Three-dimensional carbon Archimedean lattices for high-performance electromechanical actuators

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Abstract

We propose three-dimensional carbon (3D-C) structures based on the Archimedean lattices (ALs) by combining $sp^2$ bonding in the polygon edges and $sp^3$ bonding in the polygon vertices. By first-principles calculations, four types of 3D-C ALs: (4, 8\textsuperscript{2}), (3, 12\textsuperscript{2}), (6\textsuperscript{3}), and (4\textsuperscript{4}) 3D-Cs are predicted to be stable both dynamically and mechanically among 11 possible ALs, in which the notations $(p_1, p_2, \ldots)$ are the indices of the AL structures. Depending on their indices, the 3D-C ALs show distinctive electronic properties: the (4, 8\textsuperscript{2}) 3D-C is an indirect band-gap semiconductor, the (3, 12\textsuperscript{2}) 3D-C is semimetal, while the (6\textsuperscript{3}) and (4\textsuperscript{4}) 3D-Cs are metals. Considering the structural deformation due to the changes in their electronic energy bands, we discuss the electromechanical properties of the 3D-C ALs as a function of charge doping. We find a semiconductor-to-metal and semimetallic-to-semiconductor transitions in the (4, 8\textsuperscript{2}) and (3, 12\textsuperscript{2}) 3D-Cs as a function of charge doping, respectively. Moreover, the (3, 12\textsuperscript{2}) 3D-C exhibits a $sp^2$-$sp^3$ phase transformation at high charge doping, which leads to a huge 30\% irreversible strain, while the reversible strain in the (4, 8\textsuperscript{2}) 3D-C is up to 9\%, and thus they are quite promising for electromechanical actuators.

1. Introduction

Scientific and technological attempts to design and synthesize actuation materials as a building block of artificial muscle have been quite intensive during the past two decades, with wide potential applications in biomechanical robotics, and medical sciences\cite{1, 2, 3, 4}. Such an actuation material based on carbons was proposed by Baughman at al. in 1999, who synthesized carbon nanotube (CNT) yarn and demonstrated that CNT yarn can generate stress 100 times that achievable by natural muscles (~ 0.35 MPa)\cite{5}. Following this initial demonstration, many studies have sought other carbon actuator materials such as single, multilayer graphene, graphene oxide, graphitic carbon nitride, and CNT films via both experimental and theoretical approaches\cite{6, 7, 8, 9, 10, 11, 12}. However, both CNT yarn and graphene showed a small strain of about 1\% under charge (electron or hole) doping\cite{13, 14}, while the skeletal muscles provide a work cycles involving contractions of more than 20\%. The CNT yarn or the multilayer graphene are also formed by the weak van der Waals (vdW) forces, which lead to a possible mechanical failure along the direction of the vdW interactions in these material\cite{14}. To overcome this problem, we may use such CNT and graphene structures to form new three-dimensional carbon (3D-C) materials that still inherit their superb properties while simultaneously avoiding vdW interaction between different tubes or layers.

Carbon is one of the most versatile elements due to its ability to form $sp^2$, $sp^3$, and $sp^3$–hybridized bonds, resulting in various allotropes that have already been experimentally identified, such as graphite, diamond, fullerenes, CNT, and graphene\cite{15}. Recently, computational modeling based on the first-principles density functional theory (DFT) and molecular dynamics opens the possibility to predict more carbon allotropes. Some new 3D-C materials, such as 3D-C honeycomb\cite{16}, T-carbon\cite{17}, Y-carbon\cite{18}, L-carbon\cite{19}, and cubane-based 3D porous carbon\cite{20} have been either observed or proposed, where all these materials are found to be dynamically, mechanically, and thermally stable. Furthermore, the elastic constants of some of these 3D-C structures were studied. Zhao et al.\cite{21} showed that 3D-C structures have large Young’s moduli, large tensile strength, and low density. Recently, experimentalists have successfully synthesize a kind of 3D-C structures in forms of carbon honeycomb by deposition of vacuum-sublimated graphite\cite{22} and nanoporous carbon made by buckyball-like nanographene\cite{23}. This fact shows a great potential in designing the 3D-C materials and structures while retaining low density and excellent mechanical properties for electromechanical actuators. In this sense, a systematic theoretical design could be the first step to suggest possible structures suitable as electromechanical actuators so that experimentalists can synthesize the recommended structures.

Inspired by the structure of the Archimedean lattices (ALs), defined as a complete set of lattices having all vertices are equivalent, we design a class of 3D-Cs by a combination of $sp^3$ bonding in the polygon edges and $sp^3$ bonding in the polygon vertices. In this work, considering efficient computational time, 4 of 11 possible 3D-C ALs are calculated using the first-principles DFT calculations. Hereafter, the 4 possible 3D-C ALs are referred to as (4, 8\textsuperscript{2}), (3, 12\textsuperscript{2}), (6\textsuperscript{3}), and (4\textsuperscript{4}) 3D-Cs, in which the notations $(p_1, p_2, \ldots)$ are the indices of the AL structures based on the types of polygons connected at a given vertex (see Fig. 1). We find that these 3D-C ALs are both dy-
namically and mechanically stable because all of their phonon frequencies and mechanical stability conditions are positive, respectively. We will discuss the electromechanical properties of the 3D-C ALs as a function of charge doping, for both electron and hole doping cases, by considering the structural deformation due to the changes in their electronic energy bands. Three important physical phenomena in the doped 3D-C ALs are found from the calculations: (1) a semiconductor-to-metal and semimetallic-to-semiconductor transitions in the (4, 8\textsuperscript{2}) and (3, 12\textsuperscript{2}) 3D-Cs, respectively, (2) a structural transformation between \(sp^2\)-\(sp^3\) phases under heavy doping in the (3, 12\textsuperscript{2}) 3D-C, and (3) a large change of reversible strain up to 5\%, essential for the artificial muscle applications.

2. Calculation methods

We perform first-principles calculations to determine the total energy and the electronic structure of the 3D-C ALs using Quantum ESPRESSO [24]. The Rabe-Rappe-Kaxiras-Joannopoulos ultrasoft pseudopotential with an energy cutoff of 60 Ry is chosen for the expansion of the plane waves [25]. Note that the choice of 60 Ry energy cutoff is sufficient for converging the total energy calculation. The exchange-correlation energy is evaluated by the general-gradient approximation using the Perdew-Burke-Ernzerhof (PBE) function [26]. In our simulation, the \(k\)-point grids in the Brillouin-zone are employed according to the Monkhorst-Pack scheme, where \(k\) is the electron wave vector. \(9 \times 9 \times 13, 5 \times 5 \times 13, 7 \times 7 \times 9,\) and \(16 \times 16 \times 6\) \(k\)-points are used for (4, 8\textsuperscript{2}), (3, 12\textsuperscript{2}), (6\textsuperscript{2}), and (4\textsuperscript{2}) 3D-Cs, respectively. To obtain optimized atomic configurations of 3D-C ALs, the atomic positions and cell vectors are fully relaxed using the Broyden-Fletcher-Goldfarb-Shanno minimization method [27] until all the Hellmann-Feynman forces and all components of the stress are less than 5e\textsuperscript{-4} Ry/a.u. and 5e\textsuperscript{-2} GPa, respectively, which are adequate for the present work. To examine the dynamically stable of the 3D-C ALs, phonon dispersions are computed using the density functional perturbation theory (DFPT) within the linear response approximation [31]. The dynamical matrices are calculated on \(4 \times 4 \times 6, 2 \times 2 \times 6, 3 \times 3 \times 4,\) and \(8 \times 8 \times 3\) \(q\)-points for (4, 8\textsuperscript{2}), (3, 12\textsuperscript{2}), (6\textsuperscript{2}), and (4\textsuperscript{2}) 3D-Cs, respectively.

To examine the mechanically stable of the 3D-C ALs, we use the Thermo-pw code [32] to calculate the elastic constants \(C_{ij}\). The calculated elastic constants are derived from the finite difference approach and are by default averaged over the entire unit cell volumes. From the point of view of elasticity theory, it is well-recognized that the value of \(C_{ij}\) is related to the equivalent volume of the crystal. To obtain the equivalent volume of the 3D-C ALs, the wall thickness, \(d\), in the polygon vertices is considered as the interlayer spacing of graphite and multiwalled carbon nanotubes in nature based on the van der Waals interactions \((d = 3.4\ \text{Å})\) [33], where \(d\) is assumed to be independent of the small strain and charge doping [14]. From the calculated \(C_{ij}\), the polycrystalline corresponding bulk modulus \(B\) and shear modulus \(G\) are calculated using the Voigt-Reuss-Hill approximation [34], then Young’s modulus \(E\) and Poisson’s ratio \(v\) are obtained by the following relations,

\[
E = \frac{9G·B}{(3B+G)}, \quad v = \frac{(3B-2G)/(6B+2G)},
\]

respectively.

To discuss the electromechanical actuation of the 3D-C ALs, the geometry optimization is performed with the charge doping level, ranging from −0.15 to 0.15 electron per carbon atom, in which the electron (hole) doping is simulated by adding (removing) electrons to the unit cell with the same amount of uniformly positive (negative) charge in the background so as to keep the charge neutrality.

3. Results and Discussion

3.1. Structural properties

In Figs. 1a-c, we illustrate crystal structures of our proposed 3D-C ALs. Each AL is defined by a set of integers \((p_1, p_2, \ldots)\) indicating, in a unit cell, the type of polygons meeting at a given vertex (see Fig. 1a). When a polygon appears more than one time consecutively, e.g., \((\ldots, p, \ldots)\), we abbreviate the notation by writing \((\ldots, p^2, \ldots)\). For example, (4, 8\textsuperscript{2}) 3D-C means that each vertex is surrounded sequentially by one square and two octagons, as shown in Figs. 1a and 1b. This property has made them useful in the study of mathematics [35], crystallization [36], as well as metamatamers [37]. Now, the designing principle of our 3D-C structures is that the polygon vertices are formed by diamond-like \(sp^3\) bond, while the polygon edges consist of flattened \(sp^2\) bond such like that in graphene sheets, as shown in Fig. 1b. For simplicity, we focus on four allotropes of the 3D-C ALs that are found to be stable theoretically, namely, (4, 8\textsuperscript{2}), (3, 12\textsuperscript{2}), (6\textsuperscript{2}), and (4\textsuperscript{2}) 3D-Cs. We note that some ALs such as (3\textsuperscript{2}, 6) or (3\textsuperscript{2}, 4, 3, 4) might not be suitable to be a 3D-C structure because the condition for the \(sp^3\) bonding of carbon atom at each vertex is not satisfied. The details of the crystal structures, including the number of atoms in unit cell, lattice parameters and bond lengths between \(sp^2\) (green atoms) and \(sp^3\) bonded atoms (red atoms), are shown in Table 1 and Fig. 1.

3.2. Thermodynamics stability

In Fig. 2, we show the optimized total energy per carbon atom of the 3D-C ALs as a function of charge doping \(q\) ranging from −0.15 to 0.15 electron per carbon atom. This charge range is appropriate for our calculations because the limit of the experimentally accessible charge is 0.3 e/atom for graphite [13]. In the neutral condition, the total energy of the 3D-C ALs is close to that of \(C_{60}\) fullerene, 3D diamond, and

| Structure | \(N\) | \(a\) (Å) | \(c\) (Å) | \(d_{sp^2-sp^2}\) (GPa) | \(d_{sp^2-sp^3}\) (GPa) | \(d_{sp^3-sp^3}\) (GPa) |
|-----------|-----|---------|---------|-----------------|-----------------|-----------------|
| (4, 8\textsuperscript{2}) | 8 | 5.23 | 2.50 | 1.35 | 1.49 | 1.61 |
| (3, 12\textsuperscript{2}) | 12 | 8.13 | 2.51 | 1.36 | 1.47 | 1.56 |
| (6\textsuperscript{2}) | 16 | 6.34 | 4.82 | 1.38 | 1.49 | 1.63 |
| (4\textsuperscript{2}) | 6 | 2.60 | 5.98 | 1.37 | 1.54 | - |

Table 1: Number of atoms in unit cell \(N\), lattice parameters \(a\) (Å) and \(c\) (Å), and bond lengths (Å) of the 3D-C ALs.
The Archimedean lattices. (b) Perspective view of the 3D-C ALs structures in three-dimensional space. (c) The unit cell of the 3D-C ALs showing the lattice constants and the C-C bond lengths. Red atoms are \( sp^2 \)-hybridized, while green atoms are \( sp^3 \)-hybridized.

Figure 2: Optimized total energy per C-atom of the 3D-C ALs as a function of charge (electron and hole) doping per C-atom. At the charge \( q = 0.00 \) e/C-atom, total energies of individual (6, 6) SWNT [33], (6, 6) SWNT bundle [13], smallest carbon nanotubes including (2, 2), (3, 0) and (3, 1) SWNTs [33], graphite, diamond, \( C_{60} \) fullerene are also calculated and plotted here for comparison.

3.3. Dynamical and mechanical stability

In Fig. 3, we show phonon dispersions along the high-symmetry directions in the corresponding Brillouin zone of the four 3D-C ALs to discuss their lattice dynamics. All real phonon energies (positive eigenvalues from the Hessian matrix) confirm that the 3D-C ALs are dynamically stable. Three distinct acoustic modes including the in-plane longitudinal (LA), in-plane transverse (TA) and out-of-plane (ZA) modes exhibit linear dispersions near the \( \Gamma \) point, these modes are dominated by \( sp^3 \) bonding in the polygon edges. Note that, theZA mode in the 3D-C ALs is unlike that in graphene sheet since the graphene ZA mode is quadratic dispersion [42]. The highest phonon frequency in 3D-C ALs reaches 1600 cm\(^{-1}\), comparable to that in graphite [43] and diamond [44].

Next, we investigate the mechanical stability of the 3D-C ALs. To guarantee the positive strain energy during lattice distortion, the tensor components of the linear elastic constant (\( C_{ij} \)) of a stable crystal have to obey the Born criteria [45]. In the (4, \( 8^2 \)) and (4\(^4 \)) 3D-Cs, there are six independent elastic con-
Table 2: Elastic constants $C_{ij}$ (GPa), bulk modulus $B$ (GPa), shear modulus $G$ (GPa), Young’s modulus $Y$ (GPa), Poisson’s ratio $\nu$, and mass density $\rho$ (g/cm$^3$) of the 3D-C ALs, graphite, and diamond.

| Structure   | $C_{11}$ | $C_{33}$ | $C_{44}$ | $C_{66}$ | $C_{12}$ | $C_{13}$ | $B$     | $G$     | $Y$     | $\nu$ | $\rho$ |
|-------------|----------|----------|----------|----------|----------|----------|---------|---------|---------|-------|--------|
| (4, 8$^2$)  | 651      | 1026     | 260      | 30       | 90       | 67       | 303     | 179     | 449     | 0.25  | 2.33   |
| (3, 12$^2$) | 372      | 1004     | 261      | 45       | 283      | 86       | 286     | 138     | 357     | 0.29  | 1.67   |
| (6$^3$)     | 346      | 1044     | 178      | 46       | 254      | 56       | 263     | 124     | 320     | 0.30  | 1.90   |
| (4$^3$)     | 704      | 1151     | 76       | 72       | 45       | 91       | 325     | 155     | 402     | 0.30  | 2.97   |
| graphite$^a$| 1060     | 36       | 4        | 440      | –        | 15       | 8       | 440     | 1020    | 0.16  | 2.30   |
| diamond$^a$ | 1070     | 1070     | 575      | 575      | –        | 125      | 442     | 576     | 1063    | 0.10  | 3.63   |

$^a$ Reference [21][41].

3.4. Electronic properties

In Fig. 4, we show the calculated electronic structures of the 3D-C ALs along the high-symmetry directions in the corresponding Brillouin zone (see Fig. 3) for neutral and charge doping states. For the neutral case, we find that the (4, 8$^2$) 3D-C is an indirect-gap semiconductor with a band gap of about 0.96 eV, the (3, 12$^2$) 3D-C is a semimetal, while the (6$^3$) and (4$^3$) 3D-Cs are metals. The unusual electronic properties of the 3D-C ALs might originate from $sp^2$ and $sp^3$ bonded atoms (green and red atoms in Fig. 1) at the Fermi level. For the (6$^3$) and (4$^3$) 3D-Cs, the number of $sp^3$ bonded atoms is smaller than that of $sp^2$ bonded atoms in the unit cell. Therefore, most of the conducting electrons at the Fermi level are coming from the $2p_x$ orbitals of $sp^2$ bonded carbon atoms like that in graphene, which lead to the metallic properties of the (6$^3$) and (4$^3$) 3D-Cs. As for the (4, 8$^2$) and (3, 12$^2$) 3D-Cs, the numbers of $sp^2$ and $sp^3$ bonded atoms in the unit cell are the same. Therefore, both $sp^2$ and $sp^3$ bonded atoms contribute to the electrons at the Fermi level, which result in semiconducting (4, 8$^2$) and semimetallic (3, 12$^2$) 3D-Cs. Through a detailed analysis of the energy band structures under the charge doping, we can see that the energy bands change significantly under both heavy electron and hole dopings, which could affect the mechanical properties of the 3D-C ALs.

In Fig. 5, we show the energy band gap $E_g$ as a function of charge doping $q$. For the semiconducting (4, 8$^2$) 3D-C, $E_g$ decreases with both electron and hole doping cases, leading to a semiconductor-to-metal transition at $q = \pm0.15 e$/C-atom. For the semimetallic (3, 12$^2$) 3D-C, a semimetallic-to-semiconductor transition occurs when $q$ is adjusted up to 0.081 $e$/C-atom, while the (6$^3$) and (4$^3$) 3D-Cs retain their metallic states under charge doping. It is thus clear that the rigid band model, in which the effective mass and the band gap are fixed while Fermi level is shifted by charge doping, is not valid for the presented cases. As shown in Fig. 5, the rigid band model is remarkably more pronounced for the (3, 12$^2$) 3D-C in both electron and hole dopings cases and for the (4, 8$^2$) 3D-C in the electron doping case. For the (3, 12$^2$) 3D-C, $sp^3$ bonded atoms break into $sp^2$ bonded atoms under heavy electron doping, while $sp^2$ bonded atoms make new $sp^3$ bonded atoms under heavy hole doping (see the charge density in Fig. 7). The energy band structure thus changes significantly, which leads to the metallic and semiconducting properties of the (3, 12$^2$) 3D-C under heavy electron and hole dopings, respectively. For

![Image of phonon dispersions, high-symmetry points, and lines in the corresponding Brillouin zone of four different 3D-C ALs.](image-url)
the (4, 8\textsuperscript{2}) 3D-C, its electromechanical response shows a higher strain value than the (6\textsuperscript{3}) and (4\textsuperscript{4}) 3D-Cs under heavy electron doping (see Sec. 3.5), which also leads to a significant change of the energy band structure. Therefore, the rigid band model is no longer suitable for the (4, 8\textsuperscript{2}) 3D-C under heavy electron doping.

Figure 4: Energy band structures of the 3D-C ALs with different electron doping \((q = -0.15 \text{ e/C-atom})\) and hole doping \((q = +0.15 \text{ e/C-atom})\) including those with the neutral condition \((q = 0.00 \text{ e/C-atom})\). The Fermi energy (dashed line) is set to zero for all plots.

Figure 5: Energy band gap \(E_g\) plotted as function of charge doping of four different 3D-C ALs.

Figure 6: Strain as function of charge doping of four different 3D-C ALs.

3.5. Electromechanical properties

In Fig. 6 we show the charge-strain relationship of the 3D-C ALs. The strains of three 3D-C ALs, except the (3, 12\textsuperscript{2}) 3D-C, are approximately a linear function of charge doping and are isotropic in \(x - y\) plane. For the (4, 8\textsuperscript{2}) 3D-C, \(\varepsilon_{xx} = \varepsilon_{yy}\) is up to 9.0% \((-5.1\%)\) under the electron (hole) doping at \(q = -0.15 (0.15) \text{ e/C-atom}\), while \(\varepsilon_{zz} = 0.5\% (2.4\%)\). For the (6\textsuperscript{3}) 3D-C, \(\varepsilon_{xx} = \varepsilon_{yy} = 4.2\% (-4.3\%)\) and \(\varepsilon_{zz} = 3.6\% (-2.3\%)\) at \(q = -0.15 (0.15) \text{ e/C-atom}\). For the (4\textsuperscript{4}) 3D-C, \(\varepsilon_{xx} = \varepsilon_{yy} = 5.7\% (-7.8\%)\) and \(\varepsilon_{zz} = 5.4\% (-1.7\%)\) at \(q = -0.15 (0.15) \text{ e/C-atom}\).
Hole doping

Neutral

Hole doping

Electron doping

Figure 7: Charge density isosurface (0.13 e/Å³) of the (3,12²) 3D-C at neutral, heavy electron and hole dopings cases. Red atoms are sp² hybridized and blue atoms are sp³ hybridized. At heavy electron and heavy hole doping regimes, the (3,12²) 3D-C is transformed to (6¹) 3D-C and (3,4,6,4) 3D-C, respectively.

As for the (3,12²) 3D-C, the calculation results give an anisotropic strain under hole doping at q = 0.08 e/C-atom with εₓₓ and εᵧᵧ are up to 4.3% and -25.2%, respectively. For further hole doping, εₓₓ and εᵧᵧ show a finite jump that leads to εₓₓ = εᵧᵧ ∼ -31% at q = 0.081 e/C-atom. Similarly, for electron doping, εₓₓ and εᵧᵧ have a finite jump at q = -1.06 e/C-atom (εₓₓ = -0.6% and εᵧᵧ = 16.5%). We argue that a very high strain of the (3,12²) 3D-C under charge doping is a result of its chemical bond transformation. Based on the charge density plot in Fig. 7, sp³ bonds change to sp² bonds under heavy electron doping, while sp² bonds change to sp³ under heavy hole doping in the (3,12²) 3D-C. However, these sp²→sp³ transformations are not reversible. At heavy electron and heavy hole doping regimes, the (3,12) 3D-C is transformed to (6¹) 3D-C and (3,4,6,4) 3D-C, respectively. Nevertheless, the (3,12²) 3D-C is a stable structure without charge doping.

To understand the stability of the 3D-C ALs under charge doping, the mechanical stability is investigated by the Born criteria [45] at different charge states. According to the Born criteria, the elastic constants C_ij of a stable 3D-C AL structure has to satisfy the conditions C₁₁ > |C₁₂|, 2C₁₂ / C₁₁ < C₃₃(C₁₁ + C₁₂), C₄₄ > 0, and C₆₆ > 0 (see Sec. 3.3). In Fig. 8, we calculate C_ij as a function of charge doping. We find that the (4,8²), (6¹) and (4²) 3D-Cs are unstable under heavy hole doping because both C₄₄ and C₆₆ are less than zero, resulting in a negative strain energy. On the other hand, the (3,12²) 3D-C is stable under both heavy electron and hole doping cases but its structure shows an irreversible sp²→sp³ transformations under heavy charge doping (see Fig. 7). Having confirmed the stability of the 3D-C ALs, we then systematically study their mechanical properties.

Finally, Fig. 9 shows Poisson’s ratio ν of the stable 3D-C ALs as a function of charge doping. It is known that Poisson’s ratio is a measure of the Poisson effect, a phenomenon in which a material tends to expand in the directions perpendicular to the direction of compression. In the neutral condition, the ν values of three 3D-C ALs are about 0.29, except for the (4,8²) 3D-C which gives ν = 0.25. These values are larger than those of SWNT (ν = 0.07 – 0.15) and graphene (ν = 0.186), which is suitable for muscle. In the 3D-C ALs, the Poisson effect is also further enhanced by the charge doping.

Figure 9c shows the mass density ρ of the stable 3D-C ALs as a function of charge doping. At the neutral charge, (4²) 3D-C has the highest ρ = 2.97 g/cm³ and (3,12²) 3D-C has the lowest ρ = 1.67 g/cm³. These values are smaller than that of diamond (3.633 g/cm³) and SWNT (~ 3.0 g/cm³), and close to that of graphite (2.295 g/cm³) [21]. The low density of the 3D-C ALs is due to their porous structures along the axial z directions. Although the mass density of 3D-C ALs is higher than that of mammalian skeletal muscle (~ 1g/cm³), we expect that the designed 3D-C ALs with larger pore diameters along the z direction will give ultralow density. Pang et al., [51], for example, showed that the density of the (6¹) 3D-C can go down to 0.31 g/cm³ with the pore diameter of about 29.7 Å. However, both of the strength and failure strain decrease in (6¹) 3D-C with growing pore diameter [51]. We also note that for the 3D-C ALs with large pore diameter, the number of flattened sp² bonds at
the polygon edges is much higher than that of \( sp^3 \) bonds at the polygon vertices. Therefore, the 3D-C ALs might not be able to maintain their electromechanical properties when their pore diameters become larger.

4. Conclusions

By first principles DFT calculations, we have shown the feasibility of constructing stable 3D carbon materials based on the Archimedean lattices with excellent electromechanical properties. The reversible strain can be up to 9\% with stable \((4,8^2)\) 3D-C structure under heavy electron doping, which is nine times larger than that of carbon nanotube or graphene. A very high irreversible strain of about 30\% in the \((3,12^2)\) 3D-C is also found as a result of \( sp^2-sp^3 \) bond transformation. Large mechanical moduli (including the bulk modulus, the shear modulus, and Young’s modulus), large Poisson’s effect, and low mass density under charge doping may be utilized to obtain high-performance actuators with applications ranging from artificial muscle to biomedical engineering. Depending on the index of Archimedean lattices, the 3D-C ALs also show distinctive electronic properties: the \((4,8^2)\) 3D-C is an indirect band-gap semiconductor, the \((3,12^2)\) 3D-C is semimetal, while the \((6^2)\) and \((4^2)\) 3D-Cs are metals. Furthermore, we expect that the semiconductor-to-metal and semimetallic-semiconductor transitions found in the semiconducting \((4,8^2)\) and semimetallic \((3,12^2)\) 3D-Cs, respectively, under heavy charge doping will open further explorations of the 3D-C ALs in the near future.

Acknowledgments

N.T.H. and A.R.T.N acknowledge the Interdepartmental Doctoral Degree Program for Multidimensional Materials Science Leaders under the Leading Graduate School Program in Tohoku University. R.S. acknowledges JSPS KAKENHI Grant Numbers JP25107005 and JP25286005.

Supplementary information

Atomic coordinates (CIF files) of the optimized 3D-C ALs structures studied in this work.

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