Ceramics-Silikáty 62 (3), 211-220 (2018)
www.ceramics-silikaty.cz doi: 10.13168/cs.2018.0014

STRATEGIES OF FABRICATING GRAPHENE AND GRAPHENE-ANALOGOUS 2D NANOSHEETS

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Submitted December 22, 2017; accepted February 21, 2018

Keywords: Graphene, Two-dimensional nanosheets, Fabrication, Exfoliation, Strategy

Due to the unique structures and properties of graphene and graphene-analogous two-dimensional (2D) nanosheets, such as MoS_2, C_3N_4, black phosphorus nanosheets, numerous methods were developed to prepare these 2D nanosheets, however, it is still a challenge to fabricate high quality 2D nanosheets cost-effectively on a large scale. This paper reviewed the recent progress of the strategies when fabricating graphene and graphene-analogous 2D nanosheets. These strategies were divided into five main categories based on the basic principles: physical exfoliation, chemical exfoliation, electrochemical exfoliation, chemical synthesis and chemical vapour deposition (CVD). The merits of each category were summarised and analysed, and the drawbacks were highlighted as well in detail. It provides an avenue to design and fabricate different kinds of high quality 2D nanosheets cost-effectively on a large scale for energy storage and conversion, catalysis, sensors, electronics, and so on.

INTRODUCTION

Since the single layer graphene, a one-atom-thick plate of carbon, was first exfoliated mechanically from the graphite at 2004 by Geim's group, two-dimensional (2D) graphene nanosheets were studied extensively and intensively for energy storage and conversions, sensors, catalysis, electronics and many other applications, due to their ultrahigh specific surface area, excellent thermal and chemical stability, superior electrical conductivity and other properties. 2D graphene-analogous nanosheets, such as MoS_2, WS_2, black phosphorus, C_3N_4, and transition metal carbides and nitrides, and even the transitional metal hydr(oxides) were also developed and studied extensively.

Numerous efforts have been devoted to fabricating graphene and graphene-analogous 2D materials in order to controllably prepare cost-effective, ultrathin, high-quality nanosheets on an industrial scale, however, it is still a challenge to fabricate these nanosheets conveniently and cost-effectively. Numerous strategies, such as physical exfoliation, chemical exfoliation, electrochemical exfoliation, chemical vapour deposition (CVD), organic synthesis have been developed. In this paper, the strategies of fabricating graphene and graphene-analogous 2D nanosheets were reviewed, the characteristics of each strategy were analysed in detail, and a future direction of fabricating 2D nanosheets was also proposed.

THEORY AND DISCUSSION

Strategies to fabricate 2D nanosheets

Physical exfoliation

Transitional metal dichalcogenides (TMD), C_3N_4, which were layer-stacked together by van der Waals force, could be exfoliated into mono- or a few atomic layer nanosheets by physically breaking the forces between the stacked layers, where numerous physical approaches have been developed for the layer-structured materials, such as graphite.

Exfoliation by adhesive tape, sandpaper and ball milling

The first and most eminent route to fabricating atomic thin monocrystalline graphene was prepared via repeatedly peeling highly oriented pyrolytic graphite (HOPG) with adhesive tape, the exfoliated few-atoms-thick carbon film, also called graphene, displayed excellent electric conductivity and stability [1]. Pristine and high-quality thin TMD nanosheets, such as MoS_2, WSe_2 nanosheets, could also be peeled off from the bulk crystals in this way [2-5]. It is a very simple route without using complicated or expensive instruments, however, an extremely low yield was the main drawback of this approach that hindered its practical application.

Sandpaper was another effective tool to exfoliate the layer-structured materials, for instance, MoS_2 bulk

crystals could be cleaved into nanosheets by two high speed conversely orbited sandpapers – cca. 7 nm thin MoS_2 nanosheets were obtained after subsequent probe ultrasonication [6].

Ball milling could also exfoliate the layered material into nanosheets, even more, doped or functionalised nanosheets could be fabricated [7-9]. For example, edge-selective functionalised/sulfurized graphene could be prepared by ball milling a mixture of graphite and sulphur(trioxide) [8,9]. A few layer MoS_2 nanosheets could be obtained through ball milling bulk powders in NMP solutions [10].

Though nanosheets, even monolayer nanosheets, could be facilely fabricated in this way without using complicated instruments or processes, poor efficiency and low yield of a single or a few-layer nanosheets restricted its scalable application. Further research would be required to develop novel instruments or processes for scalable production of 2D nanosheets.

Exfoliation by liquid ultrasonication

In liquid solutions, ultrasonic disturbance could produce a lot of microbubbles and cavities between the stacked layers which could exfoliate the layer-structured materials. It is an effective technology which was developed and investigated intensively to fabricate nanosheets alone or combined with other methods. Great efforts have been devoted to improving the composition of the solutions, precursors of the layered materials, even an ultrasonic instrument, and so on, and great advancements have been achieved for this method. With this technology, many kinds of nanosheets, for example, graphene, MoS_2, black phosphorus, C_3N_4 and V_2O_5 nanosheets, could be successfully exfoliated from bulk powders in many kind of liquid medias, such as water, NMP, IPA, acetone, or the mixture of them [11-23].

It has been found that surface tension; Hildebrand parameters, which are related to the cohesive energy density; and Hansen solubility parameters, which are related to the dispersion, polar, and hydrogen bonding, are three important factors that affected the exfoliating efficiency. When these parameters of the solvent matched well with those of the dispersed 2D materials, the exfoliation energy was minimised, and the exfoliating efficiency would be improved greatly [19].

The solubility parameters of the solvent could be varied by simply mixing different solvents, which would enhance the exfoliating efficiency. For instance, by varying the fraction of ethanol in the ethanol/water mixed solvent, MoS_2, WS_2 and BN bulk powders were exfoliated effectively into 2D nanosheets by ultrasonication. When the volume fraction of ethanol reached 45 %, 35 % and 55 %, the exfoliating efficiency was the most superior for MoS_2, WS_2 and BN nanosheets, respectively [24]. In the NMP solution, the addition of some water enhanced the efficiency of exfoliating graphene or graphene-analogous nanosheets [11]. By tailoring the Hansen solubility parameters of the water/acetone mixed solution, concentrated graphene dispersions could be achieved by ultrasonication when the mass fraction of acetone reached 75 % [25].

As the surfactant possessed high surface activity, the surface tension of the exfoliated nanosheets would be reduced when the surfactant was introduced into the solvent, which would decrease the differences in the surface tension between the dispersed nanosheets and the solvents. For example, sodium dodecylbenzene sulfonate (SDBS) facilitated exfoliating graphite into graphene in de-ionized water by ultrasonication [26], sodium cholate was also helpful in fabricating a few-layered MoS_2, WS_2 and BN, MnO_2 and other layered material nanosheets in aqueous solutions, for MoS_2 nanosheets, the overall yield could reach up to about 17 % [27]. It is a robust and simple strategy to efficiently exfoliate layered materials into nanosheets on a large scale, however, the surfactants absorbed on the 2D nanosheets were not easy to remove and could negatively affect the properties of the nanosheets. In addition to the surfactant, some organic salts, such as imidazole and pyridinium tribromide, could also change the solubility parameters of the solvents or solute and decrease the exfoliation energy. Atomically thin 2H MoS_2 nanosheets with an extremely large area about 1000 μm^2 were obtained by tip ultrasonication in aqueous solutions [28]. Pyrene derivatives, such as 1-pyrenesulfonic acid sodium salt, could be absorbed on the basal plane of the graphite and thus reduce the surface tension of the graphene, which was helpful in exfoliating TMD and BN nanosheets [29]. It provides a great potential approach in cost-effectively and facilely fabricating atomically thin nanosheets on a large scale.

Despite of the solubility parameters, the crystal structure of the starting materials was also an important factor in influencing the exfoliating efficiency. The loosely stacked structure was one of the merits in order to start the materials for exfoliation, for example, hydrothermal synthesised nanostructured MoS_2 powders could be water-exfoliated into a few-layer 2D nanosheets by ultrasonication [30]. Metallic few-layer VS_2 nanosheets were ultrasonically exfoliated from the hydrothermally synthesised $VS_2 \cdot 3NH_3$ precursor [31]. As a precursor, nanotubes could also be used to prepare nanosheets, for example, the W-S covalent bonding of WS_2 nanotubes was broken in ethanol aqueous solutions simply by tip ultrasonication [32].

Besides, ultrasonic instrument is another factor which would affect the exfoliating efficiency. By designing an ultrasonic instrument with a stirring function, ultrasonication and agitation could be carried out simultaneously, thus a high yield, up to 95 % or more, could be obtained for superior graphene, BN, MoS_2 and WS_2 nanosheets in aqueous solutions [33].

Great advancements have been achieved for the ultrasonic exfoliation of nanosheets, so it can become a potential technology in order to fabricate 2D nanosheets

for practical applications, however, low exfoliating efficiency impedes its extensive application, further investigation is required to prepare nanosheets in low boiling point liquids without using additives which are difficult to remove.

Exfoliation by thermal expansion

For layer-structured materials, the distance between the stacked layers would be expanded, and the van der Waals force could be overcome through thermal expansion. Water freezing expansion exfoliation is a recent technology appearing to exfoliate layered materials, for instance, by exploiting the volume expansion when the liquid water was changed into ice – graphite, BN, and MoS_2 powders in water could be exfoliated into a few-layer nanosheets by repeatedly freezing and melting the water between 4°C and -20°C [34]. It was a simple and environmentally friendly method to exfoliate nanosheets that are sensitive to high temperatures, and possessed great potential for practical applications, however, it should be further improved or combined with other methods to raise the exfoliating efficiency.

Solvothermal treatment was another approach to exfoliate layer-structured materials, for example, large and excellent qualities of 6 - 10 nm thick graphene, 10 - 12 nm thick MoS_2 and 14 - 19 nm thick BN nanosheets were exfoliated by microwave-powered solvothermal treatment in IPA or NMP medias [35]. However, only comparable thick nanosheets could be obtained by this route, further optimisation was needed.

Graphene nanosheets and nanoribbons (less than 3 atomic layers thick) with a width below 10 nm with an ultra-smooth edge could be synthesised by briefly heating the expanded graphite to 1000°C under an Ar atmosphere with 3 % hydrogen and the subsequent sonication in a 1,2-dichloroethane solution [36].

Chemical exfoliation

Chemical exfoliation of the layer-structured materials was the most common strategy to fabricate graphene or TMD nanosheets. By harnessing the reaction of the layered materials with oxidants or other active compounds, the layered materials would react with the active compounds, and the formed functional groups or the intercalated ions between layers would swell the layered materials, and the distance between the layers would be enlarged, 2D nanosheets could be prepared after the subsequent mild ultrasonic treatment.

Exfoliation by chemical oxidation

2D nanosheets could be exfoliated by chemical oxidation, notable examples were the fabrication of graphene oxide (GO) nanosheets by Hummers' method, the Staudenmaier method and the Hofmann method [37-38]. Firstly, graphite was oxidised by chemical oxidants, such as $KMnO_4$, $NaNO_3$, and/or $KClO_3$, in concentrated H_2SO_4 and/or HNO_3 acid environment. Functional groups, such as -OH, =O and/or –COOH, were formed and grown on the graphite to form a graphite oxide. A layer-structured graphite was expanded between the layers by the functional groups, through stepwise mild ultrasonication, and single or a few layers of GO were formed [37-39]. Through the third step of reduction, GO nanosheets could be transformed into reduced GO (rGO), also called graphene. The oxidised degree of GO could be modulated by the concentration and/or the kinds of the oxides mentioned above [37-39], for example, –COOH groups other than the oxygen functional groups could remain for GO when the graphite was oxidised several times through Hummers' method [40]. However, the crystal structure of graphite was destroyed when it was oxidised to GO, even the crystal structure of rGO could not be recovered, which would negatively influence the electrical conductivity and restricted its application, furthermore, the strong oxidants and acid used in the process would lead to serious environmental problems.

Exfoliation by lithiation

Besides oxidation, lithiation could also be utilised to exfoliate nanosheets, especially for TMD nanosheets. For example, MoS_2 crystals were lithiated and intercalated by an organic lithium solution (such as n-butyllithium) to form Li_xMoS_2, after the subsequent reaction with water, the H_2 bubbles formed between the layers could exfoliate the MoS_2 crystals into nanosheets [41]. WS_2 and other TMD nanosheets could also be fabricated in a similar way [41-49]. Though 2D nanosheets could be synthesised by this method on a large scale, the organic lithium solution must be treated carefully under a protective atmosphere, besides, the lithation or intercalation was time-consuming, furthermore, the crystal structure of TMD was transformed from a trigonal prismatic (2H) to a metastable metallic octahedral (1T) structure during lithium intercalation, which resulted in a restack of the nanosheets during the retransformation to the thermodynamically stable 2H phase.

In order to keep the pristine 2H crystal structure, sub-stoichiometric amounts of n-butyllithium were used to edge intercalate the bulk MoS_2 crystals – the crystal structure and the semiconducting properties of the 2H phase of MoS_2 could be retained, and a 11 - 15 % mass yield of pristine MoS_2 tri-layer nanosheets (5 nm thick) were controllably achieved [50]. Although it was also time-consuming for the lithiation reaction, it was a great advancement for the exfoliation of the 2H hexagonal structured MoS_2 nanosheets without crystal destruction, and opened up a new avenue to prepare pristine TMD nanosheets by chemical intercalation and exfoliation.

Other chemical exfoliation

In spite of oxidation and lithiation, some reagents, such as alkali metals or alloys, LiOH, hydrazine, and

formamide, could also be harnessed to intercalate into the interlayers of the stacked layer-structured materials [22, 51-53]. The intercalated function would swell up the stacked layers and weaken the bonding strength between the stacked layers. For example, through intercalating with alkali metals or alloys (such as Na, NaK), and stepwise hydration reaction, MoS_2, WS_2 or BN bulk crystals were exfoliated into mono-layer or a few-layer hexagonal nanosheets [52]. Hydrazine molecules could also intercalate into the interlayer of MoS_2 crystals, the rapid radical reactions produce N_2 and H_2O formed under ultrasonication would lead to the efficient exfoliation of MoS_2 nanosheets by cavitation [53]. Few-layered V_2O_5 nanosheets were fabricated through intercalation of V_2O_5 bulk crystals by formamide molecules and thereafter ultrasonic exfoliation [22]. It is a simple and scalable liquid exfoliation method for V_2O_5 nanosheet preparation, however, the yield of a few-layer nanosheets should be further improved, and the chemicals used in the process must be treated carefully for safety and to overcome environmental problems.

Chemical exfoliation technology could fabricate single or a few layer nanosheets on a large scale with a high yield, however, the crystal structure of the exfoliated nanosheets was readily destroyed during the process, which would decrease its properties, furthermore, the reagents used in the process, such as sulfuric acid, n-butyllithium must be treated carefully, these disadvantages hinder its scalable application.

Electrochemical exfoliation

Electrochemical technology is an efficient but mild technology to exfoliate and/or intercalate layered materials either as an anode or a cathode. Electrochemical reactions occurred on the layer-structured electrode would intercalate and/or exfoliate the electrode into a few-layer nanosheets. He high quality and high yield of the 2D nanosheets could be fabricated by this strategy.

Anodic exfoliation

As an anode, tiny gas bubbles formed at the anode would swell the layered materials and the nanosheets could be exfoliated directly from the layer-structured anode, the oxidation degree of the nanosheets could be tailored by varying the oxidisability of the electrolyte. For example, by using concentrated sulfuric acid or perchloric acid as an electrolyte, the graphite anode was electrochemically intercalated and exfoliated into graphene with a certain degree of oxidation [54, 55], rich defect sites were found on the fabricated graphene due to the oxidisability of the electrolyte [56]. The oxidised degree of the exfoliated nanosheets would be decreased when the oxidisability of the electrolyte was reduced. For example, when a neutral ionic sulphate salt solution was used as an electrolyte to replace the strong oxidants of concentrated sulfuric acid, the exfoliated graphene shows a lower oxidation degree than when using an acid electrolyte, and showed higher hole-mobility than that of the electrochemically exfoliated acid electrolyte or the rGO, more importantly, a 85 % high yield was obtained for the nanosheets less than 3 atomic-layers thick, and more than 80 % of which were larger than 5.0 μm in the lateral direction [57]. It provides an efficient and environmentally-friendly approach to prepare a few-layer graphene nanosheets cost-effectively and scalable under mild conditions.

Due to the high surface activity, the anionic surfactant could be absorbed on the electrode by electrostatic attraction, thus it facilitated the exfoliation and stabilisation of the nanosheets in the liquid electrolyte under electrical potentials. For example, poly(sodium 4-styrenesulfonate) and sodium dodecyl sulphate were helpful to exfoliate graphene from the graphite anode by using a Na_2SO_4 ionic solution as the electrolyte [58]. Other surface functionalising materials, such as 9-anthracene carboxylic acid, could also be eabsordb on the graphite anode and facilitate the exfoliation of mono-layer graphene nanosheets anodically [59]. In an aqueous electrolyte, hydrophilic room temperature ionic liquid (RTIL), such as 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM][BF_4]), and lithium salts, such as $LiClO_4$, also facilitated the exfoliation of graphene, the sizes of the graphene and exfoliating efficiency could also be controlled by varying the electrochemical parameters or the kind of RTIL solutions [60].

Despite the electrical conductor, insulate materials could also be exfoliated electrochemically into nanosheets as an anode, for example, atomically thin (1.4 - 10 nm thick) and high crystalline black phosphorus nanoflakes were exfoliated electrochemically from the anode of bulk black phosphorus crystals in sodium sulphate ionic solutions, and up to 80 % high yield could be obtained [61, 62]. It opens a new window to fabricate the insulating 2D nanosheets. The fast synthesis process and the inexpensive characteristics were the advantages of this strategy for the preparation of the layered black phosphorus nanosheets.

Cathodic intercalation and exfoliation

As a cathode, layer-structured materials could be effectively lithiated and intercalated by an Li metal anode under electrical potentials. For example, a layer-structured TiS_2, TaS_2, MoS_2 cathode could be intercalated electrochemically by lithium ions, the subsequent ultrasonic treatment in water would result in the formation of the corresponding nanosheets, the yield of the single layer of MoS_2 nanosheets was up to 92 % [63-65]. Graphene less than 5 atomic layers with superior electrical conductivity could also be exfoliated from the graphite cathode, a 70 % yield of exfoliation could be achieved [66]. Though a high yield could be realised for the exfoliation of the nanosheets by cathodic lithiation, the phase of nanocrystals maybe be transformed into a

metastable phase, even more, metal lithium was used, which must be handled carefully, and the lithium ions absorbed on the nanosheets were hard to remove, these drawbacks restricted its scale application.

A layer-structured electrode, no matter whether it is conductive or not, could be fast exfoliated into nanosheets under mild conditions by anodic exfoliation, moreover, expensive instruments and complicated processes were not required, anodic exfoliation was a promising method for the scalable fabrication of high quality 2D nanosheets. Compared to anodic exfoliation, cathodic exfoliation was less effective and more complicated, however, high quality graphene with a lower oxidised degree could be obtained.

Chemical synthesis (bottom-up method)

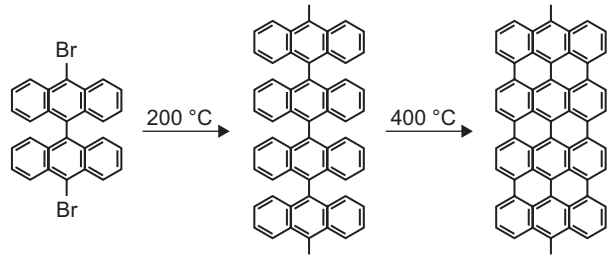
Conversely to the strategies mentioned above, nanoflakes or nanoribbons of the layered materials could also be synthesised from small molecules with a bottom-up method through synthetic reactions.

Coupling and cyclodehydrogenation reactions could be made use of to synthesise nanoribbons from the aromatic monomers [67, 68]. One famous example is the synthesis of the atomic graphene nanoribbons from 10,10'-dibromo-9,9'-bianthryl monomers through coupling and the cyclodehydrogenation reaction afterwards, the synthetic scheme and the microstructure of which were described in Figure 1 [67]. Only nanoribbons other than nanoplates could be synthesised by this way.

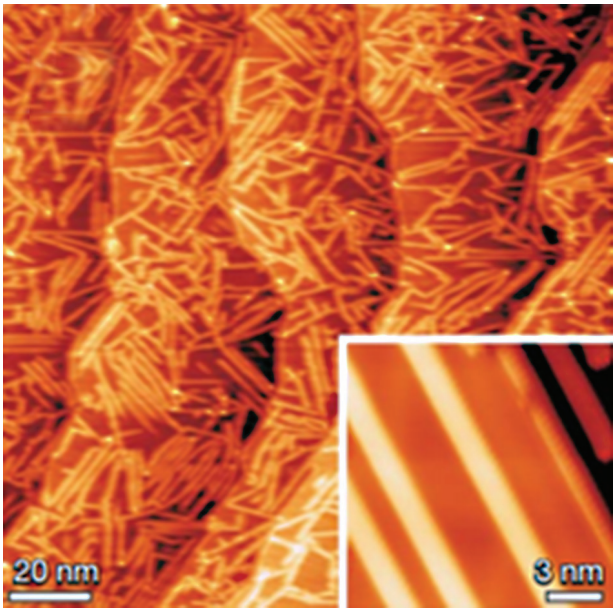
Monolayer WS_2 nanoflakes could be synthesised from the reaction of a tungsten-oleylamine (OM) complex and elemental sulphur solvated in OM [69].

A hydrothermal or a solvothermal reaction was another important approach to synthesising the nanosheets, much efforts have been devoted to this strategy. For instance, MoS_2 nanosheets could be synthesised from the hydrothermal reaction of hexa-ammonium hepta-molybdate tetrahydrate ($(NH_4)_6Mo_7O_{24}\cdot 4H_2O$) and thiourea, the defect and morphology could be controllably modulated by varying the concentration of thiourea, excessive thiourea would lead to the defect-rich and oxygen-incorporated MoS_2 nanosheets, while less thiourea would result in the oriented crystal growth of low-defect MoS_2 nanosheets [70, 71]. As an alternative Mo source of sodium molybdate (Na_2MoO_4), the process could also be hydrothermally reduced by thioacetamide to synthesize uniform MoS_2 nanosheets in water [72]. MoS_2 and WS_2 nanosheets could also be synthesised by other Mo or W sources and sulphur sources under hydrothermal or solvothermal conditions [73-77].

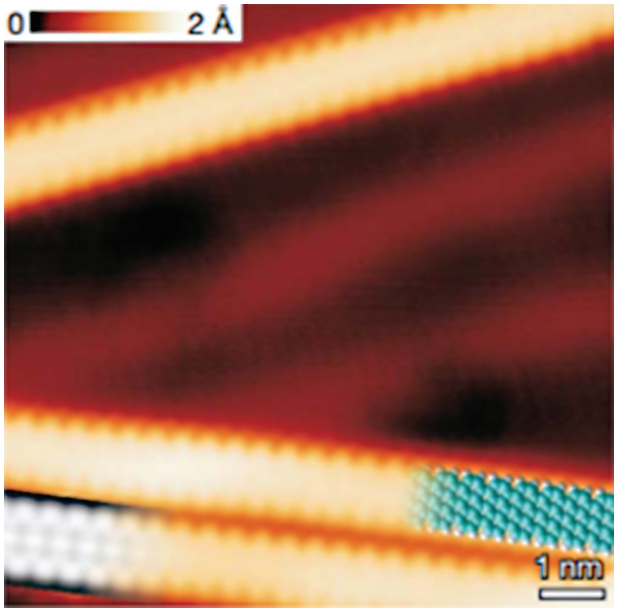
Through a decomposition reaction, MoS_2, WS_2 and other nanosheets could be synthesised from the compounds containing Mo/W/Sn and a sulphur element too. For instance, the thermal decomposition of the dip-coated $(NH_4)_2MoS_4$ on SiO_2/Si or a sapphire substrate



a)



b) STEM



c) HR-STEM

Figure 1. Reaction scheme from 10,10'-dibromo-9,9'-bianthryl monomers to graphene nanoribbons (a), STM (b), and HR-STM (c) of the graphene nanoribbons. Adapted from reference 67 with permission. Copyright 2010, Nature.

under an Ar/H_2 atmosphere and its further annealing at higher temperature would lead to the formation of a highly crystalline and a large-area of MoS_2 thin layers with uniform thickness [78]. Hexagonal SnS_2 nanosheets, with a thickness of cca. 16 nm, were synthesised by the thermal decomposition of a molecular $Sn(S_2CNEt_2)_4$ precursor in oleylamine solvents at elevated temperatures [79]. As efficient catalysts for the hydrogen evolution reaction (HER), edge-terminated MoS_2 nanosheets could be obtained from the decomposition of $(NH_4)_2MoS_4$ in N,N-dimethylformamide (DMF) by microwave heating [80].

Template-growth was also another effective approach to synthesise 2D nanosheets, and solid inorganic salts could be used as template to grow nanosheets. For instance, by using sodium chloride solid crystals as templates which were readily washed off by water, 2D nanosheets of hexagonal transitional metal oxide (MoO_3, MnO_2 and WO_3) less than 2 nm thick could be synthesised by growing and coating the corresponding transition metal containing precursors on the crystalline templates and subsequent annealing [81]. It is a scalable approach to prepare 2D transitional metal oxides which are not confined to layered materials only.

Chemical synthesis was a commonly used technology to prepare nanosheets, however, either the complicated process or the comparable thickness of the nanoplates was the disadvantage of this strategy, much effort should be devoted to simplifying the process or reducing the thickness of the nanoplates.

CVD

Chemical Vapour Deposition (CVD) is a widely used method to synthesise graphene, TMD and other 2D nanosheets by segregation, decomposition and other reactions.

Large area, highly crystalline and atomic thick TMD nanosheets could be synthesised on a solid substrate, such as a sapphire, SiO_2/Si, mica substrate in this way. In general, TMD nanosheets were synthesised by the reaction of transitional metal (oxide, chloride and iodide) and the vapour of a chalcogenide elementary substance [82-101]. The layer number and the area of TMD nanosheets could be controllably synthesised by changing the synthetic parameters, such as the temperature, partial pressure of gases, etc. For example, through controlling the amount of $MoCl_5$ and the total pressure in the synthetic setup, the layer number and atomic scale of the MoS_2 crystalline nanosheets with unprecedented uniformity could be controlled precisely by this method [102, 103]. By reacting WO_3 powders with elemental Se vapour, a large-sized monolayer of WSe_2 nanoflakes could be synthesised on sapphire [88].

Besides the horizontal nanosheets, vertical-orientated nanosheets could also be fabricated on a metal substrate by this method. For instance, vertical orientated ReS_2 nanoflakes were synthesszed on SiO_2/Si, mica, carbon nanofibers and a thin gold foil substrate by the reaction of ReO_3 and sulphur vapour [104], vertical-oriental WS_2 nanosheets could also grow on the surfaces of tungsten foil through the first oxidation of tungsten metal and the subsequent sulfurization by a sulphur vapour [105].

Though many kinds of a single or few-layered nanosheets with high crystallinity could be synthesised by CVD, the complicated process, expensive instruments and the low yield implied that CVD was not suitable for scale and cost-effective production of 2D nanosheets.

CONCLUSIONS AND PROSPECTS

In summary, the strategies of fabricating 2D nanosheets were reviewed. Based on the different principles, the strategies could be divided into five main kinds, physical exfoliation, chemical exfoliation, electrochemical exfoliation, chemical synthesis and the CVD method. Low efficiency restricted the development of the strategies of physical exfoliation, chemical synthesis and CVD, further study for improving the processes or instruments is required. Though high-quality graphene or graphene-analogous 2D nanosheets could be fabricated by the CVD method, the problems of complicated process and low efficiency should be solved for further development. Crystal destruction or transformation impeded the practical application of chemical exfoliation as well as environmental problems, electrochemical exfoliation could prepare high quality nanosheets under mild conditions without causing environmental problems, it was a promising potential strategy to fabricate 2D nanosheets cost-effectively for large scale production.

Acknowledgements

This research was financially supported by the National Science Foundation of China (No. U1504218, U1404217), Ph. D Initial Scientific Research Fund of Anyang Institute of Technology (BSJ2016006), and Scientific Research Fund of Anyang Institute of Technology (YJJ2017006).

Summary

The recent progress of strategies to fabricate graphene and graphene- analogous 2D nanosheets was reviewed. These methods include five main categories based on the basic principles: physical exfoliation, chemical exfoliation, electrochemical exfoliation, chemical synthesis and chemical vapour deposition (CVD). The advantages and disadvantages of each

method are analysed and discussed in detail. It provides one of the most effective ways to design and fabricate different kinds of high quality 2D nanosheets cost-effectively on a large scale for energy storage and conversion, catalysis, sensors, electronics, and so on.

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