

Supporting information for “Reactive molecular dynamics study of graphene oxide sheets in different saturated states: structure, reactivity and mechanical properties”

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Reactive force field validation for water modeling

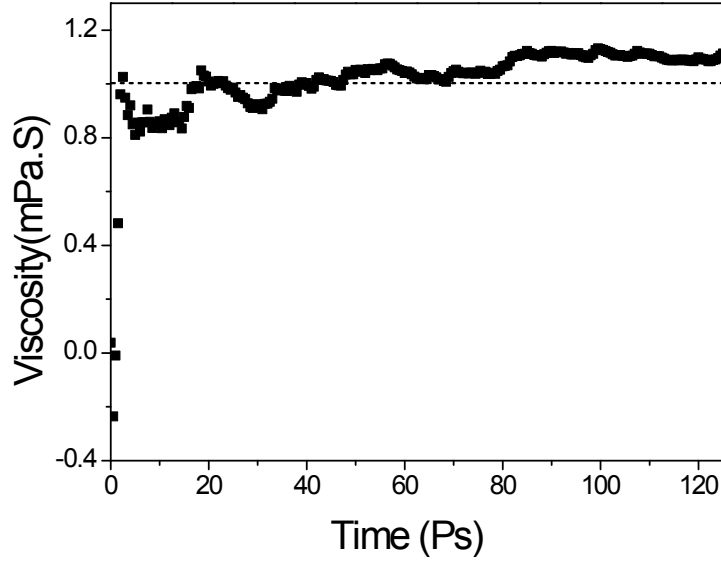
To validate the accuracy for ReaxFF in modeling water solution, reactive force field molecular dynamics was utilized to model the pure water solution. Also, based on the atomic trajectories, the viscosity, density, diffusion coefficient and resident time were calculated to compare with experimental value. The water solution model was implemented as the following procedures. First, a cubic simulation box was constructed with size of $a = b = c = 20.78 \text{ \AA}$ and 300 water molecules were randomly added to it. The water system was relaxed under NPT ensemble with temperature of 300 K and pressure of 0 atm for 200 Ps. After 300 Ps

production run, another 200 ps equilibrium run was followed under NPT ensemble for structure, dynamics and viscosity analysis. During the whole simulation process, the time step was set as 0.25 fs. The average water density is around 0.98 ± 0.02 g/cm³. This is slightly lower than the bulk water value at normal condition.

Viscosity calculation method is based on the Green-Kubo (GK) formula which relates the ensemble average of the auto-correlation of the stress/pressure tensor to eta [1] [2]. The formula is as follows:

$$\eta = \frac{V}{K_b T} \int_0^{\infty} \langle P_{\alpha\beta}(0) P_{\alpha\beta}(t) \rangle dt \quad (1)$$

where $P_{\alpha\beta}$ denotes the diagonal elements of instantaneous stress tensor, K_b is Boltzmann constant, V and T represent the volume and temperature, respectively. As shown in Figure. S1, with increasing of simulation time, the viscosity gradually increases and reaches relative constant value in the range from 1.08 to 1.14 mPa · S, which is consistent with the experimental value of 1.02 mPa · S . [3] [4]

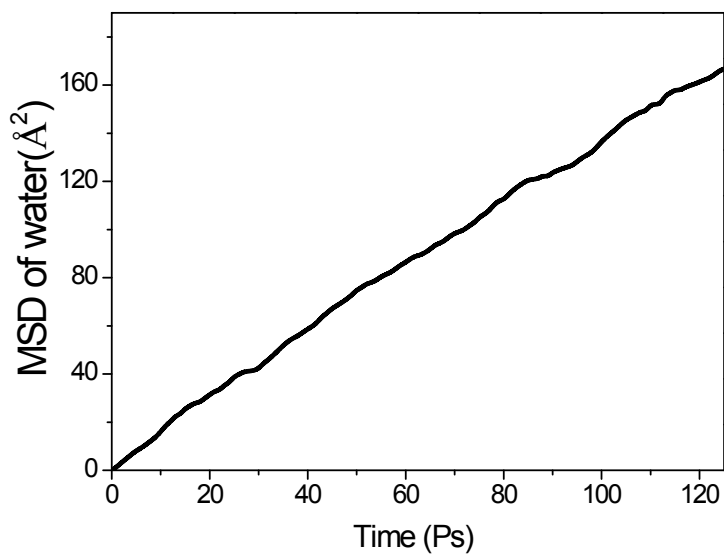


S1. Viscosity evolution as the function of time

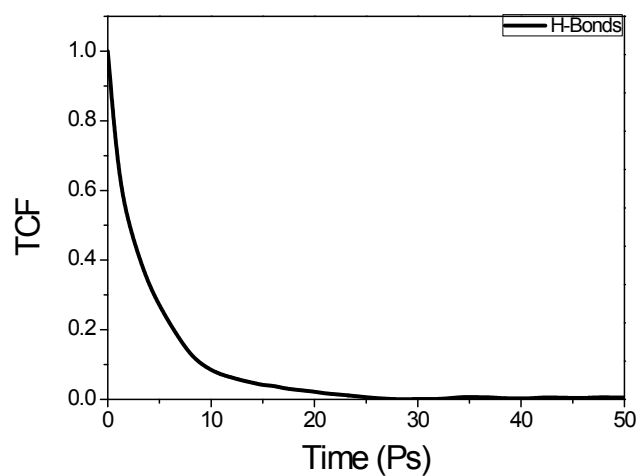
Mean squared displacement (MSD) is utilized to evaluate the dynamics properties of water molecules and is defined by the following function.

$$MSD = \langle |r_i(t) - r_i(0)|^2 \rangle \quad (2)$$

Where $r_i(t)$ represents the position of the i atom at time t , $r_i(0)$ represents the starting position of the i atom. The diffusion coefficient for water molecules is obtained by linearly fitting the MSD (t) in the diffusive regime in Figure S2. The calculated diffusion coefficient ($2.21 \pm 0.09 \times 10^{-9} \text{m}^2/\text{s}$) is consistent with the experimental value for bulk water $2.299 \times 10^{-9} \text{m}^2/\text{s}$. [5]



S2. Mean square displacement evolution as the function of time



S3. Time correlated function for the inter-molecule H-bond

The time correlated function (TCF) for H-bonds has been calculated and shown in Figure S4. By integrating the TCF during the simulation time, the resident time (τ_{res}) for ions and water can be obtained by the following equation:

$$\tau_{res} = \int_0^{\infty} C(t)dt \quad (3)$$

The resident time is utilized to evaluate the life time for the H-bond between neighboring water molecules. This reflects the stability for the H-bond stability. The hydrogen bond has a resident time of 4.57 Ps, which matches well with previous experimental finding [6].

References

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