ELECTRONIC SUPPLEMENTARY INFORMATION

Development and Application of Effective Pairwise Potentials for UO_2^{n+}, NpO_2^{n+}, PuO_2^{n+}, and AmO_2^{n+} ($n = 1, 2$) Ions with Water

Vladimir Pomogaev,[a] Surya Prakash Tiwari,[a] Neeraj Rai,[a] George Goff,[b] William F. Schneider*[c] and Edward J. Maginn,*[d]

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Table 1 Non-bonded LJ interaction parameters for AnO_2^{n+} – water interactions. LJ parameters for SPC/Fw, TIP3P, TIP4P, and TIP5P water models are taken from previous work by Rai et al [1].

	SPC/Fw	SPC/E	TIP3P	TIP4P	TIP5P
σ_{An-Ow} (nm)	0.325	0.306	0.325	0.278	0.326
ε_{An-Ow} (kJ/mol)	0.270	0.587	0.270	1.390	0.260
σ_{OAn-Ow} (nm)	0.300	0.350	0.300	0.307	0.292
ε_{OAn-Ow} (kJ/mol)	1.080	0.192	1.080	0.850	1.470

Table 2 Mean squared error (MSE) to see the quality of fitting of LJ+Coulombic functional form for AnO_2^{n+}-SPC/E water interactions to ab-initio PES

AnO_2^{n+}	UO_2^{2+}	NpO_2^{2+}	PuO_2^{2+}	AmO_2^{2+}	UO_2^{+}	NpO_2^{+}	PuO_2^{+}	AmO_2^{+}
MSE	25.6	22.4	27.3	31.0	33.1	34.3	30.2	28.3

To account for the effect of simulation box size on the hydration free energy, hydration free energies of the same ion (UO_2^{2+}) in the same water model (SPC/E) were calculated in 5 different box sizes as plotted in Fig. 1. From the plot, we see that the hydration free energy gets almost independent at large box sizes. Since we used 3.12 nm box size to calculate hydration free energies in the main paper, thus we added -2.2 kJ/mol to the calculated hydration free energies to make them box size independent. Note that the effect of box size on the final value of hydration free energy was only studied for dication in SPC/E water. We assumed that a similar trend will follow for other ions and in other water models, and the same correction of -2.2 kJ/mol was employed in all the values of hydration free energies calculated in cubic box length of 3.12 nm for the dications. A similar procedure was used to obtain a correction of -1 kJ/mol to be added to the hydration free energies for monocations.

[a] *Department of Chemical and Biomolecular Engineering, University of Notre Dame, 182 Fitzpatrick Hall, Notre Dame, IN 46556, USA.*
[b] *Los Alamos National Laboratory, New Mexico 87545, USA.*
[c] *Department of Chemical and Biomolecular Engineering, University of Notre Dame, 182 Fitzpatrick Hall, Notre Dame, IN 46556, USA. Fax: 01 574 631 8366; Tel: 01 574 631 8754; E-mail: wschneider@nd.edu*
[d] *Department of Chemical and Biomolecular Engineering, University of Notre Dame, 182 Fitzpatrick Hall, Notre Dame, IN 46556, USA. Fax: 01 574 631 8366; Tel: 01 574 631 5687; E-mail: ed@nd.edu*

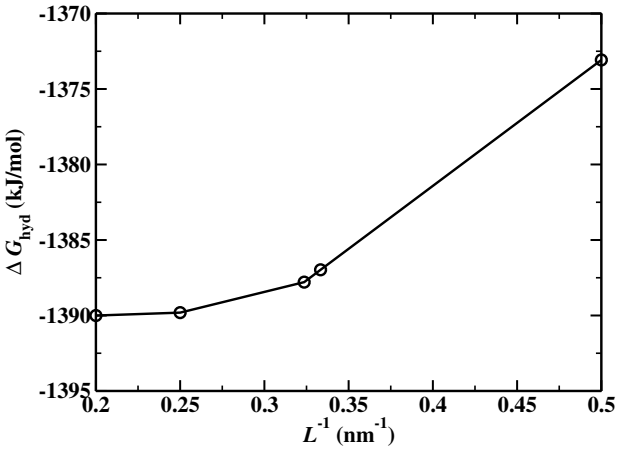

Fig. 1 Effect of cubic box length (L) used on ΔG_{hyd} of UO_2^{2+} in SPC/E water using MD simulations.

References

1 N. Rai, S. P. Tiwari and E. J. Maginn, *J. Phys. Chem. B*, 2012, **116**, 10885–97.