Designing novel carbon nanostructures for hydrogen storage

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Pillared graphene provides a stable architecture for enhanced fuel storage.

Energy consumption has reached record levels, and global demand is expected to grow by more than half over the next quarter of a century. The greenhouse effect and global warming are only two of the issues we face. In addition, fossil fuel reserves are gradually being depleted. To address these problems, we need a new, clean energy source. Hydrogen is an ideal environmentally friendly energy carrier, since the only product from its combustion is water. The main drawback limiting its wide use is the lack of an efficient storage device.

The United States Department of Energy (DOE) has established targets to be met by 2010 in order to use hydrogen as a fuel for mobile applications. Nanoporous carbon materials, like carbon nanotubes (CNTs), were initially considered ideal candidates for hydrogen storage. However, later work showed that pristine CNTs cannot store sufficient amounts of hydrogen under ambient conditions. On the other hand, doping CNTs with lithium atoms can considerably increase their capacity. Efficient storage also requires a material with high surface area and suitable pores. To fulfill these requirements, we designed pillared graphene.

As shown in Figure 1, pillared graphene is the combination of two allotropes of carbon, CNTs and graphene sheets. The entire structure looks like a building in the early stages of construction, with CNTs forming the pillars and graphene sheets forming the floors. The combined 3D material has tunable pores, in which the length, width, and intertube distance of the CNTs can be changed at will. Tunable porosity is crucial for efficient hydrogen storage.

To examine the stability of the material, we performed calculations from first principles. Optimization of the building unit without symmetry constraints showed that pillared graphene is thermodynamically stable. This conclusion was also supported by the fact that its construction is based on Euler’s generalized rule for polygons. According to this rule the number of faces, F, vertices, V, edges, E, and genus, G, obey the equation \( 6(E - F - V) = 12(G - 1) \). For a closed surface like ours, the bond surplus at the junction should be six. This bond surplus is provided by six heptagons at the junction between the CNT and the graphene sheet.

Next, we performed Monte Carlo calculations at several thermodynamic conditions to evaluate the hydrogen-storage capacity of pillared graphene. We expected and observed a

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trend similar to those known for carbon-based materials. But when we doped our material with lithium atoms, its storage capacity increased dramatically. The increase was so high that pillared graphene’s capacity far exceeds DOE targets at cryogenic conditions and roughly reaches the targets under ambient conditions. The saturation is reached at very low pressure, because we can tune the structure to the proper pore size distribution.

Hydrogen is an ideal energy carrier, and its use as a fuel could offset the depletion of fossil fuels and prevent pollution. Yet it has not been widely adopted due to the lack of efficient storage materials. We developed pillared graphene, a novel 3D network nanostructure with suitable pores targeted for hydrogen storage. Furthermore, its tunable porosity suggests that pillared graphene may be ideal for other applications, like variable heat transfer. The theoretical demonstration challenges experimentalists to take the next steps of fabricating this material and validating its storage capacity.

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References