石墨炔及其合成设计

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摘 要: 石墨炔是一种同时含有 sp 和 sp^2 杂化碳的二维全炭材料,是目前为止继石墨烯之后受到关注最多的碳的同素异形体。由于其独特的电子结构,石墨炔具有很多潜在的应用和独特的性能。本文概述了石墨炔的概念及性质,综述了石墨炔的各种独特的性能及其潜在的应用,提出了合成设计的一些方案与思路,并对石墨炔研究的前景进行了展望。

关键词: 石墨炔;性能;潜在应用;合成设计

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A review of graphynes: properties, applications and synthesis

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Abstract: Graphyne, a kind of sp-sp^2 hybrid all-carbon two-dimensional material, is currently one of the most interesting carbon allotropes besides graphene. It has potential applications and characteristic properties because of its unique electronic structure. First, the concept and properties of graphyne are summarized, then the characteristic properties of graphynes and their potential applications are reviewed, before some methods and ways to synthesize the two-dimensional structures are proposed, and finally a short perspective on the study of graphynes is given.

Key words: Graphyne; Performance; Potential applications; Design of syntheses

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1 Introduction

Carbon is one of the richest elements on the earth, almost all organisms are composed of carbon skeleton, and carbon materials are widely used in life[1]. Carbon materials are one of the earliest elements that human beings came into contact with. Since human beings learned to use fire, the use of carbon materials has been accompanied by human beings. Since the 16th and 17th centuries when graphite was found to be used for writing and painting, carbon materials have opened a wide range of relevance to human life. The unique electronic arrangement of carbon makes it have many unique properties. Carbon materials show unique characteristics different from other materials in mechanical properties, electrical properties, thermal conductivity, and optical properties and so on. Carbon also has many allotropes due to its special electronic structure and hybrid modes (Fig. 1)[2]. Among the allotropes of carbon, there are not only the hardest materials known in nature, but also the softest materials, electronic insulators, semiconductors and electronic conductors, insulating materials and good thermal conductors, and fully light absorbing materials and almost transparent materials. In addition to the graphite, diamond and amorphous carbon, carbyne[3-6], fullerene[7], carbon nanotubes[8], and graphene[9] are well known carbon allotropes.

Since the discovery of carbyne in 1960s and 1970s, fullerene in 1980s and carbon nanotubes in

1990s, the discovery of graphene at the beginning of the 21st century makes carbon materials become the focus of attention again. In the past decade or so, scientists have devoted themselves to the study of graphene, and research literatures related to the characteristics and applications of graphene have been continuously published. The discovery of graphene won the Nobel Prize in physics in 2010 for "the groundbreaking experiments regarding the two-dimensional material graphene". The discovery of graphene not only makes graphene and its derivatives (Fig. 2) become a hot spot[10], but also brings extensive attention to researchers in two-dimensional materials[11,12].

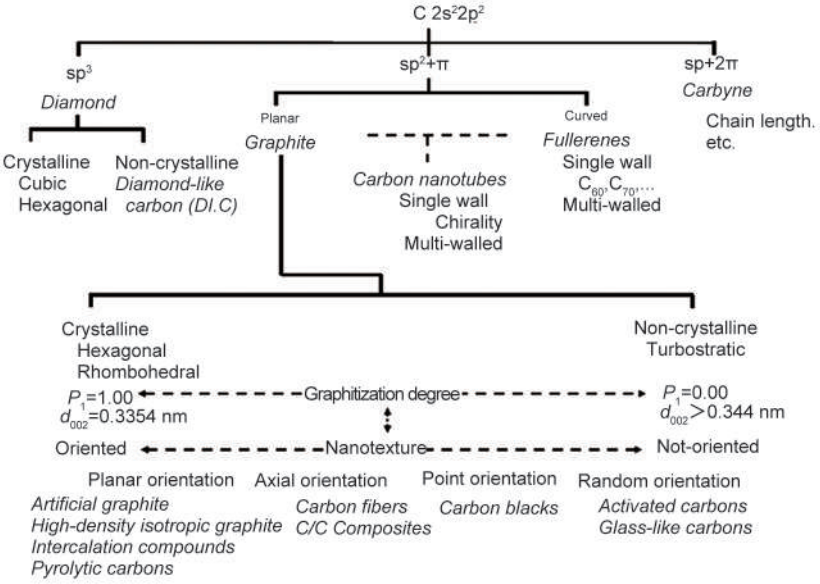

Fig. 1 Classification of carbon materials[2]. Reproduced from Elsevier Inc.

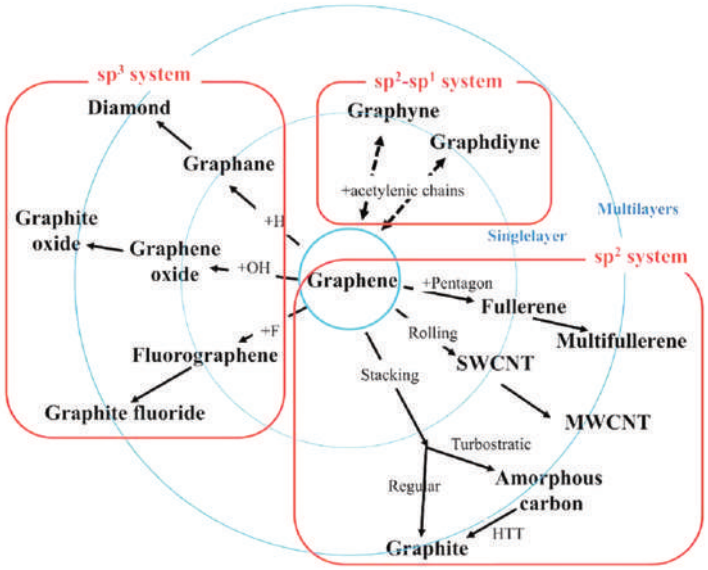

Fig. 2 Graphene derivatives[10]. Reproduced by permission of The Royal Society of Chemistry.

With the development of graphene, more and more attention has been paid to layered materials with a two-dimensional topological structure. On the one hand, due to the unique valence electron structure of carbon, the exploration of new carbon allotropes has always been the goal of researchers. The discovery of graphene has laid a solid foundation for the exploration of new carbon allotropes. On the other hand, the discovery of graphene fills in the gap of two-dimensional structure in carbon materials, which makes carbon materials become materials with zero dimensional, one-dimensional, two-dimensional and three-dimensional structures. Moreover, due to the unique topological properties of graphene as two-dimensional structural materials, researchers have carried out more extensive and in-depth research on new two-dimen-

sional topological structures. Because carbon atoms have three different hybrid modes, a variety of novel all carbon network structures and carbon allotrope structures can be designed theoretically by combining carbon atoms with different hybrid modes[13,14]. Due to the rise of graphene, carbon nanotubes and other sp^2 hybrid carbon materials, the current research on carbon materials is mainly focused on sp^2-sp^3 hybrid region, while the research on sp-sp^2 hybrid carbon materials is relatively less.

Graphyne (Fig. 2) is the representative of the sp-sp^2 hybrid carbon allotrope. It is the first carbon allotrope containing carbon-carbon single bond, carbon-carbon double bond and carbon-carbon triple bond. It is also an unnatural carbon allotrope most likely to be obtained by artificial synthesis. The carbon-carbon triple bonds formed by sp hybridization are all with a linear structure, and have the advantages of high conjugation and no cis-trans isomers. Therefore, people have been eager to obtain new carbon isomers containing the sp hybridization. Carbyne, which emerged in the 1960s and 1970s, is an important representative. In the process of synthesis new carbon materials, organic chemists have made great contributions to the structure of all carbon molecular network containing sp – sp^2 hybrid carbon atoms. Organic chemists pay more attention to the synthesis theory and the synthesis and design of monomer, dimer, oligomer, structural unit and other polymerization precursors.

The design of new all carbon network structure compounds should follow the following basic principles[15]. The designed network structure should have small tension and is not easy to convert to stable carbon allotrope graphite or diamond. The new compound should have excellent material properties. It should have potential synthesis methods, the preparation and characterization of monomer, dimer, polymer can be used to infer the structure and properties of the final network compounds. Graphyne structure has small steric hindrance and can be stable at room temperature. It has excellent electrical and optical properties. The two-dimensional extended graphyne structure can be theoretically synthesized by terminal acetylene coupling polymerization, and the final structure can be inferred by the preparation and characterization of its structural unit, monomer and oligomer. This indicates that graphyne can meet the design principle of all carbon network compounds.

It is 10 years since the first synthesis of graphdiyne, but the preparation is still a constraint to the development of graphyne. The traditional synthesis methods have their inevitable shortcomings in crystallinity and large area controllable synthesis. Actually, organic chemists have synthesized many structural units, precursors and oligomers of graphynes[16-20]. The current review of graphyne[10, 21-25] is mostly focused on the collation and synthesis of existing experimental results, and the introduction of the synthesis and application of graphyne like carbon materials containing sp-sp^2 hybrid. In this paper, the concept and properties of graphyne are elaborated. The properties and potential applications of graphyne are summarized. Some ideas for the synthesis and design of graphyne are proposed. The existing problems are briefly described and the prospect of graphyne is envisaged.

2 The concept of graphyne

The name of graphyne comes from acetylene bond and the graphite-like two dimensional unit in its structure, which was proposed by Baughman et al. at 1987[26]. Graphyne can also be seen as graphene partially substituted by carbon acetylene units (Fig. 3) to form graph-yne, graph-di-yne, graph-tri-yne and so on, depending on the length of acetylene bonds[27]. It is well known that graphene is an all carbon structure with extension of the benzene structure in two dimensional directions, both carbon-carbon single bonds and carbon-carbon double bonds exist in the molecular. While in the molecular of graphyne, carbon – carbon triple bonds exist besides single and double carbon-carbon bonds.

Actually, the structure of graphyne is not constrained in a six-fold symmetry structure composed by benzene ring and carbon-carbon triple bonds. Graphyne can exist in many structures by different constitutions of carbon-carbon bonds (Fig. 4).

3 The properties and applications of graphyne

3.1 Properties of graphyne

Although defect free graphyne still needs a long way to achieve, its properties have been extensively studied by theoretical physicists and chemists. A large number of references concerning theoretical study on graphynes greatly promote experimental works. The introduction of sp hybrid carbon – carbon triple bonds in graphene matrix brings some special properties on graphyne structures.

First, the introduction of the carbon-carbon triple bond enables a larger in - plane porosity in graphyne

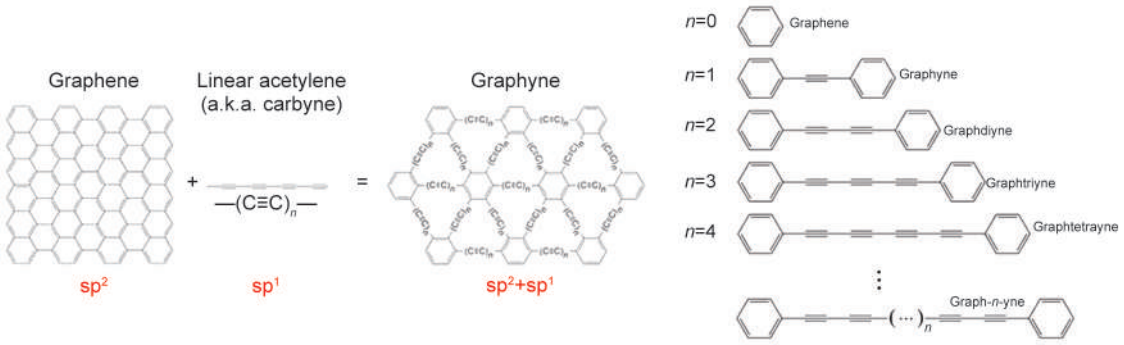

Fig. 3 Schematic of graphene to graphyne- linking aromatic groups by linear acetylene and classification of extended graph-n-ynes. Graphynes consist of a variation on of the sp^2 carbon motif forming graphene, which can be thought of as simply replacing the characteristic graphitic carbon bonds by an arbitrary number, n, of acetylene (single-and triple-bond) carbyne-like chains (pure sp^1 carbon), resulting in a hybrid sp^1 + sp^2 carbon allotrope. The length of the acetylenic carbon chains within graphynes can be variable, resulting in a family of graph-n-ynes, including simple graphyne ($n=1$) and experimentally synthesized graphdiyne ($n=2$), as well as graphtriyne, graphtetrayne, etc. As such, even graphene (without acetylene links) can be considered an allotrope of graphyne (e. g., $n=0$). The structures depicted in the right represent a single segment of the continuous hexagonal structure illustrated in the left[27]. Reproduced by permission of The Royal Society of Chemistry.

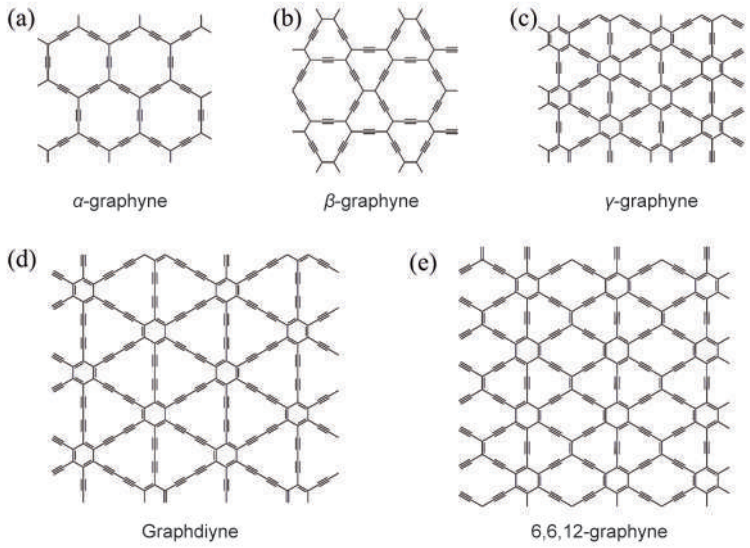

Fig. 4 All kinds of graphynes: (a) alpha-graphyne, (b) beta-graphyne, (c) gamma-graphyne, (d) graphdiyne and (e) 6,6,12-graphyne.

than graphene, and the size of pores can be adjusted by the amounts of acetylene bonds, which enables graphyne the potential use for selective molecular adsorption or separation. Secondly, the introduction of carbon-carbon triple bond reduces the number of carbon atoms per unit area, which makes the density of graphyne lower than graphene. And the more acetylene bonds in graphyne, the lighter of graphynes, which means graphyne is a lighter material than graphene. Thirdly, the mechanic properties will be different because the length of carbon-carbon triple bond is smaller than that of carbon-carbon single bond and carbon-carbon double bond, and the bond energy of carbon-carbon triple bond is the biggest. Finally, the introduction of carbon-carbon triple bond increases the amount of π bonds in the all carbon system, which enables the electronic structure and electronic property, phonic structure and thermal property of graphyne different from those of graphene. Theoretical simulation shows[28] that Dirc cones exist in all different structures of graphynes. According to the simulation of electronic property on graphynes, graphynes subvert the origin of Dirc cones in graphene and the knowledge of corresponding electronic property in graphene. The electronic structure of α-graphyne manifests that the existence of Dirc point and Dirc cone are not inherent to graphene, carbon atoms in graphene bearing the same chemical circumstance are not the precondition to the existence of Dirc cones and its corresponding electronic property. The electronic

structure of β-graphyne demonstrates that the Dirc cone can not only be produced at a high symmetrical point, but also be produced at a low symmetrical point in Brillouin zone. The electronic structure of 6, 6,12-graphyne shows that its torsional Dirc point is in relation to its four-fold symmetry structure, which means the six-fold symmetry structure is not the precondition to generate Dirc cones.

Graphyne is the most stable sp-sp^2 hybrid carbon alotrope up to now, its computational heat of formation is 12.4 kcal mol^{-1}-carbon atoms[26]. As we know that the experimental heats of formation for C_{60} and C_{70} are 10.16 and 9.65 kcal mol^{-1}-carbon atoms, respectively[15], the crystal fullerenes are theoretically stable, which can only be transformed into graphite or diamond under high temperature and high pressure. So graphyne bears the same stability[18, 26]. According to theoretical computation, graphyne and graphdiyne are stable at 300-1 000 K, which provides theoretical base to prepare graphyne or graphdiyne above ambient temperature.

The binding energy of graphyne is 7.95 eV/atom, and its optimized lattice length is 0.686 nm. While the binding energy of graphdiyne is 7.78 eV/atom, and its optimized lattice length is 0.944 nm[30]. Graphynes will be stable once they are synthesized, their binding energies are about 90% of that of graphite. Graphyne can be classified into metallic and semi-conducted according to their different stacking arrangements, and the state with semiconductor behavior and a moderate band gap is expected to be the most stable[31].

3.2 Performance of graphyne

3.2.1 Electronic performance of graphyne

The band gaps of graphynes are around 0.44-2.23 eV according to theoretical simulations[26, 30, 32-35]. The graphynes exhibit a semiconductor behavior along the zigzag direction, while exhibit a metallic behavior along the armchair direction, which is apparently different from that of graphene. And their conductivities obviously decrease with the increase of acetylene bond length in graphynes[36]. Graphdiyne sheet is predicted to be a kind of semiconductors with a band gap of 0.46 eV. The in-plane intrinsic electron mobility of a single layer graphdiyne is calculated to reach the order of 2×10^5 $cm^2 (V\ s)^{-1}$ at ambient temperature, the hole mobility of a single layer graphdiyne is predicted to be one order of magnitude lower than its electron mobility. The electron mobility of graphdiyne nanoribbons (GDNRs) at ambient temperature can also reach the order of 10^4 $cm^2 (V\ s)^{-1}$, which is far larger than its hole mobility. With the raise of width on GDNRs, the charge mobility increases, and the GDNRs with a arm chair edge have a larger mobility than GDNRs with a zigzag edge[33].

Furthermore, electronic properties and carrier mobilities of 6,6,12-graphyne nanoribbons[37], electronic properties of bilayer and trilayer graphyne in the presence of electric field[38], the electronic properties of α-graphyne nanoribbons[39], α-graphyne nanotubes[40], β-graphyne bilayers[41] are also calculated with theoretical simulation. γ-graphyne has a ~0.5 eV direct energy band gap[42], since graphene is proven to be a conductor without a band gap, γ-graphyne is expect to be appealing for electronic applications.

3.2.2 Mechanical performance of graphyne

The mechanical properties of graphyne receive extensively concerns from many researchers[43-51]. In spite of the density of graphyne is only half that of graphene, its interlayer adhesion and out-of-plane bending stiffness are at the same scale with graphene. But graphyne exhibits a nonlinear stress – strain behavior, which is different from graphene, due to its directionally acetylenic group dependent internal stiffening[45]. Graphdiyne is softer than graphyne because of less carbon-carbon bonds in the backbone of graphdiyne. And the band gap of graphdiyne can be positively adjustable through uniform strain, which is originated from less orbital overlap in carbon atoms under increased strain[47]. Graphyne can keep a large nonlinear elastic deformation up to a failure strain of 0.2, compared to graphene, it has a low in-plane Young's modulus (162 N m^{-1}) and a large Poisson ratio (0.429)[49].

3.2.3 Thermoelectric performance of graphyne

Graphyne has a very important potential application prospect as thermoelectric materials[52-58]. Compared with the corresponding graphene nanoribbons (GNRs), graphyne nanoribbons (GYNRs) have superior thermoelectric properties (the thermoelectric figure of merit is about 3-13 times of that of GNRs). The thermoelectric efficiency of GYNRs decreases with the increase of nanoribbon width, but increases monotonously with the increase of temperature[53]. Among the graphyne structures shown in Fig. 4, the thermal conductivity of 6,6,12-graphyne has the most prominent directional anisotropy because of its non-six-fold symmetry structure[55].

The existence of alkyne bond in graphyne leads to a significant decrease in thermal conductivity, which is derived from the lower atomic density in graphyne and the weaker single carbon-bond in acetylene bond[57]. The reduction of thermal conductivity in graphynes is related to the density of alkyne bonds

in their structures. The thermal conductivity of the graphyne family is not sensitive to the number of alkyne bonds, but depends on the number of benzene rings. In addition, the thermal conductivity of graphynes is also affected by the external strain and temperature. Graphyne has a high and controllable figure of merit, which makes it a more promising material as thermoelectric equipment than graphene. The external strain and increasing temperature adversely influence the thermal conductivity of both graphyne and graphene. In particular, graphyne is more sensitive to the changes of temperature than graphene, which is because graphyne has a lower stiffness and higher toughness than graphene. Graphyne is easily affected by external strain, especially at high strain. Therefore, the controllable thermoelectric performance factor of graphyne can be achieved by strain engineering[55,58].

3.2.4 Magnetic performance of graphyne

The magnetic performances of graphyne are also widely concerned[59-64]. All armchair α-graphyne nanoribbons are nonmagnetic semiconductors, and its band gap is a function of its width. The zigzag α-graphyne nanoribbons bear a magnetic semiconductor ground state with ferromagnetic order at the edge and opposite spin directions on both sides. Graphdiyne and graphyne adsorbed by transition metals are excellent materials for spintronics[59]. The adsorption of 3d transition metal atoms (V, Cr, Mn, Fe, Co, Ni) on graphdiyne and graphyne not only effectively regulates the electronic structure of the graphdiyne/graphyne system, but also introduces excellent magnetic properties, such as spin polarized semiconductors[60,63].

Pure graphyne is nonmagnetic, but the magnetic moment can be induced by the single atom hole in graphyne. The magnetic moment of α-, β-graphyne is 1.1-1.3 μB, and the value is 1.8 μB for γ-graphyne[61]. Similarly, the magnetic moment of transition metal adsorbed graphyne with hole defect is larger than that of transition metal adsorption. For example, Ni adsorbed graphyne is nonmagnetic in nature, but Ni adsorbed graphyne with holes shows half metal property, and the net magnetic moment is 2 μB, which can be used as a rotary filter[62,64].

3.2.5 Hydrogen storage performance of graphyne

Graphyne has a smaller equilibrium distance, larger binding energy with hydrogen atoms, which makes graphyne a more suitable material for hydrogen physical adsorption than graphene[65].

The hydrogen storage performance of graphyne is one of the most concerned properties at present. For the hydrogen storage performance of graphyne, more attention is paid to the hydrogen storage performance of alkali metal and alkaline earth metal decorated graphyne[66-71]. The binding energy between metal and graphyne is greater than the cohesive energy of metal itself, which means the combination of metal and graphyne is very stable and metal clusters will not be formed at the present of graphyne. But in other cases, like carbon nanotubes and graphene, the metal cluster is easily formed because the binding energy between them and metal are smaller than the cohesive energy of metals. The hydrogen storage capacity of Li-decorated single layer graphyne on one side is as high as 9.26 wt%, the hydrogen storage capacity can be as high as 15.15 wt% when Li alternately decorated on both sides of graphyne[66]. The binding energy of hydrogen molecules in metal-modified graphyne complexes is about 0.2 eV/H_2. For Ca-decorated graphyne nanotubes (GNT), the adsorption energy is 0.13-0.33 eV/H_2, almost independent of the tube diameter, and the adsorption capacity can reach 7.44-8.96 wt%. Ca-decorated GNT is a good choice for hydrogen cycle at room temperature[68].

However, the chemical adsorption of hydrogen significantly reduces the mechanical properties of graphyne[69]. In addition, besides hydrogen storage, graphyne also has the function for hydrogen purification[72,73].

3.2.6 Li^+ storage performance of graphyne

As a carbon material, graphyne possesses high mobility and high Li^+ storage capability, which makes it to be one of the best candidates for anode materials in rechargeable batteries[74-76]. The diffusion of Li^+ in graphite anode is limited in the plane, while the diffusion of Li^+ in graphyne and graphdiyne anode is not only in the plane, but also between the planes, which is resulted from the unique atomic structure in graphyne and graphdiyne. The unique structure of sp-sp^2 hybrid is beneficial not only to the diffusion of Li^+, but also to the storage of Li^+. The intercalation of Li^+ has little effect on the distance between adjacent graphyne layers, which is beneficial to the charge discharge process. Li^+ can easily pass through the graphdiyne plane with an energy barrier of 0.35 eV, which makes Li atoms well disperse on both sides of the monolayer graphdiyne[74]. The diffusion of 3D Li^+ needs to overcome the energy barrier of 0.53-0.57 eV[76].

In addition to the various properties of graphynes mentioned above, optical properties[77-80] and negative thermal expansion[81] are also concerned.

3.3 Applications of graphyne

According to the above theoretical predictions on graphyne, it can be seen that graphyne is stable and

can be fabricated from thermodynamics. In addition to the simulation of performance on graphyne, its applications are also widely considered and theoretically simulated. These applications are mainly concentrated in the fields of energy, environment and biomedicine.

According to the theoretical calculation, the application of graphyne materials is mainly on the following topics. The application of adsorption, capture and separation of carbon dioxide, carbon monoxide, ammonia, sulfur dioxide and other gases[82-84], the application of adsorption of polymer chain and acrolein and other organic matters[85, 86], the application of selective filtration and purification of water, and the application of seawater separation and desalination[87, 88], as the monoatomic catalytic substrate of noble metals and the application of stable noble metal catalysis[89, 90], and the detection of toxic and harmful substances such as hydrogen peroxide, carbon monoxide and toxic gases[91-93]. Biomedical aspects include the application of amino acid detection[94], the application of calmodulin structure and performance regulation[95], the application of promoting the extraction of cholesterol from protein[96]. In addition, there are also applications as high-density magnetic storage substrate[97].

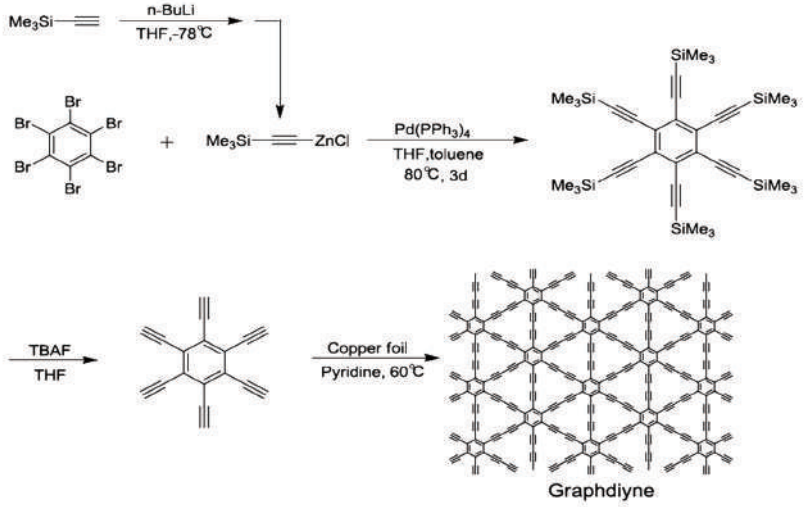

Fig. 5　The synthetic route of graphdiyne[98]. Reproduced by permission of The Royal Society of Chemistry.

In addition to the above theoretical simulation on the application of graphyne, researchers have synthesized graphdiyne materials by various methods and the main reaction process is shown in Fig. 5[98-101]. The as-prepared graphdiyne materials are mainly used in solar cells[102, 103], lithium batteries[104, 105], photocatalysis[106-108], oxygen reduction[109], field emission performance[99, 110] and real-time detection of DNA[111].

4　The design of synthesis on graphyne

Due to the potential excellent properties of graphyne, the synthesis of graphyne has attracted great attention. This has gradually attracted more researchers to participate in the experimental synthesis of graphyne.

Chemical vapor deposition (CVD) is the best method to grow graphene with few defects and large layers. Similarly, for single-layer graphyne materials, it is also a good method to deposit precursor on the surface of noble metal or catalyst metal in ultra-high vacuum or inert atmosphere by CVD method. Some precursors containing terminal alkynes have been used to obtain one-dimensional nanowires or polymers by coupling reaction of terminal alkynes on noble metal surfaces under high vacuum conditions, and the as-prepared materials have been characterized by scanning tunneling microscopy (STM)[112-117]. To synthesize graphyne by CVD, it is firstly necessary to design and synthesize new precursors according to the expected structure. Many structural units and fragments of graphyne have been synthesized[13, 15, 18, 118-122], which provides a good database for the design and synthesis of precursors. Secondly, the interaction between graphyne and metal surface should also be considered[123-125], so as to reduce the occurrence of side reactions in CVD and select the appropriate substrate. Recently, it has been reported[126] that a monolayer carbon structure containing acetylenic scaffoldings has been synthesized on the surface of silver by CVD with hexaalkynylbenzene as the precursor, which is a further step in the synthesis of graphene.

It was proposed in 1990s that graphyne and graph-

diyne can be obtained by trimerization of cycloalkenes or homopolymerization of hexaalkynylbenzene[127]. However, the preparation and design of precursors, the selection of a template or substrate and the control of various side reactions in the polymerization process are important factors affecting the structure of the products. Now, the preparation of hexaalkynylbenzene has been solved[98], and the liquid – liquid interface and gas-liquid interface are also used as polymerization templates or substrates[101]. The control of side reactions in organic polymerization, the selection and design of new polymerization templates or substrates, and the design and synthesis of new precursors are the main problems to be solved in the future.

5 Perspective

Graphyne, as a new allotrope of carbon, has attracted much attention due to its unique properties. In particular, its theoretically unique band gap structure is appealing, which makes it possible to become a star in future carbon electronics. At present, the research on graphyne materials are mainly focused on energy conversion and storage and catalysis, based on its electrical, electrochemical and catalytic properties[78,79]. In addition, the potential applications of graphyne in the fields of hydrogen storage, thermoelectric and third-order nonlinear optics have attracted much attention as a new material.

Although it has been theoretically demonstrated that graphyne has very attractive properties and potential applications in many aspects, it is still a long way to go for experimental synthesis of the defect free two-dimensional materials. The preparation of graphyne is crucial to the development of graphyne in the future.

To obtain two-dimensional graphyne materials, the most effective synthesis method is to deposit the precursor on the substrate surface by CVD, then complete the formation of carbon-carbon bond under the induction of substrate surface lattice. So the most important problem is to design and synthesize a specific precursor, which should meet the requirements of containing alkyne bond and have enough functional group tolerance, which can not only withstand certain high temperature but also certain high vacuum. Because the activity of sp hybrid carbon-carbon triple bond is higher than that of sp^2 hybrid carbon-carbon double, this process will be more challenging.

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