

RESEARCH ARTICLE

Green and Scalable Production of High-Quality Graphene by Solid Lithiation and Exfoliation

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ABSTRACT

High-quality graphene prepared by eco-friendly green methods is critical for maximizing the performance of its applications. Here, we use solid lithiation and exfoliation to achieve the green and scalable production of high-quality graphene, avoiding the use of strong oxidants. This intercalation-exfoliation process yields high-quality few-layered graphene nanosheets with an average thickness of 1.1 nm and high crystallinity. The produced graphene nanosheets have a low defect density ($I_D/I_G \approx 0.11$) and a high C/O ratio (~ 24.6), enabling large-scale dispersion and production with a yield of 26.7%. Furthermore, our graphene nanosheets show good thermal stability, remaining stable without significant weight loss up to 550°C and almost completely decomposing around 700°C in thermogravimetric analysis. In addition, the as-exfoliated graphene nanosheets were used as a conductive substrate for an oxygen evolution reaction catalyst. Cobalt-loaded graphene exhibits excellent catalytic activity and long-term stability, with an overpotential of 0.34 V at 10 mA cm⁻² and a Tafel slope of 99 mV dec⁻¹. Our work thus provides an efficient, green route to high-quality graphene, unlocking its potential for next-generation applications in electronics, energy storage, and composite materials.

1 | Introduction

Graphene has a perfect 2D crystal structure composed of sp^2 -hybridized carbon atoms, which exhibits exceptional electrical [1–3], mechanical [4–6], and thermal properties [7–9]. Graphene powder, films, or composite have practical applications in electronic devices [10, 11], batteries [12, 13], supercapacitors [14, 15], and thermally conductive materials [16–18]. The performance of graphene heavily depends on its quality, so it is important to prepare high-quality graphene with low defect density and high crystallinity. However, producing large quantities of high-quality graphene while avoiding the use of toxic chemical reagents remains challenging.

The graphene preparation methods can be categorized into two types: bottom-up and top-down approaches [19–21]. The bottom-up method can produce high-quality graphene, but its yield is low, rendering it difficult to scale up for graphene powder synthesis [22, 23]. Large-scale production of graphene is mainly achieved via top-down methods, including chemical reduction-oxidation [24], liquid-phase exfoliation [25, 26], and intercalation exfoliation [27]. The traditional Hummer's method involves oxidizing graphite with a strong oxidizing agents (e.g., H_2SO_4 , $KMnO_4$), wherein oxygen-containing groups such as $-C=O$, $-C-OH$, and $-COOH$ are inserted into the interlayer of graphite, increasing their spacing and resulting in graphene oxide nanosheets [24]. This method offers an impressive yield

and has been the dominant method for industrially preparing graphene oxide nanosheet solution [28]. Unfortunately, the Hummer's method uses strong acids, leading to environmental pollution. Additionally, oxygen-containing groups on graphene oxide nanosheets introduce numerous defects, severely impairing their properties-even after reduction to form reduced graphene oxide nanosheets [20, 28, 29].

Ion intercalation-assisted exfoliation method synthesizes graphite intercalation compounds (GICs) by inserting guest ions into the carbon layers, thereby increasing the interlayer spacing and weakening the van der Waals forces between adjacent layers. GICs react with water to produce hydrogen or spontaneously generate gas between the layers, which further promotes the exfoliation of few-layer nanosheets. For example, lithium (Li) ions are intercalated into graphite layers via an electrochemical method to form a lithium intercalated GICs (Li-GICs), enabling the controlled and efficient exfoliation of graphene [30]. However, the electrode preparation process is cumbersome, making it unsuitable for the exfoliation of graphite powder to achieve large-scale production of graphene [31, 32]. In addition, Li based chemical exfoliation has been reported using Li/liquid ammonia and biphenyl Li as intercalation sources [33, 34]. However, Li intercalation typically requires flammable reducing reagents and long reaction times, necessitating special precautions in laboratory operations. Recently, a new rapid Li intercalation method was reported using LiBH_4 as the Li source, which produced 108 g of transition metal telluride nanosheets in just 10 min via solid lithiation and exfoliation [35]. This method is simple, scalable, and green, yet it has not been previously applied to graphene synthesis.

Herein, high-quality graphene nanosheets were prepared greenly and efficiently using the solid lithiation and exfoliation method, without using any toxic reagents or strong oxidants. Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) characterizations revealed that the exfoliated graphene exhibits very high quality and excellent structural integrity ($I_D/I_G \approx 0.11$) as well as an extremely high C/O ratio (~ 24.6). The graphene nanosheets have a layer number of approximately 2–8, with a yield of up to 26.7%. The graphene prepared by this method also has potential applications in electrocatalysis. Co-loaded graphene demonstrates oxygen evolution reaction (OER) performance in 1 M KOH solution, showing long-term stability and an overpotential of 0.34 V at 10 mA cm^{-2} . Besides, our graphene shows significant weight loss only around 550°C, demonstrating excellent thermal stability.

2 | Results and Discussion

Figure 1a illustrates the schematic of solid lithiation and exfoliation of graphene. Lithium borohydride (LiBH_4), a strong reducing agent, was employed as the lithium source in this work. It was homogeneously mixed with graphite powder, followed by heating under an inert atmosphere to complete the lithiation process. Subsequently, the lithiated graphite (Li_xGr) powder was dispersed in distilled water under ultrasonication. During this process, the intercalated Li^+ ions react with water to generate hydrogen gas (H_2); the released gas exerts an expansion force on the graphite interlayers, thus realizing the exfoliation of graphene

nanosheets. Notably, in the absence of LiBH_4 , ultrasonication for 30 min produced only bright white, unexfoliated graphite aggregates (Figure S1). In contrast, the solid lithiation and exfoliation method yielded graphene nanosheets deposited on a silicon substrate with a distinct purple hue, indicative of ultrathin nanosheets having lateral dimensions of approximately 1–2 μm . This finding confirms the critical role of LiBH_4 in enabling efficient graphene preparation. The as-exfoliated graphene was subsequently purified via thorough washing, vacuum-dried, and weighed, giving a calculated yield of 26.7%. It is worth noting that the Li intercalation process is accompanied by electron injection, resulting in the formation of negatively charged graphene nanosheets [36–38]. Scanning electron microscopy (SEM) was utilized to characterize the microtopography and overall morphology of the pristine graphite powder (Figure 1b). Compared with the well-stacked lamellar structure of pristine graphite, the exfoliated graphene nanosheets display a distinct flexible, wrinkled, and translucent morphology, with lateral sizes in the range of 1–2 μm (Figure 1c; Figure S2). Figure 1d shows a photo of a batch of graphene dispersions prepared by solid lithiation and exfoliation, demonstrating their potential for scalable production.

To determine the thickness of the obtained graphene, we performed further atomic force microscopy (AFM) measurements on silicon wafers substrates (Figure 2a–d; Figure S3a) and conducted statistical analysis (Figure S3b). The thickness of graphene was measured to be as low as 0.7 nm. Approximately 50 different samples were randomly observed using AFM, and the thickness distribution of graphene exfoliated by solid lithiation and exfoliation was statistically analyzed, showing an average thickness of about 1.1 nm, equivalent to 3–4 layers. High-resolution Transmission electron microscopy (HRTEM) characterization of the graphene nanosheet edges reveals a well-defined layered structure, which can be distinguished by dark lines at the edges. The typical layer count of graphene nanosheets is 2–8 layers, with the majority being 4 layers (Figure 2e–h; Figure S4), which agrees with the AFM results. TEM images were employed to reveal the crystalline characteristics of the graphene nanosheets, which further confirm their ultra-thin and high crystallinity nature (Figure 2i). The HRTEM image of the graphene nanosheets displays the 2D honeycomb lattice structure of carbon atoms, with a lattice spacing of 0.21 nm, which is consistent with the (100) crystal plane of graphene (Figure 2j,k). Additionally, the excellent crystallinity of the graphene nanosheets was verified by selected area electron diffraction (SAED, Figure 2l); the typical hexagonal symmetric diffraction pattern confirms the high crystallinity of the exfoliated graphene [39].

To further assess the structural quality of graphene, we conducted X-ray diffraction (XRD) and Raman spectroscopy measurements. It is well-documented that LiBH_4 undergoes thermal decomposition at 400°C, producing Li, H_2 , and B_2H_6 according to Equation (1) [40]. When LiBH_4 : Graphite = 0.5, two distinct diffraction peaks centered at 24.1° and 25° were detected, which can be unambiguously assigned to the characteristic reflections of LiC_6 and LiC_{12} , respectively (Figure S5). In contrast, the Li_xGr sample with a 1:1 LiBH_4 -to-graphite ratio exhibits a prominent peak at 24.1°, corresponding to the (001) plane of LiC_6 (Figure 3a) [41, 42]. This observation provides direct evidence of interlayer spacing expansion induced by lithium ion intercalation into the graphite lattice. Additionally, the diffraction peaks located at

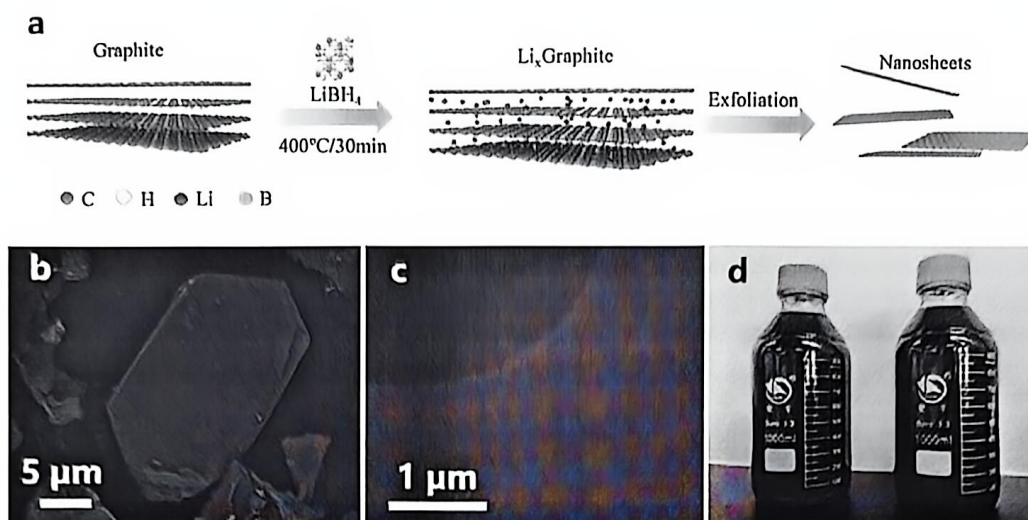


FIGURE 1 | Preparation and morphology characterization of graphene nanosheets. (a) Schematic of the solid lithiation and exfoliation process for the preparation of graphene nanosheets. (b) SEM image of the original graphite powder. (c) SEM image of exfoliated graphene nanosheet. (d) Photograph of two 1 L bottles of graphene dispersion.

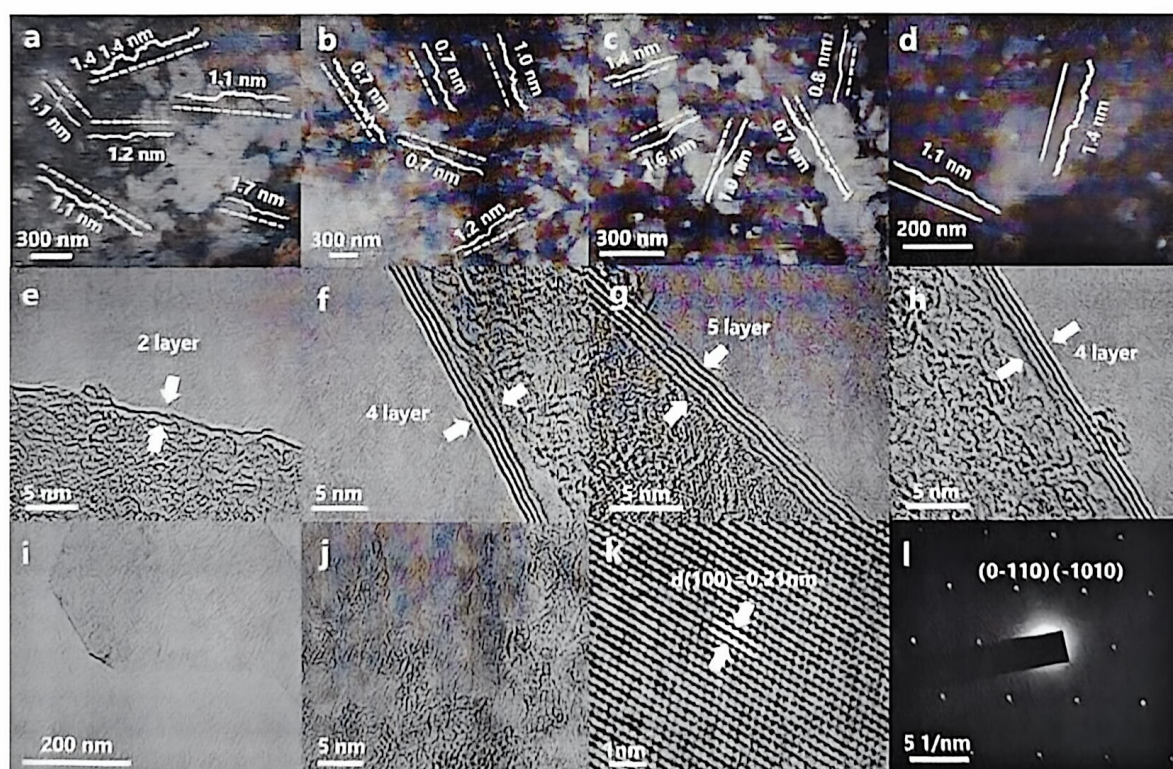


FIGURE 2 | Thickness analysis of the exfoliated graphene nanosheets. (a-d) AFM images of graphene nanosheets (insets are the height profiles of nanosheets). (e-h) HRTEM images of the edge of graphene nanosheets. (i) Low-magnification TEM image of graphene nanosheet. (j,k) HRTEM images of graphene nanosheet. (l) SAED profile of graphene nanosheet.

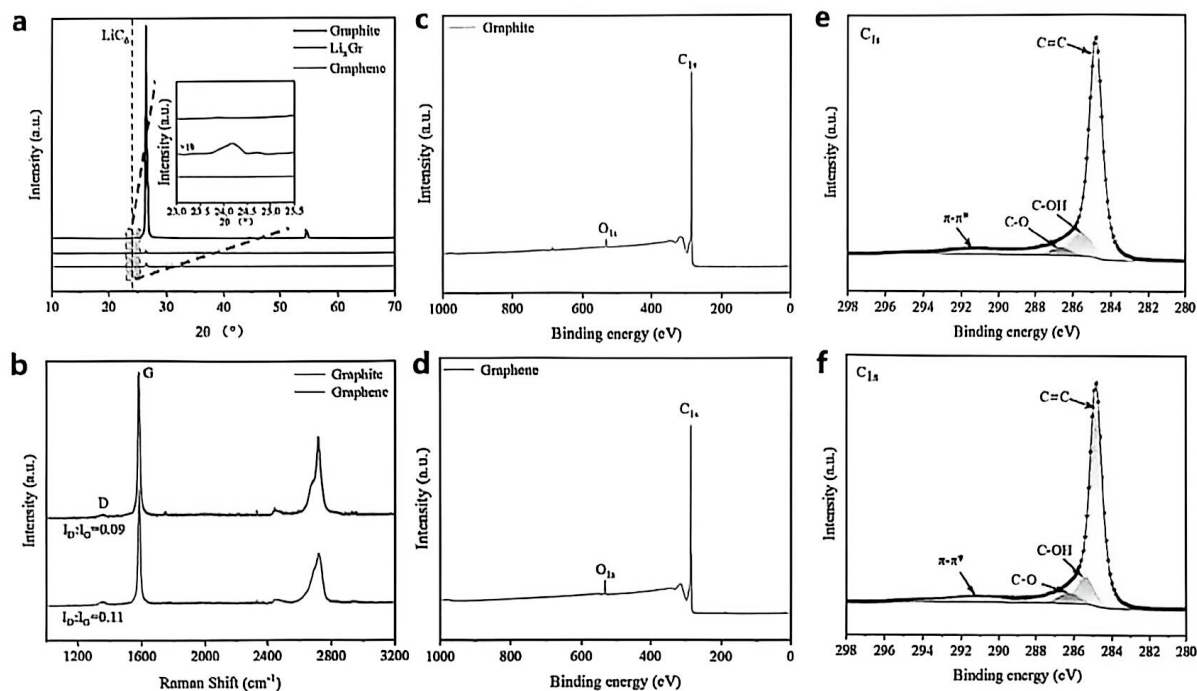
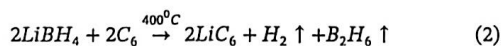
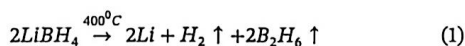


FIGURE 3 | Structural characterizations of the exfoliated graphene nanosheets. (a) XRD patterns of raw graphite, Li_xGr, and graphene nanosheets. (b) Raman spectra of raw graphite and graphene nanosheets. (c,d) XPS spectra of (c) raw graphite and (d) graphene nanosheets. (e,f) C1s XPS spectra of (e) graphite and (f) graphene nanosheets.

44.5° and 54.6° are attributable to the (101) and (004) lattice planes of pristine graphite [41]. On the basis of these results, the lithium intercalation reaction into graphite can be described by Equation (2).



In contrast, the intensity of this peak is significantly reduced in graphene, suggesting high efficiency of our solid lithiation exfoliation method. At the same time, the (002) peak position of graphene remained unchanged after exfoliation, indicating that the layer spacing and structural integrity were maintained [34]. As shown in Figure 3b, the characteristic peaks centered at 1352, 1582, and 2718 cm⁻¹ correspond to the defect-induced D band, the G band, and the 2D band, respectively. Specifically, the G band and 2D band are associated with the in-plane vibrational modes of sp²-hybridized carbon atoms and second-order zone-boundary phonons, respectively; while the D band is excited by first-order zone-boundary phonons, which originate from defects or edge effects in graphene [43–46]. The intensity ratio of the D to G bands (*I_D/I_G*) is an important metric for the defect density of carbon materials. The *I_D/I_G* value of the prepared graphene is 0.11, which is close to the original graphite (*I_D/I_G* = 0.09), indicating the low defect density of exfoliated graphene. Furthermore, a comparative analysis between this work and previously reported studies (Table S1) confirms the high quality of the graphene prepared in this work.

Figure 3c,d shows the XPS survey of the original graphite powder and graphene [47]. XPS results show that the exfoliated graphene has an oxygen content of 3.91% and a high C/O molar ratio (C/O ratio of 24.6%), indicating that few functional groups were introduced during the solid lithiation exfoliation process. High-resolution C1s XPS peaks (Figure 3e,f) were deconvoluted into three peaks of 284.8, 285.3, and 286.2 eV, corresponding to the C=C, C–OH, and C–O bonds, respectively [48, 49]. The oscillation of the peak at 291.08 eV related to the π–π* transition, which is clearly visible after exfoliation, indicates that the conjugated aromatic structure is preserved, which is characteristic of carbon materials with a high concentration of sp² carbon [34, 49]. In addition, Figure S6 presents the surface morphologies of graphene solutions with and without polyvinyl pyrrolidone (PVP) addition. It can be observed that the graphene solution without PVP exhibits a reflective film on its surface, whereas no such reflective film is detected in the PVP-containing graphene solution. Collectively, these observations further confirm the high quality of the as-prepared graphene nanosheets.

Graphene nanosheets with 2–8 layers were successfully prepared using a solid lithiation and exfoliation method. To achieve the optimal yield, we further investigated the concentration and yield of graphene nanosheet dispersions obtained by varying the dosage of LiBH₄, as well as the heating time and temperature (Figure 4a–c). The results demonstrate that the maximum yield of graphene nanosheets (26.7%) is achieved when LiBH₄ and graphite are mixed at a molar ratio of 1:1, heated at 400°C for 30 min, followed by centrifugation of the resulting graphene

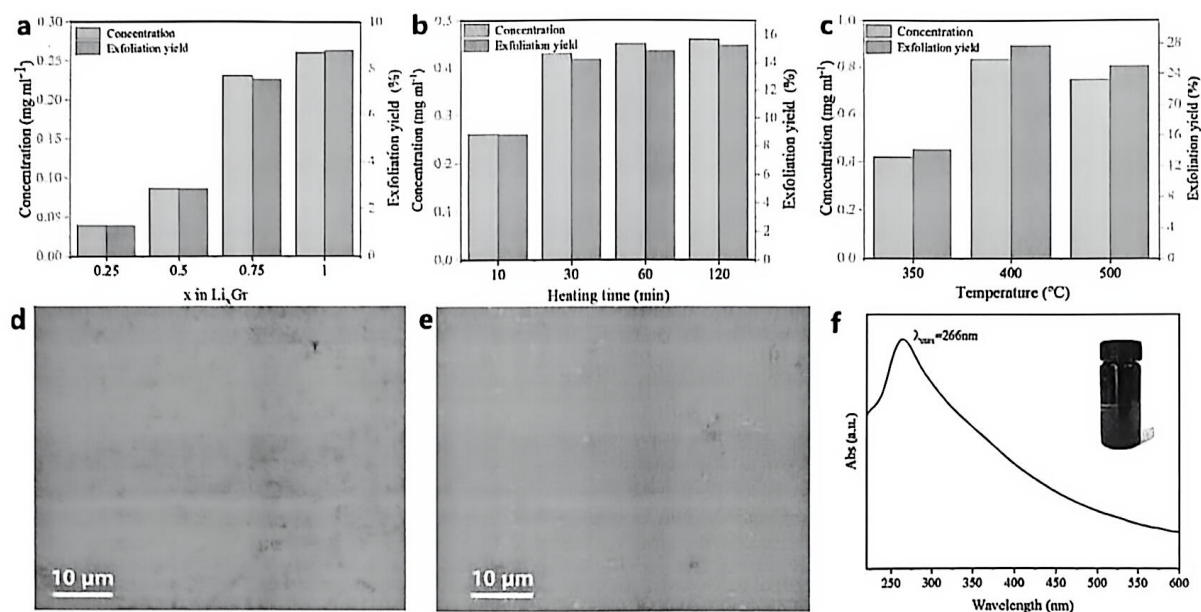


FIGURE 4 | Characterization of the influence relationship between the degree of lithiation of bulk Graphite and graphene yield in solid lithiation and exfoliation. (a–c) Graph of the concentration and yield of graphene dispersions at (a) different molar ratios of LiBH₄ and bulk graphite powder, (b) different lithiation heating time, and (c) different lithiation temperature. (d,e) Laser scanning confocal microscopy (LSCM) images of graphene prepared (d) with and (e) without PVP in. (f) UV-vis spectrum of prepared graphene (Inset: Digital photograph of a graphene dispersion with a pronounced Tyndall effect).

dispersion at 1000 rpm for 10 min to remove unexfoliated graphite particles.

During the exfoliation process, the addition of PVP to the graphene dispersion significantly enhances its dispersion stability, as evidenced by the increased presence of more purple graphene nanosheets (Figure 4d,e; Figure S7). This improvement is attributed to the surface energy matching between PVP as a surfactant and graphene, which efficiently facilitates nanosheet dispersion [50]. Notably, PVP can be readily removed through washing. TEM characterization of the graphene before and after washing (Figures S8 and S9) confirms that PVP residues, originally clustered in molecular chains on the nanosheets, are completely removed, yielding very clean graphene nanosheets. Figure S10 shows the lateral dimension distribution of the graphene nanosheet, with an average transverse size of $0.85 \pm 0.5 \mu\text{m}$. The UV-vis spectrum of the graphene dispersion after centrifugal washing in absolute ethanol is shown in Figure 4f, where the graphene dispersion exhibits an absorption peak at 266 nm, corresponding to the conjugated π -plasma electronic transition. In addition, this peak is also relatively symmetrical and sharp, indicating that graphene has a relatively complete sp^2 carbon network structure [47, 51]. The graphene is dispersed more uniformly and stably in the dispersion liquid, with no significant agglomeration observed, and the colloidal solution of graphene nanosheets displays a distinct Tyndall effect (Figure 4f inset).

To demonstrate the potential application of the prepared graphene in energy conversion, Co-loaded graphene (Co-Graphene) was fabricated by heating $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and Li_4Gr in NMP for 24 h (see Experimental Section). Its OER performance

was evaluated under alkaline conditions. TEM-EDS of Co-Graphene shows the presence of C and Co elements (Figure S11a–c), which are evenly distributed, indicating that Co has been successfully loaded onto graphene surfaces. The spacing of the lattice fringes measured from the HRTEM image is 0.23 nm, which is consistent with the (002) plane of $\text{Co}(\text{OH})_2$, and the SAED pattern of the sample (Figure S11d) indicates its polycrystalline nature [52, 53]. The content of Co is relatively lower than that of C, which also explains why only graphene peaks appeared in XRD results (Figure S12). The XPS spectrum of Co 2p (Figure S13) shows two characteristic peaks with binding energies of 781.78 and 797.68 eV, corresponding to Co 2p_{3/2} and Co 2p_{1/2} [53]. The position and distance between these two peaks ($\Delta = 15.9 \text{ eV}$) indicate that Co exists in the form of $\text{Co}(\text{OH})_2$ [52]. OER tests (1 M KOH) revealed Co-Graphene had the best activity (overpotential 0.34 V at 10 mA cm^{-2}), smallest Tafel slope (99 mV dec^{-1}), and lowest charge transfer resistance (Figure S14a–c). It maintained stability for 50 h at 100 mA cm^{-2} and 24 h at 10 mA cm^{-2} without decay. All of the above indicates that the material has excellent stability (Figure S14d). Besides, we compared the overpotentials and Tafel slopes of our prepared Co-Graphene and related catalysts at 10 mA cm^{-2} , highlighting the superior performance of our synthesized catalysts (Table S2).

Moreover, we further characterized the thermal stability of exfoliated graphene powder by thermogravimetric analysis (TGA, Figure S15a). No significant weight loss was observed from room temperature to 550°C , and complete decomposition occurred around 700°C , far outperforming graphene oxide [54, 55]. This confirms the high quality of the prepared graphene and its potential for thermal-related applications. The only initial weight

loss was attributed to water evaporation, with no weight loss from oxygen-containing group removal, indicating excellent thermal stability. The lyophilized graphene powder was black and fluffy (inset of Figure S15a), suggesting a low degree of oxidation. In addition, the vacuum-filtered graphene film exhibited good electrical conductivity (Figure S15b) with a resistance of only 4.5 Ω , which could successfully light a small bulb (Figure S15c,d). Further, the electrochemical impedance spectroscopy (EIS) semicircle of the graphene nanosheets in the high-frequency region is smaller, indicating lower resistance. Figure S16 shows the output characteristics (I_{ds} - V_{ds}) of graphene at a gate-source voltage (V_{gs}) of 0, with the drain-source current (I_{ds}) plotted against the drain-source voltage (V_{ds}). The inset shows a schematic of the field-effect transistor (FET) device with interdigitated electrodes. The linear I_{ds} - V_{ds} curve indicates good ohmic contact between the metal electrodes and graphene. The drain-source resistance (R_{ds}) is approximately 212 Ω , which is lower compared to conventional graphene FET on SiO_2 substrates, where R_{ds} is usually 100–50 k Ω [56–58]. The graphene we prepared exhibits a lower R_{ds} , demonstrating excellent conductivity.

3 | Conclusion

In summary, we have developed a solid lithiation and exfoliation method that can efficiently enable the scalable production of high-quality few-layer graphene nanosheets from raw graphite. This strategy not only addresses the problem of environmental pollution associated with the traditional Hummer's method, but also preserves the honeycomb lattice structure of the exfoliated graphene, possessing low defect density ($I_D/I_G \approx 0.11$), and a high carbon-to-oxygen ratio (~ 24.6). Using this method, 2–8 layer high-quality graphene was obtained with an average thickness of 1.1 nm. Moreover, the as-prepared graphene exhibited excellent OER performance and superior thermal stability, highlighting its tremendous application potential in catalysis, energy storage, thermal management, and other related fields.

4 | Experimental Section

4.1 | Materials

The materials used in this study were sourced as follows: graphite (99.99%, 200 to 325 mesh) was supplied by Guangzhou Metallurgy. PVP was purchased from Sigma-Aldrich Co., Ltd, while LiBH_4 (95%), $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (98%) were provided by Shanghai Aladdin Biochemical Technology Co., Ltd. N,N-dimethylformamide (DMF, 99.8%) and NMP (99.5%) were purchased from J&K Scientific Co., Ltd.

4.2 | Exfoliation of Graphene Nanosheets

In the graphene preparation process, LiBH_4 was prone to moisture absorption in ambient air, so all mixing operations were performed in an argon-filled glove box. Specifically, LiBH_4 and bulk graphite powder were uniformly mixed at a molar ratio of 1:1 (with 60 mg of graphite). The mixture was heated to 400°C for 30 min under vacuum to form Li_xGr . Subsequently, Li_xGr and PVP were added to deionized water to prepare a solution with a

PVP concentration of 1 mg/mL. The mixture was gently sonicated for 30 min to form a graphene colloidal suspension, where PVP acted as a surfactant to reduce the hydrophobicity of graphite and prevent the aggregation of graphene nanosheets. After exfoliation, the suspension was centrifuged at 1000 rpm for 10 min to remove unexfoliated graphite. The resulting graphene solution was further washed with deionized water, ethanol, and DMF, followed by centrifugation at 10 000 rpm. The obtained graphene was redispersed in ethanol for subsequent characterization.

4.3 | Synthesis of Co-Graphene

60 mg of graphene nanosheets were mixed with 110 mg of LiBH_4 . The mixture was directly heated at 400°C for 30 min in a chemical vapor deposition tube furnace to obtain Li_xGr . Subsequently, 120 mg of $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ was added to the as-prepared Li_xGr in 20 mL of anhydrous NMP solution. The mixture was magnetically stirred to form a homogeneous suspension, which was then transferred into a sealed autoclave and heated at 80°C for 48 h. After the hydrothermal reaction, the obtained product was thoroughly rinsed sequentially with four distinct solvents, namely NMP, isopropanol, ethanol, and deionized water, followed by vacuum drying for 12 h.

4.4 | Electrochemical Measurements

All electrochemical tests were performed at room temperature using a standard three-electrode system for the Corrtest potentiostat (Wuhan Corrtest Instrument Co., Ltd). Catalyst powder (5 mg) was added to a mixture of anhydrous ethanol (500 μL) and Nafion solution (25 μL , 5 wt.%), followed by sonication for 1 h to generate a homogeneous dispersion as the catalyst slurry. Subsequently, a drop of catalyst ink (200 μL) was drop-cast onto a carbon paper with an area of 1 $\times$ 1 cm^2 . A standard three-electrode electrolyzer with a 1.0 M KOH solution was used for all tests, with carbon paper, Hg/HgO electrodes, and graphite rods as working electrodes, reference electrodes, and counter electrodes, respectively. The LSV test has a scan rate of 5 mV s^{-1} . Convert all measured potentials to a reversible hydrogen electrode (RHE) scale using the following formula: $E(\text{RHE}) = E(\text{Hg}/\text{HgO}) + 0.059 \times 14 + 0.098 \text{ V}$. The EIS frequency range is from 100 kHz to 0.01 Hz.

4.5 | Materials Characterization

The morphology of graphite and graphene nanosheets was analyzed using SEM (S-4800, Hitachi). The crystalline phases of the graphite powder and nanosheets were identified through XRD (Ultima IV, Rigaku Corporation). The thickness of the graphene was characterized by AFM (SmartSPM 1000, AIST-NT). XPS results were obtained with Thermo Scientific K-Alpha. Raman spectroscopy was determined by LabRAM HR800. The lattice structure of graphene was obtained by the TEM (FEI Tecnai F20). TEM image of Co-Graphene was characterized by Tecnai F30. UV-vis spectroscopy of graphene was conducted using a Shimadzu UV-2700 spectrophotometer. TGA testing was performed using a high-temperature thermogravimetric and differential thermal analyzer (STA449F5, NETZSCH-Gerätebau

GmbH). Electrical measurements were carried out using a semiconductor analyzer (Agilent B1500A, Agilent Technologies).

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

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