

Carbon materials: structures, properties, synthesis and applications

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Received: 28 January 2023 / Accepted: 18 June 2023

Abstract. As one of the most versatile elements, carbon materials occupy the most plentiful allotropes composed of pure or mixed hybridization orbitals of $sp^1/sp^2/sp^3$. The design and synthesis of new carbon materials may be stimulated based on a deeper understanding of underlying structures and related properties. In this review, the initial early discoveries of carbon materials are examined based on their hybridization of orbitals. According to the type of hybridization, the discovered carbon materials are firstly classified and introduced in detail based on their crystal structures. Secondly, its physical and chemical properties, mainly including mechanical properties, optical properties and electronic properties, are reviewed. Thirdly, the existing methods of predicting carbon structure and synthesizing carbon materials are classified and summarized, and some typical carbon materials predicted or prepared are discussed respectively. Then, the main applications of newly synthesized carbon materials in the last two decades are classified and summarized, and the microstructure is linked with the macro properties and specific applications. Finally, the future research opportunities for carbon materials and their potential applications are prospected from the aspects of the gap between theoretical prediction and preparation, the current research hotspot of carbon materials and the incomplete application of carbon materials. It is the authors' intention for this review paper to serve not only as a valuable reference for research into carbon materials and related composites, but also as a guidance for novel materials design at the atomic level.

Keywords: Carbon materials / sp^1 sp^2 / sp^3 hybridization / Atomic structure / Microstructure and property / Mechanical property

1 Introduction

Carbon (C) is one of the most versatile elements in the Periodic Table, known for its tetravalent shell. Its unique electronic structure makes it difficult to completely gain or lose electrons for forming fully occupied/unoccupied orbitals in chemical reactions. By combining various hybridizations of sp^3 , sp^2 and sp^1 or embedding other foreign atoms in carbon hybrids the desired properties can be modified, and researchers working with carbon materials have been doing this for a considerable time [1]. Besides, with the aid of driving forces under extreme conditions, including high pressure and/or high temperature, the energy barrier for destroying the chemical bonds and forming new metastable phases can be overcome in order to obtain almost infinite performance in scientific and engineering fields [2].

For designing and fabricating the carbon materials based on the hybridized orbitals or structural units, the underlying mechanism of each hybridized orbital/state should be revisited. In general, a large proportion of sp^3 orbitals in a carbon material means that the material has improved mechanical properties. For example, diamond, which is made of pure sp^3 bonds, is known for its excellent hardness [3]. Moreover, the degenerated state of sp^2 generally represents good compatibility with other atoms, which could be conducive to for developing two dimensional/ one dimensional (2D/1D) carbon materials with excellent electronic/optical/ frictional/ catalytic properties [4].

In this review, new carbon materials obtained in recent decades are summarized according to the hybridized states of the carbon orbitals. Then, the physical and chemical properties of the materials including their mechanical, electronic and optical properties and other aspects such as thermal are examined. Additionally, the theoretical design and experimental synthesis of carbon materials are

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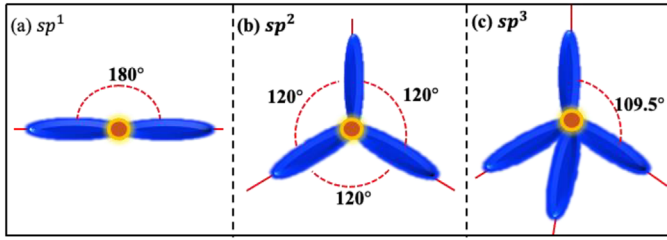


Fig. 1. Schematic configurations for (a) sp^1 , (b) sp^2 , and (c) sp^3 .

discussed systematically. Finally, this review points out the future research opportunities for carbon materials and their potential applications.

2 Atomic structure of carbon materials

The versatile configurations of C hybridization of orbitals, including sp^1 , sp^2 and sp^3 (Fig. 1), could provide a fertile ground for discovering and designing new C materials. For example, the configuration composed of sp^3 endows a strong σ bond to an adjacent atom wherein every C atom is tetrahedrally surrounded by four neighboring C atoms with a 109.5° directed bond, as shown in Figure 1c [5]. As for the three-fold coordinated sp^2 configuration, three of the four valence electrons enter trigonally directed sp^2 orbitals with formations of σ bonds in a plane style. The fourth electron of the sp^2 atom lies in a $p\pi$ orbital, which lies normal to the σ bonding plane. This π orbital forms a weaker π bond with a π orbital on one or more neighboring atoms (Fig. 1b). For further degeneration of the sp^1 configuration, two of the four valencing electrons enter σ orbitals, each forming a σ bond directed along the $\pm x$ -axis, as presented by red line in Figure 1a, and the other two electrons enter $p\pi$ orbitals in the y and z directions [6].

Fueled by studies on a typical 2D C material graphene, extensive research efforts have been directed towards the discovery and synthesis of novel two-dimensional (2D) carbon allotropes. For convenience, timeline of new development of carbon material since fullerene has been provided in Fig. 2. In the following, we introduce recent research results on the structure of carbon allotropes in terms of the combination of different hybrid states.

2.1 sp^1 and sp^2 hybridization

There is little research on carbon materials containing only sp^1 hybridization since sp^1 hybridization generally coexists with hybridization of sp^2 and/or sp^3 . Representatively, three carbon allotropes (Graphynes, net- π , and Ct8) that are made of sp^1 and sp^2 hybrids or pure sp^2 hybrids were introduced as follows:

2.1.1 Structure of Graphynes (GDY)

GDY [26–28], which are composed of sp^1 - and sp^2 -hybridized carbon atoms with different stacking sequences, are considered as a series of new 2D carbon allotropes. The specific feature of GDY is that the structural units consist of two adjacent sp^2 hybrid carbon atoms linked together by

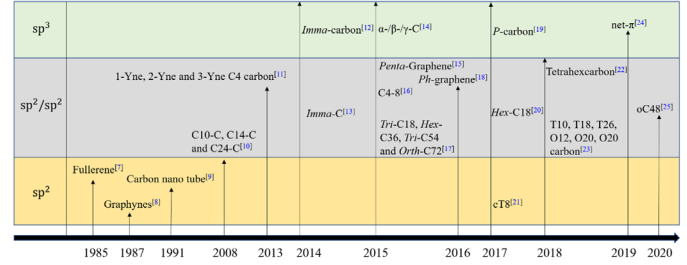


Fig. 2. Timeline of new development of carbon materials.

n " $C\equiv C$ -" bonds. It is worth noting that their different structural characteristics in GDY could be modified by the different stacking sequences of sp^1 - and sp^2 -hybridized carbon atoms, that is to say, different kinds of single-layer graphynes have different crystal structures and symmetries. For example, most of the GDY have hexagonal symmetry like graphene (GR), meanwhile, the original cell of graphyne (6,6,12) with has a rectangular shape and symmetry with in-plane anisotropy. Among them, replacing all the carbon-carbon bonds in GR using acetylenic ($-C\equiv C-$) linkages could endow a α -graphyne. Further, β -graphyne (γ -graphyne) results from the replacement of two-third (one-third) carbon-carbon bonds in GR by acetylenic linkages.

2.1.2 Structure and property of net- π

net- π [24], which consists of quadrilateral, pentagonal, hexagonal, octagonal and decagonal rings, possesses a $Pmmm$ symmetry (space group D_{2h}^{12}) with an orthogonal lattice, as shown in Figure 3. The optimized lattice parameters for the unit cell of net- π are $a = 9.95 \text{ \AA}$ and $b = 5.77 \text{ \AA}$, as represented by a dashed rectangle in Figure 3. It is found that there are six unequivalent carbon atoms (labeled C1-C6 in Fig. 3) in the unit cell of net- π . The bond lengths of net- π is very close to that ($1.42 \text{ \AA}$) of GR, wherein the C1-C2, C2-C3, C2-C4, C3-C6, C4-C5 and C5-C6 bonds are $1.388 \text{ \AA}$, $1.440 \text{ \AA}$, $1.492 \text{ \AA}$, $1.421 \text{ \AA}$, $1.453 \text{ \AA}$ and $1.412 \text{ \AA}$, respectively, indicative of the existence of a double C-C bond. However, the range of bond angles of net- π is 108° – 149° , which differs significantly from the 120° of GR, implying that the strain in the net- π is higher than that in graphene and probably leads to an enhancement in total energy. It is also found that net- π has a low lithium ion migration barrier, which is comparable to graphene, and is a promising carbon allotrope anode material with potentially high performance.

2.1.3 Structure and property of cT8

cT8 [21], which is unlike the two previously described carbon allotropes (GDY and net- π), is a pure three-dimensional (3D) sp^2 -hybridized carbon crystal. From Figure 4, one can see that the cT8 structure crystallizes in a tetragonal cell with the space group of $I4_1/amd$ and the Wyckoff site of $16f$ (0.2500 0.3855 0.8750). Observed along the top view (Fig. 4b), there are four chiral helical carbon chains via linkages of the double C-C bonds along

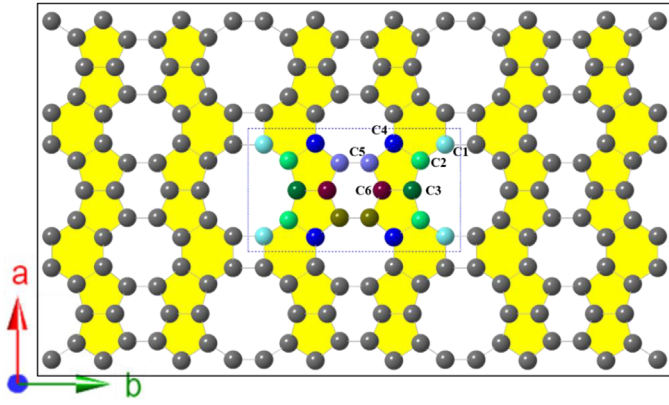


Fig. 3. Atomic configuration of net- π [24]. Different colors in the picture represent unequal carbon atoms.

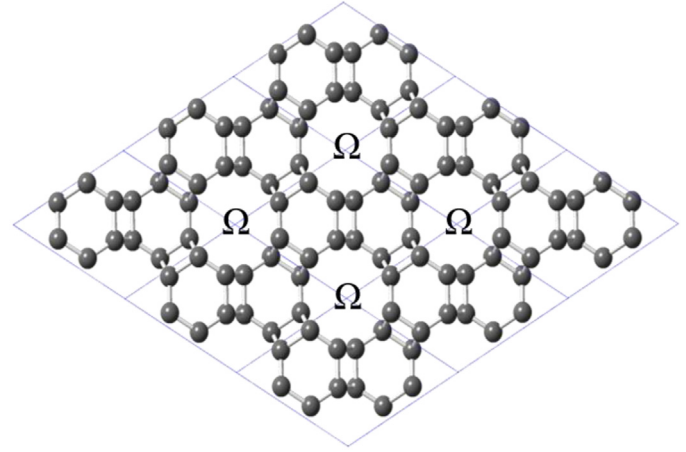


Fig. 5. Top view of the structures for the α -C [14].

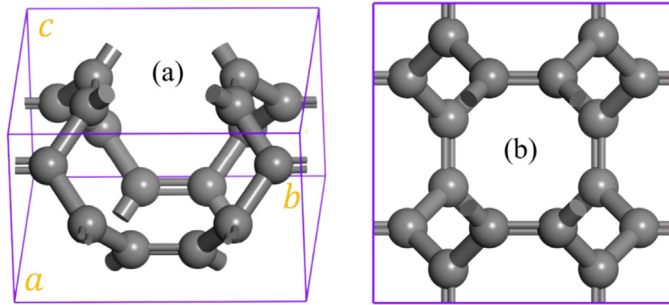


Fig. 4. Crystal structure of cT8 carbon (a) side view, and (b) top view [21].

perpendicular a and b crystalline directions. The relaxed lattice constants of cT8 are $a = b = 5.901$ Å and $c = 3.760$ Å, wherein the bond length of the single C–C is 1.476 Å and that of all the double C=C bonds is 1.341 Å. The cT8 has negative linear compressibility and is promising as a shock-absorbing material or even body armor.

2.2 sp^3 hybridization

Generally, the configuration of sp^3 is the primary candidate for designing the new carbon materials, with the aid of consideration of possible stacking sequences and the extreme conditions of high pressure/high temperature. As well known, the new carbon materials could not be energetically stable in comparison to the graphite and diamond, which usually are defined as being in a metastable phase. However, the metastable phase, which always exhibits the unexpected properties in comparison to the stable phase, could stably exist at the room temperature under the specific conditions. The acronyms of predicted carbon materials were usually defined according to the symmetry or the matrix. In the following, three typical carbon allotropes with sp^3 hybridization, including α -C, P-carbon and *Imma*-carbon, will be introduced in terms of structure and property.

2.2.1 Structure and property of α -C (Obtained from compressed Carbon Nanotubes (CNTs))

CNTs are one-dimensional quantum carbon materials with special structure. CNTs are a coaxial hollow seamless tubular structure formed by a single or multilayer graphite sheet coiled around the center at a certain Angle. Most of the wall tubes are composed of hexagonal carbon atom grids [9]. The transverse compression of carbon nanotubes could produce three sp^3 -hybridized carbon allotropes (α -, β -, and γ -C) [14]. The atomic structure of the α -C with symmetry of $P6_3/m$ is shown in Figure 5. One can see that each carbon interacts with four neighboring carbon atoms to form the sp^3 -hybridized bonds. Accordingly, the α -C could be obtained by transversely compressing the smallest (3, 3) CNTs with same directions and positions. The average bond length d (unit:) is 1.556 Å, which is similar with that (1.54 Å) of diamond. And α -C has an equilibrium density of 1.50 g/cm³ and a Vickers hardness of over 70 GPa. With its light weight and high hardness, α -C has the potential to be a versatile room-temperature superhard material.

2.2.2 Structure and property of P carbon

P carbon [19], occupies a tetragonal symmetry of $I4/mmm$ with the optimized lattice parameters of $a = b = 11.11$ Å and $c = 2.5$ Å. The atomic structure of P carbon in $2 \times 2 \times 1$ configuration is presented in Figure 6, one can see that there are two types of micropores (4 Å and 5.2 Å in diameter) in a radial arrangement, marked with red and green atoms, respectively. The average bond length of the C–C bond was 1.54 Å, which is equal to the diamond bond average length. P-carbon has superhard and robust properties (Vickers hardness reaches 86.7 GPa) with relatively low density. In addition, electronic analysis shows that P-carbon is a semiconductor with a direct band gap of 3.52 eV. Its unique combination of electrical and mechanical properties makes P-carbon a potential material in the semiconductor industry.

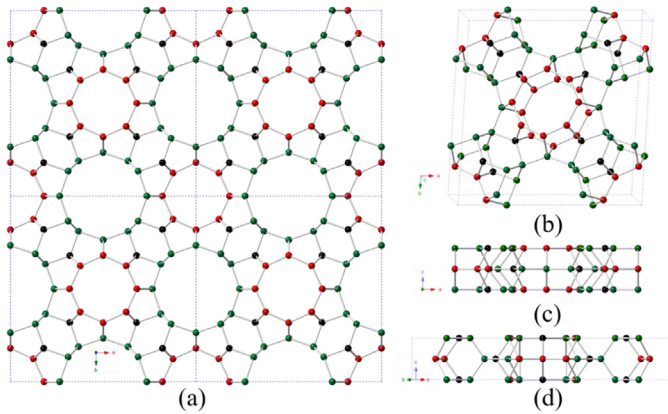


Fig. 6. (a) $2 \times 2 \times 1$ configuration of *P*-carbon with a microporous structure [19]. Two types of micropores with different diameters

2.2.3 Structure and property of *Imma*-carbon

With the aid of theoretical analysis, some novel carbon materials could be predicted under various conditions including pressure *etc.* For example, *Imma*-carbon [12] was identified using a developed methodology on the theoretical design of superhard materials based on the CALYPSO (Crystal structure AnaLYsis by Particle Swarm Optimization) by algorithm. The 2D projection of *Imma*-carbon with symmetry of *Imma* along the two different directions is presented in Figure 7. One can see that there are 24 carbon atoms with the $4+6+8$ even membered patterns in a unit cell, marked by the dashed rectangle. The optimized parameters of *Imma*-carbon at 0 GPa are $a = 7.5147$ Å, $b = 4.2363$ Å, and $c = 4.3173$ Å. Energetically, *Imma*-carbon is predicted to be more stable than graphite under pressures above 12.9 GPa, indicating that the metastable phase may exhibit distinct stability behavior in comparison to the stable phase under extreme conditions. *Imma*-carbon is a semiconductor with a direct band gap of ~ 2.6 eV. In addition, it has a high bulk modulus of 444.7 GPa and a Vickers hardness of 83.5 GPa. *Imma*-carbon provides a new idea for designing new strongly covalent superhard materials.

2.3 sp^2 and sp^3 hybridization

Both theoretical calculations and experimental investigations show that 3D assembly is an effective method to overcome the shortcomings of low dimensional materials. So, mixed hybridization of sp^2 and sp^3 configurations in one kind of carbon material has always been the main direction of scientific researchers. From this perspective many predicted results, wherein some results have also been proven experimentally, have been achieved with unexpected properties [29–31].

Viewed from the symmetry point, the six-membered ring is primarily employed as building units due to the good stability and lowest ring strain energy. For example, the hexagon is the only building block for carbon phases from zero-dimensional (0D) nanoflakes or nanorings to 1D nanotube, 2D graphene, and 3D graphite and metallic

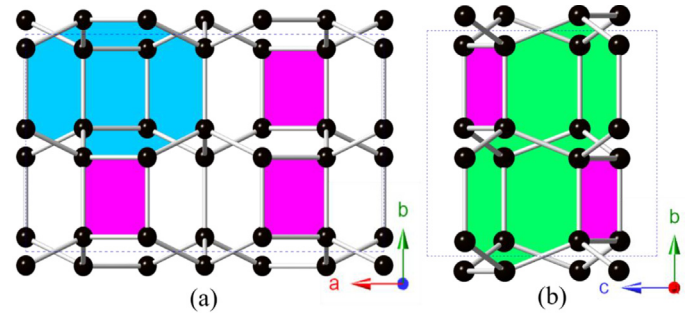


Fig. 7. (a,b) Polyhedral views of the crystal structure of *Imma*-carbon along two different directions, respectively [12].

carbon phases [32,33]. Extended carbon topology consisting only of pentagons is rare, and other forms of carbon shape are almost absent. Meanwhile, carbon pentagons are usually considered as topological defects or geometrical frustrations [34]. As stated in the well-known “isolated pentagon rule” (IPR) for fullerenes, the pentagons must be separated from each other by surrounding hexagons to reduce the steric stress [35]. For instance, fullerene C_{60} consists of 12 pentagons separated by 20 hexagons forming the shape of a soccer ball, which is a perfect footnote to IPR [7]. Moreover, the carbon pentagons are also separated by heptagons from each other in some cases [34].

In the following, carbon materials with sp^2 and sp^3 hybridization, such as Penta-Graphene, *Ph*-graphene, Tetrahexcarbon, C4-8, Hex-C18, *Imma*-C and oC48, will be introduced in detail.

2.3.1 Structure of Penta-Graphene

Penta-Graphene [15,36], which is composed entirely of pentagons and resembling the Cairo pentagonal tiling, is produced by stripping a single layer in T12-carbon. That is to say, Penta-Graphene is a 2D metastable carbon allotrope. The atomic structure of Penta-Graphene occupies the symmetry of $P-421m$ with a tetragonal lattice, which can be considered as a multidecker sandwich. The optimized lattice constants of the 2D Penta-Graphene are $a = b = 3.64$ Å.

2.3.2 Structure and property of *Ph*-graphene

In contrast to Penta-Graphene, *Ph*-graphene [18], is a new 2D pentagonal composition with a small percentage of C6-ring. As shown in Figure 8a, a quasi-2D membrane can be constructed by the formation of covalent bond between C_{36} fullerenes wherein there are two-layers thickness of membrane marked by red and black atoms. So, *Ph*-graphene can be constructed by cleaving the C_{36} quasi-2D membrane along the direction parallel to the basal plane (Fig. 8b). *Ph*-graphene possesses symmetry of $P6mm$ with the lattice constant of $a_0 = 7.05$ Å and the thickness of 1.54 Å, wherein the residual hexagons release the stress of

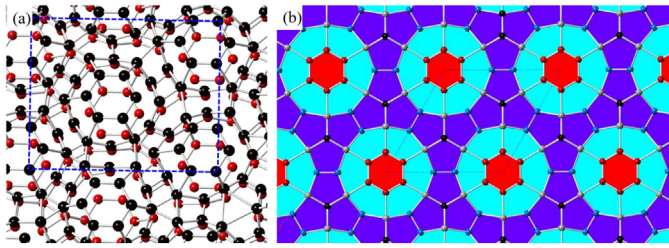


Fig. 8. Schematic representation of solid C_{36} quasi-2D membrane (a) and *ph*-graphene (b). The dotted lines indicate the region of the primitive cell [18].

pentagons. *Ph*-graphene has anisotropic negative Poisson's ratio and semiconductor properties, which are promising for flexible electronic devices.

2.3.3 Structure of tetrahexcarbon

Tetrahexcarbon [22], which is composed of tetra and hexarings of carbon atoms, is a novel 2D sheet. Unlike Pentagraphene, which is composed of only pentagons, Tetrahexcarbon exhibits a layer-type-structure with sp^3 -hybridized atoms (C2) sandwiched between two layers of sp^2 -hybridized (C1) atoms. The optimized lattice parameters of tetrahexcarbon are $a = 4.53$ Å and $b = 6.11$ Å. And the corresponding bond length between C1 and C2 are C1–C2 = 1.535 Å; C1–C1 = 1.34 Å. The angle between C1 and C2 is $\theta_{C1-C2-C1} = 135.5^\circ$ whereas the total layer thickness is 1.167 Å.

2.3.4 Structure and property of C4-8

In addition to carbon allotropes made of rings of more than five members, researchers have also looked at ternary or four-membered rings. But the results were not ideal, and the only dynamically stable carbon allotrope found was C4-8 and its derivatives [16]. C4-8, which is composed of C–C single and triple bonds linking cyclobutadiene or cyclopropene radicals, is formed by successful experiments, as shown in Figure 9. According to calculation of Density Functional Theory (DFT), the lattice constants (a or b) increase from 3.44 to 6.02 and 8.59 Å when the C4-8 formed by cyclobutadiene or cyclopropene radical ring linkage is optimized. And the C–C bond length of the cyclobutadiene ring is slightly longer than the bond length for the parent cyclobutadiene, but the C–C bonds connecting the cyclobutadiene rings show decreased bond lengths of 1.371 Å. C4-8 shows almost linear dispersion in the electron band diagram and is expected to have a small effective mass, suggesting that C4-8 should have good conductivity.

2.3.5 Structure and property of Hex-C18

In addition to the above structures, researchers have also found many carbon allotropes based on theoretical prediction from computer software of CALYPSO, which also have good properties and a wide range of application prospects. For example, Hex-C18 [20], which contains 18 atoms per unit cell, has a hexagonal structure with symmetry of $P6_3/mcm$. The optimized lattice parameters

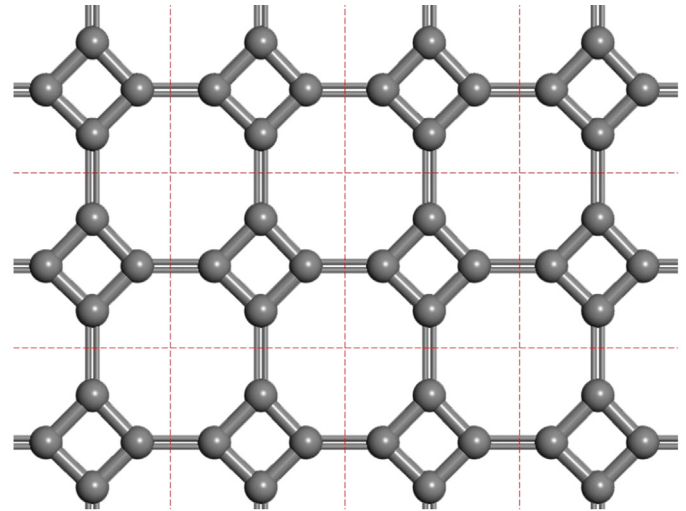


Fig. 9. Chemical structures of the C4-8 materials studied composed of octatonic- and four-membered rings [16].

of Hex-C18 are $a = b = 8.36$ Å, and $c = 2.46$ Å. Seen from Figures 10(a), 10(b), two chemically nonequivalent carbon atoms are presented in the primitive cell: one is the sp^3 -hybridized carbon atoms, colored in yellow, and the other is the sp^2 -hybridized carbon atoms, colored in red. So, Hex-C18 could be considered as the hybridization of the sp^3 and sp^2 carbon chains in a 3D assembly ([Fig. 10(c)]). Based on DFT calculations, the bond length among the sp^2 -hybridized zigzag chains is 1.41 Å, which is very close to that of graphene (1.42 Å), as well, the average bond angle of the sp^2 -hybridized carbon atoms is 120.5° , which is also very close to that (120°) of graphene, indicative of intrinsic similarity of 2D materials. Meanwhile, the bond length among carbons atoms in sp^3 -hybridized chains is 1.54 Å, very close to that of diamond (1.53 Å), while average bond angle of the sp^3 -hybridized carbon atoms is 110.5° , which is quite close to the ideal bonding angles of 109.5° in diamond. The hybridization of sp^2 and sp^3 leads to the high energetic stability of Hex-C18. The theoretical specific capacity of Hex-C18 is 496mAh/g, and the energy barrier of lithium ion diffusion is 0.018–0.142eV, indicating that Hex-C18 is a promising anode material for lithium ion batteries.

2.3.6 Structure and property of *Imma*-C

Imma-C [13] occupies the orthorhombic carbon crystal with symmetry of *Imma*. Based on the DFT calculation, the optimized lattice parameters of *Imma*-C are $a = 2.515$ Å, $b = 4.885$ Å, and $c = 4.154$ Å. As shown in Figure 11, the atomic arrangement of *Imma*-C, which has eight atoms in the unit cell, can be considered as being formed by '4-membered rectangles' sharing one d_2 bond, which are then connected by two d_3 bonds forming the '6-membered rings'. And each of them is four-fold coordinated with two d_1 bonds of 1.527 Å, one d_2 bond of 1.593 Å, and one d_3 bond of 1.479 Å. Through first-principles calculation, it is found that *Imma*-C is a kind of indirect band-gap crystal with a gap of 2.68 eV. Although it shows anisotropy, the hardness of Vickers reaches 75.6 GPa, which is expected to become a superhard semiconductor material.

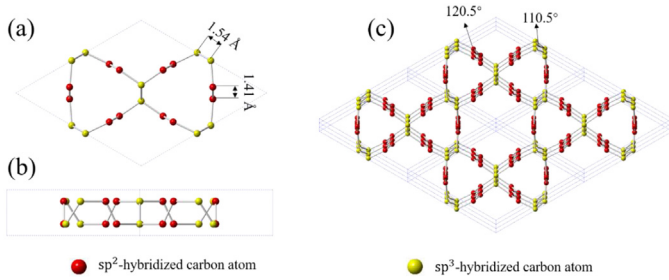


Fig. 10. (a) Top view and (b) side view of the optimized structure of Hex-C18; (c) A schematic illustration of a possible synthesis strategy for Hex-C18 by using C-H chains via dehydrogenation and assembly [20].

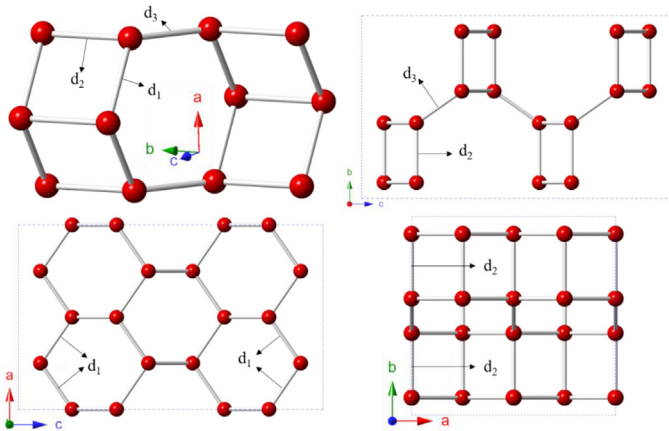


Fig. 11. Crystal structure of the new orthorhombic carbon ($Imma$ -C) [13]: (a) three-dimensional view of the unit cell; (b) view along the a axis, (c) view along the b axis, and (d) view along the c axis of the $2 \times 1 \times 2$ supercell.

2.3.7 Structure and property of oC48

oC48 [25], which was hybridized by sp^2 - and sp^3 - orbitals, has 48 carbon atoms in the unit cell, as shown in Fig. 12. The optimized lattice parameters of oC48 are $a = 9.4076$ Å, $b = 4.3435$ Å, and $c = 7.3597$ Å, obtained from DFT calculations. There are two chemically nonequivalent carbon atoms in the primitive cell, colored by black and purple spheres. Moreover, oC48 has four types of bond length, including: 1.4782 Å, 1.6551 Å, 1.5790 Å, and 1.3336 Å. It is worth noting that the black atom is four-coordinated, whereas purple atom is three-coordinated, indicative of the hybridization of sp^2 and sp^3 for carbon allotrope oC48. oC48 is predicted to have a hardness of 73.3 GPa, and band analysis shows that it is an indirect band gap semiconductor with a band gap of 1.464 eV, which is promising as a superhard material.

2.3.8 Others

New carbon allotropes can be predicted by modifying the type of bond in the original structures by adding atoms or connecting multiple existing structures with covalent bonds. The original structures could be predicted theoretically or prepared experimentally. For examples, Tri-C₁₈,

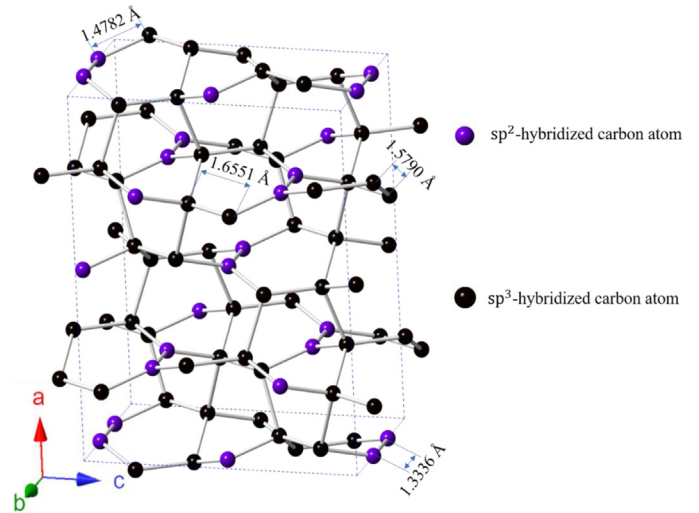


Fig. 12. Global view of the crystal structure of oC48 in a unit cell [25].

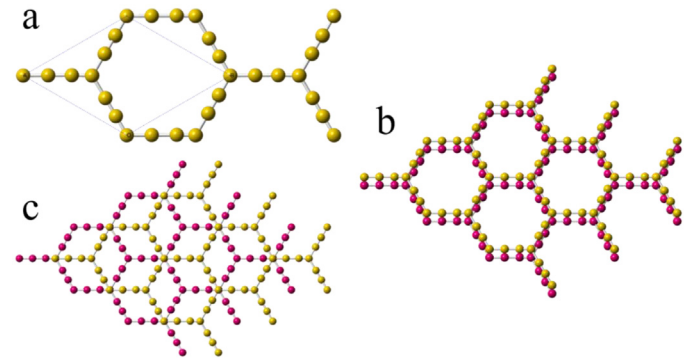


Fig. 13. (a) Top view α -graphyne monolayer ($N=2$); (b) and (c) stand for the AA stacked and AB stacked of bilayer α -graphyne with $N=2$. The yellow and pink atoms indicate the top and lower layer in-plane positions, respectively [17].

Hex-C₃₆, Tri-C₅₄ and Orth-C₇₂ could be designed by graphdiyne based on the prediction of CALYPSO [17]; 1-Yne, 2-Yne and 3-Yne C₄ carbon are predicted by substituting carbon-carbon bonds for carbon-carbon triple bonds on the basis of the structure of body centered tetragonal (Bct) C₄, wherein the triple bond of carbon is called the yne-bond [11]. α -graphyne could be constructed by replacing all the covalent bonds of graphene by sp^2 - sp^1 linkages, as shown in Figure 13a. Furthermore, the different stacking patterns AA and AB could enrich the system via weak van der Waals interactions, which is similar to the case of graphene bilayers (Fig. 13b, c) [37].

In addition to exploring new carbon allotropes, the researchers also studied transitions between different hybrid states. The two main methods of transforming graphene into two-dimensional diamond (sp^2 - hybridized to sp^3 - hybridized) [38–39] are chemical functionalization and pressure [40]. The graphite-to-diamond transformation is governed by the formation of nanoscale coherent interfaces (diamond nucleation), which, under static compression, advance to consume the remaining graphite

Table 1. Bulk modulus B (GPa), shear modulus G (GPa), Young's modulus E (GPa) and Vickers hardness H_v (GPa) of some carbon allotropes.

Name		B	G	E	H_v
Diamond [44]		467.4	550.1	1185.3	94.0
cubic-BN [44]		403.0	411.5	921.0	66.3
Imma-carbon [12]		444.7	498.3	1088.4	83.5
γ -C [14]		342		759	72.4
P-carbon [19]		334.2	360.1		86.7
Hex-C18 [20]		282.9	254.6	1122.9	80.8
	O12	433	520	1113	96.1
O-type carbon [23]	O20	362	294	695	48
	O28	382	351	806	59.2
T-type carbon [23]		407	369	851	62.6
<i>oC48</i> [25]		353	340	773	73.3

(diamond growth) [41]. Moreover, the metastable phase which appears during the transition process and always occupies the specific crystalline structure, should be further investigated in order to uncover the underlying mechanisms and discovering new carbon materials with unexpected properties.

3 Properties of carbon materials

The free combination of different hybrid states of carbon or the embedding of other atoms in carbon hybrids can create a wide variety of structures and hence properties. In this section, the latest progress in mechanical, electronic, and optical properties of carbon materials are introduced.

3.1 Mechanical properties

As well known, a large number of carbon allotropes with sp^3 bonds, like diamond [42], tend to be extremely hard but have poor toughness properties, this could be attributed to the strong covalent bonds among them. For a large number of carbon crystals containing sp^2 bonds, the mechanical properties include generally excellent flexibility. Typically, hardness of graphite, is relatively lower than that of diamond due to weak van der Waals interaction between the layers [39]. Moreover, the study shows that Young's modulus of graphene decreases with the increase of acetylene bond percentage [27]. It is found that the doping of different atoms in sp^2 hybrid states will also have a significant impact on its stiffness. For example, the doping of N atoms will increase the stiffness of carbon materials while the doping of B atoms will decrease its stiffness [43]. Through the combination of sp^2 / sp^3 hybrid states and being composite with other materials, carbon materials have made a great breakthrough in mechanical properties while maintaining high stiffness and hardness to achieve the maintenance of other properties. Like P-carbon, the yield stress reaches 109.33 GPa, the bulk modulus reaches 344.2 GPa, and the hardness reaches 86.7 GPa, while the direct band gap is 3.52 eV, making it a material with great

potential in the semiconductor industry. Some of the high hardness/stiffness carbon materials discovered in the last decade and their specific properties are listed in Table 1.

3.2 Optical properties

At present, the attentions on the optical properties of carbon materials is mainly paid on the solar cells where carbon materials are generally used as the absorption layer, since optical absorption is one of the most fundamental optical properties of materials [45]. The quantum efficiency of a solar cell is strongly dependent on the optical absorption coefficient, and the hybrid sp^2 - sp^3 carbon allotropes have larger light absorption coefficients and higher efficiencies in comparison with other known light absorbers. Also carbon quantum dots have also made great progress in photovoltaics [46,47].

3.3 Electronic properties

Graphene is a zero bandgap semiconductor material with a Dirac cone. Inspired by graphene research, great success has been achieved in developing the electronic properties of carbon materials. It has been found that most carbon materials have a great potential in conductivity. At present, the following methods are mainly adopted to measure the electronic properties of materials: electron localization function (ELF) [20,48], carrier velocity by PBE band [20] and cyclic voltammetry (CV) [49–56]. The first two are generally used to measure only carbon materials and carbon semiconductors that have not been prepared, while the latter are generally used to measure carbon materials or carbon composites that have been prepared. ELF is a uniquely defined dimensionless quantity, and only takes values in the range between 0 and 1, where 1 refers to the fully localized electrons, and 0.5 corresponds to fully delocalized electrons, while 0 represents very low charge density [57]. Moreover, mobility is a key parameter that characterizes the electrical properties of semiconductor materials, it also determines the applications of semiconductor materials in the field of electronics. Carbon

materials with highly anisotropic transport properties can conduct electricity [20]. Additionally, the doping of carbon material with other atoms would also have a significant impact on their electronic properties. For example, the doping of N can accelerate the conductivity [58–60]. The results confirmed the excellent performance of carbon composites in conducting electricity through the CV method calculation, thus indicating their great potential in electronic fields such as supercapacitors.

3.4 Other properties

In addition to the above performance, carbon materials also have quite superior performance in heat, catalysis, adsorption and other aspects. Most carbon materials have a porous structure [61], which leads to their unique advantages in adsorbing solid impurities and even gas separation [27]. Due to their intrinsically high stability and ability to combine with various atoms, carbon materials also have excellent performance in heat conduction [62] and catalysis [63].

4 Design and synthesis of carbon materials

4.1 Theoretical prediction of the new carbon allotropes

For the time being, there are two main methods used to find new carbon allotropes. One is to predict new structures based on algorithms, among which the main algorithms are Particle Swarm Optimization (CALYPSO) [13,17,20,25], Genetic Algorithm (GA) [64], Universal Structure Predictor Evolutionary Xtallography (USPEX) [65], Ab Initio Random Structure Searching (AIRSS) [66] and the multi-objective inverse band structure design method [45]. CALYPSO, is a code designed by Prof. Yanming Ma Group in Jilin University to search for stable or transferable crystal structures using only the chemical composition of a given compound at a given pressure. The CALYPSO software is free to get [67]. GA is similar to Darwin's theory of "survival of the fittest by natural selection", in which space groups represent the environment and crystal coordinates represent evolving DNA [64]. USPEX is a powerful tool for crystal structure prediction, wherein the energetically stable structure and a set of low-energy structures at given pressure-temperature conditions could be provided [68,69]. Similarly, AIRSS could also predict stable and metastable structures of crystals and clusters including atomic positions at point defects in solids by relying on the repeated generation of random structures near the interface [70–72].

The other method is to apply pressure to an existing structure to change it into a new one [73]. In both theory and experiment, compression is a good way to find new carbon allotropes. In theory, a variety of lightweight superhard carbon allotropes can be obtained by arranging stacked carbon nanotubes in different ways and adjusting the compression position and angle [14]. In the experiment, superhard carbon conductors could also be obtained by

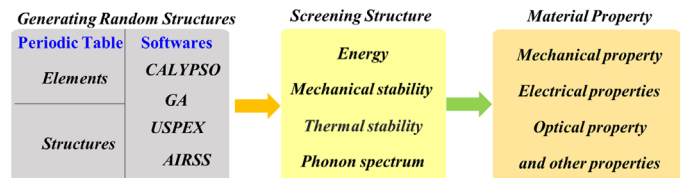


Fig. 14. The process of predicting new materials

Fig. 14. The process of predicting new materials.

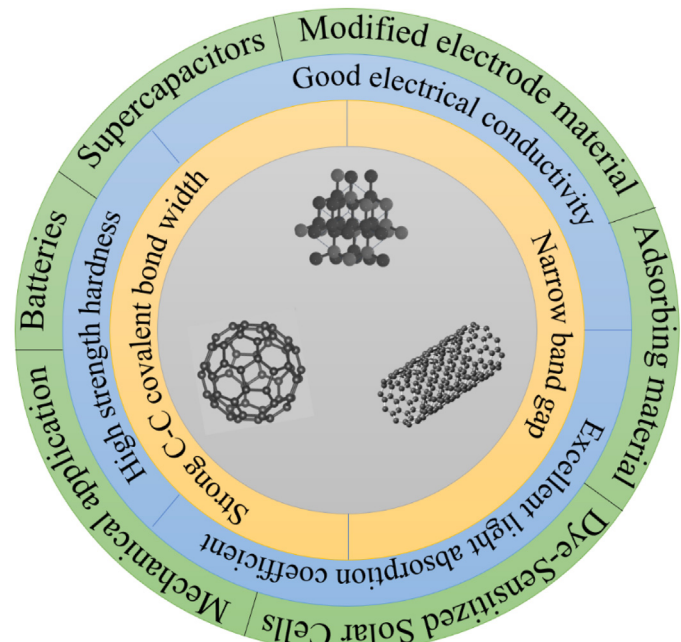


Fig. 15. Schematic illustration of the intrinsic properties and their applications.

compressing the mixture of carbon black and tetraethylene, these can even be used as multi-functional materials under extreme conditions [74].

There are generally three steps for theoretical prediction of new carbon allotropes. Firstly, the candidates of random structures were generated according to the chosen elements and lattice structure; Secondly, the candidates were screened from various criteria including energy, mechanical property, thermal stability and phonon spectrum. Generally, the “Energy” is always considered as the primary factor, that is, the stable materials occupy the relatively minimum total energy. Finally, the materials property of predicted carbon materials could be further determined based on the analysis of total energy and calculation of electrons distribution at the ground state. The general process of theoretical prediction of new materials was shown in Figure 14.

4.2 Synthesis of carbon materials

Great progress in the theoretical study of carbon materials have been made, but turning theory into reality is a significant problem for carbon materials researchers. Early

preparation methods either produced carbon materials with poor performance, or carbon materials with satisfactory performance but expensive which were not suitable for use in normal production. Recently the following methods are commonly used in preparing carbon materials with desirable properties: template synthesis [75–84], hydrothermal [85–87], pyrolysis [88,89] and chemical activation [80,90].

4.2.1 Template synthesis

Template synthetic approaches are extensively applied to synthesize nanoporous carbon materials. In general, the template synthetic procedure for producing mesoporous carbon, which was first proposed in 1999 [91,92], could be finished as following procedures: (i) Preparation of the mesoporous template; (ii) Removal of the surfactant; (iii) Synthesis of a carbon precursor/silica nanocomposite; (iv) Carbonization; and (v) Removal of the template [84].

In this method, the template and precursor should be determined by the desired properties of the material. Distinct properties of the carbon material could be obtained by adopting a different carbon precursor [93–95]. For example, taking sucrose as a carbon precursor, materials tend to have a high volume of micropores and uniform distribution of the holes in the network [96]. In contrast, the carbon precursors of polyacrylonitrile can improve the capacity performance of materials [97–99].

4.2.2 Hydrothermal

The hydrothermal method is to apply a high temperature and high pressure aqueous solution to make those insoluble or insoluble substances under atmospheric conditions to dissolve or react to generate the dissolved products of the substance. This is done by controlling the temperature difference of the solution in the autoclave to produce convection to form an oversaturated state and precipitation of crystal growth. In the field of carbon materials, this method is often used as a preparation for atomic doping or as a bridge between nano-carbon materials [85–87].

4.2.3 Pyrolysis

During the pyrolysis process, a dry solid carbon source with pre-carbonization is broken down into constituent materials and impurities at high temperature. The resulting mixture needs to be rinsed with deionized water or some other solvent to get the desired final material. However, this requires that the materials to be prepared have good thermal stability [79,89], this method is often used to prepare materials with good adsorption properties [88,100].

4.2.4 Chemical activation

High purity of carbon materials often does not have as good conductive properties as expected, which seriously hindered their applications in electrode materials. Generally, two main methods are proposed for improving

its electrical conductivity as an electrode material: one is to couple carbon materials with other materials, the other is to activate the carbon materials via chemically functional groups [101]. Chemical activation generally refers to the activation of reactants in the way of chemical reactions, so that its performance in a certain aspect is enhanced. For example, the activated carbon is basically microporous when preparing activated carbon with flamboyant pods as raw material and NaOH as activator [80].

4.2.5 Other synthesis methods

In addition to the general methods, the researchers also found some suitable preparation, such as electrophoresis [102,103], embedding method [104], acid etching [105], etc. These methods all have unique advantages for the preparation of the carbon materials after a variety of solutions. Unfortunately, some preparation methods contain the potential for contaminated waste which should be avoided [106]. For example, a new type of activated carbon material can be extracted from oil sludge, which could be adopted for removing toxic dyes from an aqueous solution [107]. Furthermore, the multi-walled carbon nanotubes (MWNTS) were used to remove methylene blue from wastewater [108].

4.3 Gap between theoretical prediction and experimental fabrication

Presently, the achievements of theoretical prediction in designing carbon materials is far ahead of the experimental counterparts. The gaps between theoretical prediction and practical production can be roughly summarized as following two aspects:

1) New carbon materials were usually predicted under the extreme conditions, such as high pressure (hundreds of GPa, even 1 TPa) and high temperature (above 2000 K), which is difficult for fabrication in experiments. For instance, the pressure is usually adopted to be the driving force for discovering the new carbon materials, which is corresponding to the environments in the Earth's inner core (150 GPa, 250 GPa, and 350 GPa) [109]. However, the diamond anvil cell, which has been widely applied in experimental compression, only provided pressure with tens of GPa [110].

2) The theoretically predicted carbon materials are usually in format of single crystal, perfect without any defects. And the defects could not be avoidable in experimental aspect, which would affect the thermal stability of expected materials. Moreover, the size of single crystal with mm scale, which is the fundamental requirements for engineering applications, was difficult for experimental preparation in comparison to the polycrystalline counterparts.

For accelerating the development of experimental fabrication, more attentions should be paid on not only the progress of new technology in devices and equipments, but also the theoretical investigations of new materials with defects or in format of polycrystalline.

5 Applications of carbon materials

As described in Section 3, the multitudinous properties of carbon materials render them with wide application in various fields as shown in Fig. 15. In this section, the typical applications of carbon materials in lithium-ion batteries, supercapacitors, solar cells, adsorbent materials, and mechanical applications are roughly introduced.

5.1 Lithium ion battery

Lithium ion batteries have attractive advantages due to high energy density, low maintenance, and relatively low self-discharge, etc. In order to improve the performance of lithium ion batteries, it is urgent need to design and synthesis of novel electrode materials that have excellent lithium storage behavior. Traditionally, enlarging the high surface area is an effective strategy since it is favorable for the storage of lithium ions. Moreover, the discovery of graphene, which can promote the transportation of lithium ions and electrons, also made a great step in improving high conductivity. In recent decade, porous graphene frameworks can further enhance the electrochemical stability of the electrodes due to extra-space for accommodating the large volume change during cycles of charge-discharge. Consequently, these porous graphene materials hold promise as electrode materials for lithium ion batteries [61]. Building on their work with graphene, researchers have found two more carbon materials suitable for use as negative terminals in lithium-ion batteries. Due to the inherent metallic demeanor and regular porosity, Hex-C18 has a lower lithium ion diffusion barrier and a higher lithium ion capacity than graphite (the theoretical specific capacity of Hex-C18 is 496 mAh/g, which is 1.3 times as large as that of graphite) found in new lithium-ion batteries [20,111,112]. All above carbon materials have shown their great application potential in the field of lithium-ion batteries.

5.2 Fuel cells

C element has a four-electron in shell, and the kinetic transfer process is sluggish. So, it is key to discover highly active catalysts for fuel cells and metal-air batteries with high-efficiency [61]. For example, Au nanoparticles can be directly carbonized by zinc oxide nanorods to modify porous hollow tubular carbon materials (PHTCs@Au) [113]. It was found that the modified enzyme electrode shows not only good catalytic activity towards glucose oxidation but also excellent long-term stability; the Ni-Pd/Cr₂O₃-C electrocatalyst has a higher hydrogen desorption peak due to the promotion of metal oxides on graphite carbon [106]. The fuel cell electrodes prepared by the above two methods have high catalytic activity and have great advantages in the preparation of fuel cells in future. Finally, the carbon coating is also an important application of carbon materials in fuel cells to protect the electrodes. For example, the surface of 316L stainless steel coated with a nanocomposite carbon layer can make fuel cells more efficient, which can be attributed to lower interfacial

contact resistance even in corrosive anodic and cathodic environments [114].

5.3 Other batteries

In addition to the common use of carbon materials in the above two types of batteries, carbon materials are also favored in sodium and chloride ion batteries because of their good corresponding ion storage characteristics and good stability. Finding and optimizing the appropriate cathode materials in the electrolyte are critical to the development of chloride ion batteries. For example, the prepared positive material of PANI/50CNTs, which were prepared by in situ chemical oxidation polymerization, had uniform distribution of element and a maximum reversible capacity, indicating that the modified material can be used as cathode material for a chloride ion battery [115]. Moreover, reduced graphene oxide/carbon nanotubes, which is fabricated via a simple freeze drying and subsequent thermal treatment in a nitrogen atmosphere, has been applied as an anode for sodium-ion batteries for the first time [116].

5.4 Supercapacitors

Supercapacitors, which have a much higher energy density than electrostatic or electrolytic capacitors, allow for the ultrafast and highly reversible storing and releasing of energy. However, the limited energy and power densities has seriously hindered the widely applications of supercapacitors. It is thus of high demand for discovering electrode materials with high capacity [61]. Generally, the performance of electrode materials for supercapacitors is intimately dependent on both the accessible specific surface area and the pore structure [117]. In this respect, carbon materials undoubtedly have a huge advantage [118]. As mentioned in Section 3.4, many carbon materials have a highly open porous structure that allows the electrolyte to enter the surface of the porous frame [26, 27,119]. Also carbon materials generally have low density and high strength to weight ratio, as well as its ability to combine with various atoms through different hybrid orbitals (sp^1 , sp^2 and sp^3) [120], which makes their application prospects in supercapacitors very wide [10,121]. In addition, it also has excellent stability, at a high current density of 50A/g for more than 200,000 cycles, the capacitance retention rate can reach 94.6%.

5.5 Modified electrode material

In addition to being used as an electrode in batteries, carbon can also be used as an electrode material in measuring instruments. Many of today's most sensitive sensors are made of carbon materials [55,122]. Carbon materials sensors can often be sensitive, selective, simple, low-cost and stable due to the excellent stability of carbon materials [80]. In the process of detecting biological small molecules it also showed high sensitivity, good stability, and a strong anti-interference ability [113,123]. Moreover,

as a new electrode material for the electrochemical determination of hydroquinone (HQ) and catechol (CC), graphene carbon nanosheets (GCN) have been found to obtain satisfactory recovery rates for the determination of HQ and CC in real water samples [124].

5.6 Adsorbing material

Coagulation, flocculation, biodegradation, membrane separation, ion exchange, oxidation/advanced oxidation, adsorption methods, *etc.*, are generally adopted for treatment of all kinds of pollutants treatment methods and processes [125,126]. Carbon materials are often used as adsorption materials because of their lower density, higher strength to weight ratio, specific surface area, and porous structure with a variety of morphology and interesting properties [127]. In addition, carbon materials have the ability to combine different hybrid orbitals with various atoms, so carbon materials are not only useful for adsorption of solid pollutants [117,128], but also can be used in gas separation in addition to heterogeneous ions [61]. For examples, carbon fiber modified with Mn and Mo can experimentally purify arsenic in arsenic solution with a low concentration; the decontamination effect of 3-chlorophenol solution in the heterogeneous Fenton-process is improved using Fe/Fe oxide modified carbon fiber as a catalyst [129]. Also carbon materials can even be used as a hydrogen storage material because of their strong adsorption capacity [130].

5.7 Solar cell films

Carbon materials are also mainly used in solar cells as electrode materials (described above) or as solar absorbers based on their excellent light absorption coefficients [114]. For example, five carbon allotropes (C10-C, C14-C, C24-C, C20-D and C24-D) with direct or quasi-direct band gaps ranging from 1.01 eV to 1.58 eV have been found for photovoltaic applications recently. In the range of visible light, the optical absorption coefficients of C10-C and C24-C are one order of magnitude higher than that of GaAs, indicative of its potential application as the absorption layer materials of thin film solar cells [45]. In addition, carbon quantum dots have been developed as a new hole transport material for perovskite solar cells in the nanometer field.

5.8 Mechanical applications

Just like in Section 3.1, the free combination of sp^2 and sp^3 allows carbon materials to have good mechanical properties while also having excellent properties in other aspects, making super-hard materials that can be used in extreme conditions a possibility [131]. In addition, many materials with negative compression coefficients have been found, which can become promising shock absorbers [21,132], also some carbon materials have properties close to the human cortical bone, making this material a potential candidate for fixation of long bone fractures [133]. The mechanical applications of carbon material is also an important aspect

in consideration of strong bonding nature of sp^3 hybridization. For instance, Diamond-like carbon (DLC) coating has been widely applied in protecting the devices or matrix [134]. And, the mechanical property is generally considered to be the fundamental requirement for designing carbon-based materials. Once the new carbon-based materials are predicted or fabricated, the mechanical properties, including the yield stress (σ_s), Young's modulus (E), and hardness (H_v), etc, are primarily assessed for further applications.

6 Conclusions and outlook

As summarized above, carbon materials have been widely used in various fields because of their diverse structure and various properties. Although there have been great advances in various aspects of carbon materials, several shortcomings remain to be solved.

- The theoretical research on the structure of carbon materials is far ahead of the experimental research. How to realize the theoretical structure in the experiment/engineering fields will be the main problem that researchers have to go through. Current techniques for predicting carbon materials are relatively sophisticated, but the mature methods for producing them are still crude. It is important to develop new sophisticated instruments for producing carbon materials.
- Porous structure is one of the characteristics of carbon materials. However, it is a challenging task to accurately and uniformly prepare pores. The design and preparation of precise porous carbon materials has become an inevitable problem to significantly combine carbon materials with other materials and atoms. Moreover, achievement of the exact sequencing of different atoms in the same carbon material will be an important direction of porous carbon materials research in future.
- The exploration of nanoscale carbon materials is insufficient. At present, the latest research on carbon materials at the nanoscale level is the study of carbon quantum dots (QDS). However, the development of QDS is not perfect, and only partial properties of QDS in the field of electronics and optics have been studied [46–47].
- Although carbon is already widely used, it should be even more useful as nature's most attractive element. In spite of wide applications in electronic devices, optical films and other fields, it is not generally used in medical treatment, prosthesis, implants, decoration and other new industries. Moreover, carbon materials can play a multi-coupling role even in previously mature fields with continued research and the discovery of new properties.

Based on the current development of carbon materials, future research should be focused on:

- 1) The fundamental theory and algorithms for designing the desired properties of carbon materials including strength, ductility, optical, and electrical. For example, a new strength standard has been recently presented for considering structural defects [135]. Also the algorithm should be deduced from the atomistic information of the structural unit composed of $sp^1/sp^2/sp^3$.

2) For application of carbon materials in the medical field, sensors, prosthesis and implants made of carbon materials can have a high sensitivity and stability for the detection of small biological molecules. A recent article on the use of carbon materials in cancer treatment as seeds for brachytherapy undoubtedly opens a new door for this field of research [136].

3) Research on nano-carbon materials [62,137]. Although carbon nanotubes were proposed in 1991 [9] there have been few applications. Carbon nanotubes are already used in liquid antifreeze [62], to improve the mechanical properties of composite materials [138] and in capacitors [139]. Their excellent performance is also believed to have a huge application potential in optics, electronics and other fields. The related research on QDS is undoubtedly more important in this aspect [46]. The latest research on QDS proves that they can be widely used in optical fields as well as in electronic fields [47].

Therefore, nanoscale carbon materials have excellent properties in various fields. What we need to do in the future is to fully develop their properties and apply them in appropriate situations.

Based on the research work summarized in this review, it is not difficult to see the extraordinary potential of carbon materials. With their rapid development research and synthesis, they have shown excellent performance in the fields of electronics, thermal science and others, which leads us to believe that with continued research, their commercial use will be more fully realized in future.

Funding information

This review is supported by the National Natural Science Foundation of China (Grant No. 11972097).

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Cite this article as: Jiang Li, Deqiang Yin, Yi Qin, Carbon materials: structures, properties, synthesis and applications, *Manufacturing Rev.* **10**, 13 (2023)