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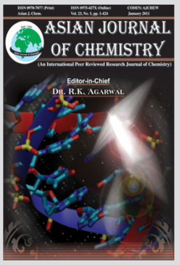

Boron Fullerene B_{80} Doped with Hydrogen and Phosphorus Atoms

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The incorporated H and P atoms in B_{80} fullerene, is examined within the first-principles density functional theory (DFT) calculations for possible applications as single quantum bits (qubits) in solid-state quantum computers as well as molecular electronics devices. It was observed atomic displacement of both H and P atoms from the center of the cage so that the wall B atoms can form covalent bonds with encapsulated atoms making them unavailable for hyperfine contact coupling to the nuclear spin. Hence, B_{80} fullerene nanocage may not be suitable for the solid-state quantum computers. However, the electronic structure for incorporated nanocages is significantly modified by the reduction of the band gap, inducing a metallic character to the encapsulated fullerene. This open the way towards the assessing the possibility to create molecular electronic nano-devices.

Key Words: B_{80} Fullerene, Quantum computers, Molecular electronics, Encapsulation, Density functional theory.

INTRODUCTION

In the last decade, interaction between fullerenes and a variety of atoms is becoming a new field of cluster research. So far, numerous theoretical and experimental studies for endohedrally doped fullerenes with foreign atoms have been undertaken[1-10]. On the other hand, the work of recent years has uncovered a large number of possible solid-state systems in which quantum computing might be achieved by using nuclear spin 1/2 donor atom dopants incorporated into fullerenes as solid-state single qubits[11,12]. Quantum computers have become well known due to the integer factoring which allow the execution of certain tasks in much fewer numbers of steps by replacing the quantum states of a two-level system (qubit) instead of ones and zeros (bits) algorithm of classical computers and have complexities much better than their classical counterparts[11,13-16]. One bottleneck in developing solid-state quantum computers is to find feasible materials that can accommodate dopant atoms. Recently, Srivastava *et al.*[11] examined the reaction of a ^{1}H atom with the internal binding sites of fullerenes *via* the *ab initio* local density functional method. They showed that, the encapsulated ^{1}H atom is stable in its atomic state at the center of the $C_{20}D_{20}$ (deuterated) fullerene[11].

Boron-based fullerenes (B_{80}) which discovered by Szwacki *et al.*[17] may also be promising solid-state quantum computer media, since they possess unusual high stability. Based on first-principles feasibility simulations, in this context the interaction of B_{80} fullerene with nuclear spin 1/2 donor atom dopants is a timely subject and its investigation may pave the way for the realization of potential applications for solid-state quantum computers.

EXPERIMENTAL

All the computations were performed within the DFT framework by using the *ab initio* program package SIESTA[18,19]. The generalized gradient approximation (GGA) with the PBE functional was adopted for the exchange-correlation functional[20] and the electron-ion interactions were modeled by the norm-conserving Troullier-Martins pseudopotentials[21] in the Kleinman-Bylander factorized form[22]. We have used Sankey finite range pseudo atomic orbitals (PAOs)[23] as the split-valence double-ζ basis set with polarization (DZP)[24] for valence electrons. The geometries were considered optimized by means of conjugate gradient (CG) method, until the remanent force becomes smaller than 0.02 eV/Å.

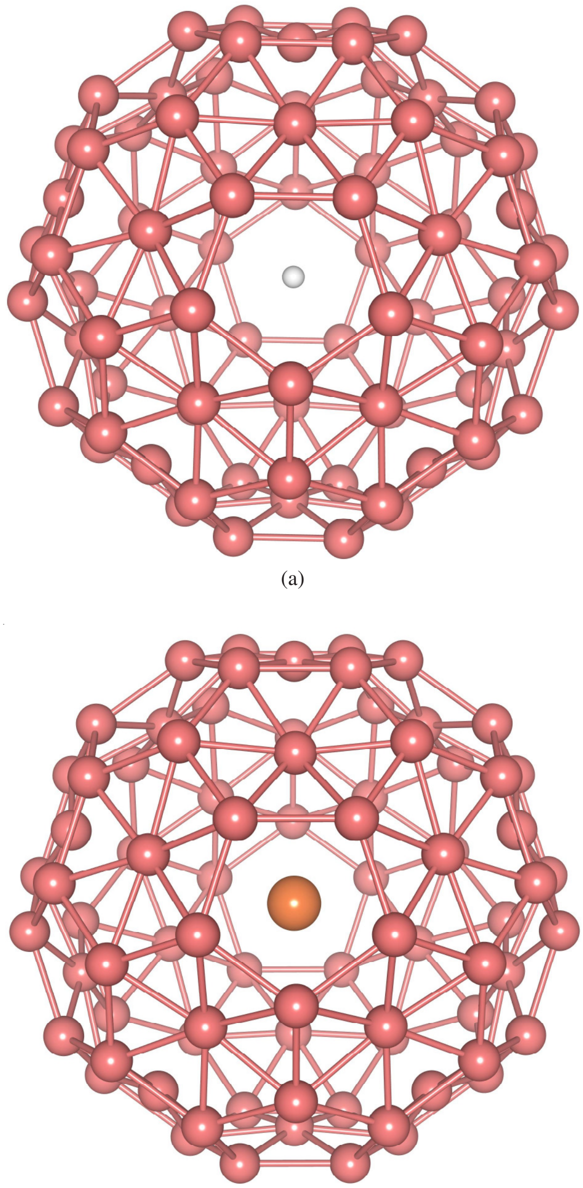
From the well known expression for calculating the molecular binding energies, E_b are obtained for various cases of present study.

$$E_b = E_{ful-x} - E_{ful} - E_x$$

where E_{ful-x} = total energy of the fullerene with a encapsulated X atom, E_{ful} = total energy of the pure fullerene and E_x = total energy of the isolated X atom. The reliability of present method has been confirmed by the previous works[25-32] thus reinforcing the reliability of theoretical method used in the present work.

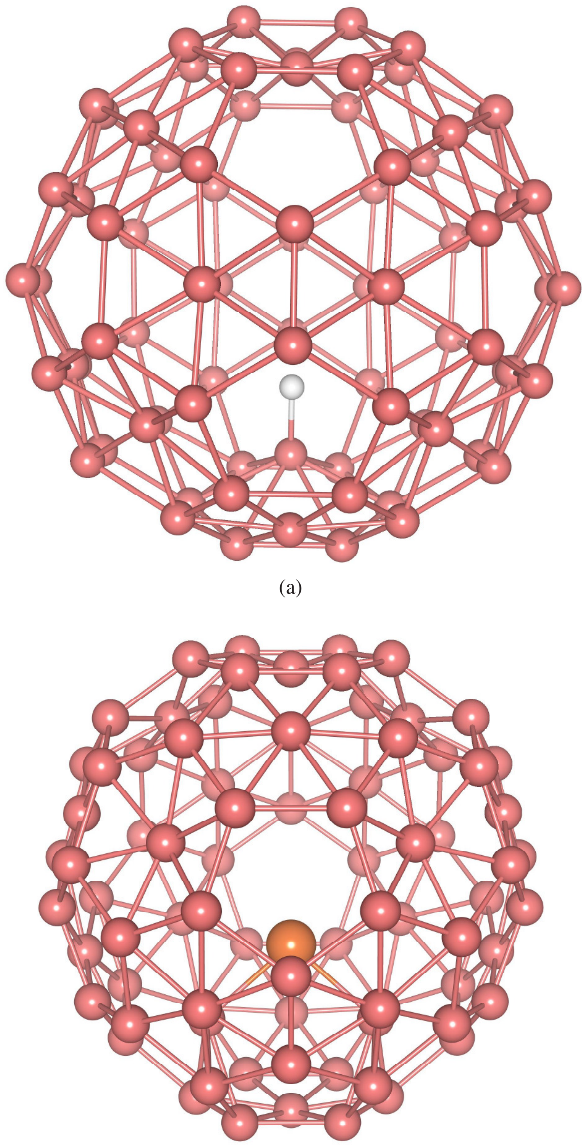
RESULTS AND DISCUSSION

The central objective in the present study is to examine the stability of encapsulated ^{1}H and ^{31}P atoms into the cavity of B_{80} fullerene nanocage (X@B80 complex). Fig. 1 represents the initial configuration of the incorporated atoms into the B_{80} nanocage. For this propose, we will first begin by discussing the geometrical properties of $^{1}H@B_{80}$ molecule interactions. The optimized B_{80} fullerene was used for the atom adsorption. To find the stable adsorption configuration, we carried out the full structural optimization of the initial configuration of Fig. 1. Our first-principles calculations on the $^{1}H@B_{80}$ complex yielded atomic displacement of H from the center. The results show that the binding energy and equilibrium distance between the closest atom between fullerene and H are about -4.13 eV (-85.09 kcal mol^{-1}) and 1.217 Å, respectively. The small distance of adsorbed H atom from the interior side-wall of the cage, the negative binding energy of -4.13 eV, indicate strong interaction of H atom with the inner surface of the B_{80} fullerene[25-33]. The optimized geometric structure of the $^{1}H@B_{80}$ complex is represented in Fig. 2(a). As it can be seen from the figure, a new bond was formed between the encapsulated H atom and B atom of the cage hence, the only available valance electron will be used up in the reaction making it unavailable for hyperfine contact coupling to the nuclear spin. Therefore, one can conclude that a B_{80} fullerene cage may not be suitable for the qubit application in conjunction with an ^{1}H atom.


(a)

(b)

Fig. 1. Model for H and P atoms encapsulated into B_{80} fullerene nanocage, (a) $^{1}H@B_{80}$ and (b) $^{31}P@B_{80}$. Atom colours: red-boron, white-hydrogen and orange-phosphorus

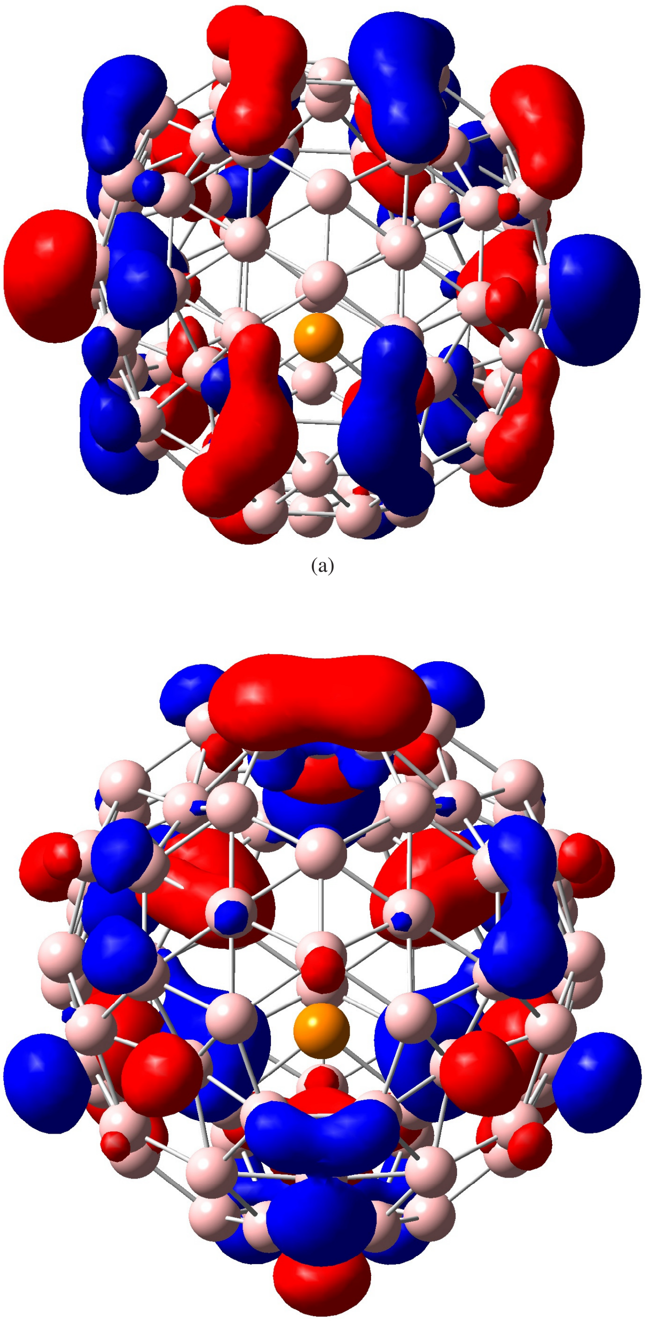

(a)

(b)

Fig. 2. Schematic representation of the optimized geometries of (a) $^{1}H@B_{80}$ and (b) $^{31}P@B_{80}$ structures of Fig. 1

To further evaluate the feasibility of B_{80} nanocage for the qubit applications, the incorporation of the less reactive ^{31}P atom into the cage has also been investigated (Fig. 1(b)). After full structural optimization of the considered system it is find also that the encapsulated atom prefers to be bound strongly to the inner side-wall of the cage with binding energy of about -5.59 eV. The schematic representation of the optimized geometric structure of the $^{31}P@B_{80}$ complex is given in Fig. 2(b). It can be seen that the P atom moves along the interior surface of the fullerene and forms new bonds with two B atoms of the cage, again making it unavailable for hyperfine contact coupling to the nuclear spin. Consequently, from the present result it is found that B_{80} fullerene nanocages are not suitable material for solid-state quantum computers.

To understand better the adsorption properties of incorporated atoms into the nanocages, we now look in detail at the electronic structures of the $^{31}P@B_{80}$ complex. Fig. 3 represents the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) electron density of the considered complex. They were obtained with the similar Troullier-Martine Pseudo-Potentials OpenMX[44] computer code. As it can be seen from the figure, the electron density is located only on the fullerene indicating a weak interaction between the incorporated atom and fullerene nanocage. This effect can be explained by analyzing the charge transfer between the interacting molecules. Charge analysis shows 0.07 and 0.08 e charges transferred from the cage to the H and P atoms, respectively, indicating that both of the encapsulated atoms are electron acceptors.


(a)

(b)

Fig. 3. HOMO and LUMO isosurfaces for $^{31}P@B_{80}$ complex where 0.03 a.u. was used as an isovalue of contour level

Table-1 contains the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energies for the considered complexes. The obtained results indicate the bare fullerene to behave as a wide band gap (1.03 eV) semiconductor or insulator. By incorporation of H atom, the band gap diminishes to 0.84 eV while for the case of P atom it changes to 0.30 eV. Therefore, in the case of $^{31}P@B_{80}$ complex the band gap is significantly reduced, inducing a metallic character to the encapsulated fullerene. Hence, despite the unfeasibility of this endohedrally naocage for solid-state quantum computers the present results open the way towards the assessing the possibility to create nanoconductors and connections between different parts of nanodevices for molecular electronics.

TABLE-1
HOMO AND LUMO ENERGIES OF A BARE B_{80} FULLERENE, $^{1}H@B_{80}$ AND $^{32}P@B_{80}$ COMPLEXES AT DZP LEVEL BASED ON DFT-GGA OPTIMIZED GEOMETRIES

	HOMO (eV)	LUMO (eV)	ΔE (eV)
B_{80}	-5.18	-4.15	1.03
$^{1}H@B_{80}$	-4.96	-4.12	0.84
$^{31}P@B_{80}$	-4.99	-4.69	0.30

Conclusion

Incorporated ^{1}H and ^{31}P atoms into the boron fullerene nanocage ($^{1}H@B_{80}$ and $^{31}P@B_{80}$) were studied using *ab initio* calculations based on density functional theory. The fullerene selected was B_{80} nanocage, which possess unusual high stability. The results for the electronic and structural properties were found to be similar appreciably. It is observed that the atomic displacement of both H and P atoms from the center of the cage so that the wall B atoms can form covalent bonds with encapsulated atoms. Thus, the only available valance electron will be used up in the reaction making it unavailable for hyperfine contact coupling to the nuclear spin. Consequently, B_{80} fullerene nanocage may not be suitable for the solid-state quantum computer applications. However, the electronic structure for incorporated nanocages is significantly modified by the reduction of the band gap, inducing a metallic character to the encapsulated fullerene. This open the way towards the assessing the possibility to create nanoconductors and connections between different parts of nanodevices.

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