

# Electrical Contacts to Graphene by Postgrowth Patterning of Cu Foil for the Low-Cost Scalable Production of Graphene-Based Flexible Electronics

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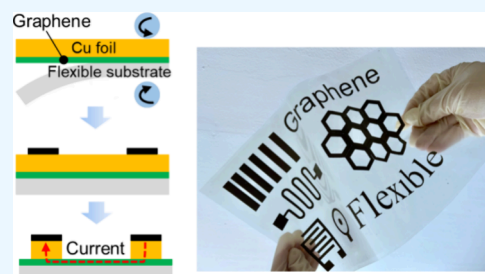
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**ABSTRACT:** Numerous studies have focused on graphene owing to its potential as a next-generation electronic material, considering its high conductivity, transparency, superior mechanical stiffness, and flexibility. However, cost-effective mass production of graphene-based electronics based on existing fabrication methods, such as graphene transfer and metal formation, remains a challenge. This study proposes a simple and efficient method for creating electrical contacts with graphene. The method involves patterning a Cu foil after graphene growth, enabling the low-cost scalable production of graphene-based flexible electronics. The fabricated graphene devices exhibited linear current–voltage characteristics, indicating good electrical contact between the postgrowth-patterned Cu electrodes and graphene. The proposed postgrowth patterning method allows for the fabrication of Cu-contacted graphene devices on large areas and various flexible substrates, including ultrathin and stretchable films ( $<10\ \mu\text{m}$ ). The feasibility of the proposed method for electronic devices was demonstrated by implementing gas and flexible force sensors. The proposed approach advances the field of graphene-based electronics and holds potential for practical applications in various electronic devices, paving the way for scalable, cost-effective, and flexible technology solutions.



## INTRODUCTION

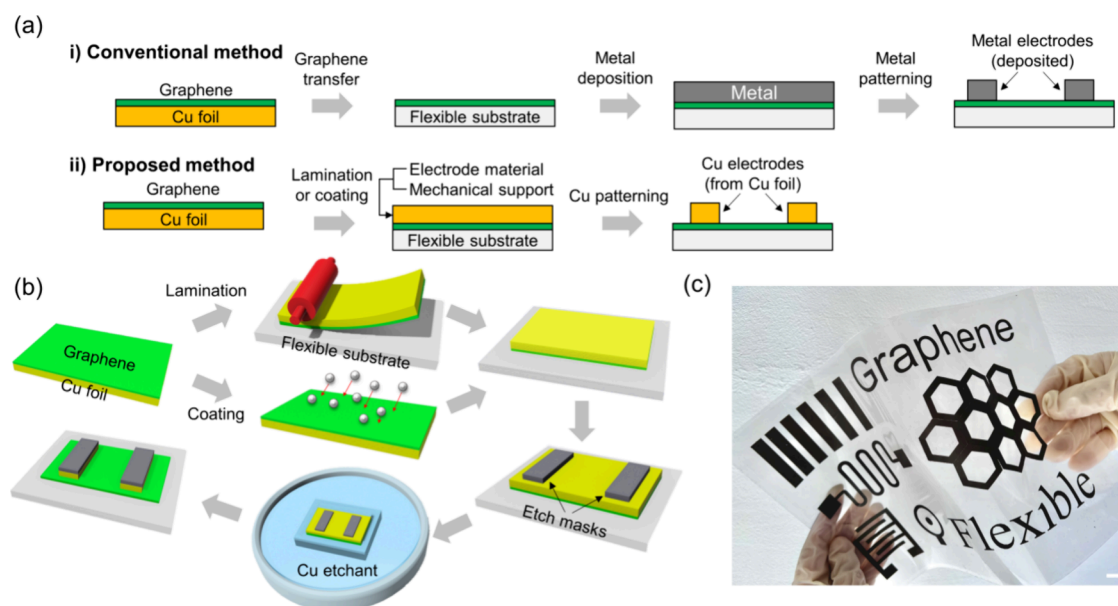
Graphene, a honeycomb-like sheet of carbon atoms, has gained significant research attention owing to its potential as a next-generation electronic material with diverse applications, considering its high conductivity, transparency, superior mechanical stiffness, and flexibility.<sup>1–4</sup> Since the discovery of graphene sheets through the mechanical exfoliation of graphite in 2004,<sup>1</sup> various preparation methods of graphene have been developed, such as liquid-phase exfoliation,<sup>5,6</sup> desorption of Si from SiC single-crystal surfaces,<sup>7,8</sup> and chemical vapor deposition (CVD).<sup>9–11</sup> Among these, the CVD method produces high-quality large-area graphene films on Cu foils, reaching sizes of centimeters or meters,<sup>12,13</sup> as a self-limited process facilitated by the low solubility of carbon in Cu. CVD enables scalable production of large-area monolayer graphene without adlayers (i.e., bilayer or multilayer regions),<sup>14,15</sup> a crucial requirement for industrial applications of graphene-based electronics. CVD-grown graphene on Cu foil is typically processed into graphene-based electronics through two fabrication steps: graphene transfer and electrode formation. Graphene film is then transferred to a target substrate, usually with an insulating layer. The wet transfer method with a carrier layer (i.e., a polymer) coated on CVD-grown graphene is commonly used owing to its advantage in maintaining the integrity of the graphene film.<sup>16–18</sup> The polymer/graphene stack is transferred to the target substrate after etching the

growth substrate (i.e., Cu foil) with a liquid etchant. The graphene-coated substrate is completed by removing the polymer. Details of the transfer methods are provided in review papers.<sup>19,20</sup> After the graphene transfer process, metal electrodes are formed on the graphene-coated substrate for electrical contacts with graphene by combining photolithography and metal deposition processes, such as evaporation and sputtering.<sup>21–23</sup> Various attempts at Cu recycling have been made for graphene transfer.<sup>24–27</sup> A mechanical peeling method called “dry transfer”<sup>24,25</sup> and electrochemical process called “bubbling transfer” using hydrogen bubble generated during water electrolysis process<sup>26,27</sup> delaminate the graphene from the Cu foil without etching, enabling Cu substrate reuse. Various studies have demonstrated the direct growth of graphene on target substrates, such as SiO<sub>2</sub>,<sup>28,29</sup> polyimide (PI),<sup>30,31</sup> and polyethylene terephthalate (PET).<sup>30,32</sup>

However, achieving low-cost, scalable fabrication of metal electrodes on flexible substrates for graphene-based electronics

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**Figure 1.** Electrical contacts to graphene using postgrowth-patterned Cu foil. (a) Comparison of electrical contacts between the conventional and proposed methods for fabricating graphene-based flexible electronics. In the conventional method, a Cu foil (i.e., the growth substrate of CVD-grown graphene) is completely etched (or delaminated) for graphene transfer, and metal is deposited on the graphene for electrical contacts. In the proposed approach, the Cu foil is used as an electrode to create an electrical contact with graphene by patterning the Cu foil into the desired electrode shape without additional metal deposition on the graphene. (b) Schematic of the fabrication process of electrical contacts with graphene using postgrowth-patterned Cu foil. (c) Graphene-based flexible device with postgrowth-patterned Cu electrodes fabricated on a 12.9-in. silicone/PET film. Scale bar: 1 cm. Photograph courtesy of “Lee Kyung Bae”. Copyright 2024. Reusing the image needs permission.

remains challenging. Metal deposition processes require the operation of large vacuum equipment and the consumption of metal sources, potentially increasing the cost of disposable graphene sensors, such as patches and tattoos.<sup>33–36</sup> In addition, ultrathin and stretchable substrates, prone to wrinkling or curling,<sup>37–39</sup> require additional processing steps, such as attaching the flexible substrate to a handling substrate and lift-off (or delamination) of the substrate after the completion of fabrication,<sup>40–42</sup> potentially affecting the production cost.

This study proposes the concept of creating electrical contacts with graphene by patterning a Cu foil after graphene growth for the low-cost scalable production of graphene-based flexible electronics. Rather than implementing additional metal deposition, Cu foil, a graphene growth substrate, was used as the metal electrode. A flexible substrate was covered with a graphene film synthesized on a Cu foil, and the Cu foil was patterned into individual electrodes through Cu etching with masking layers. Combining large (up to 12.9 in) graphene/Cu films and a “cut-and-paste” method of masking materials onto the Cu foil yielded a scalable array of graphene devices on a flexible substrate. The proposed approach leverages a copper substrate to facilitate a cost-effective and efficient method for patterning graphene without the necessity of additional metal deposition, thereby distinguishing it from existing research that primarily focuses on synthesis and transfer techniques.<sup>12</sup> The Cu-contacted graphene devices exhibited linear current–voltage ( $I$ – $V$ ) characteristics, indicating good electrical contact between the Cu foil and graphene. The contact resistance between the Cu-based contacts and graphene was determined using a test pattern at various distances. The proposed postgrowth patterning of Cu foil was applied to various flexible materials, including ultrathin stretchable films, such as silicone/PET, thermal release tape (TRT), polydimethylsilox-

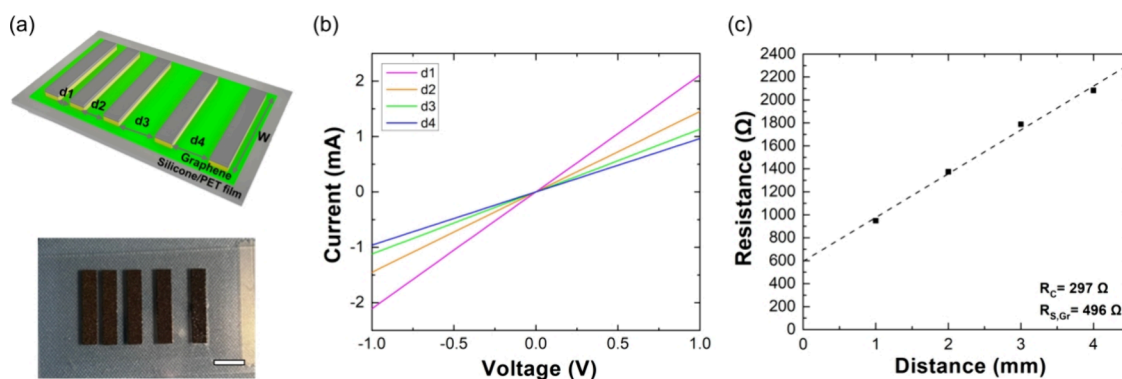
ane (PDMS) film, PI tape, parylene, and polyurethane (PU). Electrical contacts to graphene created through the postgrowth patterning of Cu foil were used to implement gas sensors for detecting  $\text{NH}_3$  and  $\text{NO}_2$  gases and flexible force sensors capable of responding to bending tests.

## RESULTS AND DISCUSSION

**Proposed Concept: Electrical Contacts with Graphene by Postgrowth Patterning of Cu Foil.** In conventional methods for fabricating graphene-based electronics (Figure 1a, left), CVD-grown graphene on a Cu foil is transferred to a target substrate by etching or delaminating the Cu foil. Subsequently, metal evaporation or sputtering is performed to form metal electrodes on the graphene. In the proposed approach (Figure 1a, right), the additional process of metal deposition is eliminated by utilizing the Cu foil (i.e., growth substrate) as electrical contacts or electrodes for graphene. The proposed method does not require metal sources and vacuum equipment used in metal deposition, reducing process costs and simplifying fabrication.

Gold is commonly used as an electrode but does not support in situ graphene growth through chemical vapor deposition (CVD), requiring instead complex transfer processes from other substrates.<sup>43</sup> Copper, by contrast, allows direct CVD-based graphene growth, simplifying fabrication and enhancing electrode stability and performance, making it highly suitable for scalable, cost-effective graphene-based applications.<sup>12,44,45</sup>

Figure 1b shows the fabrication process of graphene devices through the postgrowth patterning of the Cu foil. The top side of the graphene on the Cu foil was covered with a silicone/PET film, and the back side was etched through an  $\text{O}_2$  plasma process (100 W for 90 s). The flexible substrate used in this study is a silicone/PET composite, with a silicone layer added to improve PET’s thermal stability and insulation, making it



**Figure 2.** Electrical characteristics of postpatterned Cu contacts. (a) Schematic (top) and optical image (bottom) of the test pattern with various spacing distances for determining the contact and sheet resistances. A CVD-grown graphene on a silicone/PET film was electrically contacted with the postgrowth-patterned Cu electrodes at various distances. Scale bar: 1 cm. Photograph courtesy of “Lee Kyung Bae”. Copyright 2024. Reusing the image needs permission. (b) Current–voltage characteristics. (c) Resistance versus distance.

more suitable for applications involving temperature variations. Although not illustrated here, the top graphene layer on the Cu foil could be patterned to define the active area using an etching mask through  $O_2$  plasma etching. The thin film of the Cu foil was transformed into Cu electrodes through a conventional patterning process: placing etch masks and etching with a Cu etchant (0.3 M APS solution for 90 min). Cu-contacted graphene devices can be fabricated using different masking materials, categorized as nonconducting or conducting materials (Table S1). Conductive carbon tape was selected as the masking material owing to its suitability for economical and large-scale processes. In this study, conductive carbon tape was securely adhered to the copper foil, ensuring no infiltration of APS solution during the etching process. The strong adhesion of the tape effectively prevented solution seepage, maintaining precise etching boundaries, and ensured minimal contact resistance, allowing reliable electrical performance measurements. However, it is worth noting that the conventional photolithography process using a photoresist can be employed to achieve high-resolution patterning if required (Figure S1).

Figure 1c shows an example of the fabricated Cu-contacted graphene device. An array of graphene devices was fabricated on a flexible substrate (silicone/PET film) with an area of up to 12.9 in. This configuration demonstrates the feasibility of using Cu foil as a growth substrate and an electrode material, showcasing its potential for flexible electronics where cost efficiency and process simplicity are essential.

#### Electrical Characteristics of Cu Foil-Based Contacts.

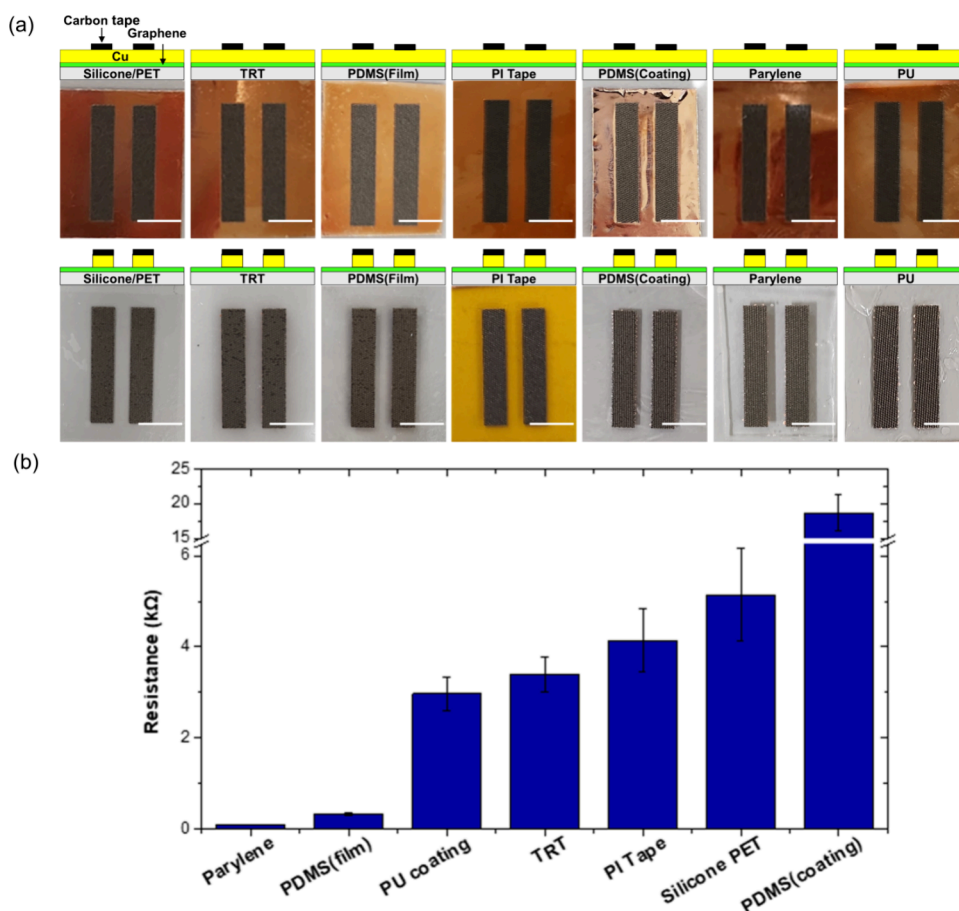
The electrical characteristics of the Cu-contacted graphene devices were evaluated by determining the contact resistance between Cu and graphene. A test pattern with various distances between the Cu electrodes was used (Figure 2a,b). This method, known as the transfer length method (TLM), is widely used for determining the contact resistance of graphene device.<sup>46,47</sup> The graphene was patterned via an  $O_2$  plasma etching process to define the active area and ensure accurate TLM measurements. The top graphene layer on the Cu foil was covered with a patterned carbon mask, and the Cu foil was subsequently etched using a Cu etchant. This patterning process is crucial to isolate individual graphene channels and minimize measurement errors associated with unpatterned sheets. The equation,  $R_{\text{tot}} = 2R_C + R_{S,Gr}(d/W)$ , was used, where  $R_C$ ,  $R_{S,Gr}$ ,  $d$ , and  $W$  denote the contact resistance between the Cu-based contact and graphene, graphene sheet

resistance, distance between the contacts, and width of the graphene channel, respectively. The fabricated graphene devices exhibited linear  $I$ – $V$  characteristics (Figure 2b), indicating good electrical contact between the Cu substrate and graphene. The distance between the Cu electrodes influence the resistance values (Figure 2c). From the linear fitting of the resistance ( $R$ ) as a function of distance ( $d$ ), the calculated contact resistance of Cu/graphene and sheet resistance of graphene were 297  $\Omega$  (386  $\Omega$  mm) and 496  $\Omega/\square$ , respectively. The contact resistance of the Cu/graphene device is similar to the values ( $\sim 100$   $\Omega$  mm) of the contact resistance between graphene and other metal contacts (Ti, Ti/Au, and Cr/Au).<sup>48</sup> The contact resistance of the proposed device is relatively high, which makes the proposed Cu/graphene contacts unsuitable for high-performance transistor applications. Acknowledging this limitation, the focus has shifted to the cost-effective fabrication of disposable sensors. The total resistance of graphene sensors is expressed as  $R_{\text{tot}} = 2R_C + R_{Gr}(d/W)$ . The graphene sensor should be designed with  $R_{Gr}(d/W)$  larger than  $R_C$ , effectively allowing for the measurement of a change in the graphene channel resistance when subjected to an external stimulus.

The graphene-Cu-graphene configuration not only effectively isolates the copper foil from environmental oxygen, thereby preventing oxidation, but also significantly enhances the electrical conductivity and durability of the contacts. This encapsulation reduces the contact resistance, critical for high-performance applications, as demonstrated by the improved interface quality in our SEM, Raman spectroscopy, and XPS analyses.<sup>49</sup> Furthermore, CVD-grown graphene provides robust protection for Cu surfaces against oxidation at elevated temperatures, ensuring long-term stability and reliability under operational conditions.<sup>50</sup> Additionally, thicker copper foil may improve oxidation resistance, potentially extending device lifetime by minimizing surface degradation. This phenomenon has been observed in previous studies where increased copper thickness led to enhanced stability against oxidation, especially in high-temperature or reactive environments.<sup>51</sup> Therefore, using thicker copper foil in our proposed device could further stabilize contact resistance over prolonged operational periods, supporting its application in disposable sensors and other flexible electronic devices.

**Postgrowth Patterning of Cu Foil Applied to Various Flexible Materials.** The proposed postgrowth patterning method can be applied to various flexible substrate materials.





**Figure 3.** Postpatterning of Cu foil on various flexible substrates. (a) Optical images of the graphene devices on different flexible substrates before (top) and after (bottom) postpatterning of the Cu foil. Scale bar: 1 cm. Photograph courtesy of “Lee Kyung Bae”. Copyright 2024. Reusing the image needs permission. (b) Resistance values for the Cu-contacted graphene devices with various flexible substrates.

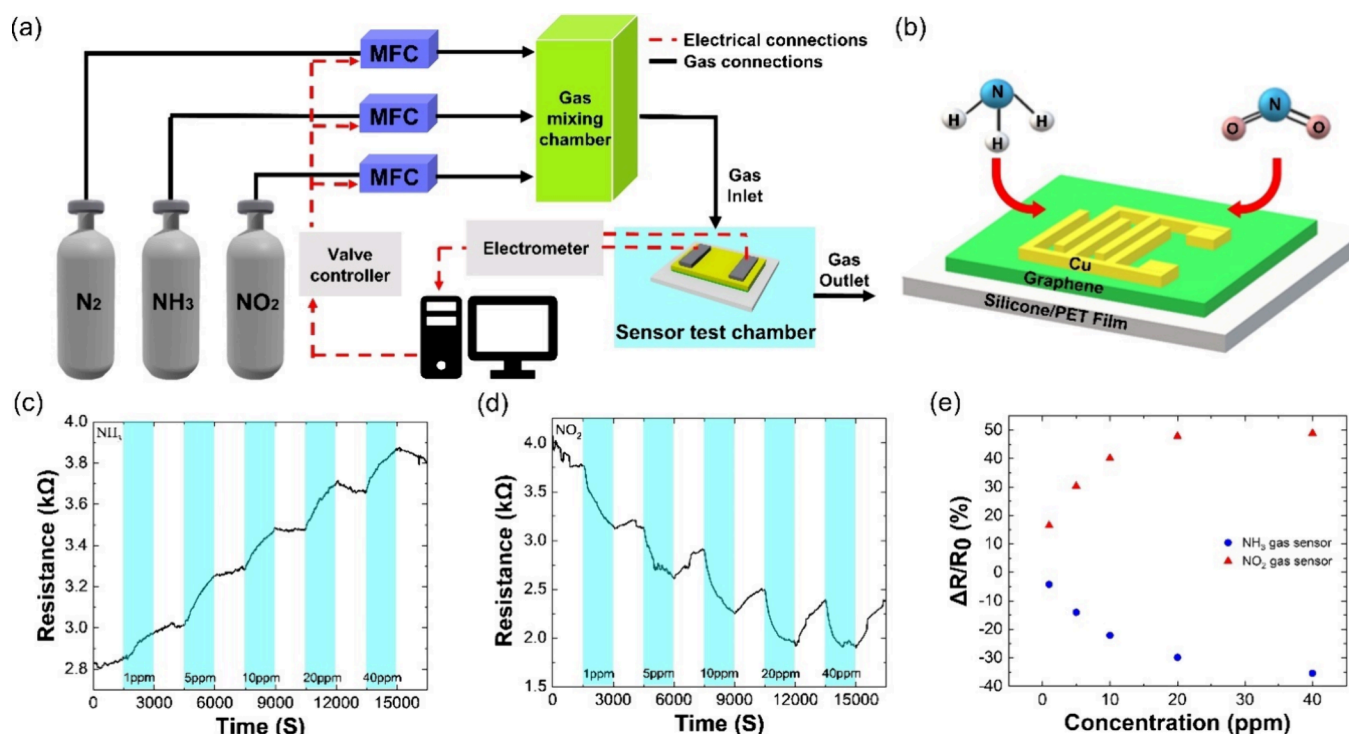
CVD-grown graphene on Cu foil was attached to flexible substrates using two different methods: (1) lamination of graphene/Cu foil with a flexible substrate and (2) coating of a precursor polymer onto graphene/Cu foil through spin-coating or CVD. The detailed processes for preparing the multilayer structures of the flexible substrate/graphene/Cu foil are described in the Supporting Information (Figure S2) or Methods. Figure 3a shows the postgrowth patterning of the Cu foil applied to laminated substrates, such as silicone/PET, TRT, and PDMS films. These flexible substrates have sufficient adhesion suitable for graphene attachment and adequate thickness ( $>100\ \mu\text{m}$ ) for mechanical handling during fabrication. The direct lamination of graphene/Cu foil onto these flexible substrates is compatible with roll-to-roll processing,<sup>52,53</sup> facilitating industrially scalable production of graphene-based electronics. The postgrowth patterning method was also applied to graphene/Cu foil coated with polymer thin films, such as parylene and PU (Figure 3a). These ultrathin and stretchable films are too thin ( $<10\ \mu\text{m}$ ) to handle mechanically, requiring attachment of the film on a handling substrate with a sacrificial layer and the subsequent lift-off process after fabrication.

However, in the postgrowth patterning method, the Cu foil serves as a handling substrate to mechanically support the graphene and ultrathin polymer film, enabling patterning without additional attachment and lift-off processes.

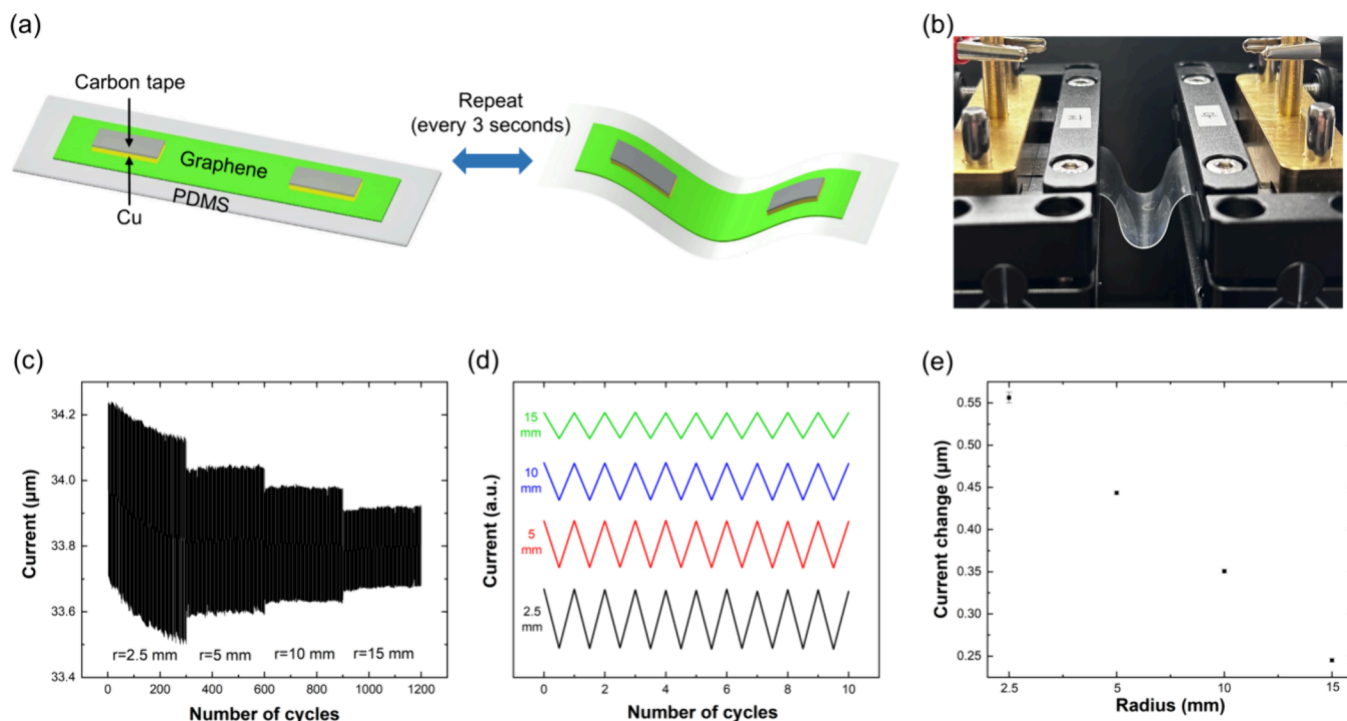
Figure 3b shows the resistance values of the graphene devices (Figure 3a) on various flexible substrates. All devices

exhibited linear  $I$ – $V$  characteristics (Figure S3), confirming that current flowed between the Cu electrodes through the graphene channel. Flexible substrates can be fabricated using the lamination and coating methods. The resistance of graphene-based devices varies based on the substrate material. The interfacial material affects the conductivity of graphene. Interfacial defects that deteriorate device performance may occur at the interface between the graphene and substrate material. If the trapped charge occupies the majority of the moving charge in the graphene channel, and the recharging process is repeated, the change in the Fermi energy is significantly hindered, degrading performance.<sup>54</sup> Applying various substrate materials to graphene is beneficial for identifying substrates with excellent interfacial properties. Specifically, the higher currents observed on parylene can be attributed to its superior insulating properties and excellent compatibility with high-quality graphene. This compatibility improves electron mobility by forming a high-quality interface with graphene, thereby enhancing device performance.<sup>55</sup> Compared to PDMS film, devices formed on PI and PU films exhibit lower currents owing to their higher dielectric constants and greater charge scattering, which reduce electron mobility.<sup>56</sup> The excellent flexibility, low dielectric constant, and high chemical resistance of PDMS make it more suitable for maintaining the integrity of the graphene layer and facilitating better charge transport.<sup>57</sup>

**Application (1): Gas Sensors.** The ability of the postgrowth-patterned Cu contacts to detect changes in the



**Figure 4.** Gas sensing using graphene devices with postpatterned Cu contacts. (a) Schematic of the gas test setup. (b) Schematic of the interaction between the gas molecules ( $\text{NH}_3$  and  $\text{NO}_2$ ) and graphene device. (c) Resistance versus  $\text{NH}_3$  concentration. (d) Resistance versus  $\text{NO}_2$  concentration. (e) Normalized resistance versus gas concentration.



**Figure 5.** Bending test of graphene devices with postpatterned Cu contacts. (a) Schematic of the bending test procedure. (b) Optical image of the bending test setup. Photograph courtesy of “Lee Kyung Bae”. Copyright 2024. Reusing the image needs permission. (c) Current versus time curves for repetitive bending with bending radii ( $r$ ) of 2.5, 5, 10, and 15 mm. (d) Magnified waveforms of the bending profiles at different bending radii. (e) Change in current versus  $r$ . The error bars represent the average difference in current between contraction and relaxation phases over ten cycles, using the values from Figure (d) for each bending radius.

signal (i.e., resistance) from graphene-based sensors was evaluated. Gas-sensing experiments were performed using graphene devices and postgrowth-patterned Cu contacts

(Figure 4a,b). The resistance of graphene increased upon exposure to  $\text{NH}_3$  gas (Figure 4c). By contrast, exposure to  $\text{NO}_2$  gas reduced the resistance of graphene (Figure 4d). The

graphene devices exhibited concentration-dependent resistance changes for  $\text{NH}_3$  and  $\text{NO}_2$  gases (Figure 4e). The similar response trends for  $\text{NO}_2$  and  $\text{NH}_3$  gases are likely caused by the intrinsic electronic properties of CVD-grown graphene, which exhibits high sensitivity to both electron donor and acceptor gases under specific conditions.<sup>58</sup> Additionally, the Cu electrode plays a catalytic role in enhancing the gas sensor response.<sup>59</sup> When  $\text{NH}_3$  molecules adsorb onto the Cu surface, they participate in reduction reactions, releasing electrons that subsequently increase the resistance in the graphene layer by reducing charge mobility.<sup>60</sup>

Conversely,  $\text{NO}_2$  molecules on the Cu surface undergo oxidation, accepting electrons that lower the resistance in the graphene.<sup>61</sup> This catalytic activity of the Cu electrode accelerates electron transfer rates, enhancing the sensitivity and response time of the sensor for low-concentration gases.<sup>62</sup> These experimental results align well with previous studies on graphene-based gas sensors, where exposure to  $\text{NH}_3$  increases resistance and  $\text{NO}_2$  decreases it, as illustrated in Figure 4c,d.<sup>63,64</sup> Upon stopping the  $\text{NO}_2$  gas flow, the sensor gradually returns to its baseline resistance due to the desorption of  $\text{NO}_2$  molecules from the graphene surface.<sup>65</sup> While recovery remains incomplete, additional energy sources like high temperatures or light can facilitate the desorption process.<sup>66,67</sup> Gas sensing signals primarily originate from the Cu/graphene contacts, where the Cu layer's catalytic properties enhance detection capabilities by promoting faster electron transfer rates and improving sensitivity and response time for low-concentration gases.<sup>68–71</sup> To ensure accurate evaluation of sensor response, the gas sensing experiments were conducted under controlled environmental conditions, including humidity, temperature, and airflow, to minimize the influence of external factors on sensor performance. The high gas flow rate of 1000 sccm was maintained to allow rapid and consistent replacement of the testing atmosphere, ensuring reproducible sensor measurements.

**Application (2): Flexible Force Sensors.** The electrical characteristics of the postgrowth-patterned electrodes in a graphene-based sensor was evaluated through mechanical bending. The process involved repeated mechanical contraction and relaxation upon application of external forces. The bending test was performed by measuring the current values based on the bending radius ( $r$ ) as the Cu-contacted graphene device using a PDMS substrate was bent (Figure 5a). As  $r$  decreased or the degree of bending increased, a significant difference in the current values was observed when the substrate contracted and relaxed (Figure 5c,d). Bending the substrate more significantly increased the stress on the device, resulting in a larger change in current (Figure 5e). The flexible Cu-contacted graphene devices demonstrated applicability in electronic skins,<sup>72,73</sup> responding to mechanical stimuli. The mechanical deformation of graphene caused by bending the device alters the electronic band structure of graphene, which affects the mobility of charge carriers, resulting in variations in the current.<sup>74</sup> In addition, the strain sensing mechanisms of graphene-based flexible sensors primarily originate from the geometrical effect and the piezoresistive effect of the graphene material.<sup>72</sup> The mechanoelectrical transduction properties observed in our bending tests validate the potential of these sensors for practical force sensing applications. When subjected to mechanical bending, the induced strain changes the electrical properties of graphene-based sensors, specifically their resistance. This strain-induced change in resistance is a

direct measure of the mechanical force applied to the sensor, thereby demonstrating the capability of the sensor to detect and measure force.

## CONCLUSIONS

This study demonstrated the concept of postgrowth patterning of Cu foil, enabling the formation of electrical contacts with CVD-grown graphene by utilizing the Cu foil as the electrode material. The postgrowth patterning method involved etching of the Cu foil into desired electrode patterns using a masking layer (i.e., conductive carbon tape), applied on the Cu foil/graphene/flexible substrate through a simple “cut-and-paste” method. The proposed approach enables the cost-effective production of graphene devices by eliminating the metal deposition process used in conventional approaches. Moreover, it facilitates scalable fabrication of a large-area device array on a flexible substrate approximately the size of the Cu foil. The Cu-contacted graphene devices exhibited non-rectifying linear  $I$ – $V$  characteristics with a contact resistance of 445.5  $\Omega$  mm. The proposed postgrowth patterning method proved to be a universal method applicable to various flexible substrates, such as silicone/PET, TRT, PDMS, parylene, and PU. In this approach, the Cu foil functions as an electrode material and a mechanical support. This allows for the formation of electrodes on ultrathin and stretchable films, which are typically difficult to manipulate without proper handling of the substrate. The Cu-contacted graphene devices were applied for gas and force sensing applications. The postgrowth patterning of Cu foil is a promising fabrication method for achieving low-cost scalable production of graphene-based electronics for flexible and disposable sensors.

## METHODS

**Materials.** CVD-grown monolayer graphene sheets on Cu foil (thickness = 18  $\mu\text{m}$ ) were purchased from Charmgraphene Co., Ltd. (South Korea). Ammonium persulfate (APS) (248614) was purchased from Sigma-Aldrich (United States). A commercial screen protector (silicone/PET film) for iPad Pro 12.9-in. was purchased from Skoko (South Korea). Thermal release tape (TRT) was purchased from Jinsung Chemicals Co., Ltd. (South Korea). A 200  $\mu\text{m}$  thick polydimethylsiloxane (PDMS) film was purchased from CSCRIE Co., Ltd. (Japan). A 70  $\mu\text{m}$  thick PI adhesive tape was purchased from iNexus, Inc. (South Korea). The PDMS base and curing agent were purchased from Dow Corning (United States). A conductive carbon tape (EST12250) with a width of 50 mm was purchased from LK Lab Korea (South Korea).

**Multilayer Structures of Flexible Substrate/Graphene/Cu Foil.** Two methods were employed in preparing a multilayer flexible substrate/graphene/Cu foil: (1) lamination of the graphene/Cu foil with a flexible substrate and (2) coating of a precursor polymer onto the graphene/Cu foil. The CVD-grown monolayer graphene sheet on the Cu foil was laminated onto a flexible substrate, such as a silicone/PET film, TRT, adhesive PI tape, and PDMS film, using a laminator. Subsequently,  $\text{O}_2$  plasma etching (100 W for 90 s) was performed to remove the back-side graphene on the Cu foil. For the coating process, a PDMS elastomer mixed with an elastomer base and a curing agent at a ratio of 10:1 was cured onto the graphene/Cu foil and heated at 150  $^\circ\text{C}$  for 15 min on a hot plate. To prepare the ultrathin polymer film, a 1  $\mu\text{m}$  thick



polyene C was coated onto the graphene/Cu foil through CVD. Subsequently, O<sub>2</sub> plasma etching (100 W for 90 s) was performed to remove the back-side graphene on the Cu foil. To prepare the ultrathin and stretchable films, a tetrahydrofuran (THF)-based solution with a PU pellet (5%) was spin-coated onto the graphene/Cu foil at a rate of 1,000 rpm at room temperature. These ultrathin and stretchable substrates attached to the Cu foil can be mechanically handled for device fabrication without an additional supporting substrate.

**Postgrowth Patterning of Cu Foil.** The postgrowth Cu foil was patterned into electrodes in two steps: (1) forming a masking layer on the Cu foil/graphene/flexible substrate and (2) Cu etching. The conductive carbon tape used as a masking material was cut into electrode shapes using a computer-controlled CO<sub>2</sub> laser (Universal Laser Systems VLS 2.30DT) with a power of 20 W and scan rate of 20 in/s. After peeling off the unwanted carbon tape from the release liner, the designed mask patterns of the adhesive carbon tape were laminated onto the Cu foil/graphene/flexible substrate. This is similar to the “cut-and-paste” method for the rapid prototyping of soft electronics.<sup>75</sup> The formation of masking patterns with additional materials is presented in the [Supporting Information](#). A 0.3 M ammonium persulfate (APS) solution was used to remove the Cu foil unprotected by the masking patterns, forming separate Cu electrodes in contact with graphene. The removal of the carbon tape masking patterns on the Cu electrodes can be omitted for the electrical characterization of the fabricated graphene devices because the carbon tape is highly conductive (0.3 Ω/□).

**Gas Sensing.** The electrical characteristics of the Cu-contacted graphene devices were measured using a semiconductor parameter analyzer to verify the electrical reaction between the graphene and gases (NO<sub>2</sub> and NH<sub>3</sub>) over time. After placing the postgrowth-patterned Cu contacts in the gas test chamber, the concentration of NO<sub>2</sub>/NH<sub>3</sub> gas was gradually increased from 1 to 40 ppm (1, 5, 10, 20, and 40 ppm) ([Figure 4a](#)). The total gas flow rate was fixed at 1000 sccm. The gas injection and N<sub>2</sub> purging were alternated at 25 min intervals. The high flow rate of 1000 sccm helps achieve rapid response times by quickly replacing the gas around the sensor, ensuring the sensor quickly reaches a steady state when exposed to different gas concentrations. Additionally, the high flow rate ensures consistent gas concentration throughout the sensing chamber and contributes to the reproducibility of sensor measurements, resulting in reliable and reproducible sensor performance.<sup>76</sup>

**Bending Test.** The bending tests involved measuring the resistance change of the Cu-contacted graphene devices using a PDMS substrate through repeated contraction and relaxation. After fixing the postgrowth-patterned Cu contacts in a motion controller (ST-BJS-0810-SSU, Koreaopto (South Korea)), the electrical properties were measured using a SourceMeter (Keithley 2612 B) by varying the radius (*r*) of the bent portion to 2.5, 5, 10, and 15 mm using a motion controller (STM-1-TS, ST1 (South Korea)). The contraction and relaxation were performed 300 times for each radius.

## ■ ASSOCIATED CONTENT

### Data Availability Statement

Data supporting the findings of this study are available from the corresponding author upon request.

## ■ Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.4c09156>.

Additional experimental details, methods, and results (DOC): Selection of masking materials; postgrowth patterning of Cu foil using photolithography; fabrication processes for the postgrowth patterning of Cu foil; and *I*–*V* characteristics of the Cu-contacted graphene devices applied to various flexible materials ([PDF](#))

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### Author Contributions

L.K.B., S.G.S., and J.H.A. conceived and designed the experiments. L.K.B., S.G.S., S.C.P., W.G.P., K.K., H.J.L., D.B., S.H.L., and I.S.K. fabricated the device. L.K.B. and S.G.S. performed the electrical measurements and data analysis. L.K.B., S.G.S., and J.H.A. organized the figures and wrote the manuscript. J.H.A. supervised the study. All authors approved the final version of the manuscript.

### Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

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