

First-Principles Study of Hydrogen Storage on Li₁₂C₆₀Qiang Sun,^{*,†,‡} Puru Jena,[‡] Qian Wang,[‡] and Manuel Marquez[§]

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Abstract: Solid state materials capable of storing hydrogen with high gravimetric (9 wt %) and volumetric density (70 g/L) are critical for the success of a new hydrogen economy. In addition, an ideal storage system should be able to operate under ambient thermodynamic conditions and exhibit fast hydrogen sorption kinetics. No materials are known that meet all these requirements. While recent theoretical efforts showed some promise for transition-metal-coated carbon fullerenes, later studies demonstrated that these metal atoms prefer to cluster on the fullerene surface, thus reducing greatly the weight percentage of stored hydrogen. Using density functional theory we show that Li-coated fullerenes do not suffer from this constraint. In particular, we find that an isolated Li₁₂C₆₀ cluster where Li atoms are capped onto the pentagonal faces of the fullerene not only is very stable but also can store up to 120 hydrogen atoms in molecular form with a binding energy of 0.075 eV/H₂. In addition, the structural integrity of Li₁₂C₆₀ clusters is maintained when they are allowed to interact with each other. The lowest energy structure of the dimer is one where the Li atom capped on the five-member ring of one fullerene binds to the six-member ring of the other. The binding of hydrogen to the linking Li atom and the potential of materials composed of Li₁₂C₆₀ building blocks for hydrogen storage are discussed.

Hydrogen has been recognized as an ideal energy carrier and has the potential to reduce our dependence on fossil fuels which are not only limited but also are harmful to the environment.^{1–4} The success of a new hydrogen economy, however, depends on the ability to find materials that can store hydrogen reversibly with high gravimetric and volumetric density and operate under moderate temperatures and pressures. Although a great deal of effort has recently been devoted to searching for such materials, none are known to meet the above conditions.^{5–11} The difficulty lies in tailoring the chemistry of hydrogen as it interacts with materials.

Hydrogen molecules are known to interact with materials in one of two ways: Hydrogen is either physisorbed in molecular form or chemisorbed in atomic form. In the former case no transfer of charge takes place between the surface and the hydrogen, resulting in weak binding. In the latter case, transfer of electrons from the surface to the antibonding orbital of the

H₂ breaks its molecular bond and atomic adsorption ensues, resulting in strong bonding. Thus, there are two classes of materials: one where large amounts of hydrogen can be stored, but it is difficult for hydrogen to desorb (e.g., CH₄), and the other where hydrogen can desorb easily, but not much of it can be stored (e.g., carbon nanotubes).^{12–14} An ideal storage system would be one where hydrogen binds molecularly but with a binding energy that is intermediate between the physisorbed and chemisorbed state. We show that coating of C₆₀ fullerenes with suitable metal atoms may lead to the synthesis of novel hydrogen storage materials. In particular, we show that the unusual ability of Li₁₂C₆₀ to bind 60 hydrogen molecules stems from the unique chemistry at the nanoscale.

To demonstrate this chemistry we recall the results of some earlier calculations.¹⁵ It was shown that a neutral Li atom is able to bind only *one* hydrogen atom with a binding energy of 2.49 eV, while a Li⁺ ion can bind at least 12 hydrogen atoms in molecular form. The binding energy of the first H₂ molecule to Li⁺ ion is 0.253 eV and decreases only slightly as more H₂ molecules are bound. For example, the binding energy of the sixth H₂ molecule to a Li⁺ ion is 0.202 eV. This “insensitivity” in the binding energies as a function of number of adsorbed H₂ molecules is a consequence of the mechanism that binds the hydrogen. The positive charge of the cation polarizes the hydrogen molecule, and the bonding results from an electrostatic

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interaction. This mechanism has also been demonstrated for transition metal ions where the binding of hydrogen to the metal ion is even larger, namely about 0.5 eV/H₂ molecule.¹⁶ Thus, the key to molecular adsorption of hydrogen with binding energies intermediate between physisorbed and chemisorbed states is to find a system where the metal atom can remain in a positively charged state.

Our rationale for studying Li-coated C₆₀ is two-fold: (i) Since the C₆₀ fullerene has a large electron affinity (2.66 eV) which is comparable to that of Cl (3.62 eV), the Li atom would partially donate its valence electron to the fullerene resulting in strong bonding. The charge transfer from Li to the fullerene cage would leave the Li atom in a cationic state which can then bind hydrogen in a molecular form due to the polarization mechanism^{15,16} discussed above. Since this bonding does not require charge transfer, the amount of hydrogen that can be stored on a Li₁₂C₆₀ is limited mainly by steric hindrance. Recently three theoretical groups^{17–19} have studied the possibility that Sc-, Ti-, and Ni-coated carbon fullerenes and nanotubes may be able to store large amounts of hydrogen. They have indeed found that hydrogen binds in molecular form with a binding energy of the order of 0.5 eV/H₂ molecule and with a gravimetric density of 6.8–9.0 wt %. In these calculations the authors, however, assumed that the metal atoms remain isolated. We have recently shown this not to be the case.²⁰ In fact it is energetically favorable for Ti atoms to cluster on C₆₀, thus resulting in a significant reduction in the weight percentage of stored hydrogen. The situation in Li₁₂C₆₀, however, is expected to be different. The cohesive energy of Li is substantially smaller than those of transition metals, and Li atoms bind more strongly to C than to itself. Thus, Li atoms are not likely to form clusters on the fullerene surface. (ii) Experiments based on mass spectroscopy of Li-decorated C₆₀ fullerene have shown that Li₁₂C₆₀ is a very stable cluster.²¹

Our density functional calculations fully support the above conjectures. We find that the binding energy of a Li atom to the five-member ring of C₆₀ is 1.80 eV which is larger than the cohesive energy of bulk Li, namely 1.63 eV. In the equilibrium geometry of Li₁₂C₆₀, the 12 Li atoms remain isolated and bind on the pentagonal rings. The Li atom in the Li₁₂C₆₀ cluster carries a positive charge of +0.5/Li, and 60 H₂ molecules can be bound to the Li₁₂C₆₀ cluster with the total adsorption energy of 4.5 eV, corresponding to 0.075 eV/H₂ molecule. Note that the room temperature corresponds to 0.026 eV. In the following we discuss our results in detail.

The above results are based on first principles spin polarized calculations using density functional theory with generalized gradient approximation for exchange and correlation. To calculate the optimized structure of Li₁₂C₆₀ with and without adsorbed hydrogen, we used a supercell approach where the cluster was surrounded by 15 Å of vacuum space along *x*, *y*, and *z* directions. The Γ point is used to represent the Brillouin

zone due to the large supercell. The total energies and forces and optimizations of geometry were carried out using a plane-wave basis set with the projector-augmented plane wave (PAW) method as implemented in the Vienna Ab initio Simulation Package (VASP).²² The PW91 form was used for the generalized gradient approximation to exchange and correlation potential. The geometries of clusters were optimized without symmetry constraint using a conjugate-gradient algorithm. The energy cutoff and the convergence in energy and force were set to 400 eV, 10⁻⁴ eV, and 1 × 10⁻³ eV/Å, respectively. The accuracy of our numerical procedure for C₆₀ and hydrogen was well tested in our previous papers.^{20,23} For the Li₂ molecule, the bond length and binding energy are found to be 2.71 Å and 0.95 eV, in good agreement with experimental values (2.67 Å and 1.03 eV).²⁴ For the LiH molecule, the corresponding values are 1.61 Å and 2.38 eV, which also agree well with the experimental values of 1.60 Å and 2.50 eV.²⁵

We begin with the results of a single Li atom interacting with C₆₀. We studied the relative stability of the Li atom bound on the pentagonal and hexagonal rings and found that the binding energies for these two sites are nearly degenerate. However, in agreement with previous work,²¹ we find that as more Li atoms are attached to the fullerene, the pentagonal face clearly becomes the preferred site. In Figure 1, we show (a) the geometry of Li situated on top of the pentagonal face, (b) the geometry of the adsorbed hydrogen molecule, and (c) the valence charge density distribution.

When one Li atom is capped on one of the pentagons, the C–C bond length in the five-member ring is slightly enlarged from 1.457 to 1.461 Å and the Li–C distance is 2.229 Å. The charge transfer from Li 2s to the fullerene cage makes the Li atom positively charged resulting in the interaction energy of 1.80 eV. When one H₂ molecule is introduced to this system, due to the polarization interaction between the charged Li atom and the H₂, the H–H bond length expands slightly from 0.740 Å to 0.755 Å (Figure 1b). The equilibrium Li–H distance is found to be 2.070 Å, which is very close to the value (2.040 Å) found in the case of a free Li⁺ ion interacting with H₂.¹⁵ Because of the presence of H₂, the Li–C distance changes only a little, namely, 2.236 Å, but the C–C bond length in the five-member ring remains unchanged. The binding energy of H₂ to LiC₆₀ is found to be 0.18 eV, which is comparable to that in Li⁺H₂, namely 0.25 eV.¹⁵ In addition, the Li–H and H–H bonds in LiC₆₀ cluster in Figure 1 are, respectively, 2.07 Å and 0.755 Å. In the Li⁺H₂ cluster these distances are, respectively, 0.75 Å and 2.04 Å. This close agreement suggests that, in the LiC₆₀ cluster, Li remains as a positively charged ion. This is also seen from the valence charge density distribution given in Figure 1c. We see that because of the charge transfer to the fullerene cage, the valence charge around the Li atom is not visible. In fact, the Mulliken charge analysis²⁶ indicates that the Li carries a charge of +0.75, suggesting that the bonding feature between Li and C₆₀ has significant ionic character. On the other hand,

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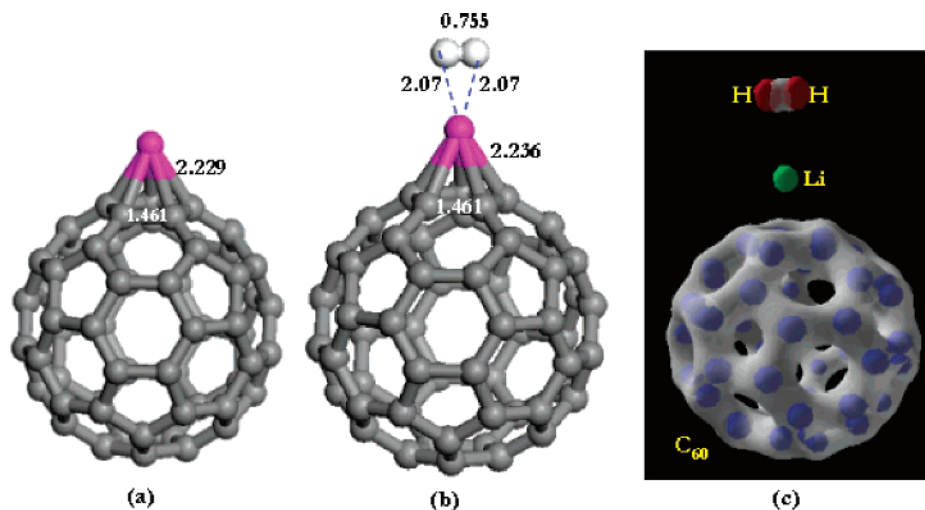


Figure 1. Geometries of (a) LiC_{60} and (b) $\text{LiC}_{60}\text{H}_2$. The valence charge density distribution (in white) in $\text{LiC}_{60}\text{H}_2$ is given in (c).

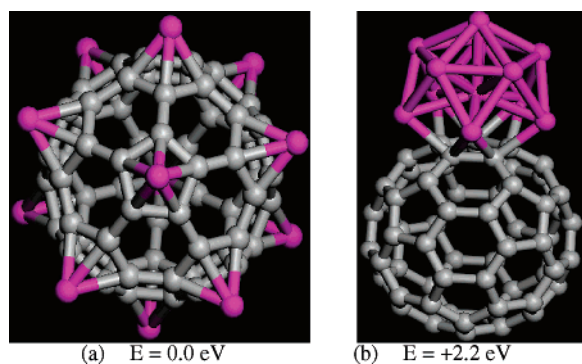


Figure 2. Two configurations of $\text{Li}_{12}\text{C}_{60}$: (a) Li atoms are kept isolated and (b) Li atoms are allowed to cluster. The relative energy ΔE is evaluated referring to configuration (a).

the interaction between H–H and C–C remains covalent as the charges are concentrated on the bonds between H–H and C–C.

Now we discuss the structure and properties of $\text{Li}_{12}\text{C}_{60}$. As stated above, the binding energy of a Li atom to C_{60} (1.80 eV) is larger than that in the Li_2 dimer (0.95 eV) and the Li bulk cohesive energy (1.63 eV). Thus at low Li coverage, the clustering of Li atoms would not be expected. In fact, for $\text{Li}_{12}\text{C}_{60}$, the isolated configuration (Figure 2a) is 2.2 eV lower in energy than the clustered configuration (Figure 2b). In the stable structure, 12 Li atoms remain isolated on the 12 pentagons resulting in a high symmetric geometry. This is also in agreement with previous studies.²¹ The average binding energy per Li atom with C_{60} in $\text{Li}_{12}\text{C}_{60}$ is 1.78 eV, which differs very little from that in LiC_{60} , namely, 1.80 eV. The Mulliken charge analysis shows that each Li atom in $\text{Li}_{12}\text{C}_{60}$ carries the charge of +0.5, very close to the Mulliken charge on Ni in Ni-C_{60} cluster.¹⁹ Thus the bonding between Li atoms and C_{60} in $\text{Li}_{12}\text{C}_{60}$ is partly ionic and partly covalent. It is worth pointing out that the equilibrium geometry of $\text{Li}_{12}\text{C}_{60}$ is totally different from that of $\text{Ti}_{12}\text{C}_{60}$ where clustering of Ti was shown to lower the energy.²⁰

We now enter the next phase of our calculation, namely, to study the possible number of hydrogen atoms that can be bound to the $\text{Li}_{12}\text{C}_{60}$ structure and the nature of hydrogen bonding. It is known that four hydrogen molecules can be bound to each transition metal atom monodispersed on a C_{60} fullerene. Since

a Li ion is much smaller in size than a transition metal ion, we began by studying the structure and stability of 60 hydrogen molecules—5 molecules for each Li atom in $\text{Li}_{12}\text{C}_{60}$. The starting configuration for geometry optimization was taken by attaching 60 hydrogen molecules around Li atoms above the 12 five-member rings, as shown in Figure 3a. The resulting optimized structure is given in Figure 3b. Here we note that hydrogen atoms remain molecular with a bond length of 0.753 Å. The total interaction energy of 60 H_2 molecules with $\text{Li}_{12}\text{C}_{60}$ is 4.5 eV which yields an average binding energy of 0.075 eV/ H_2 molecule. We want to emphasize that the sequential binding energy of a H_2 molecule to $\text{Li}_{12}\text{C}_{60}$ is expected to be a slowly varying function as was seen in the case of the $\text{Li}^+(\text{H}_2)_n$ cluster.¹⁵ The maximum energy needed to remove a H_2 molecule from $\text{Li}_{12}\text{C}_{60}$ is 0.18 eV, and the minimum energy to remove a H_2 molecule is 0.06 eV. Although these binding energies are significantly smaller than that in Li^+H_2 , namely, 0.253 eV, they are much above that at room temperature (26 meV). The gravimetric density of H stored on $\text{Li}_{12}\text{C}_{60}$ is 13 wt %, and the volume density can be estimated by calculating the volume of the $\text{Li}_{12}\text{C}_{60}\text{H}_{120}$ cluster in Figure 3b which has a radius of 7.2 Å. The volumetric density of 128 g/L is then obtained by dividing the mass of 60 H_2 molecules with the above volume.

While the above results are promising for an isolated $\text{Li}_{12}\text{C}_{60}$ cluster, one has to wonder about its potential as a hydrogen storage material. For example, is it possible to synthesize a cluster assembled material composed of $\text{Li}_{12}\text{C}_{60}$ clusters as building blocks where the structural identity of individual $\text{Li}_{12}\text{C}_{60}$ clusters remain? If so, do the clusters retain their original capacity to store hydrogen? Although the eventual structure of a bulk material composed on $\text{Li}_{12}\text{C}_{60}$ is difficult to determine, we should note that the Li atoms on the fullerene are positively charged and as two $\text{Li}_{12}\text{C}_{60}$ clusters are brought into the vicinity of each other, the repulsion between Li atoms may prevent their coalescence. Hence a solid material composed of $\text{Li}_{12}\text{C}_{60}$ may be possible just as a solid made of zintl ions is known to exist.²⁷ However, it is also likely that the Li atom capped onto a pentagon face of one C_{60} may bind with the hexagon face of the other and lead to some structural distortion.

To address these questions, we studied the interaction between two $\text{Li}_{12}\text{C}_{60}$ clusters in two steps. First, the geometry optimiza-

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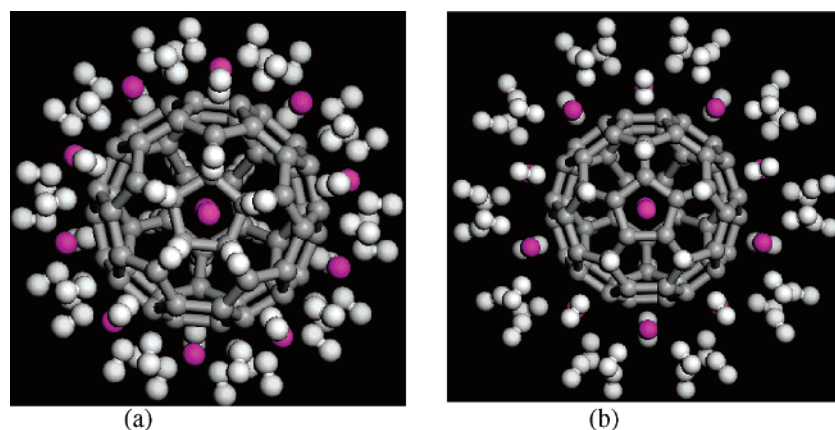


Figure 3. (a) Initial and (b) optimized geometry of $\text{Li}_{12}\text{C}_{60}(\text{H}_2)_{60}$.

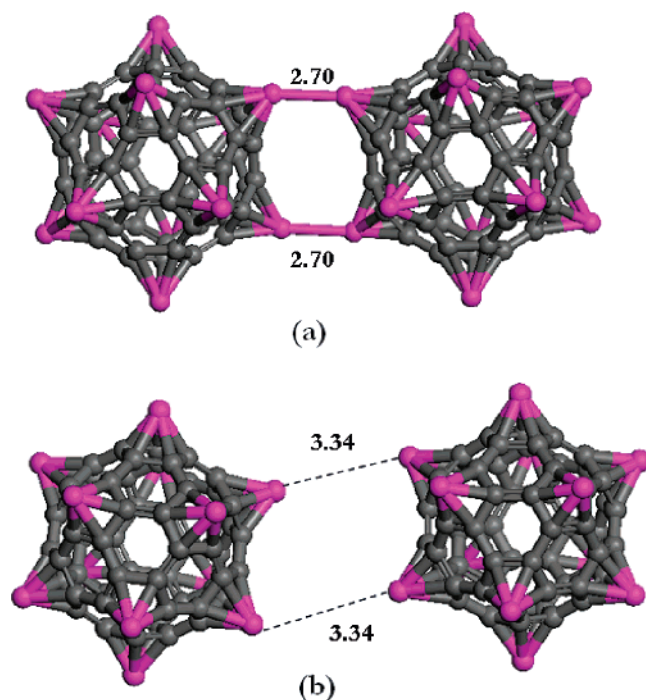


Figure 4. Initial geometry (a) and fully optimized geometry (b) for dimer with the interaction energy 0.4 eV in the final configuration.

tion of a $(\text{Li}_{12}\text{C}_{60})_2$ dimer was performed by starting with an initial configuration where the distance between two Li atoms was set to 2.70 Å, which is the equilibrium bond length of the Li_2 dimer (see Figure 4a). However, after full symmetry unrestricted geometry optimization, we found the structure of the $\text{Li}_{12}\text{C}_{60}$ dimer to be that in Figure 4b. Note that the structures of the individual $\text{Li}_{12}\text{C}_{60}$ clusters are unchanged from that in Figure 2a. However, due to the repulsion between the like charges, the two clusters rotated and moved away from each other. In the equilibrium configuration, the distance between the two Li atoms is 3.34 Å and the binding energy of the $\text{Li}_{12}\text{C}_{60}$ dimer is 0.40 eV. Thus, $\text{Li}_{12}\text{C}_{60}$ clusters not only not coalesce but also maintain their structural identity. If the structure of a bulk material composed of $\text{Li}_{12}\text{C}_{60}$ units would be such that the Li atoms remain far apart, the hydrogen storage capacity of the bulk material may not be lowered too much from that of the isolated cluster.

Second, we started with an initial configuration where the Li atom capping the pentagonal face of a C_{60} interacts with the

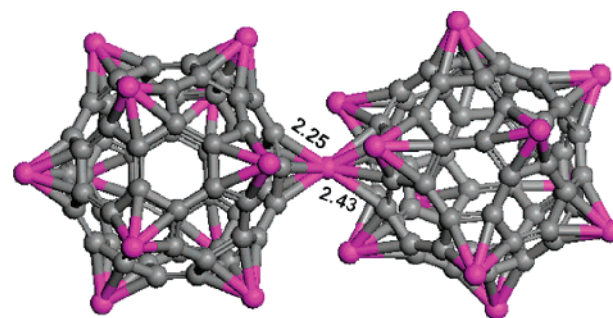


Figure 5. Optimized geometry for dimer with the hollow site configuration. The interaction energy is 1.18 eV.

hexagonal face of the other $\text{Li}_{12}\text{C}_{60}$. The resulting optimized structure is shown in Figure 5. This structure has a binding energy of 1.18 eV which is substantially larger than 0.40 eV corresponding to the structure in Figure 4b. Note that, in comparison, the binding energy of a Li atom to C_{60} is 1.68 eV. Interestingly, the integrity of the geometry of $\text{Li}_{12}\text{C}_{60}$ in the dimer remains, although there are minor changes in the bond lengths. This suggests that the $\text{Li}_{12}\text{C}_{60}$ cluster can form the building blocks of a new kind of solid similar to that of crystals of C_{60} with one major exception. In the fulleride crystal, the hexagonal faces interact with each other, while, in a material composed of $\text{Li}_{12}\text{C}_{60}$, the interaction is between the hexagonal and pentagonal faces. Thus, the crystal structure of $\text{Li}_{12}\text{C}_{60}$ is likely to be different from that of the fulleride. While it will be useful to determine the possible structure of $\text{Li}_{12}\text{C}_{60}$ cluster assembled material, computational limitations do not permit us to undertake this problem at this time.

We have, however, examined if the Li atom linking two $\text{Li}_{12}\text{C}_{60}$ clusters can bind hydrogen. First, we optimized the structure of one H_2 molecule placed in the vicinity of the linking Li atom in the $(\text{Li}_{12}\text{C}_{60})_2$ dimer (see Figure 6a). The resulting optimized structure is given in Figure 6b, where the H_2 molecule is seen to move away and bind molecularly to an adjacent Li atom. The binding energy of this H_2 molecule is 0.18 eV with a H–H bond length of 0.766 Å and Li–H bond length of 1.99 Å. Note that these values are very close to those in Figure 1b, where a single H_2 molecule interacts with a single LiC_{60} cluster. We next started with an initial configuration where five H_2 molecules were placed in the vicinity of the linking Li atom (Figure 7a). The optimized structure is shown in Figure 7b. Once again, the H_2 molecules moved away and attached to adjacent Li atoms molecularly. The binding energy of hydrogen in Figure

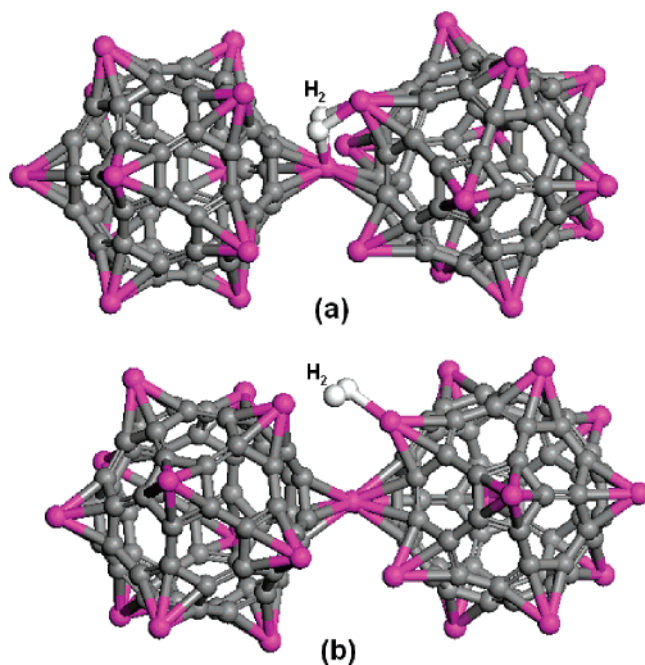


Figure 6. Initial geometry (a) and fully optimized geometry (b) for H_2 on dimer with the absorption energy 0.18 eV.

7b is found to be 0.18 eV/ H_2 molecule, and the H–H and Li–H bond lengths are, respectively, 0.765 Å and 1.98 Å. These are nearly identical with the results obtained in Figure 6b. These calculations show that although the binding of the H_2 molecule to the linking Li atom is not preferred, the binding energy of H_2 to other Li atoms is not adversely affected. Thus the ultimate hydrogen storage capacity of $\text{Li}_{12}\text{C}_{60}$ material would depend on how many Li atoms form the linkage between clusters. This is difficult to say at this stage without detailed calculations. We could visualize, just from the symmetry, that four Li atoms could form the linkage. This would lower the hydrogen storing capacity of each $\text{Li}_{12}\text{C}_{60}$ from 60 H_2 molecules to 40 H_2 molecules yielding an approximate gravimetric density of hydrogen of 9 wt %. It should also be emphasized that the binding energy of the H_2 molecules to $\text{Li}_{12}\text{C}_{60}$ is small, and the system may have to be operated at lower than ambient temperature.

In conclusion we have shown that $\text{Li}_{12}\text{C}_{60}$ is a promising system to store hydrogen in molecular form with high gravimetric and volumetric densities. Equally important is the fact that this system can also store more hydrogen per weight and volume than many of the light metal hydrides such as alanates, borohydrides, amides, and imides. However, unlike the case for these light metal hydrides, the sorption of hydrogen in $\text{Li}_{12}\text{C}_{60}$ would not require any catalysts. The stability of the storage

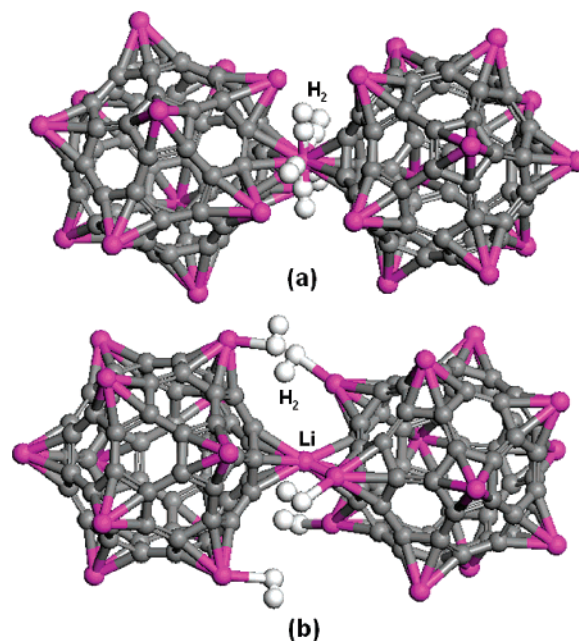


Figure 7. Initial geometry (a) and fully optimized geometry (b) for five H_2 molecules on dimer with the absorption energy 0.18 eV/ H_2 .

medium, i.e., $\text{Li}_{12}\text{C}_{60}$ following repeated hydrogen cycling is also not expected to be a problem as the Li atoms are bound strongly to the surface of C_{60} and are prevented from clustering. In addition, the structural identity of the $\text{Li}_{12}\text{C}_{60}$ cluster is maintained when allowed to interact with each other, and the dimer is found to bind within an energy range of 0.40 and 1.18 eV. However, some of the Li atoms that link $\text{Li}_{12}\text{C}_{60}$ clusters upon assembly may not be able to store hydrogen, thus lowering the gravimetric and volumetric densities calculated for the isolated cluster. In addition, it should also be emphasized that the binding energy of the H_2 molecules to $\text{Li}_{12}\text{C}_{60}$ is small, and the storage system may have to be operated at lower than ambient temperature. We believe that the predicted results are interesting enough that attempts should be made in the laboratory to study the hydrogen storing capacity of an isolated $\text{Li}_{12}\text{C}_{60}$ cluster and to assemble these clusters to synthesize bulk materials. We also hope that these results may lead researchers to a new line of thinking and to the eventual design and synthesis of ideal storage materials.

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