Supplementary Material: Density Artefacts at Interfaces Caused by Multiple Time-Step Effects in Molecular Dynamics Simulations

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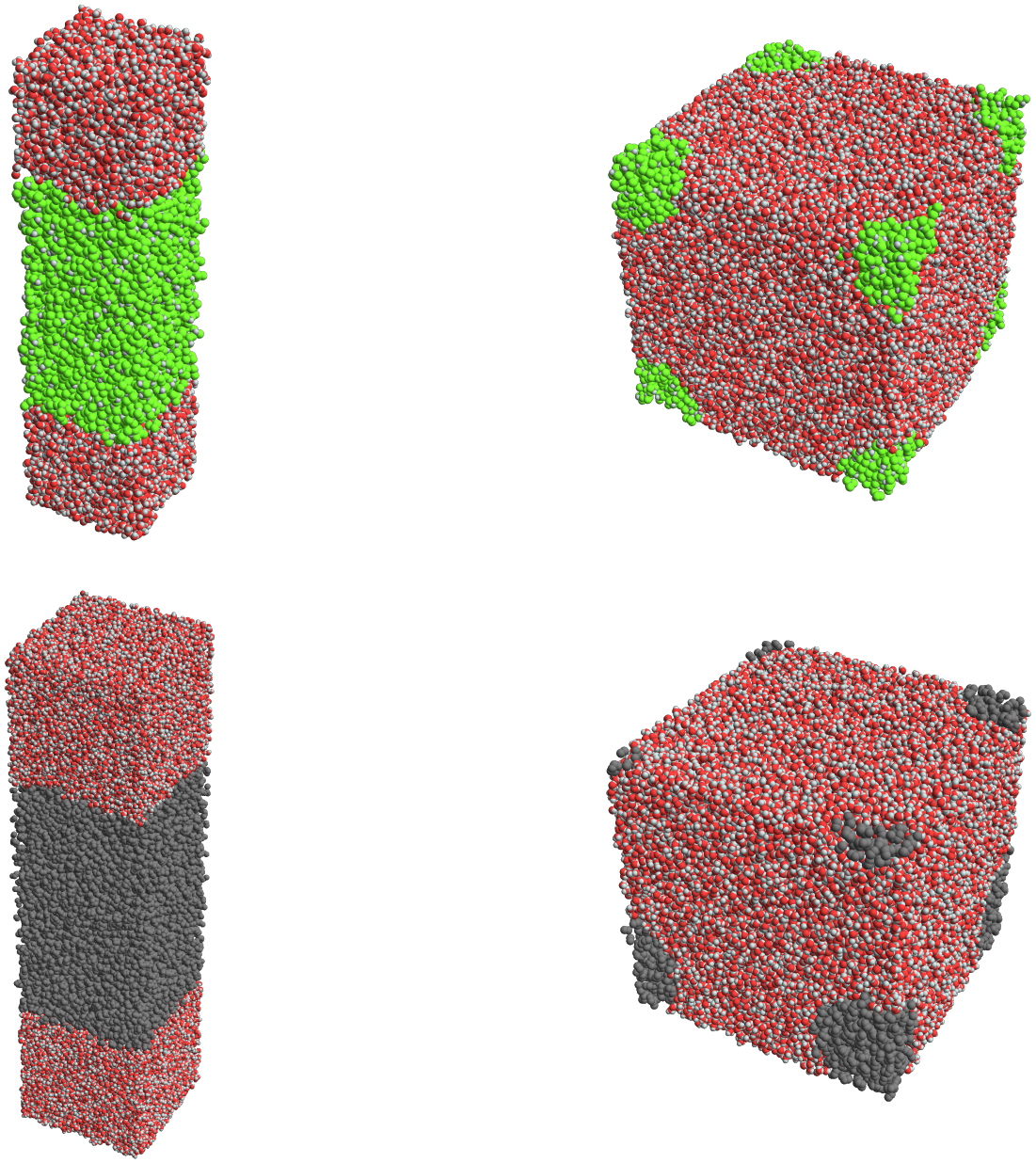


Figure S1: Snapshots of the layer (left) and droplet (right) system setups with water-chloroform (top) and water-cyclohexane (bottom).

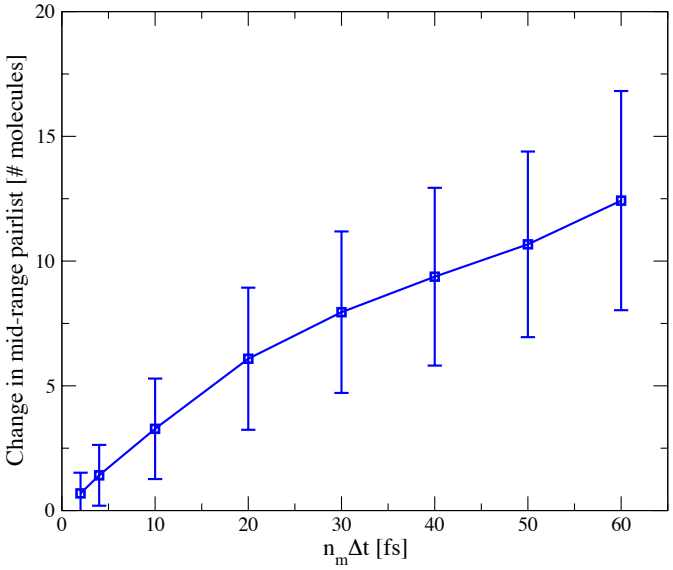


Figure S2: Mean change (in number of molecules) of the mid-range pairlist between pairlist updates (i.e. every n_m steps). A cubic SPC water box of 1000 molecules was simulated under NVT conditions using the TR CFA algorithm. The mid-range shell was bound by $R_s = 0.8$ nm and $R_l = 1.4$ nm. After 0.4 ns equilibration time, all molecule coordinates were written out every outer time step $n_m \Delta t = n_p \Delta t$ during 1000 consecutive pairlist updates. One random molecule was selected and the average difference between two consecutive pairlist updates was tracked for the mid-range pairlist. The average number of water molecules in the mid-range pairlist is 305.

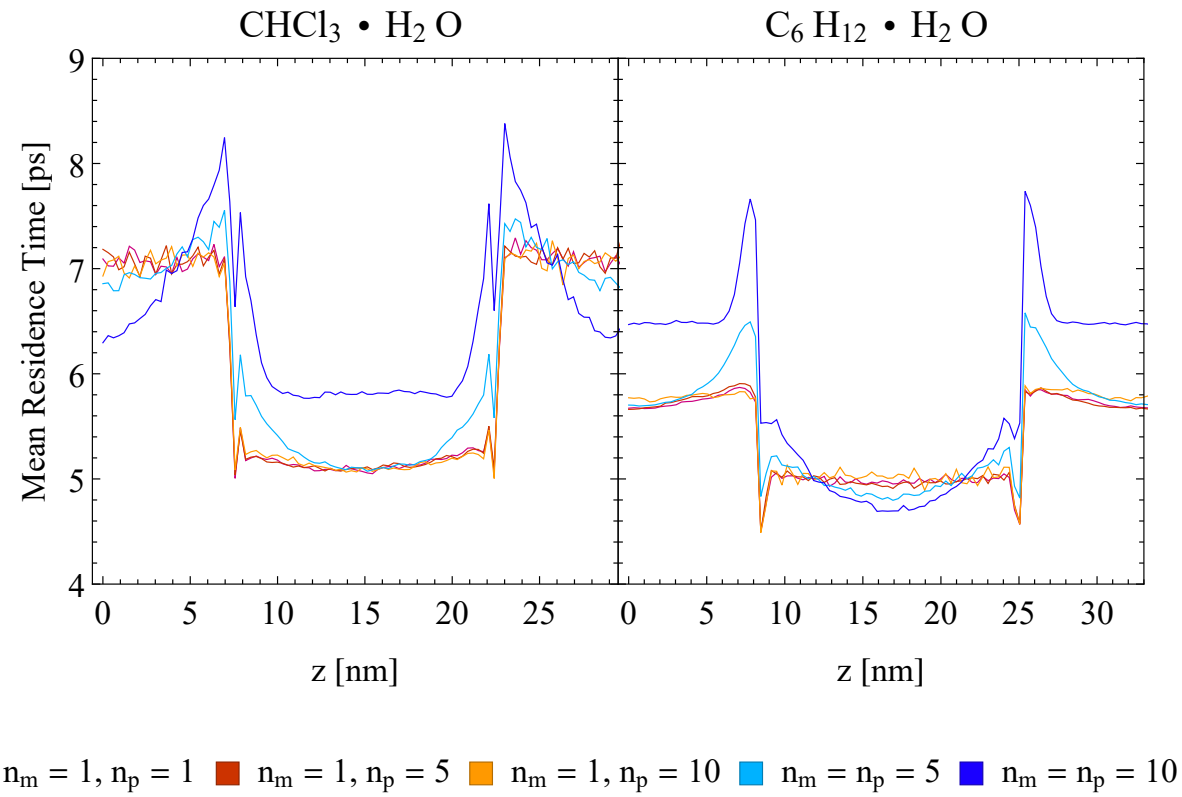
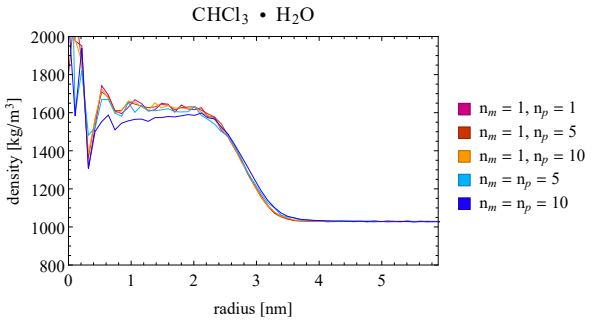


Figure S3: Mean residence time as a function of the z-coordinate for the two investigated layer systems with water-chloroform (left) and water-cyclohexane (right). 100 vertical slices of 0.3 nm width were used. Simulations were performed under NVT conditions over 1 ns with the SR scheme (red, blue and orange lines) and the TR CFA scheme (green and purple lines) with three different update frequencies ($n_p = 1, 5, 10$).

(a) $CHCl_3$ - Droplet



(b) C_6H_{12} - Droplet

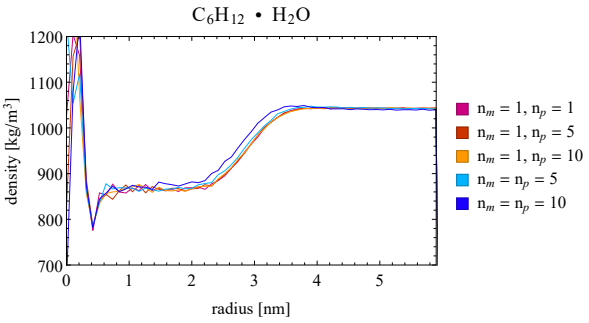


Figure S4: The density profile with respect to droplet radius is shown for the chloroform in water (left) and for the cyclohexane in water droplet (right). Note that the transition region between the two phases is broadened, in comparison with the planar systems. This broadening is caused by deviations of the droplet from its ideal spherical symmetry during runtime.