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REVIEW

Modulation of the lattice structure of 2D carbon-based materials for improving photo/electric properties

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Abstract

Reliable, inexpensive, environment-friendly, and durable properties of carbon materials with unique and outstanding photoelectric performance is highly desired for myriad of applications such as catalysis and energy storage. Since lattice modulation is a vital method of surface modification of materials, which form by an external force during the synthesis process, causing the internal compression and stretching, leading to lattice sliding event. In this review, we present a summary of different methods to tailor the lattice modulation in 2D carbon-based materials, including grain/twin boundary, lattice strain, lattice distortion, and lattice defects. This overview highlights the implication control of the diverse morphologies of nanocrystals and how to tailor the materials properties without adding any polymers. The improvement in the performance of 2D carbon materials ranges from the enhancement of charge transport and conductivity, structural stability, high-performance of light absorption capacity, and efficient selectivity promote the future prospect of 2D carbon materials broaden their applications in terms of energy conversion and storage. Finally, some perspectives are proposed on the future developments and challenges on 2D carbon materials towards energy storage applications.

Keywords Lattice structure · 2D carbon materials · Improving performance · Energy conversion · Energy storage

1 Introduction

Recently, vast development of the globalized economy has triggered significant implications to meet the huge demands of industrial production. Current daily consumption that globally necessities deserve special attentions to accommodate a large population, which in turn promotes the huge consumption of traditional fossil fuels in the world. This has caused irreversible environmental problems, such as global warming, acid rain, and water pollution [1–4]. To reduce greenhouse gas emissions and energy consumption, a high demand of "carbon neutrality" by using sustainable, green, and low-cost method of production is imminent. The use of solar energy or electrochemical is the most ideal way to solve the imminent problems such as photoelectric catalytic water splitting, photoelectric oxygen evolution, methane oxidation and etc. [5–7]. However, durability and practicality are the important aspect that needs to be developed toward durable, low-cost, and sustainable catalysts [8–10]. Two-dimensional materials can be categorized for those compounds with the thickness of a single atomic layer (~ 1 nm or even less) or few atomic or molecular layers. The interlayers are connected by the strong covalent or ionic bonds, whereas the interlayers are combined by the weak van der Waals forces [11, 12]. Two-dimensional materials have peculiar properties and functions due to their unique 2D structure [13]. At present, 2D optoelectronic materials include graphene (GN) [14, 15], topological insulators

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(TI) [16, 17], transition metal chalcogenide compounds (TMDCs) [18, 19], black phosphorus (BP) [20, 21]. Among them, two-dimensional carbon materials are famous for their excellent properties: electrical conductivity, large specific surface area, environmental friendliness, high durability, excellent adsorption properties, and good thermal conductivity [22, 23]. Carbon nanomaterials are very stable due to their metal-free, chemically stable properties, and have a suitable energy band structure [24]. However, the disordered structure and high exciton binding energy resulted in low efficiency of charge transfer performance [25]. In addition, the use of polymer binders could hamper their photoelectric properties leading to uncontrollable material morphology. Thus, this is unavoidable since it is mostly used in the carbon materials production. [26]. The field of environment and photoelectric catalysis cannot be further expanded, which greatly limits the solar hydrogen production. In the past decades, researchers have made relentless efforts to modify the structure of carbon nanomaterials by improving the lattice structure to improve their optoelectronic properties, and physicochemical characteristics. For example, several reports by introducing lattice defects [27, 28], poly grain boundaries [29, 30], heterostructures [31, 32], crystalline [33, 34] are pursued. This paper discussed recent strategies that reported the relationship between photoelectric properties and the morphological state of lattice structure modification in two-dimensional carbon nanomaterials. On the other hand, the advantages of modified lattice structures in the application of hydrogen production is also discussed. We systematically classify the lattice structure into the following four aspects as, (i) twin boundaries, (ii) lattice strain, (iii) lattice distortion, and, (iv) lattice defects shown in Fig. 1. This review summarizes the structure properties, characteristics, and the advantages of 2D carbon materials for specific applications in hydrogen production aimed to guide the modification of 2D materials toward high photoelectric performance and efficient catalysts to alleviate the energy crisis.

2 Method of lattice modulation of 2D carbon nanomaterials

Several conventional structures such as orthorhombic, monoclinic, and triclinic 2D carbon-based materials have shown great potential in electrical, optoelectronic, and other catalytic applications driven by their low symmetry. This is mainly governed due to strong anisotropy that manifest in their physical properties. However, there is still a lack of proven methods for preparing low-symmetry materials, and their industrial production still needs to be further explored. Lattice modulation, including the lattice strain (such as compression or tension), lattice defects create different grain boundaries and crystal plane distortions to alter the distance between surface atoms. It is effective pathway to tailor the surface electronic structure of 2D carbon materials, promoting tunable control of the catalytic activity and defects of high symmetry profile.

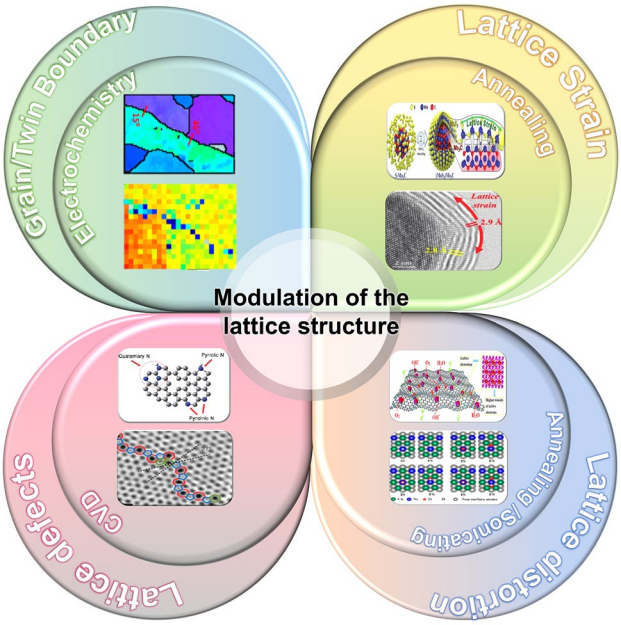


Fig. 1 Four methods of modulation of the lattice structure [43, 47, 52, 53, 58, 61]

2.1 Grain/twin boundary

The strength and ductility of nanomaterials are largely determined by 1D defects (dislocations) and 2D defects, including grain boundaries (GBs) and twin boundaries (TBs). Both of these structural parameters are mostly found in stable defects of some metal surfaces and those can serve as active sites in catalytic reactions (e.g., CO_2 electro-reduction) [35, 36]. The difference is that the GBs can accumulate dislocations, but it is difficult to further improve the malleability [37]. In contrast, TBs can not only accumulate dislocations but also it increases simultaneously the strength and ductility of nanomaterials. In general, nanomaterials are characterized by the presence of in-plane covalent bonds and weak van der Waals bonds between planes. Therefore, grain boundary sliding tends to occur on the plane and thus can slide on the "edge" plane. It is well known that 2D crystals with hexagonal unit cells, such as graphene, have preferential low-energy edges, commonly referred to as armchair (ac) and zigzag (zz) [38–40], which are considered dislocation motions. However, when these plane dislocations are vertically stacked led to the perfect stacking and symmetrical mirror image of twin boundaries occurred [41].

Because of large specific surface area, excellent electronic conductivity, and robust electrochemical stability of 2D $Ti_3C_2T_x$ MXenes nanosheets and negatively-charged of $Ti_3C_2T_x$ nanosheets surface are suitable to cover with a layer of positively charged poly (diallyldimethyl-ammonium chloride) (PDDA) to adjust the charge properties of the surface and consequently induce grain boundary allowing Pt assembled as "nano-worms" (Fig. 2a–b). Pt NW/PDDA-$Ti_3C_2T_x$ catalysts exhibit superior durability and anti-toxicity in environmental applications and photocatalysis compared to the conventional Pt NP/C, Pt NP/CNT, Pt NP/RGO, and Pt NP/$Ti_3C_2T_x$ catalysts. For instance, the CO adsorption energy of Pt NWs is − 2.21 eV, which is significantly smaller than that of Pt NPs (− 2.49 eV) (Fig. 2c). The difference in adsorption energies indicates that the modified MXenes nanosheets have significantly improved toxicity tolerance to CO, and shown more promising electronic structure and good electronic conductivity [42].

To investigate the role of the grain boundary structure of carbon nanomaterials in extreme environments, Yule et al. [43] used scanning electrochemical cell microscopy (SECCM) to investigate the structure-dependent cathodic activity of low carbon steel in aqueous sulfuric acid (pH ≈ 2.3). It can be seen from Fig. 2d that the hydrogen evolution capacity at the edge of the grain boundary is greatly improved. It is contributed that there are abundant surface defects at the grain boundary and contain a large number of unsaturated sites that can be used as active sites in electrocatalytic applications. It is also proven to apply to electrochemical CO_2 reduction during atmospheric corrosion (i.e., in the presence of acid rain).

The carbon coating during material synthesis cannot guarantee that each MnO nanoparticle is effectively embedded. This can be solved by forming embedded of a carbon matrix in their mesoporous MnO/C microsphere (Fig. 2e–f). These unique grains boundaries have high tap density and structural stability, which generate active centers to distribute more uniformly, and mediate the electron transport and lithium ions through 3D pores, speeding up the transmission of Li ions of stable and durable lithium batteries and lay a promising prospect toward electric vehicle industries [44].

2.2 Lattice strain

Lattice strain is considered as interface reconstruction mediated by the nano-effects. The lattice strain on the surface is imposed by the external stress of the heterogeneous substrate (such as UV lamp irradiation). It is often formed at the interface between different crystal components. The generation of lattice strain is complicated and time-consuming, and typically the phenomenon is degraded from the interface [45]. Lattice strain, especially the strain effect of metal and alloy catalysts, can be used to tune the electronic and geometric structures of catalysts to improve their performance [46]. From the perspective of Cheng et al., they found that the lattice parameters of the edge of NiFe-MOFs along the [100] direction were expanded from 11.6 Å to 11.8 Å, 12.0 Å, and 12.1 Å after ultraviolet irradiation. Due to different lattice

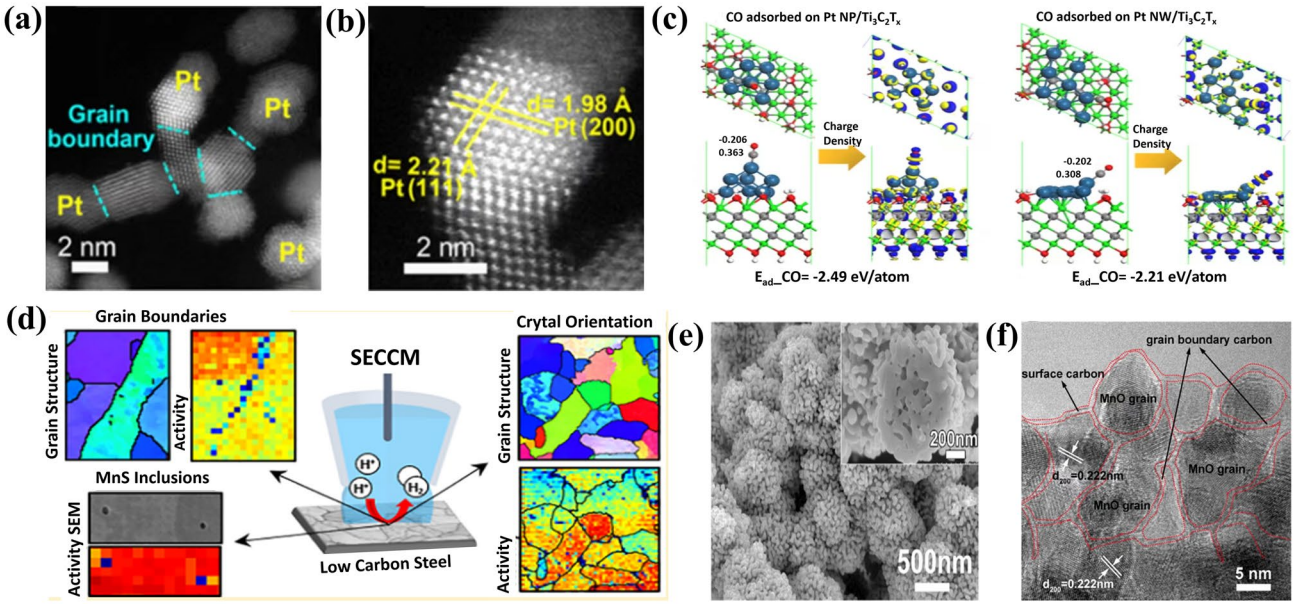


Fig. 2 Lattice microstructure modeling and characterizations **a–b** STEM images of the crystal structures of $Ti_3C_2T_x$ nanosheets and Pt nano worms, **c** relaxed atomic structures for the CO adsorption on Pt NP/$Ti_3C_2(OH)_x$ and Pt NW/$Ti_3C_2(OH)_x$, respectively [42], **d** EBSD map of a "cathodically active" grain boundary and schematic of the SECCM hopping-mode [43], **e–f** SEM and TEM images GBMOC microspheres [44]

expansion ratios, 1.7%, 3.6%, and 4.3%, respectively. The lattice strained MOF nanosheets demonstrated an excellent OER and ORR electrocatalytic activities, and this contribute to the emergence of intermediates that accommodate the realization of Ni^{4+} active sites of superoxide *OOH for efficient hydrogen evolution (Fig. 3a) [47]. From point of the structure, especially in 2D materials, bulk lattice strain can achieve photoexcitation to separate charge carriers, therefore inhibiting the formation of electron–hole pairs. Based on Tiwari et al. [48], the core shell structure for MoS_2 on Mo_2C, and the lattice strain in the MoS_2 sheet provides efficient active site catalytic activity for electrons (different from the plane of the original MoS_2), while the core of Mo_2C provides a conductive channel for fast electron transfer that showing the efficient formation of oxygen radicals, and excellent bifunctional electrocatalysis in ORR and OER (Fig. 3b–d). However, lattice strain does not always produce positive effects, Zhang et al. constructed a Cu@FeNC core–shell model to calculate lattice strain. Theoretical calculations corroborate that both the formation and binding energies produce lattice tensile and compressive stresses, which are detrimental to the stability of FeNC electrocatalysts. Interestingly, when Fe atoms is introduced in ORR, lattice strain plays a positive role and this can greatly enhance the ORR activity of FeNC electrocatalysts (Fig. 3e–f) [49].

2.3 Lattice distortion

Another mechanism that has been considered important for lattice tuning is lattice distortion. The formation of lattice distortion is important to consider because the defects on the surface disrupt the equilibrium state along the GBs that involved how atoms, local tension regions and compression regions interact to each other. The atoms could deviate from the equilibrium position such that increasing their potential energy, and the free energy. As a consequence, the mechanical strength of the crystal has improved, however the trade-off is that the structural stability is reduced, which affects a series of physical and chemical properties of the crystal. For example, the insertion of various vacancies and atomic defects could cause lattice distortions, lead to electronic variation [50, 51].

Zhang et al. [52] found that the formation of defects and the number of intercalated atoms are closely related to the magnitude of the lattice distortion of the carbon material such as graphene. A large amount of lattice distortion can be observed in the vicinity of the defect, some bonds are stretched (brown or red) and other bonds are compressed (blue or purple), whereas those sites which was further away from the defect, generates the smaller lattice distortion. For example, this is observed in Stone Wales (SW) and V2 (divacancy defect in graphene is the three-pentagons and three-heptagons)-V4 (the most favorable structure of tetravacancy defect) defects (Fig. 4a). The other bonds are representing a compressed region in some directions (cyan) and stretched in others (light green). The lattice distortion in Single vacancy (SV) and V6 (obtained by removing a hexagonal ring from the graphene) corresponds to the formation of dangling bonds, which are more localized. In addition to the in-plane distortion, Stone Wales (SW), A2 (structure for embedding two atoms that contains a pair of joined pentagonal carbon rings placed between a pair of heptagonal rings), and A4 (structure for embedding four atoms consist of a pair of adjacent five-membered rings, a hexagonal ring and another five-membered ring, surrounded by the three seven-membered rings.) defects also have significant contribution along the vertical distortion. These defects introduce locally electronic states in graphene, affecting the electron transport properties of graphene. Intercalating atom is a useful strategy to adjust the microstructure condition of the electrode materials. Compared with the undoped 2D carbon-based materials, the impact on the lattice distortion is pronounced for 2D carbon-based materials than its counterparts. Consequently, the defect density in this scenario is prominently increased. In addition, the larger interlayer distance can enhance the electrochemical performance of anode materials, particularly, in high-rate sodium-ion batteries (SIB) [53].

This new finding supports a whole new class of research toward new graphene batteries. Currently, the battery industry is in the bottleneck stage of the development of lead-acid batteries and traditional lithiums. After the successful development of graphene energy storage equipment, with the assumption that the wonder material can be mass-produced, new changes in the battery industry could elevate to the electric vehicle industry.

Tuning the electronic structure in the bimetallic electronic structure through lattice distortion can enhance activity and function of the catalytic performance. Ahsan et al. reported a sp^2 carbon framework with trifunctional groups of bimetallic nickel-copper (NiCu) alloy nanoparticles. The unit cell volume value of the bimetal decreases with the onset potential (trend Ni0.50Cu0.50/C > Ni0.75Cu0.25/C > Ni0.25Cu 0.75/C), indicating lattice shrinkage. Two different metal lattices are mixed to form a bimetallic system, lattice mismatch occurs at the interface of the defective metal, and as a result lattice distortion occurs. This is accompanied by a pronounced lattice shrinkage that contributes to the distortion of the lattice parameters. While the catalytically active sites for hydrogen adsorption are located in the hollow positions of the NiCu (111) surface, the appearance of lattice distortion can maximize the catalytic performance of HER, ORR, and OER, as well as optimize the interaction of reactants and intermediates (Fig. 4b–c) [54]. $FeNi_3$-based alloys are considered to be the most promising oxygen electrocatalysts, although the catalysts show excellent OER activity, the ORR activity is still unsatisfactory. Based on DFT calculations, it

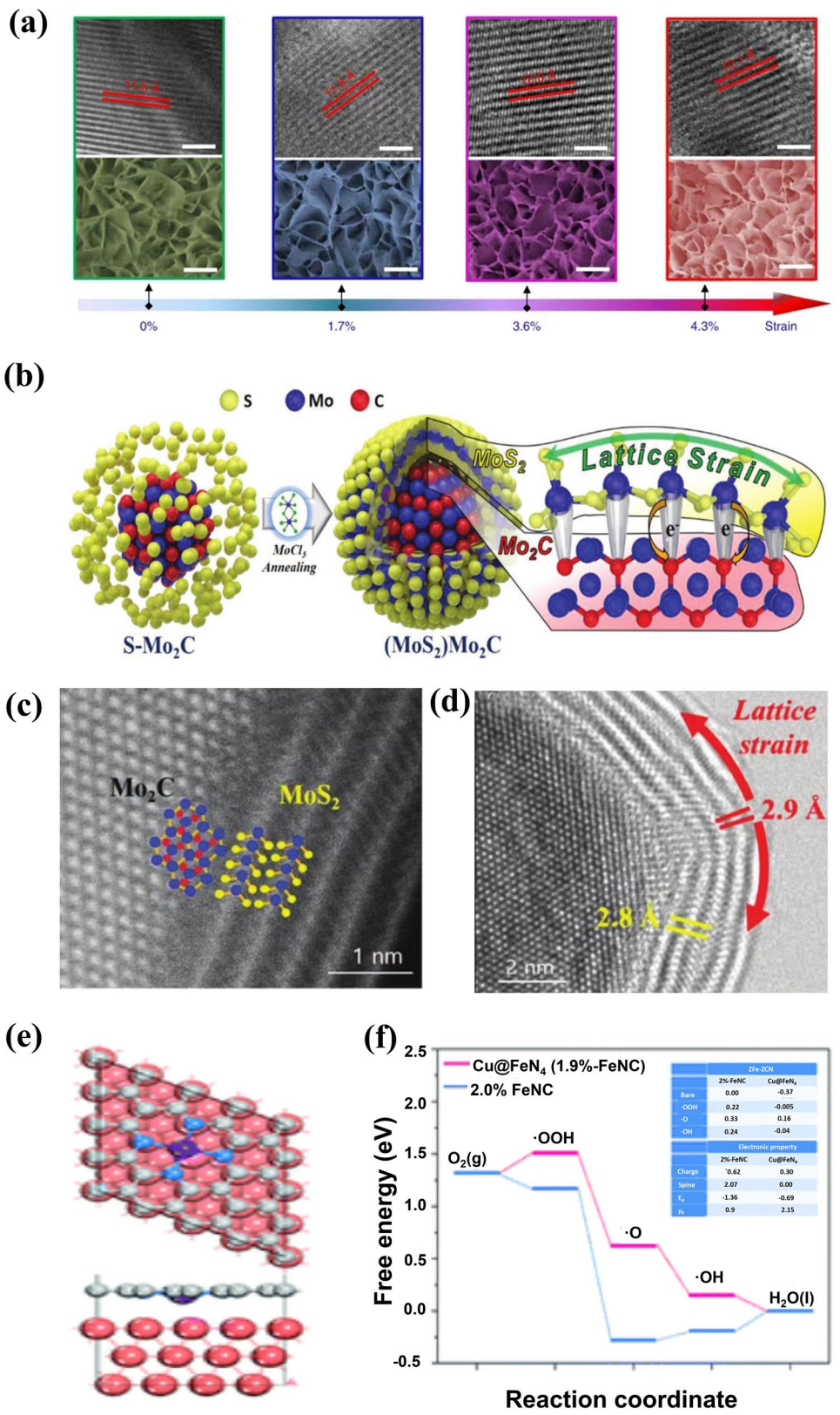


Fig. 3 Detailed lattice characterizations and analysis of hydrogen production performance. **a** HRTEM and SEM of structural characterizations of lattice-strained MOFs [47], **b** schematic illustration for the synthesis of (MoS_2) Mo_2C core–shell structure, **c–d** HRTEM and SEM of Mo_2C and MoS_2 [48], **e** the top and side view of optimized adsorption structures of 2*OH on FeNC slab, **f** the free energy diagram and property difference for FeNC [49]

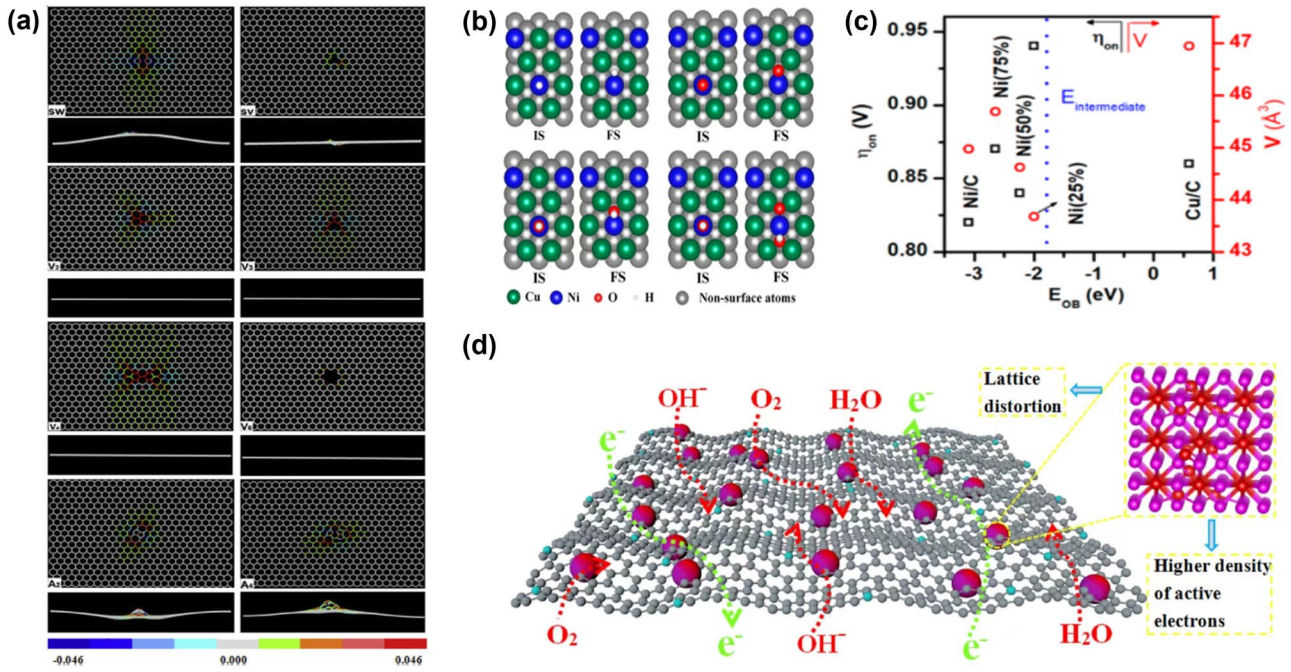


Fig. 4 The role of lattice distortion in hydrogen production. **a** Bond length distortion concerning that of perfect graphene (1.42 Å) induced by the defects [52], **b** H*, O*, OH* and, OOH* adsorbed on the surface of NiCu (Ni0.25Cu0.75), **c** ORR onset potential and unit cell volumes as a function of the DFT-calculated EOB [54], **d** Crystal structure model and schematic of the Fe-enriched-$FeNi_3$/NC for bifunctional oxygen electrocatalysts of Fe-enriched-$FeNi_3$ [55]

is demonstrated that lattice distortion can promote a higher density of active electrons near the Fermi level, and the foreign-incorporated cations suppress the lattice structure through lattice distortion, resulting in enhanced interaction between Fe and Ni atoms. Therefore, the improvement of electrocatalytic performance can greatly make up for the deficiency of Fe-enriched $FeNi_3$ inter-metallic nanoparticle/nitrogen-doped carbon (Fe-enriched-$FeNi_3$/NC) $FeNi_3$-based material, making it have good bifunctional oxygen activity (Fig. 4d) [55].

2.4 Lattice defects

Lattice defects are generated from the structural deviations of microscopic atomic arrangements of a substance due to the thermal motion of atoms, impurity filling, and other conditions during the formation of crystals [56]. The main defect types are (i) point defects, commonly referred to as "lattice vacancies" and "interstitial atoms", that is, vacancies caused when certain atoms in the lattice move away from their lattice nodes and transfer to lattice gaps [57]; (ii) Line defects, i.e., "dislocation lines", are also interface parts of lattice sliding [58]; (iii) plane defects, i.e., grain boundaries and sub-grain boundaries are easily formed under high temperature and pressure [59]. According to the second law of thermodynamics, defects have a large effect on the structural properties of a crystal, led to topology or curvature response, and thereby changing its structure. Lattice defects manipulation could become an important tuning parameter for photocatalysts applications by improving the photocatalytic performance through defect engineering. These efforts have been gradually been recognized in recent years. For example, the strategy is essentially to employ the energy band structure manipulation of semiconductor materials by controlling their defects density. In this approach, the surface defects can be transformed into highly active sites for mediating catalytic reactions towards the enhancement of photocatalytic activity [60]. Graphene has many unique physical and chemical properties because of its perfect hexagonal single-layer carbon atomic sheet structure, but it is difficult to avoid the formation of defects during the synthesis process, which alter the geometric symmetry and perfection of graphene nanoribbons (Fig. 5a) [61]. On the other hand, the gaps formed by defects and open additional ion transport pathways and enhance the interaction between atoms and graphene, which is helpful for the application of graphene-carbon-based composites [62]. Graphene is not a magnetic material, but defective graphene exhibits a response signal. Yan Wang et al. reported that graphene oxide exhibited ferromagnetism at 400 °C and room temperature at 600 °C. This ferromagnetism is formed by eliminating intrinsic defect with oxygen-containing functions and changing the bond length of the interatomic valence bond [63]. Most of the research have pointed out that atomic defects of nitrogen

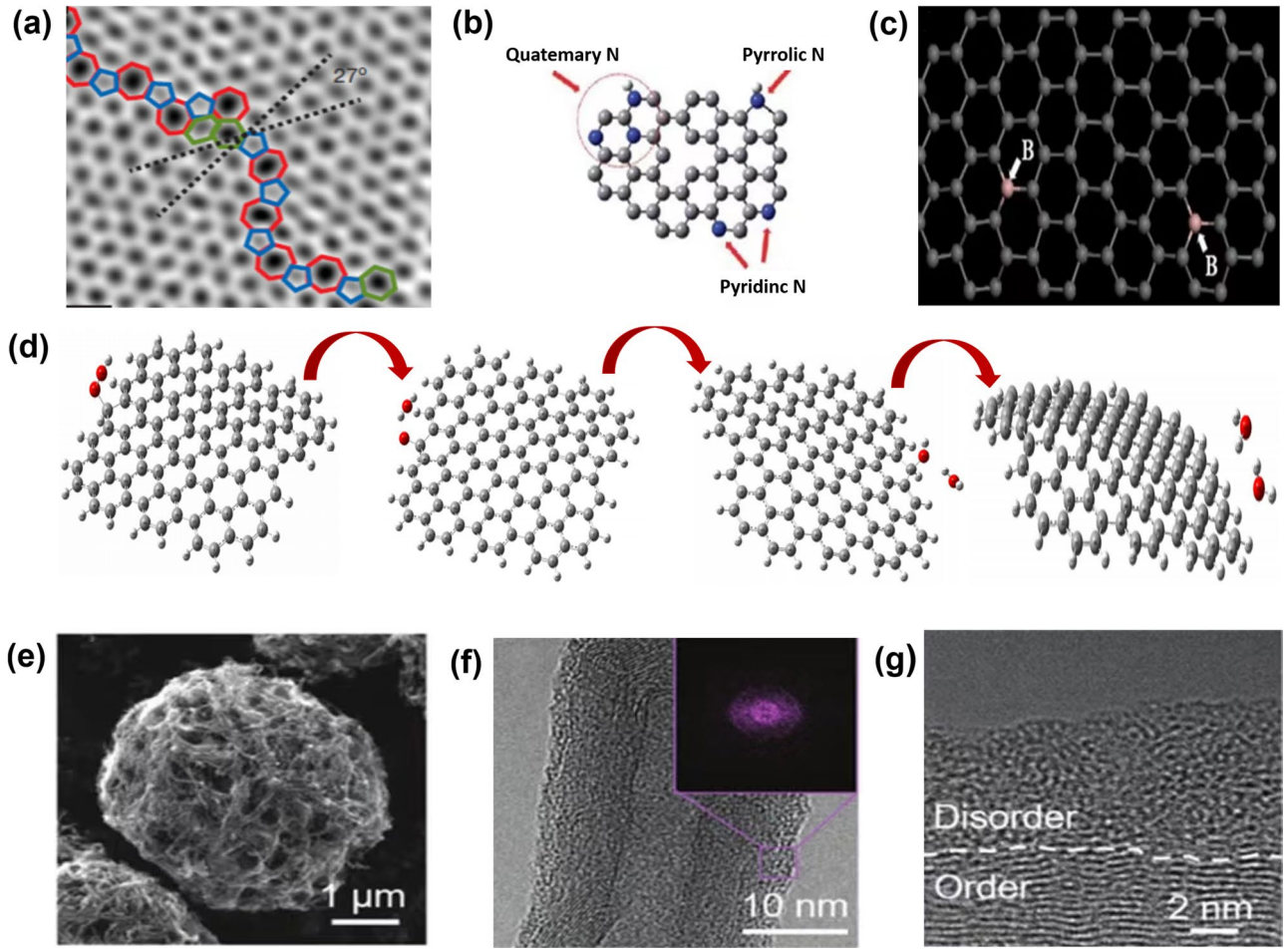
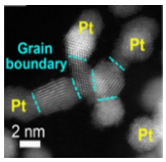
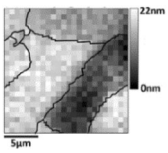
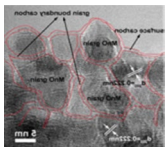
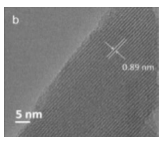
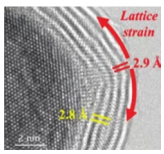
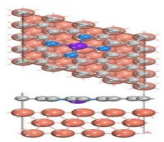
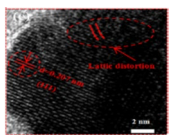
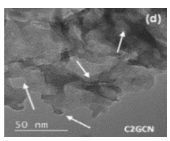
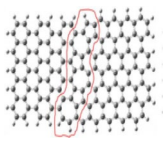
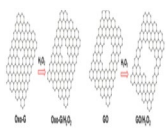


Fig. 5 Location of lattice defects. **a** ADF-STEM of line defects in graphene [61], **b–c** graphene in-plane heteroatom substitution defect model of nitrogen defects and boron defects [64], **d** four electron transfer processes of ORR on graphene (the larger grey, red, and smaller white balls denote to carbon, oxygen and hydrogen atoms, respectively) [65], **e** SEM images of PCNTM, and **f** TEM image of ePCNTM-20, **g** AC-HRTEM image of ePCNTM-20 [66]

and boron atoms can improve the electrical conductivity of graphene. Therefore, graphene is widely used in gas sensors to detect gas concentration and has strong adsorption energy for CO, NO, and NO_2 molecules, which can effectively reduce greenhouse gases and alleviate the impact of excessive consumption of traditional energy from the source (Fig. 5b–c) [64].

To meet the needs of different applications for the physical and chemical properties of materials, the electronic structure of a material can be tailored by regulating lattice defects. Zhang et al. [65] studied the impact of catalytic activity by the DFT method, on the midpoint and line defect in graphene clusters. As shown in the Fig. 5d, OOH is initially adsorbed on the carbon atom, then the O–O bond breaks to form water molecules, followed by the formation of OH molecules. Finally, two water molecules leave the graphene surface. Therefore, by controlling the defect location and adjusting the active site, then the enhancement of material performance can be realized effectively. In addition, lattice-defected graphene could be utilized as a promising route for cathode catalyst fuel cells applications. To date, the recent development of graphene which is generated via a simple and scalable spray-drying method and "Sauna" activation has been reported by Zhang et al. [66]. They fabricated a porous microsphere structure that composed of defect-rich interweaving atomic defects inherently found in carbon. The carbon nanotubes are endowed with an enhanced ability to react with sulfur and higher catalytic activity. The samples were reported to exhibit certain resonance signals at $g = 2.003$, indicating the presence of unpaired electrons in both pristine and etched carbon structures. However, an incremental change of EPR is found as the increasing of etching time and simultaneously the peak intensity is preserved for a long time, which is a clear indicator of carbon defect accumulation. This "sauna" (water-steam etching under high temperature) not only able to activate defects

Table 1 Applications and advantages of lattice-tuning methods in the field of new energy

Lattice structure methods	Carbon-based materials	Structure feature	Applications	Advantages	Ref.
Grain/twin boundary	$Ti_3C_2T_x$ nanosheets		Methanol oxidation	Improved electronic structure, and good electron conductivity	[42]
	Carbon steel		Hydrogen evolution	Rich coordinatively unsaturated sites	[43]
	MnO/C microspheres		Lithium-ion battery	Creates uniform interconnected 3D networks and fastens the speed of Li-ion and electron transport	[44]
	Polymeric carbon nitride		Solar H_2 production and CO_2 reduction	Improved quantum efficiency, and optical absorption	[67]
Lattice strain	Mesoporous carbon matrix		Hydrogen evolution	Modify the work function of the carbon matrix, resulting in the DGH* close to zero	[48]
	FeNC		Oxygen reduction reaction	Improved stability, add extra coordination	[49]
Lattice distortion	Fe-enriched-$FeNi_3$/NC		Zn-air battery	Improved charge distribution and durability performance	[55]
	C–g–C_3N_4 nanosheets		Photocatalytic hydrogen generation	Increased photoactive sites and improved photocatalytic efficiency	[68]
Lattice defects	Graphene clusters		Fuel cells	Improve oxygen reduction reaction activity, and electrocatalytic capability in energy conversion	[65]
	N-Graphene		H_2O_2 production	Increased selectivity for H_2O_2 production	[69]

that intrinsic topological carbon lattice but also promote the excellent adsorption and catalytic activity of porous carbon nanotube microspheres (ePCNTM). Secondly, the intrinsic lattice defect engineering in carbon-based materials decorated by the sulfur host can solve problems such as the excessive polysulfide shuttling and their slow reaction kinetics.

3 Conclusions and perspectives

In this review, we summarize four different lattice tuning approaches to optimize the optoelectronic properties, durability, and stability of carbon-based materials. These four lattice manipulations have their advantages and applicable applications, as shown in (Table 1) [42–44, 48, 49, 55, 65, 67–69]. The modulation of the lattice mainly occurs in the process of synthesizing the material, the edges of the nanosheets are curling, stretching, or sliding in the lateral direction at the boundary where the lattice changes intensely. It is also the specific location in which the active sites are populated. The interior of the lattice is twisted and compressed when subjected to external forces such as ultraviolet light irradiation, high temperature, pressure, and vibration caused by microwave radiation. During the photocatalytic reaction process, these active sites can adsorb oxygen (oxidant) enhancing the electron transport. In addition, the catalytic efficiency and generation of defects caused by the lattice changes is favorable via atomic doping, as consequence, strengthening the chemical and physical interaction between different metals to avoid the poor electrical conductivity of traditional 2D carbon materials.

Multiple in-depth studies of 2D have shown great progress in the development of photocatalysis. In summary of current relevant research in the past decades, the work on modulation of the lattice structure of 2D carbon-based materials is likely to have an impact on many disciplines. First, on the case of a polycrystalline nanosheets or nanoparticles, alternate different lattices or crystal planes are beneficial to form poly grain boundaries. In addition, a large number of active sites of 2D carbon-based materials can enhance the adsorption of oxygen in photocatalytic methane conversion and photocatalytic hydrogen production applications, and thus product selectivity can be enhanced. In particular, for carbon materials, which are relatively stable and symmetrical in their structures. Twin boundaries are special boundary structures, forming close to the linear defects. The mutual interaction of twin and poly grain boundaries could promote the carbon materials that are structurally-stable, and this can be applied at high temperature, pressure, and a wide range of pH in photocatalytic reactions.

Second, the lattice strain of 2D carbon-based materials can be categorized as a structural modification of lattice sliding. Here, different lattices are squeezed and pulled up each other such that the cavities formed into a regular rectangle. More importantly, the mechanical strength is an excellent property of carbon materials and the lattice distortion on carbon materials can improve the mechanical strength and thereby promoting the field of mechanical devices.

Compared to the metal-based materials, carbon materials have relatively poor electrical conductivity, and the defects within their structures could be considered to promote active sites for atomic doping. For example, the electrical conductivity of carbon materials can be improved via doping metals. However, doping atoms through lattice manipulation still faces many fundamental challenges, including the number of single-atom doping sites registry, the reduction of the chemisorption energy barrier, and precise control of electron cloud structures, and chemical bond variations.

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Declarations

Conflict of interest The authors declare that they have no competing financial interest.

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