 14.7 Å. The free energy was obtained as $A' = -125.5 \pm 0.7$ kcal mol^{-1} using the whole cell, and correcting for contributions from the inside sphere only gave -117.9 kcal mol^{-1}. Comparison of the free energies obtained with the two systems of different sizes indicates that the Born correction estimate is a lower bound to the outside contributions since the difference between the simulations' contribution, 7.5 kcal mol^{-1}, is about twice the corresponding difference between the Born corrections. Incidentally, this indicates that waters at 10.4–14.7 Å distance from an ion still cannot be fully considered a dielectric continuum — a similar conclusion was reached by Straatsma and Berendsen [33] on a different model.

Combination of the estimates of the multibody and long-range terms for Na^+ thus provides an upper limit of at most 10.6 kcal mol^{-1} for these two corrections, significantly falling short of the discrepancy between the calculated and experimental solvation free energies. For the Li^+ ion, the combined estimate is -1.9 kcal mol^{-1}, making the comparison even worse.

Since both the ion–water potential and the polarization models are essentially ab initio, and the shortfall is rather large, it is reasonable to assume that there must be an additional neglected contribution. One such candidate is the possibility of charge transfer. This hypothesis was prompted by recent ab initio calculations showing that a sodium ion is likely to lose about 5% of its charge in a polysaccharide crystal hydrate [34]. If charge transfer between the ion and waters is significant, then the use of a full +1 charge on the ions will introduce a distortion to the system consisting of a reduced charge separation between the ion and the waters. Thus one would expect both the ion–water and the water–water electrostatic energies to be different from the situation where no charge transfer has occurred. In the current parameterization, the ion–water energies have been fitted to the ab initio values that incorporate the charge transfer occurring between each pair, the effect of such charge transfer is not taken into account for the calculation of water–water interactions. As a result, the system would display lower energies and this is indeed what is observed.

To obtain a quantitative characterization of the extent of charge transfer, we extracted from the configurations used for the estimation of the multibody contribution the first shell around Na^+ and Li^+, and from similarly generated configurations in chloroform, and performed ab initio calculations on them. The average loss of charge of Li^+ in water and in chloroform was 0.32 ± 0.04 and 0.57 ± 1.19, respectively. For Na^+ the average loss of charge in water and in chloroform was 0.27 ± 0.06 and 0.29 ± 0.05, respectively.

Thus these ab initio results confirm the presence of significant charge transfer. The fact that more extensive charge transfer was observed for Li^+ where the shortfall is larger lends further support to our proposition that the inadequate treatment of charge transfer significantly contributed to the overestimation of the solvation free energies in the simulations. However, quantitative estimation of the effect of the charge transfer on the solvation free energy requires further study involving not only additional simulations but development of an appropriate methodology.

5. Conclusions

The solvation thermodynamics of the sodium and lithium ions in water and chloroform was studied with nearly linear and polynomial thermodynamic integration. In the aqueous systems it was shown that the residual error in the solvation free energy cannot be explained by the lack of multibody effects in the potential and by the neglect of contributions from waters beyond the simulation cell alone. It was shown that the use of a water potential parameterized for the neat liquid in combination with an ion–water potential fitted to ab initio energies can lead to the type of discrepancy observed in this study when significant charge transfer is present. Since ab initio calculations on selected solvation shells indicated that charge transfer does indeed occur it was proposed that the inadequate treatment of charge transfer might be responsible for the additional shortfall. Our results thus suggest that for best results in modeling ionic solvations the potential function should be able to accommodate not only polarization effects but charge transfer as well.

The solvation of these ions in chloroform is found to be much weaker (as expected) resulting in much smaller solvation free energies and less structure in the ion–solvent radial distribution function when compared with the respective aqueous case. The calculations also provided additional demonstration of the efficiency and thermodynamic integration over the nearly linear and especially over the polynomial path.

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