

# Interfacial Engineering for High-Conductivity Copper/Graphene Composites by Combined CVD and Sputtering

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## Abstract

Copper is a fundamental material used in energy and electronic systems. However, achieving a balance between strength and conductivity in existing materials remains challenging, and Joule heating loss under high current restricts the development of high-power equipment. Therefore, it is crucial to develop new copper-based materials with high strengths and conductivities. Graphene is regarded as an ideal reinforcement material for constructing high-performance copper-based composites, owing to its excellent intrinsic properties. In this study, an innovative preparation strategy was proposed: high-quality graphene was grown in situ on copper foil by chemical vapor deposition, and a copper nanofilm was deposited by magnetron sputtering to construct a copper/graphene/copper composite sandwich structure, followed by densification via hot-press sintering. The results demonstrated a synergistic improvement in the electrical and mechanical properties of the composite: the conductivity reached 105.27% IACS, and the tensile strength was 240.32 MPa, both significantly better than those of pure copper. This study confirms that the material properties can be simultaneously optimized through precise interface and structural engineering, providing a new technical approach for developing efficient conductive materials for energy and electronic applications.

## Keywords

Chemical Vapor Deposition; Magnetron Sputtering; Copper-Graphene Composite Materials.

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## 1. Introduction

As a core basic conductive material, copper has expanded applications with the rapid development of high-power electrical and electronic devices, rail transit, and new energy technologies. [1–3] However, the energy loss and heating caused by the inherent Joule heating effect significantly constrain the performance enhancement of high-power equipment[4–6]. To address energy consumption and optimize the overall performance, researchers commonly employ alloying or the introduction of carbon materials for strengthening[7–9]. Traditional alloying often sacrifices conductivity to gain strength, and carbon-reinforced copper-matrix composites represent one of the most active research areas. In recent years, the focus has shifted from simple compositing to precise interfacial control[10,11].

Among the various carbon reinforcements, graphene, owing to its ultrahigh electron mobility and excellent mechanical properties, has attracted widespread attention and is considered an ideal reinforcement that simultaneously improves both the strength and conductivity of copper-based composites[12,13]. To date, researchers have explored various methods to prepare Cu/Gr composites. For instance, Yue et al.[14] prepared graphene nanosheet-reinforced pure copper matrix composites

via ball milling and hot-press sintering. Li et al.[15] completely coated copper powder with several layers of high-quality graphene by chemical vapor deposition (CVD), followed by composite preparation by vacuum hot-pressing. Based on a powder metallurgy strategy, Zhang et al.[16] constructed a three-dimensional continuous graphene network within a copper matrix via thermal stress-induced welding between graphene-like nanosheets grown on copper powder surfaces. These studies have shown that graphene can significantly improve the mechanical properties of copper-based composites, and Cu/Gr composites prepared by in situ growth of graphene on copper substrates often exhibit better electrical conductivity, approaching nearly 100% of the International Annealed Copper Standard (IACS). Because the actual enhancement of copper conductivity by graphene is closely related to the graphene quality and the copper-graphene interfacial bonding state, CVD is considered the optimal choice because of its ability to controllably prepare high-quality, large-area graphene with high cost-effectiveness[17–19]. The extremely low carbon solid solubility and self-limiting growth characteristics of copper substrates are particularly conducive to the in-situ growth of high-quality graphene via CVD, thereby avoiding contamination and damage from transfer processes and enabling the potential for simultaneous performance optimization[20,21].

However, the significant difference in the thermal expansion coefficients between copper and graphene induces considerable interfacial thermal mismatch stress during high-temperature processing. This stress often leads to the local exfoliation of graphene, forming a wrinkled network structure and compromising the physical continuity of the interface. Such interfacial defects weaken the mechanical load-bearing capacity, act as strong electron scattering centers, and hinder the efficient cross-interface transmission of load and charge, thereby limiting the comprehensive performance of the composite[22–24]. To mitigate the adverse effects caused by thermal mismatch and actively construct high-performance interfaces, this study introduces magnetron sputtering, a physical vapor deposition technique, as a solution. Controllable deposition of nanoscale film thickness, composition, and microstructure can be achieved by precisely adjusting sputtering power, pressure, and time[25,26]. This strong interfacial bonding can effectively alleviate stress concentration and inhibit graphene instability under thermal stress, which is significant for simultaneously improving the mechanical and electrical properties of the composite. Therefore, high-quality graphene was grown in situ on a copper foil surface via CVD to achieve good initial interfacial contact. In combination with magnetron sputtering, copper nanofilms (NFCu) were accurately deposited on the graphene surface to construct a Cu/Gr/Cu sandwich structure. Finally, diffusion bonding and overall densification of the structure were achieved via hot-press sintering.

Fig. 1(a) shows a schematic of the preparation process for Cu/Gr/Cu composites, including copper foil pretreatment, annealing, CVD growth of nanofilm graphene, sputter deposition of a copper film, and hot-press sintering densification.

### 1.1 Copper Foil Pretreatment and Graphene Synthesis

Copper foil (commercial, 99% purity) was used as a substrate for graphene growth. It was ultrasonically cleaned in acetone and absolute ethanol, followed by electrochemical polishing in 5% nitric acid solution. The treated copper foil was then heated to 800°C in an atmosphere with an argon/hydrogen flow rate of 400/80 sccm, and held for 60 min for annealing. Subsequently, the temperature was increased to 1050°C and a carbon source gas mixture (C<sub>2</sub>H<sub>4</sub>:Ar = 1:5) at 20 sccm was introduced and maintained for 10 min. Finally, the Ar gas flow rate was increased to 500 sccm and the furnace was cooled to room temperature.

### 1.2 Magnetron Sputtering Process for Copper Films

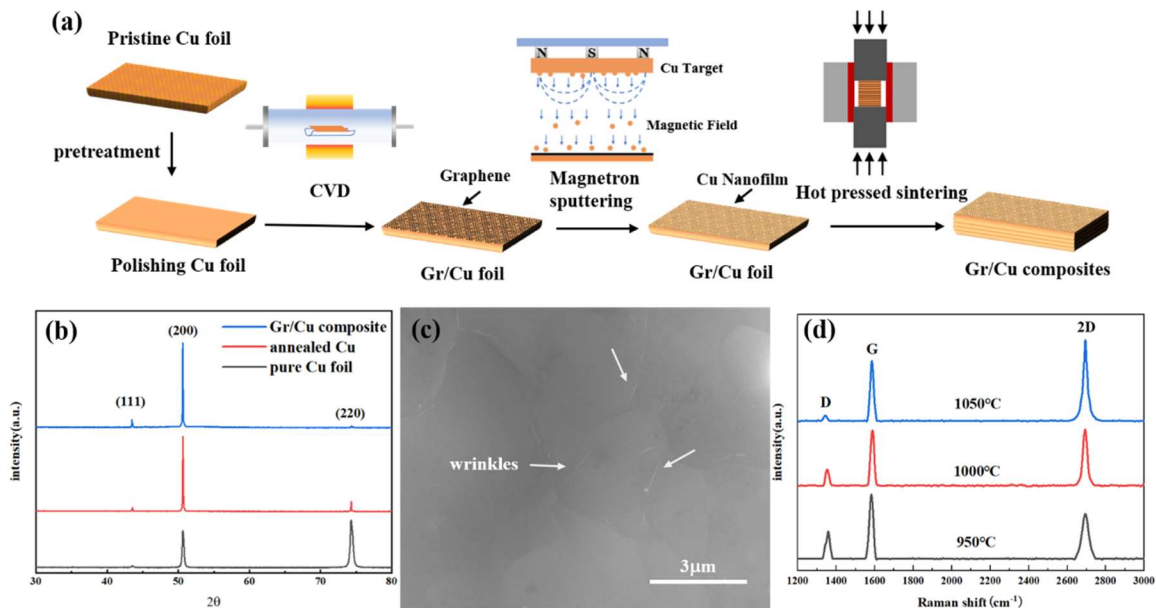
A nano-copper film was deposited by magnetron sputtering using a copper target (99.99% purity). When the vacuum chamber pressure was below  $3 \times 10^{-5}$  Pa, argon was introduced as the protective gas. The rotation speed of the sample stage was set to 5 rpm and the sputtering power was 200 W. After pre-sputtering the copper target surface for 30 s to remove impurities, deposition commenced and lasted for 10-60 minutes to complete the coating process.

### 1.3 Preparation of Cu/Gr Composite Materials

The Cu/Gr composites were prepared via hot-press sintering. NFCu/Gr/Cu composite copper foils were stacked in a hot-press furnace. Under vacuum, the temperature was increased to 800°C at a heating rate of 10°C/min, maintained at a pressure of 50 MPa for 30 min, and then cooled to room temperature at a cooling rate of 10°C/min. Finally, Cu/Gr composites with a diameter of 30 mm and height of 10 mm were obtained.

### 1.4 Characterization Study

X-ray diffraction (XRD) analysis of the composites was performed using an X-ray diffractometer (SmartLab 9kw). The graphene quality was assessed by Raman spectroscopy (B&W Tek) with a 532 nm laser source. The surface morphology and microstructure of the samples were analyzed by scanning electron microscopy (SEM, Quanta450) and transmission electron microscopy/high-resolution transmission electron microscopy (TEM/HRTEM, Talos F200X). The TEM samples were prepared by mechanical grinding to a thickness of 50 μm, followed by ion-beam thinning. Room-temperature tensile tests were conducted on an Instron-1186 tensile testing machine using dogbone-shaped specimens with gauge lengths of 15 mm, gauge widths of 2 mm, and thicknesses of 1 mm. The conductivities of the samples were measured using a four-probe square resistance/resistivity tester (Ningbo Ruike Analytical Instrument Co., Ltd.). The test samples were wire-cut to the dimensions of 20 mm × 4 mm × 1 mm.



**Fig. 1** (a) The schematic diagram of the preparation process of Cu/Gr/Cu composites (b) XRD patterns of pure Cu, annealed Cu, and Gr/Cu structure (c) SEM images of the graphene (d) Raman spectra of the graphene

## 2. Results and Discussion

### 2.1 Preparation and Characterization of Graphene

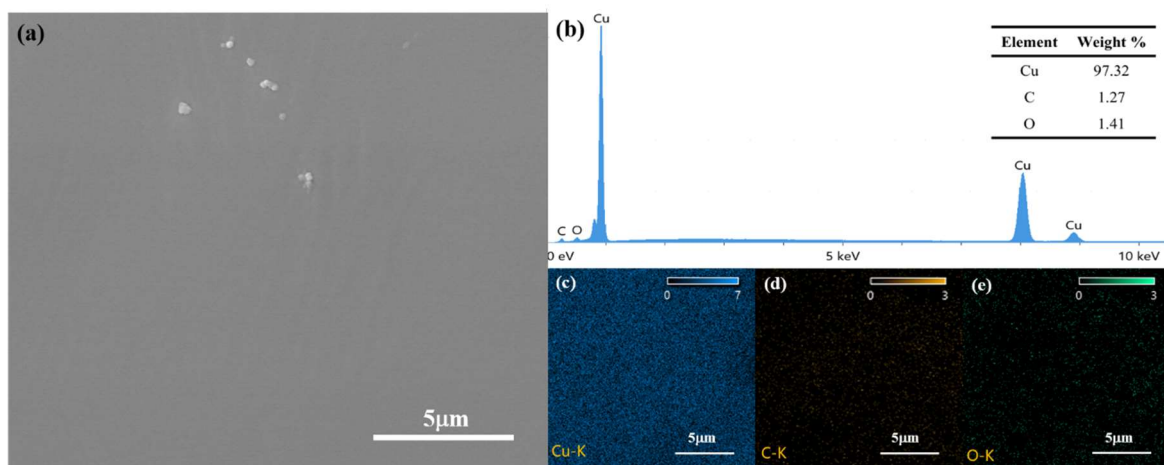
Graphene quality is a key factor determining the electrical conductivity and mechanical properties of the composites, and a suitable growth substrate is a prerequisite for obtaining high-quality graphene. As shown in Fig.1(b), this study systematically analyzed the effect of copper foil crystallization on graphene growth quality by comparing the XRD patterns of the original copper foil, annealed copper foil, and CVD-grown graphene samples. The results show that after annealing, the intensity of the Cu (220) diffraction peak of the copper foil significantly decreased, while the intensity of the Cu (200) peak significantly increased, and its full width at half maximum (FWHM) decreased. This change indicates that the annealing process promotes structural rearrangement of the copper foil surface,

leading to more consistent grain orientation, increased grain size, and the formation of a substrate with high crystal quality and low surface energy defects. This structure is conducive to the uniform adsorption and diffusion of the carbon source on the copper surface, providing ideal conditions for the subsequent growth of high-quality graphene with large grain size, few grain boundaries, and complete structure[27].

Fig.1(c) shows the characterization results of the prepared graphene microstructure by SEM. It can be observed that a continuous and complete graphene film was successfully formed on the copper foil surface. In the grain boundary regions of the copper foil, graphene exhibited smooth and uniform film characteristics, visually confirming its ability to grow continuously across grain boundaries. Simultaneously, the film surface showed typical wrinkled structures generated during cooling because of the significant difference in the thermal expansion coefficients between graphene and the copper substrate. This morphological feature indicates that the prepared graphene film has a good structural integrity and can continuously cover the substrate. The formation of wrinkles is attributed to the strong interaction between the film and substrate and the thermal mismatch stress, indirectly indicating the formation of a close interface between graphene and the copper foil, rather than simple physical adhesion[28]. These morphological characteristics are typical indicators of high-quality low-defect graphene films.

Fig. 1(d) shows the Raman spectra of the graphene grown at different temperatures. The D, G, and 2D peaks are located near  $1350\text{ cm}^{-1}$ ,  $1585\text{ cm}^{-1}$ , and  $2695\text{ cm}^{-1}$ , respectively. The D peak reflects the degree of structural disorder in the graphene, the G peak represents the lattice structure, and the 2D peak intensity indicates the stacking state[29]. With increasing growth temperature, the D peak intensity gradually decreased, indicating that heating effectively promoted surface migration and orderly arrangement of the carbon precursor, thereby reducing the graphene defect density and improving the crystal quality. The 2D peak intensity increased significantly and its FWHM narrowed. Finally, at the optimal temperature, a sharp and symmetrical Lorentzian line shape was observed. This feature is highly consistent with the typical spectrum of few-layer graphene, confirming the effective layer number control[12]. This demonstrates that by adjusting parameters such as the growth temperature and gas flow rate during the CVD process, graphene films with controllable layer numbers, high crystal quality, and low defect density were successfully prepared on copper foil substrates, providing an ideal material foundation for subsequent composite property research.

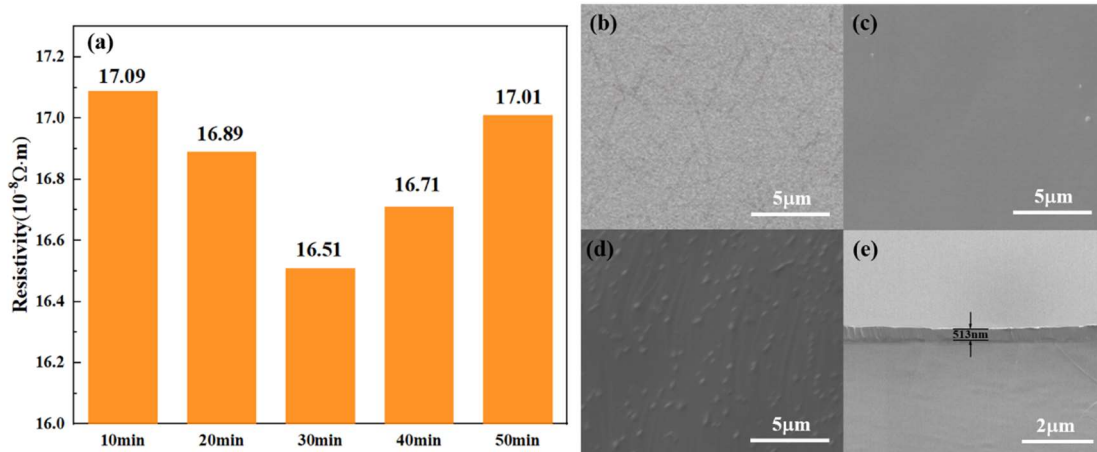
## 2.2 Process and Characterization of Magnetron Sputtering of Copper Nanofilms



**Fig. 2** (a) Scanning electron microscope images of sputter deposited copper nanofilms ;(b –e) shows the energy spectrum element mapping of Cu/Gr/Cu composite structure after copper deposition.

Fig. 2(a) shows an SEM image of the copper nanofilm layer deposited at 200 W for 30 min. It can be clearly observed from the image that the deposited copper film had no obvious defects such as holes or cracks. Simultaneously, influenced by the underlying graphene wrinkles, some micro-ridge

structures were formed, displaying a highly dense, uniform, and continuous-coverage state. Fig. 2(b-e) presents the EDS elemental distribution analysis results of the deposited copper film. The content of C and O elements is very low, at 1.27 wt% and 1.41 wt%, respectively, with no aggregation of C or O elements in the corresponding elemental maps. This result indicates that the prepared copper nanofilm not only completely covered the graphene layer but also possessed high intrinsic quality and excellent compactness. The uniform distribution of trace oxygen is mainly attributed to surface physical adsorption caused by brief exposure to the atmosphere after deposition, rather than the deposition process itself.



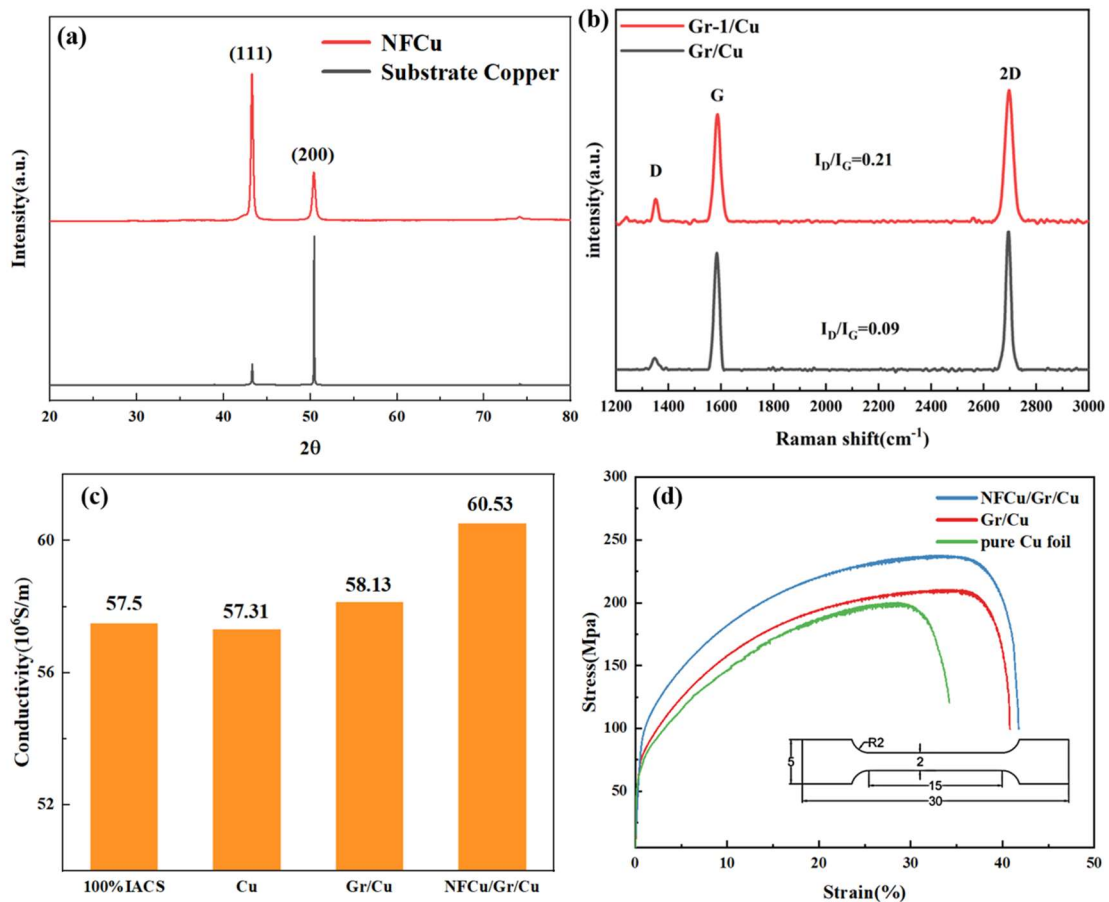
**Fig. 3** (a) The resistivity of NFCu / Gr / Cu at different sputtering time;(b-d) SEM images of copper films prepared by sputtering time of 10 minutes, 30 minutes and 50 minutes;(e) Cross-sectional SEM images of deposited nano-copper films.

To systematically study the influence of magnetron sputtering time on the conductivity of the composites, we tested the resistivity of Cu/Gr/NFCu composite foils prepared at a sputtering power of 200 W for times ranging from 10 to 50 min. The results are shown in Fig. 3(a). As the sputtering time was increased, the resistivity of the composite initially decreased significantly. This is primarily because a longer sputtering time allows for a more sufficient deposition of the copper nanofilm, improving the film continuity, compactness, and crystalline integrity, thereby forming a more efficient electron transport channel and reducing the interfacial contact resistance. When the deposition time increased to 50 min, the resistivity rebounded. An excessively long sputtering time may lead to the overgrowth of the copper film, which can easily form a rough and uneven surface morphology. The increase in surface roughness significantly enhances electron scattering during transmission and hinders directional migration, resulting in increased resistivity. Fig. 3(b-d) compare the surface SEM morphologies of the copper films under different sputtering times. It was verified that at a deposition time of 10 min, the deposited copper film was thin and structurally loose, resulting in an incomplete conductive path and a high resistivity. When the deposition time was increased to 30 min, the Cu film exhibited uniform, continuous, and dense morphology. When the deposition time reached 50 min, cluster-like amorphous copper particles and excessive growth protrusions appeared on the surface, destroying the uniformity and compactness of the film and leading to an increased resistivity. Based on electrical testing and morphological analysis, 30 min was determined to be the optimal sputtering time for achieving the best conductivity in this deposition system.

Fig. 3(e) shows a cross-sectional SEM image of the copper nanofilm deposited on the substrate via magnetron sputtering for 30 min. The results show that we successfully prepared a dense copper film with a uniform thickness of approximately 513 nm. The cross-sectional image clearly shows that a continuous and compact interface was formed between the copper film and the underlying substrate, without obvious voids or delamination, exhibiting a layered composite structure with strong bonding and an orderly architecture. The deposition of the copper film not only provides effective physical

protection for the underlying graphene but also significantly improves the electrical contact and mechanical coupling between graphene and metallic copper by forming strong interfacial bonding[30].

In this study, the residual stress state and crystallographic orientation of magnetron-sputtered copper nanofilms were systematically analyzed using grazing incidence X-ray diffraction (GIXRD). The corresponding GIXRD patterns are shown in Fig. 4(a). Compared to the diffraction signal from the copper substrate, the diffraction peak intensity of the (111) crystal plane of the deposited copper film was significantly enhanced, indicating that the copper nanofilm exhibited a strong (111) texture on the graphene/copper substrate. Good lattice matching was observed between the hexagonal lattice of graphene and the (111) plane of Cu. The (111) preferred orientation of the copper film greatly optimizes interfacial crystallographic matching, effectively reducing interfacial defects and stress concentration caused by lattice mismatch[31]. This optimized interface matching, combined with the good crystallinity of the copper film itself, further enhances the overall structural uniformity and regularity of the composite.



**Fig. 4** (a) GIXRD measurement of substrate copper and NFCu/Gr/Cu;(b) Raman spectroscopy of Gr/Cu and Gr-1/Cu;(c) the electrical conductivities of composites;(d) comparative tensile test curves, the insert figure shows the detailed dimension of the sample for tensile test.

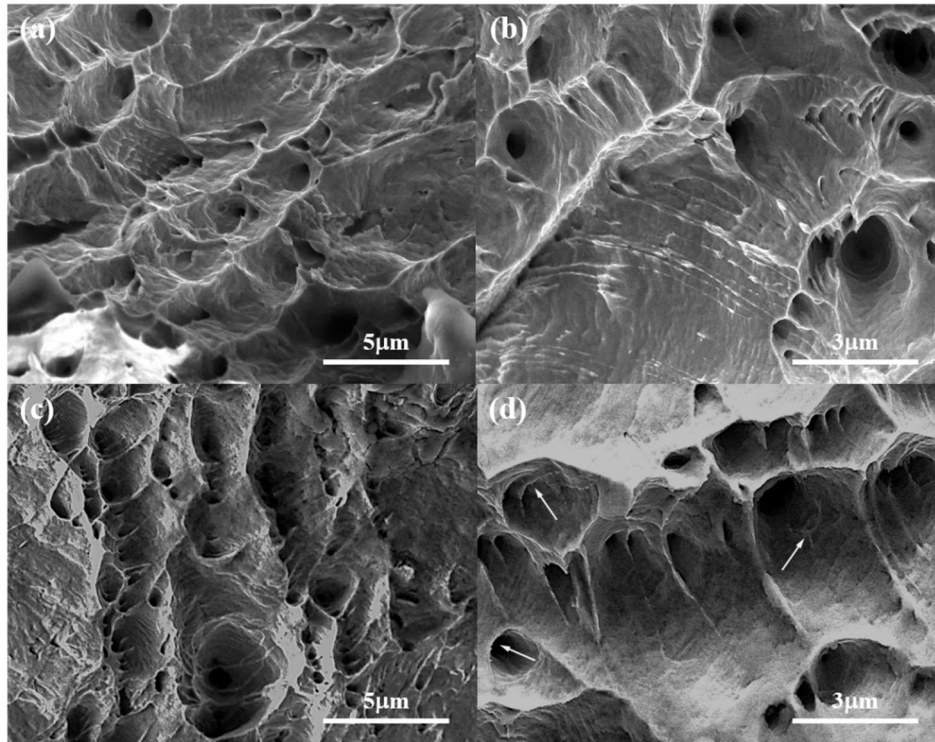
To explore the potential influence of the sputter-deposited copper nanofilm on the quality of the underlying graphene, the copper nanofilm on the surface of the prepared NFCu/Gr/Cu sample was etched to obtain a Gr-1 sample (Gr-1 represents graphene after removing the copper nanofilm with a copper etchant). A comparison of the Raman spectra is presented in Fig. 4(b). For the original Gr/Cu sample without a deposited Cu film, the  $I_D/I_G$  ratio, indicative of structural disorder, was 0.09, indicating that the graphene grown in situ by the CVD process had a high crystalline quality. After

depositing the copper film by magnetron sputtering and its subsequent removal by etching, the  $I_D/I_G$  ratio of the obtained graphene sample increased to 0.21. An increase in this ratio typically corresponds to an increase in defect density within the graphene lattice. This result confirms that the magnetron sputtering process causes a certain degree of microscopic damage to the underlying graphene. This damage is mainly due to bombardment by the high-energy backscattered argon atoms generated during sputtering. During the sputtering process, some argon ions gain a kinetic energy exceeding 100 eV after being reflected by the target. When they strike the graphene surface, they are sufficiently energetic to break  $sp^2$  carbon bonds and introduce point defects, vacancies, or lattice distortions[25]. Although Raman spectroscopy confirmed an increase in the defects, the resistivity test results showed that this damage did not severely deteriorate the overall conductivity of the composite. This may be due to the relatively dispersed spatial distribution of the damage and the extremely high intrinsic carrier mobility of graphene, which is not highly sensitive to a certain concentration of point defects.

### 2.3 Microstructure, Mechanical and Electrical Properties of Graphene/Copper Composites

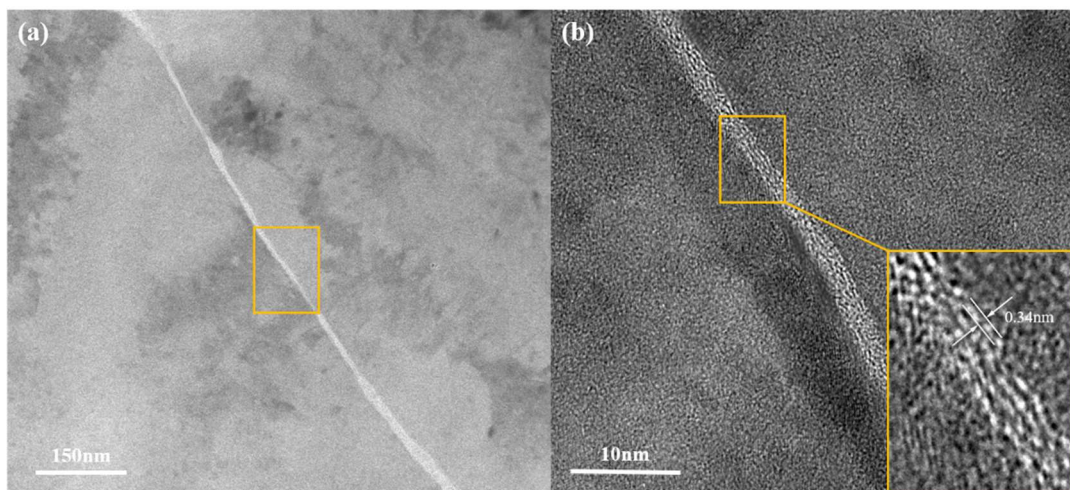
In this study, graphene was prepared by CVD, followed by magnetron sputtering of a copper film, and Cu/Gr composites were successfully fabricated via hot-press sintering. We tested pure copper, graphene-only reinforced Gr/Cu composites, and Gr/Cu composites that were further enhanced using magnetron-sputtered copper films. A comparison of the conductivities is shown in Fig. 4(c). The test results show that the conductivity of pure copper foil after the same hot-press sintering process is  $57.31 \times 10^6$  S/m, which is very close to the reference value of  $57.50 \times 10^6$  S/m for the International Annealed Copper Standard (IACS), confirming that the sintering process used in this study effectively maintains the high conductivity of the copper matrix. For Gr/Cu composites containing only CVD graphene, the electrical conductivity increases to  $58.13 \times 10^6$  S/m, equivalent to 101.1% IACS, showing a slight enhancement. This improvement can be attributed to the excellent carrier mobility of graphene and its grain-boundary modification effect on the copper matrix. The conductivity of the Gr/Cu composites with the sputtered copper film was further increased to  $60.53 \times 10^6$  S/m, equivalent to 105.27% IACS. Compared to the pure copper reference sample, the conductivity increased by approximately 5.3%. The results indicate that the introduction of the magnetron-sputtered copper film had a positive strengthening effect on the conductivity of the composite. This is because the dense copper film deposited by sputtering not only protects the graphene structure but also constructs a strong, well-matched bonding interface between graphene and the copper matrix. This interface greatly optimizes the cross-interface electron transmission between the graphene and metal phases, reduces the contact resistance, and thus synergizes with the intrinsically high conductivity of the graphene[23,32].

Mechanical properties are important considerations for the practical application of composites. The stress-strain curve in Fig. 4(d) shows that the ultimate tensile strength and elongation of the Gr/Cu composites and magnetron-sputtered copper film-reinforced Gr/Cu composites were significantly improved compared with those of the unreinforced copper foil. This is attributed to graphene, as a strengthening phase, effectively bearing and transmitting loads during stretching, thereby delaying fracture and improving material strength and ductility. Among them, the magnetron-sputtering-enhanced Gr/Cu composite exhibited the best performance, with its ultimate tensile strength further increasing to 240 MPa. This is mainly attributed to the strong interfacial coupling formed by the sputter-deposited dense copper film between graphene and the copper substrate. The copper film not only acts as an additional load-bearing phase, but its high-energy deposition characteristics also significantly enhance the interfacial adhesion between graphene and the copper matrix. This strong interface ensures that tensile stress can be transferred more efficiently from the plastic copper matrix to high-strength graphene, thereby fully utilizing the enhancement potential of graphene and realizing the synergistic strengthening of the composite[33].



**Fig. 5** SEM morphology of the fracture surface (a) and (b) pure Cu;(c) and (d)NFCu/Gr/Cu.

Fig. 5 compares the tensile fracture morphology of the magnetron-sputtered copper-film-reinforced Cu/Gr composites with that of the hot-pressed pure copper. As shown in Fig. 5(a) and (b), the fracture surface of pure copper is evenly distributed with large and deep dimples, resulting from sufficient plastic deformation and exhibiting typical ductile fracture characteristics. In contrast, the microstructures of the composite fractures in Figs. 5(c) and (d) are significantly different: pull-out and fracture traces of lamellar graphene can be clearly observed inside the dimples. This phenomenon indicates that good interfacial bonding is formed between graphene and the copper matrix, enabling graphene to be effectively loaded and fractured during tensile processing rather than simply debonded from the matrix. During tensile fracture, graphene can withstand a certain load and share part of the load on the copper matrix, thereby delaying composite fracture. It can also be inferred that the tensile fracture of the composite was primarily an caused by matrix tearing and graphene fracture[34].



**Fig. 6**(a) TEM image of the composite;(b) HRTEM image of the graphene in Cu matrix corresponding to the region enclosed by yellow solid rectangle of (a).

Fig. 6 shows transmission electron microscopy images of the copper/graphene composite. As shown in Fig. 6(a), the graphene film was evenly distributed between the copper layers, forming a continuous, regular sandwich structure that provided an ideal geometric configuration for electron transport and load transfer. The high-resolution TEM image in Fig. 6(b) further confirms that the embedded phase is graphene by measuring an interplanar spacing of 0.34 nm. More importantly, no common defects, such as amorphous impurities, oxides, or micropores, were observed in the interface region between the graphene and copper matrix, indicating the formation of a clean and tight interfacial bond. This complete interface structure can effectively reduce electron scattering and promote stress transfer, thus explaining the mechanism by which the composite achieves simultaneous synergistic improvements in the mechanical strength and conductivity[22].

## 2.4 Discussion

In this study, high-quality single-layer graphene was grown in-situ on a copper substrate via CVD, and copper nanofilms were deposited via magnetron sputtering to successfully construct an NFCu/Gr/Cu composite sandwich structure. As a low-temperature physical vapor deposition process, magnetron sputtering can maximally preserve the intrinsic quality of the graphene. The high-energy particle flux generated during sputtering facilitated the formation of strong bonds and the close contact between the copper nanofilm and graphene. This effect not only promotes interfacial bonding but also effectively alleviates the residual thermal mismatch stress.

In the subsequent sintering process, the sputter-deposited copper nanofilm acts not only as a functional layer to construct electron transport channels but also as a protective layer to isolate high-temperature shock and enhance interfacial bonding. This structural design and process combination ensures a low-contact resistance and high-stability interface connection between copper and graphene, thereby laying a microstructural foundation for the excellent conductivity of the composite.

## 3. Conclusion

In summary, this study successfully fabricated copper-based composites with synergistic enhancement in both electrical and mechanical properties through an integrated process of CVD, magnetron sputtering, and hot-press sintering. The composite achieved an electrical conductivity of 105.27% IACS and tensile strength of 240.32 MPa, thereby presenting a novel strategy for developing high-performance graphene/copper composites.

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