Formation of water bilayer on graphene surfaces

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Water wettability of graphitic surfaces is of great interest to fundamental understanding of graphene surfaces and for practical applications of graphene-based materials since the wettability is closely related to whether the surface is hydrophobic or hydrophilic. One of the characteristic measures of the wettability is a contact angle that is the angle of the edges of a water droplet placed on target surfaces. While graphitic surfaces are believed to be hydrophobic, in recent studies, it has been reported that the water contact angle of the graphene surfaces becomes relatively small by removing surface contamination [1]. This observation is also confirmed by evaluating the water contact angle with molecular dynamics (MD) simulations [2]. The wettability of pristine graphene surfaces is controversial.

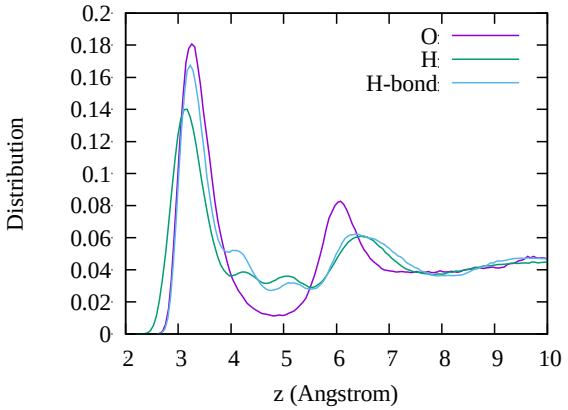

Fig 1. The density distribution of oxygen atoms, hydrogen atoms, and hydrogen bonds of water as a function of the distance z from the graphene surface.

We have examined the structure of water molecules on graphene surfaces with MD simulations. As the number of water molecules increases, the droplet covers the surface and the layered-structure of molecules on the surface is formed. This indicates that, at the level of an empirical model, a graphene surface is capable of wetting.

The formation of water layers on the surface can be seen by the density profile of water molecules. Figure 1 shows the probability density distribution of oxygen atoms, hydrogen atoms, and hydrogen bonds of water along the normal direction to the graphene surface. For the oxygen atoms, the density distribution has two peaks, indicating the bilayer of water molecules is definitely formed. Moreover, the distinct peaks of the hydrogen atoms and the hydrogen bonds distributions indicate that the oxygen-hydrogen (O-H) bonds are not randomly formed but are certainly oriented. Our analyses on the angle distribution of O-H bonds show that hydrogen bonds are formed within and between the water layers. Above the second layer, there are no dangling hydrogen bonds that are pointing perpendicular to the layer plane, which means that a graphene surface covered with the water double-layer has a hydrophobic character.

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