Supporting Information for

Adaptive Resolution Simulation of Supramolecular Water: The Concurrent Making, Breaking, and Remaking of Water Bundles

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MD SIMULATION DETAILS

Simulations are performed with the ESPResSo++ software package [1]. For the integration we use the standard velocity Verlet with a time step of 1fs. We use orthorhombic simulation box with periodic boundary conditions and minimum image convention. The simulation box size is $11.2\times2.8\times2.8$ nm^3. The temperature is maintained at 300 K with a local Langevin thermostat, with the value of the friction constant equal to 5.0/ps. The geometry of the water molecules is constrained with SETTLE [2]. The cutoff distance for the nonbonded interactions is $r_c = 1.2$ nm. The reaction field method [3] is used for the electrostatic interaction beyond the cutoff, with dielectric permittivity of inner and outer region equal to 1 and 80, respectively. Production runs for all simulations are 10 ns, whereas the equilibration runs are 1 ns. The AdResS results are in relevant areas compared to the fully atomistic simulations of the SPC [4] and bundled-SPC [5] water and to the fully coarse-grained simulation of the MARTINI [6] water. All simulations are performed under identical conditions.

ADDITIONAL RESULTS

To show that the clustering algorithm and the AdResS scheme do not introduce any spurious orientational alignments with the transition regions we plot in Figure 1 average orientation of water molecules. We consider two orientational order parameters $\eta^{(1)}$ and $\eta^{(2)}$, defined as

$$\eta^{(1)} = \langle \cos\alpha \rangle, \tag{1}$$

$$\eta^{(2)} = \frac{1}{2}\langle 3\cos^2\alpha - 1 \rangle, \tag{2}$$

where α denotes the angle between the dipole moment of water molecule and the normal vector pointing towards the CG region. A random orientation of water molecules corresponds to $\eta^{(1,2)} = 0$. The AT and ATwB regions show no preferential orientation, while in the HY region slight ordering is observed. Similar effect was noticed also in previous AdResS simulations [7–9].

The disparate local order of the unconstained and constrained SPC water can be clearly seen from the distributions of the tetrahedral order parameter (Figure 2). We

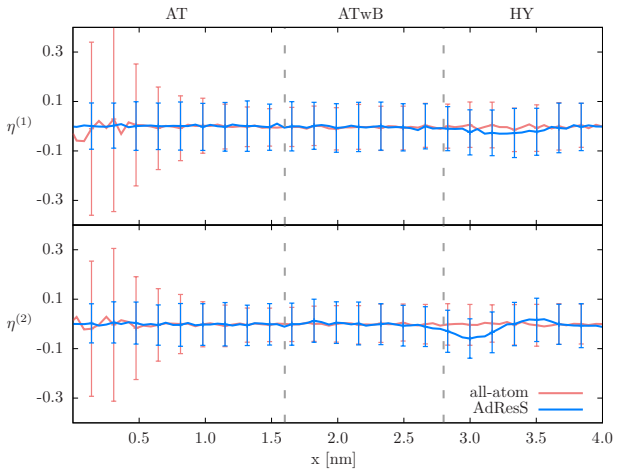

FIG. 1. Water order parameters $\eta^{(1)}$ and $\eta^{(2)}$ as a function of the x coordinate of the simulation box. Resolution region boundaries are denoted with the vertical gray lines.

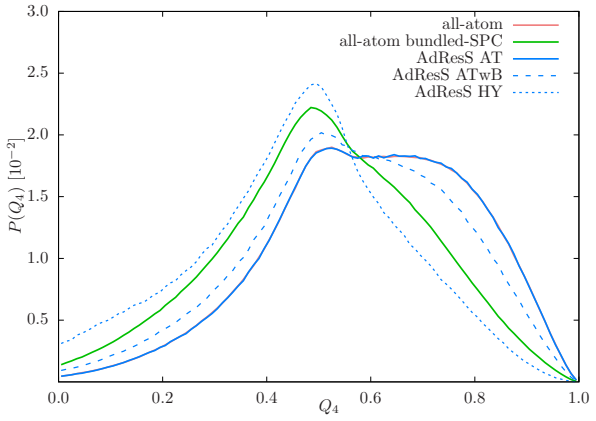

FIG. 2. Distribution of the tetrahedral order parameter $P(Q_4)$ for the all-atom SPC and bundled-SPC water. The order in the AdResS simulation is shown separately for the AT, ATwB, and HY regions.

distributions are computed separately the for the water molecules located in AT, ATB, and HY regions. The profile in AT region and the reference all-atom simulation profile match very well. Figure 3 shows the bundling energy U_B distributions of bundles located in the ATwB and HY regions.

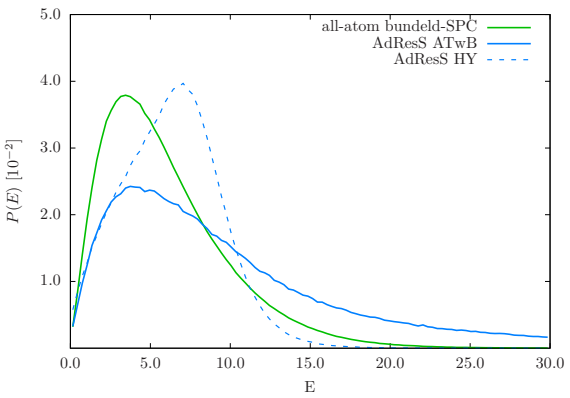

FIG. 3. Bundling energy U_B probability distributions for different regions of the AdResS multiscale simulation. For comparison we also plot the profile for the all-atom bundled-SPC simulation.

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