

Supporting Information

The Interfacial-Organized Monolayer Water Film (MWF) Induced “Two-Step” Aggregation of Nanographene: Both in Stacking and Sliding Assembly Pathways

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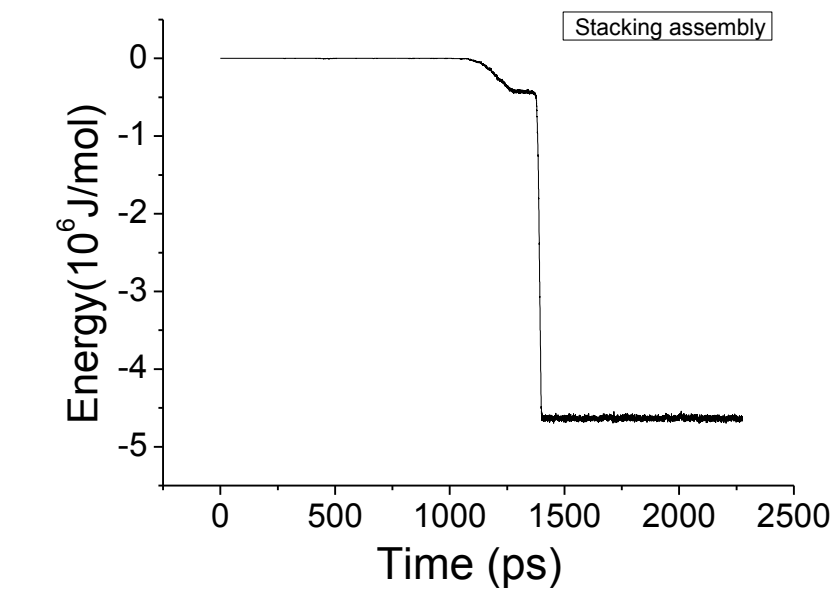
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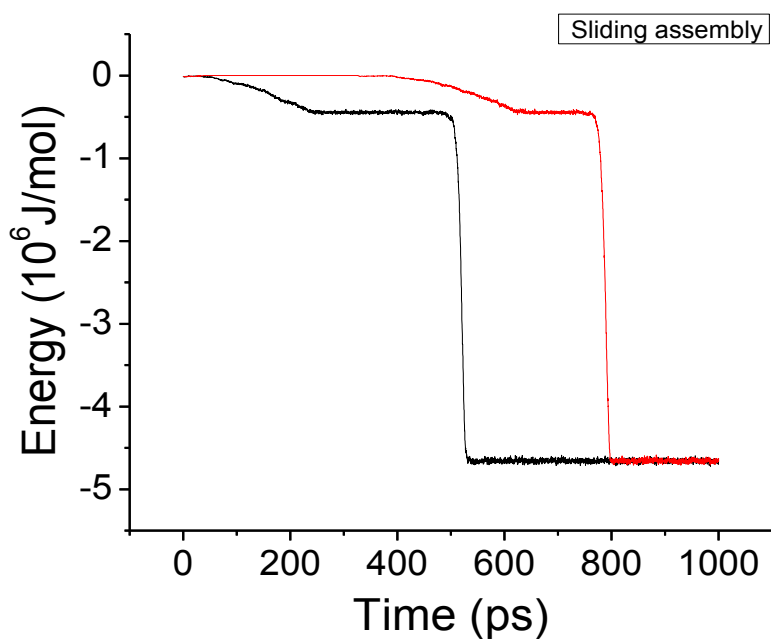
The evolution of interaction energy for two graphene nanosheets assembly in stacking (a) and sliding (b) pathway was plotted in **Figure S1**. The time evolution of three dimension distance for stacking assembly of two graphene nanosheets with the edge-edge orientation of 45° was plotted in **Figure S2**. The initial orientations of graphene nanosheets in three simulations (edge-edge distance in x-direction (d_x) was 0.3 nm, but in z-direction (d_z) was 0.0 nm, 0.4 nm and 0.7 nm, respectively) were shown in **Figure S3**. The snapshots of the evolution of hydration shells during the sliding assembly of nanographenes were shown in **Figure S4**, with the separation of two graphene nanosheets in z-direction is (a) 0 nm and (b) 0.7 nm, respectively.

The process of two graphene nanosheets assembly in stacking pathway was

shown in **Movie S1** as video. The process of two graphene nanosheets (with a separation of 0.7 nm in normal direction) assembly in sliding pathway was shown in **Movie S2** as video. The dynamical evolution of interfacial water during the sliding assembly of nanographene was shown in **Movie S3** as video. The process of extruding the monolayer water film (MWF) out of the interplate of two graphene nanosheets was shown in **Movie S4** as video. **Movie S5** displays that the graphene–water–graphene sandwiched structure was successfully maintained during a 10 ns MD simulation.



(a)



(b)

Figure S1. The interaction energy vs. the time for the assembly of two graphene in (a) stacking pathway and (b) sliding pathway (two lines represent two repeated simulations).

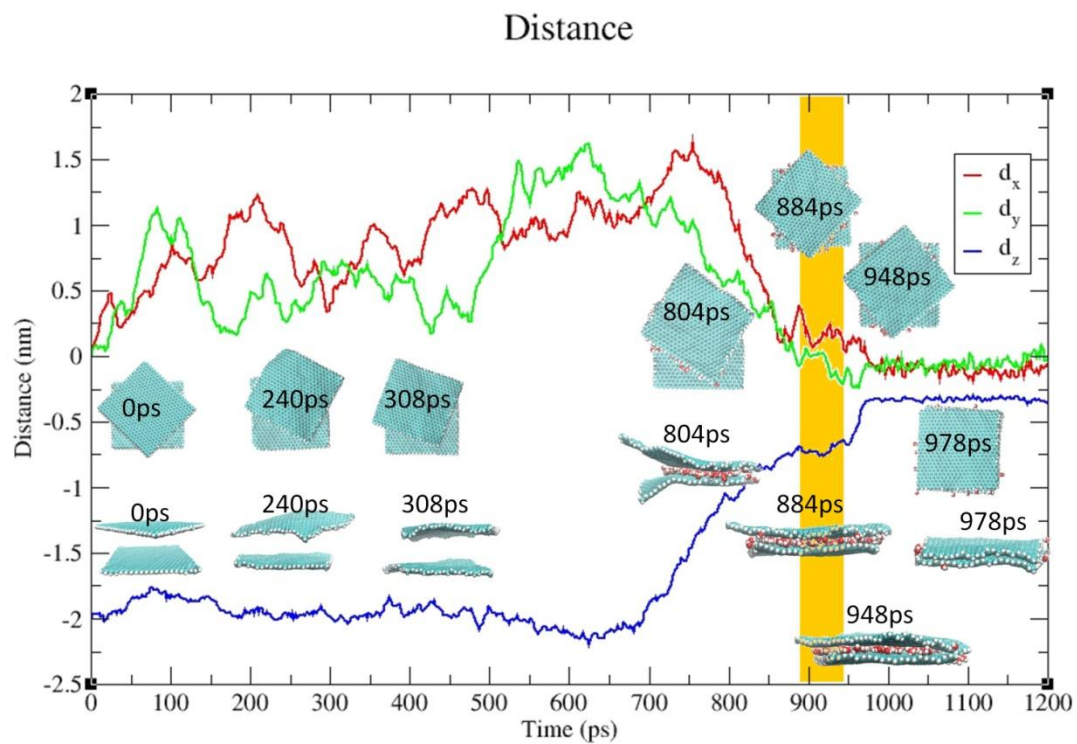


Figure S2. The time evolution of three dimension distance for stacking assembly of two graphene nanosheets with the edge-edge orientation of 45° . The critical conformation snapshots during stacking assembly were plotted as insets. The width of orange band indicates the duration of graphene-water-graphene sandwiched structure (GWGSS).

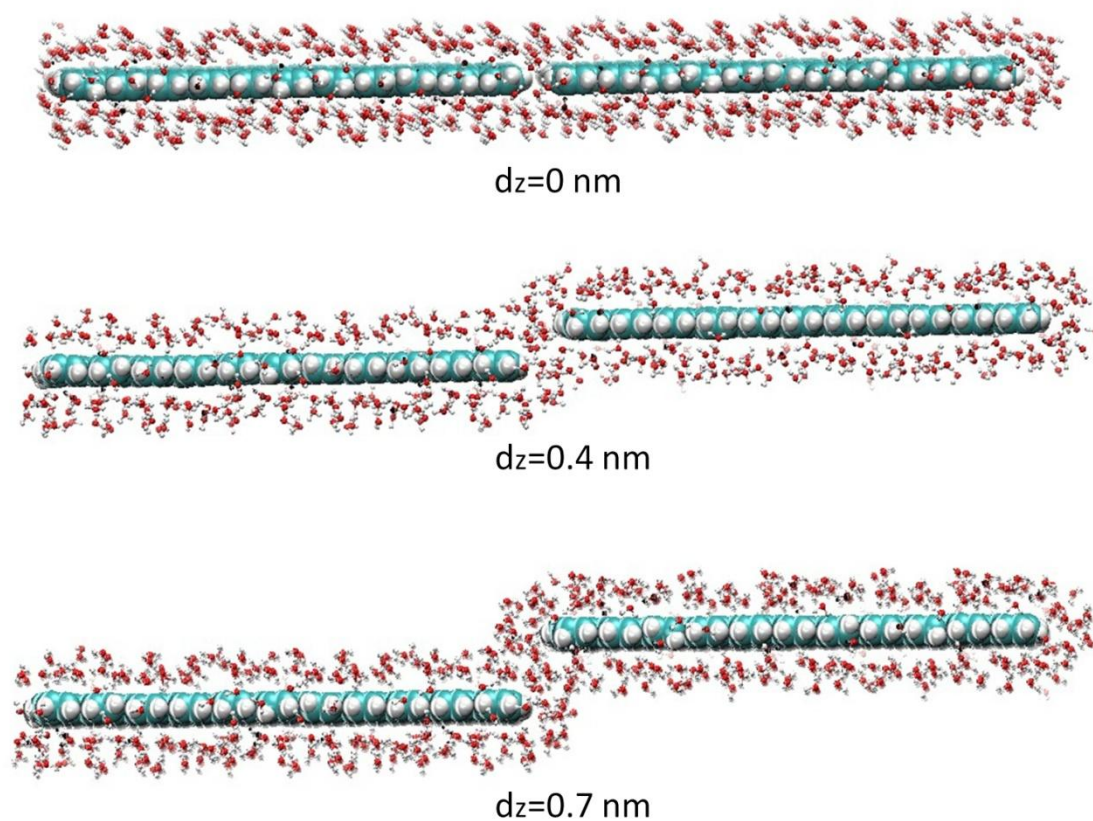


Figure S3. The initial orientations for two graphene nanosheets with different interfacial water in three simulations. Only the interfacial water (within 0.5 nm of graphene) was presented.

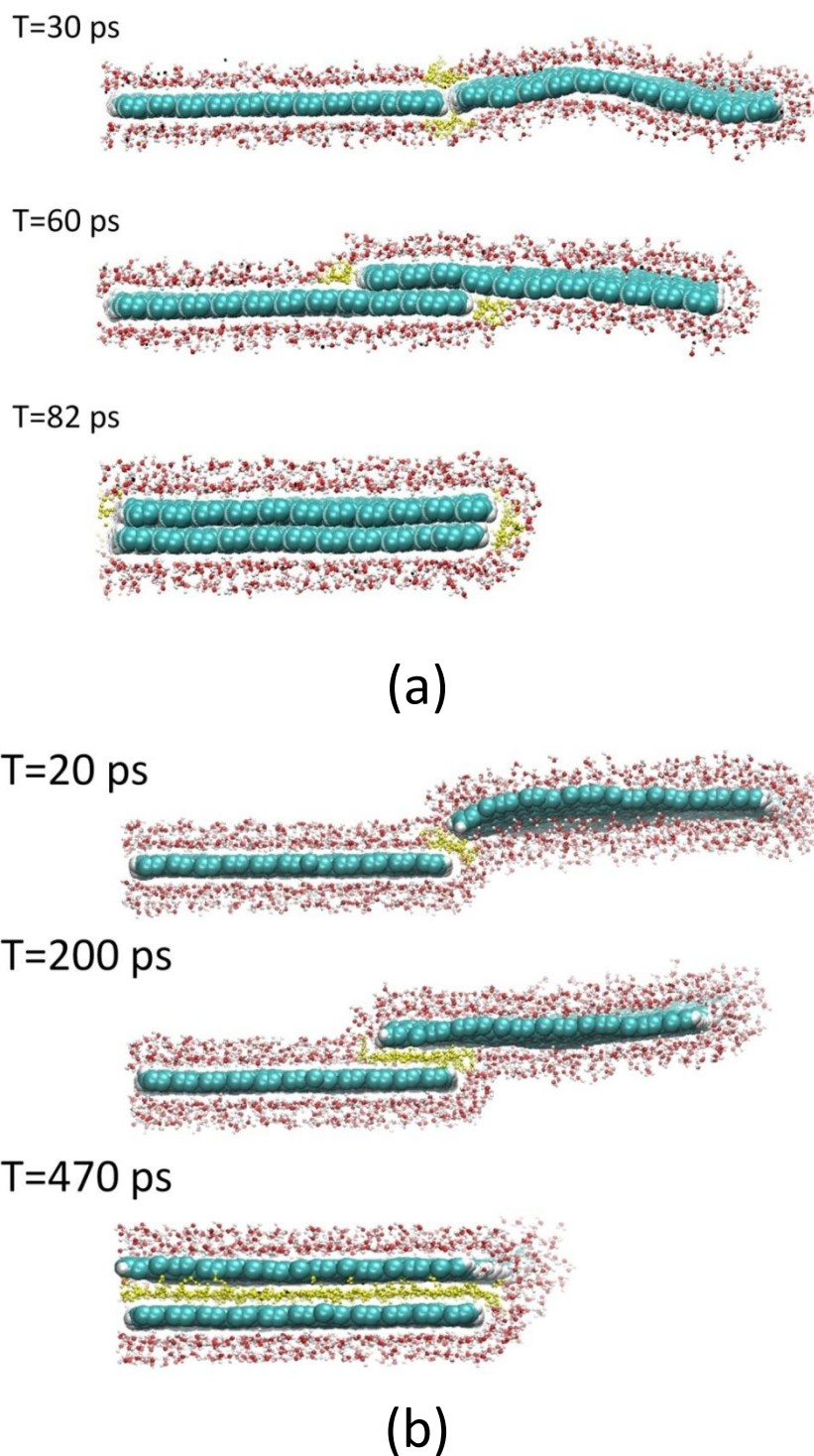


Figure S4. The evolution of hydration shells during sliding assembly of two nanographenes with the separation of two graphene nanosheets in z-direction of (a) 0 nm and (b) 0.7 nm. The interfacial water (within 0.6 nm to graphene nanosheets) is drawing in “CPK model”, and the hindering water at the contact region (within 0.5 nm to both graphene nanosheets) is highlighted in yellow; the graphene nanosheets are drawing in “vdW model”, colored in cyan.