Research Progress: Characterization and Repair of Graphene Defect

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Abstract: Graphene has extremely high theoretical properties and wide range of applications in metal-based composites. However, due to defects on its surface, the actual performance was far lower than the theoretical. In addition, graphene containing defects could easily react with a matrix alloy (such as Al) to generate brittle phase and hydrolyzed phase, which further reduced the performance of the composite. Therefore, studying the repair of graphene defects was an important area of graphene research. The repair methods reported in the paper are mainly CVD repair, doping repair, liquid phase repair, external energy graphitization repair and alloying repair. Moreover, a detailed analysis and comparison on above methods are carried out, and the characterization methods of graphene are also introduced in this paper. The mechanism, important research value and future outlook of graphene repair have also been discussed in depth.

Keywords: Graphene; Metal Matrix Composites; Defect Repair; Characterization; Repair Mechanism.
1. Significance of Graphene Defect Study for Composites

Graphene is a unique two-dimensional carbon material with a honeycomb-like lattice structure and delocalized electrons. In the ideal state, it has been widely valued by scientific researchers because of extremely high strength [1], electrical conductivity [2], and thermal conductivity [3]. Especially in the field of metal matrix composites, due to the high performance of graphene, it could be added to enhance the properties of the composites.

In the research area of Al matrix composite, graphene was particularly widely used. Bastwros et al. [4] reported that a maximum enhancement of 47% in flexural strength was observed when compared with the reference Al6061. Zhang et al. [5] explored that the enhancements of 15.4% in thermal conductivity, 9.1% in specific heat capacity, 21.1% in hardness, and 25.6% in compressive strength were achieved with only 0.3wt% graphene addition into pure Al. The experimental results showed that the graphene could greatly improve the overall performance of Al matrix composites. However, it was found that the properties of graphene reinforced Al matrix composites were directly related to the defects of graphene. As the content of graphene defects increased, the load transfer enhancement effect of graphene was destroyed, and the performance of composite materials was also decreased [6]. Al₄C₃ was also a harmful phase for graphene-reinforced Al matrix composites. Xin et al. [7] and Liu et al. [8] have reported that the generation of Al₄C₃ would destroy the interfacial bonding of C-Al, resulting in degradation of composite properties. Ci et al. [9] and Zhou et al. [10] found that Al₄C₃ was preferentially generated at the defects of graphene structure.
Therefore, in order to improve the load transfer ability of graphene, reduce the
generation of interface phase Al₄C₃, and improve the structural integrity of graphene has
become an important research direction.

2. Characterization of Graphene Defect

The defects in graphene are mainly divided into functional group defects and
lattice defects. Functional group defects are mainly oxygen-containing functional
groups attached to the surface of graphene, such as hydroxyl, carbonyl, and carboxyl
groups, which are widely present in graphene oxide. The distribution of functional
groups in graphene is unevenly island-like. Erickson et al. [11] observed that the
isolated and complete sp² structure was surrounded by a large number of sp³ regions
destroyed by oxygen-containing functional groups. The presence of oxygen-containing
functional groups would greatly reduce the conductivity of graphene, increase the
thickness of single-layer graphene, change the graphene interface structure, and improve
the hydrophilicity of graphene.

Lattice defects were mainly manifested as holes and edge defects in the graphene
plane. The carbon atom was mainly sp³ C, which destroyed the delocalized electronic
distribution and leaded to the degradation of graphene performance.

The macroscopic characterization of graphene defects was mainly achieved
through Raman (Raman Spectra), XPS (X-ray Photoelectron Spectroscopy), IR
(Infrared Spectrometry) and conductivity performance testing. The microscopic
characterization was mainly realized through STM (Scanning Tunneling Microscope),
AFM (Atomic Force Microscope) and TEM (Transmission Electron Microscope)
techniques.

2.1 Raman

Raman is a reliable means of reflecting the defects of graphene. Graphene has four main peaks in the Raman characterization, which are the D, G, D+G and 2D peaks (2D peaks were also written as G' peaks). The D peak appearing around 1400 cm\(^{-1}\) reflected asymmetric lattice vibration, and the G peak appearing around 1500 cm\(^{-1}\) reflected symmetrical lattice vibration, which could be considered to be the characteristic peak of the sp\(^3\) and sp\(^2\) C atom, respectively. Many researches used \(I_D/I_G\) to reflect the defect content in graphene, and as the number of graphene layers were increased, the peak position of the G peak would red shift \([12, 13]\). The 2D peaks appearing at around 2700 cm\(^{-1}\) were related to the number of layers, and the shape variety of the 2D peaks in the graphene with a small number of layers (1-5 layers) was particularly obvious. Wu et al. \([14]\) systematically summarized the characteristics and physical significance of Raman peaks. The second-order Raman peak (2D peak) with single Lorentz peak type was a simple and effective method for determining single-layer graphene, and multi-layer graphene had a 2D peak with multiple Lorentz peaks due to the electronic band structure split \([15]\). In addition, as 2D peaks were red-shifted when electrons were doped and blue-shifted when holes were doped, Raman could also be used as a criterion for determining the doping type and concentration of graphene \([16]\). Researches usually used \(I_D/I_G\) and \(I_{2D}/I_G\) to reflect the change of defects, and the surface distribution of defects could be directly reflected by surface scanning \([17, 18]\), as shown in Fig.1. The D+G peak around 2960 cm\(^{-1}\) was related to the defect density, and could also reflect the
defect content in graphene. In the Raman characterization of defective graphene, a special D' peak appeared near 1620 cm⁻¹. Eckmann et al. [19] found that the D' peak was related to the type of defect. For defects generated by sp³ C, $I_D/I_{D'}$ was the largest about 13. For gap type defects, this ratio was about 7. And for graphene edge type defects, this ratio was the smallest about 3.5.

![Image](image.png)

Fig.1 (A) Schematic illustration of the reduction of graphene oxide and single vacancy defect healing during CVD treatment. (B) Scanning electron micrograph of rGO flakes on Si/SiO₂. Raman maps of the same area (100 × 200 μm) showing $I_D/I_G$ ratio of rGO before (C) and after CVD annealing (E). CVD annealing conditions: 750°C, 990 sccm Ar, 5 sccm H₂, 5 sccm IPA, 10 min. Raman maps showing fwhm (2D) of rGO before (D) and after CVD annealing (F). Scale bar in (C−F) is 20 μm. Arrows indicate the positions (1, 2, 3) of single Raman spectra of monolayered rGO [18].

In addition, Raman data could assist in estimating the defect distance ($L_D$) [20, 21], the defect density ($n_D$) [22] and the crystal size ($L_a$) [13, 23], which was helpful for quantitatively characterizing the distribution of defects. Raman could also reflect the two states of the graphene defect distribution. When the average defect distance $L_D>3$ nm, the sp² C was the main form of C atom, and $I_D/I_G$ increased as the defect density increases. When $L_D<3$ nm, large number of defect structures were distributed on the
surface of graphene, and $I_D/I_G$ decreased with increasing defect density.

2.2 XPS

XPS is a commonly used surface analysis method. It could accurately calibrate the valence and content of atoms by analyzing chemical shifts. It has been often used in the field of graphene to characterize the carbon/oxygen ratio (C/O) and carbon atom hybrid state of materials, as shown in Fig.2. Daukiya [24], Hafiz [25], Lesiak [26], Dwivedi [27] and Xie [28] studied the binding energy of different carbon structure, respectively. It was concluded that the binding energy of $sp^2$ C was about $284.4 \pm 0.3$ eV, and the binding energy of $sp^3$ C was about $285.2 \pm 0.3$ eV. The oxygen-containing functional group could also be characterized by XPS. Generally, the binding energy of -OH, -C-C=O, and C=O were around 285.7, 287.5, and 288.7 eV, respectively [25]. What’s more, IR is also one of the methods to help characterize functional groups, which was widely used in the field of graphene oxide reduction characterization [29].
Fig. 2 Global measurements (XPS and ARPES) of graphene before and after the reaction with FMAL molecules (dipping duration 80 h). (a and b) Deconvoluted C 1s spectra before and after dipping, respectively. The successful reaction of FMAL molecules is represented by the change in the sp² component which is associated with graphene and sp³ (dotted lines). (c) F 1s spectra corresponding to the CF₃ groups of the FMAL molecule, and (d) N 1s components corresponding to the to C-O – N-O-C group, which ensure that the molecule is intact. (g) Evolution of the sp3 component of C 1s spectra and F 1s spectra as a function of dipping time. (e and f) Band structure measured by ARPES for monolayer and bilayer graphene before and after the D-A reactions, respectively. The constant momentum dispersion curve shows a dip in the intensity as a tendency for opening of a gap. (h) Evolution of the Fermi velocity for ML graphene as a function of the immersion time. [24].

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2.3 STM, TEM and AFM
Both STM and AFM are excellent means of characterizing the number of graphene layers. The thickness of graphene could reflect the number of graphene layers and surface functional groups. Generally speaking, the thickness of graphene was about 1 nm when single-layer graphene had oxygen-containing functional groups on its surface. When there was no functional group on the surface of single-layer graphene, the thickness was close to the theoretical value of 0.34 nm. The thickness of single-layer graphene could be used to judge the effect of functional group defect repair. In addition, Rozada et al. [30] also found that STM as more conducive to analyzing the defect repair mechanism of graphene. As STM was more sensitive to electrons, some structural defects were not seen by AFM could be observed in atomic STM. TEM is also a useful tool to assist in determining the structure of graphene [31]. It was difficult to directly analyze the number of graphene layers through STM and AFM in composite as the graphene was dispersed in matrix. Generally, choosing TEM to directly observe the structure of graphene was appropriate [29], as shown in Fig.3. The graphene surface observation results are shown in Fig.4. Rozada [30] observed and analyzed the hole defects on the surface of graphene by STM.
Fig. 3 TEM images of (a) FLGO and (b) FLG sheets. The insets show GO and rGO layers at the edges of nanosheets [29]. Copyright © 2017 Elsevier B.V.

Fig. 4 Nanometer-scale STM image of a highly reduced graphene oxide sheet folding over itself supported onto the pristine HOPG surface after annealing at 1923 K (a). Schematic illustration of the possible configurations of a pit in the region of overlapping of highly reduced graphene oxide sheets (b). Nanometer-scale (c) and atomic-scale (d) STM images of the region of overlapping shown in (a). Nanometer-scale image of highly reduced graphene oxide sheets deposited on top of an HOPG substrate previously exposed to dielectric barrier discharge (DBD)-generated plasma prior (e) and after annealing at 2073 K (f). The insets to (e) and (f) are detailed images of the HOPG substrate modified with the DBD-generated plasma before and after annealing at 2073, respectively. Imaging conditions: 0.2 nA (tunneling current) and 800 mV (bias voltage) (a), 0.3 nA and 500 mV (c), 1.5 nA and 5 mV (d), 1 nA and 100 mV (e, f) [30]. Copyright © 2017, Royal Society of Chemistry.
3. Research Progress of Graphene Defect Control

The repair of functional group defects (especially oxygen-containing functional groups defects) has been extensively studied. Reducing agents such as hydrazine hydrate [32], alcohols [33], and sodium borohydride [34] were used for thermal reduction. This reduction method has been widely used in the large-scale preparation of reduced graphene, and the stable dispersion and surface modification of graphene sheets were achieved while the oxygen-containing functional groups were removed. However, the reducing agent could not improve the integrity of the graphene skeleton structure, and new defects would be introduced during the reduction process. Therefore, in order to obtain higher quality graphene, further lattice defect repair was needed.

There were many ways to repair the defects of graphene lattice defects. The main ones that have been reported are CVD repair (Chemical Vapor Deposition), doping repair, liquid phase repair, external energy graphitization repair (such as high temperature, microwave, and irradiation) and alloying repair.

3.1 CVD Repair

The CVD repair process mainly repairs holes through the decomposition of molecules in the gas source or the carbon source generated by the plasma at high temperature. Due to the higher reactivity of the carbon atoms at the defect, the repair and growth of the graphene holes and edges was preferentially performed [35]. The reaction temperature was from 500 °C to 1000 °C. The choice of gas source was usually methane [36, 37], hydrogen [18], ethylene [38], ethanol [39] and their plasma.

Zhu et al. [36] used H₂ to assist CH₄ for graphene repair, and found that CH₄ would
decompose at high temperature to generate $\text{CH}_x \! (x = 1, 2, 3)$ plasma, which had a high repair activity. Specially, Zhu [36] used $\text{H}_2$ to corrode the edges of graphene, promote the growth of new graphene, and inhibit the formation of $\text{sp}^3 \text{C}$, thereby achieving better defect repair effects. The introduction of $\text{H}_2$ could also effectively reduce the temperature of the CVD process, reduce instrument requirements, and save experimental costs. Zhou et al. [40] explored that $\text{CF}_4$ could also be connected with graphene-defective C atoms to form the high-dispersive fluorinated GO (FGO) with tunable atomic ratio of F/O (RF/O), and the RF/O could be readily manipulated just by adjusting the reaction time.

López et al. [38] used conductivity and Raman to characterize the graphene defects, and observed that the conductivity of graphene repaired by CVD has significantly improved, which proved that the defects in graphene were carbon vacancies and could be filled by CVD. The $I_D/I_G$ in Raman has still been greatly improved. It was due to the large mismatch between the newly grown CVD graphene and the original lattice structure. Lattice mismatches could cause defects, leading to the increase of D peaks in Raman. In addition to direct gas source repair, Kim et al. [41] used CVD to grow graphene balls on the Ni surface, and used graphene hollow spheres after removing Ni to repair defects as shown in Fig.5. The material had an electrical conductivity of 18620 $\text{S/m}$ and a specific surface area of 527 $\text{m}^2/\text{g}$. 

![Diagram of graphene repair process](image-url)
3.2 Doping Repair and Liquid Phase Repair

The process of liquid phase repair and doping repair was that the atoms diffused into the graphene defects under the liquid phase condition, thereby preparing carbon atom filled or heteroatom doped graphene structures.

Cao et al. [42] used ethanol molecules to intercalate between graphene layers and performed heat treatment in a microwave environment. It was found that the graphene layer was separated during the decomposition of ethanol. The carbon atoms in ethanol were calibrated by means of isotope tracing. After treatment, the content of $^{13}$C atoms in the graphene structure increased significantly, proving that the carbon atoms in ethanol entered the graphene lattice and played a repairing role. The study also found that methanol did not repair the graphene under the same process. It was because under the experimental conditions, methanol could not break down like ethanol to repair graphene defects. It also proved that the repair of graphene defects was by C atom rather than molecular.

Tung et al. [43] used poly (1-vinyl-3-ethylimidazolium bromide) as the raw material to prepare an N-doped graphene structure, and the repaired graphene had a large area and a complete lattice structure by TEM observation. XPS proved N doping into the graphene lattice structure. Omidvar et al. [44] prepared GO/Pd, characterized the repair status of oxygen-containing functional groups by IR, and found that the doping of Pd caused the original functional groups to be redshifted, which reflecting the interaction between Pd atoms and graphene. From the photoluminescence spectrum test,
the luminous intensity of GO/Pd was significantly higher than that of GO. It reflected the increase in the sp² structure in graphene, which indirectly proved that Pd has repaired the defect in the graphene.

The liquid phase repair and doping repair process has the advantages of simple equipment, high yield, and low price. The process conditions are similar to those of the thermal repair process, so it is often performed together with functional group repair. It is currently widely used in the field of large-scale reduction of graphene oxide. However, these two repair methods can only partially fill the holes, and cannot achieve atomic diffusion and rearrangement in the defect area. Therefore, the theoretical strength of the graphene repaired by this method still has a large gap.

3.3 External Energy Graphitization Repair

High temperature repair of graphene structure was a stable and reliable method for repairing lattice defects. Through self-diffusion of carbon atoms in high temperature (usually higher than 2000 °C) environment, the structure could be graphitized, and defects such as holes in graphene could be repaired.

Xin et al. [45] prepared graphene sheet fibers under high temperature conditions. Performance showed that the thermal conductivity was increased from 400 W/(m·K) to 1300 W/(m·K) after heating, and the electrical conductivity was increased from 0.8×10⁻⁵ S/m to 2.2×10⁻⁵ S/m. Both the tensile strength and Young's modulus were greatly improved. Moreover, the D peak of the samples treated at 2850°C could not be observed from the Raman characterization, which proved that the graphene defects were basically disappeared at high temperatures. Ruan et al. [46] have also done graphene
thermal repair research in the field of electrochemistry, and found that samples repaired at high temperature showed higher cyclic stability and lower AC (Alternating Current) impedance. It was also found in Raman and XPS that the defects of graphene have basically disappeared, as shown in Fig.6.

Fig.6 (a) Typical SEM images of 2850CNTs–Gra; (b) Raman spectra and (c–d) XPS curves of the 2850CNTs–Gra and CNTs–Gra [46]. Copyright © 2018, Royal Society of Chemistry.

Rozada et al. [47] used two-step heat treatment to perform graphene repair, and made a detailed study on the thermal defect repair mechanism of graphene. It was found that with the increase of the heat treatment temperature, $I_D/I_G$ was decreased significantly, whereas $L_c$ (the crystal size calculated by XRD) and $L_a$ (the crystal size calculated by Raman) both were increased significantly. The performance of the graphene treated at the highest temperature (HG1500-2700) was close to that of highly oriented pyrolytic graphite, and almost all defects were repaired. Obvious stacking
structures were found under high-resolution STM characterization, and a 3° difference existed between the angle of the stacked graphene and the original region. It was suggested that the graphene structure graphitization process was continuous and layer-by-layer crystallization.

In addition, Sun et al. [48] carried out related research on Ni etching-assisted graphene thermal repair, and found that Ni atoms adsorbed around the saturated hydrocarbon structure and destroyed the C-H bond to act as a catalyst to promote the growth.

In addition to traditional heating, irradiation and microwave heating were also used to provide energy for graphene repair. Chen et al. [49] found that the interlayer spacing of GO was decreased and amorphous C-C was increased after electron-beam irradiation. Shi [50], Xu [51] and Zhang [52] et al. reported that γ-ray could etch the edge defects of graphene, and promoted the combination of graphene with metals or polymers to control edge defects. Under the action of γ-ray, it could also help the restoration of sp² structure. Shi et al. [53] prepared a graphene-reinforced PVA material and characterized its properties. As the radiation intensity increased from 0 to 100 kGy, it was found that the tensile properties of the material were gradually improved, and 2D peaks were also observed in graphene. This reflects the lattice structure of graphene was repaired. Xu et al. [54] found that irradiation could cause graphene $I_D/I_G$ values to rise and oxidize, which could be beneficial to the large-scale functionalization of graphene. However, the wrong irradiation conditions could also cause graphene defects to rise [55]. Voiry et al. [56] used microwave to repair the defects of graphene oxide (Mw-rGO). It was found
that the repair effect of Mw-rGO was significantly higher than that of ordinary chemically reduced graphene, and slightly lower than that of CVD-grown graphene and highly oriented pyrolytic graphite, which showed that the microwave repair method had an excellent repair effect. The original graphene structure was irregular, and there were oxygen-containing functional groups and holes on the surface, which affected the comprehensive performance of graphene. The repaired graphene had a very ordered lattice structure, and functional group defects and lattice defects were basically disappeared. This excellent repair effect was attributed to the rapid heating of GO during the microwave process, which achieved the decomposition of functional groups and the rearrangement of atoms in the plane.

The high-temperature graphitization repair process is one of the most effective repair methods for graphene skeleton defects, and is suitable for repairing most carbon material structures. However, high temperature has high requirements on the instrument, which limits the large-scale application and mass production of high temperature repair.

3.4 Alloying Repair

Alloying repair is a special repair method existing in composite materials. In composites, the interface is one of the key factors determining strength, and matrix alloying can achieve the segregation of elements at the interface. The segregated elements will be spontaneously adsorbed on the surface of graphene, filling graphene defects, and improving the interfacial bonding of graphene-matrix.

Shao et al. [57] found that in the graphene-reinforced 5083 Al, the Mg element was enriched spontaneously to the interface in the matrix, as shown in Fig.7. The structure
of Mg@GNPs was formed. After the Mg element was adsorbed on the surface of graphene, the production of Al₄C₃ was greatly reduced, and the tensile strength of the composite material was further improved. Guan et al. [58] researched Ni@graphene reinforced Al. The mechanical properties of composite materials were greatly improved. Growing graphene in metal matrix by CVD could also form a metal@graphene structure. Liu et al. [59] and Wang et al. [60] prepared Ni and Cu @GNS/Al by CVD, respectively. It could be clearly seen under TEM that Ni and Cu had a good combination with graphene. The mechanical properties of the CVD-grown graphene composites were much higher than those of the graphene-added composites. As the CVD-grown graphene was covered by Ni and Cu elements, its lattice structure had higher integrity. Complete graphene had a stronger load transfer effect and higher enhancement efficiency than graphene with defect.

Fig.7 Elements distribution in the GNPs/5083Al composite. (a) High-angle annular dark field (HAADF) image of the GNPs/5083Al composite and the corresponding distribution of the (b) Al, (c) C, and (d) Mg.
There are fundamental differences between alloying repair and three other repair methods, CVD repair, doping repair, liquid phase repair and external energy graphitization repair. The other four types of repair methods mainly introduced foreign atoms or promoted internal rearrangement of graphene, so as to reduce the defects of graphene and improve the integrity. However, alloying repair did not substantially reduce the number of graphene defects in fact. Alloying repair is through the adsorption of alloying elements on the surface of graphene, filling the defects of graphene and improving the integrity of graphene as a whole, so that the performance of defective graphene is close to that of complete graphene.

4. **Graphene Repair Mechanism**

It was reported that defects had different behaviors at different temperatures. According to the research results of Bagri [61] and Sun [62] et al., under low temperature conditions (about 300 ~ 400 K), the oxygen functional groups began to decompose, but some functional groups would be converted into stable functional groups (such as carbonyl groups). The stable functional groups began to decompose above 1300 K. When the functional groups on the graphene surface were thermally decomposed, the C atoms left the graphene lattice in the form of CO and CO$_2$, resulting in the generation of vacancies. C atom vacancies destroyed the integrity of the graphene lattice, and resulted in the generation of graphene lattice defects.

Graphene's defect change process at high temperature is shown in Fig.8. Rozada et al. [30] treated highly reduced graphene, middly reduced graphene and unreduced
graphene at high temperature. The graphene structure of the non-reduced samples was greatly damaged after high temperature heat treatment, and only small pieces of structure remained on the substrate. There were a large number of functional groups on the surface of unreduced graphene, and carbon atoms in the crystal lattice was taken away during thermal decomposition, which caused the graphene structure to be greatly damaged. It was also found that the edges of the graphene sheet were smoother after heat treatment, which was the result of the high temperature diffusion of carbon atoms. There are two forms of hole defects movement in the process. One of the process was hole defects merge together to form larger defects, and another was hole defect movement to the edge disappears.
Fig. 8 Nanometer-scale AFM images for the unreduced (a, d, g), mildly reduced (b, e, h) and highly reduced (c, f, i) graphene oxide sheets deposited on top of an HOPG substrate, prior to the thermal treatment (a–c) and annealed at 1773 K (d–f) and 2073 K (g–i) [30]. Copyright © 2015, Royal Society of Chemistry.

Cao et al. [42] studied the graphene repair mechanism with C source through $^{13}$C isotope tracer reaction. The repair process for hole defects is shown in Fig. 9. GO was rehabilitated with ethanol and methanol under microwave environment. Ethanol and methanol both were evaporated into gas and reduced the oxygen-containing functional groups on the surface and edges of GO. Ethanol cracked into carbene radicals and healed the defects by a radical–radical reaction with active carbon atoms from carbene. However, methanol could not crack into carbene radicals and interacted with the active
carbon atoms at the edge of the defect sites, thus the defects still remained. From the results of these studies, it could be seen that active atoms were the key factor in repairing the graphene lattice structure.

Fig. 9 (a) A schematic illustrating the graphene layer healed by 13C atoms. Original 13C atoms (blue), new 13C atoms (red) and single vacancy defects (green areas). (b) 1H-13C CP and direct 13C pulse MAS spectra of 13C-GE and GE. (c) Schematic illustrating of the CDE process. Focus defect (orange bond), carbon atoms for patching (shiny red ball), two kinds of mixed gases (green and violet) and healed graphitic structure (shiny red bond). Expansion shows the detail of the healed focus defect [42]. Copyright © 2014, Royal Society of Chemistry.

5. Conclusions

With the rapid development of graphene material performance, the impact of graphene defects on performance has gradually been valued. The characterization and repair of defects has become the focus of graphene research. At present, repair of graphene has been implemented, and macroscopic and microscopic representations are becoming more comprehensive. There are still two major problems in graphene research:
One of the key problems was the process conditions for graphene repair. The best graphitization repair requires extremely high temperatures. However, high temperatures exceeding 2000 K cannot be directly applied to composite and other fields, which would cause damage to other structures. Although the CVD repair and liquid phase repair are mild, the process is complicated, and the repair effect is not ideal. The advantage of Allo-ying repair is that it can be easily and directly applied to composite material systems, but the selection of elements has higher requirements.

Another problem was that the characterization of graphene in composite materials was still difficult. Graphene added to composites cannot be characterized by AFM and STM, and it was difficult to separate. Although it was reported in the paper that graphene could be removed by corrosion, the corrosion process would cause secondary damage to graphene. Due to the shielding and reflection effects of the base alloy in the matrix, the signals of Raman and XPS was also affected. Therefore, accurately characterizing the existence of graphene in the matrix was an important research direction for the future.

High-quality, few-layer graphene is costly and yields low. If low-quality graphene can be repaired so that its performance can reach the effect of high-quality graphene, it would dramatically reduce production costs and revolutionize the development of existing graphene products. With the deepening of research, it is believed that graphene repair would be improved and developed, and higher performance would continue to emerge.

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