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Towards industrial applications of graphene electrodes

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Abstract

Since the first isolation of graphene in 2004 by mechanical exfoliation from graphite, many people have tried to synthesize large-scale graphene using various chemical methods. In particular, there has been a great number of advances in the synthesis of graphene using chemical vapor deposition (CVD) on metal substrates such as Ni and Cu. Recently, a method to synthesize ultra-large-scale (~30 inch) graphene films using roll-to-roll transfer and chemical doping processes was developed that shows excellent electrical and physical properties suitable for practical applications on a large scale. Considering the outstanding scalability/processibility of roll-to-roll and CVD methods as well as the extraordinary flexibility/conductivity of graphene films, we expect that transparent graphene electrodes can replace indium tin oxide in the near future.

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(Some figures may appear in colour only in the online journal)

1. Introduction

In the last few decades, advances in electronic devices have focused on higher speed, larger capacity and smaller size. However, now people are looking for more comfortable electronics devices that are flexible, foldable or even wearable. Thus, the paradigm of electronics is shifting from technology-oriented to user-oriented devices with various types of human interfaces. One of the key materials needed to realize such a new paradigm of electronics is a flexible transparent conductor, which is essential for solar cells, light-emitting diodes (LEDs), organic light-emitting diodes (OLEDs), touchscreens, smart windows and LCD displays [1]. Currently, indium tin oxide (ITO) is mainly used as transparent conducting electrodes in various electronic devices, since it shows an excellent sheet resistance of $10\text{--}50\ \Omega\ \text{sq}^{-1}$ at ~90% transmittance as well as virtually unlimited scalability [2, 3]. However, typical flexible polymer substrates such as polyethylene terephthalate (PET) are easily deformed at the usual deposition temperatures of ITO

($T_g < 200\ ^\circ\text{C}$). Therefore, ITO needs to be sputtered and annealed near room temperature for flexible applications based on polymer substrates, leading to amorphization and degradation of electrical properties due to its low carrier concentration and high defect densities [4]. Moreover, amorphous metal oxide thin film materials such as ITO are brittle and fragile, which is unsuitable for applications that require high flexibility. In addition, since ITO is sensitive to both acidic and basic environments, it is inappropriate to use ITO for an application that needs chemically inert electrodes such as dye-sensitized solar cells (DSSC) [1]. Over the last few years, the production cost of ITO has increased a few times because of the imbalance between demand and supply, which makes it more difficult to use ITO for future electronic applications.

Figure 1 shows possible applications of flexible transparent electrodes for corresponding sheet resistance ranges. Most of the industrial applications require sheet resistance lower than $500\ \Omega\ \text{sq}^{-1}$ for transparent electrodes. For example, the sheet resistance for OLED displays and solar

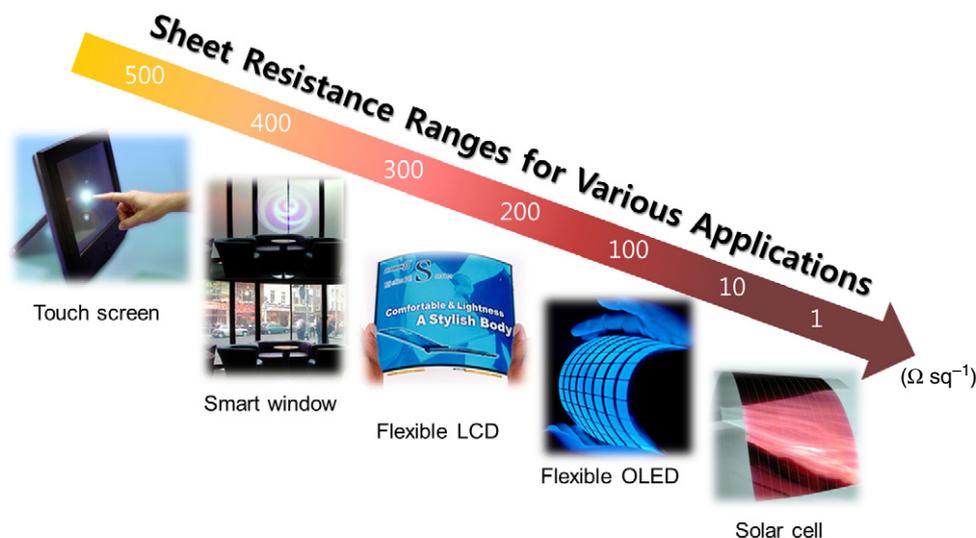


Figure 1. Potential graphene applications and corresponding resistance range required for each application.

cells should be less than $\sim 50 \Omega \text{sq}^{-1}$, and for touchscreens a sheet resistance of $200\text{--}500 \Omega \text{sq}^{-1}$ is acceptable.

Recently, various alternative conducting transparent electrodes to ITO were developed, which include metallic mesh films [5, 6], conductive polymer films and carbon nanotube (CNT) [7–9] films. However, none of the alternative electrodes have been used practically due to performance limits as well as higher costs of production. For example, it is very difficult to separate metallic CNTs from semiconducting CNTs, which limits the sheet resistance of CNT films. The conductive polymers also have many drawbacks such as complicated synthesis processes, low solubility that limits processibility, large absorption in blue ranges and chemical instability in ambient conditions. Unlike the shortcomings of the aforementioned materials, graphene shows outstanding properties to be used as transparent conductors as we will discuss later.

2. Scaling-up graphene electrodes

Graphene is made of a single layer of carbon atoms that are densely packed into a honeycomb lattice. It is an attractive material with outstanding electrical, mechanical and chemical properties that can be used as transparent electrodes for various applications in flexible electronics [10–17]. Since the discovery of the first isolated graphene obtained by mechanical exfoliation of bulk graphite [10], many chemical approaches for obtaining large-scale graphene have been developed, including epitaxial growth of graphene on silicon carbide [18, 19] and ruthenium [20] as well as the approach to produce (reduced graphene oxide, RGO) sheets by a simple solution dispersion process [21–24].

Table 1 summarizes four representative methods for obtaining graphene films. The graphene obtained from the mechanical exfoliation method shows excellent properties but it is impossible to utilize it for large-scale practical applications. Epitaxial growth allows us to produce high-quality multilayer graphene samples. However, SiC substrates are relatively expensive, and the scale of synthesis is limited by the size of SiC wafers. In addition, it is difficult

to transfer graphene from SiC to an arbitrary substrate, which is essential for the use of graphene for transparent electrodes.

The self-assembly of dispersed graphene sheets in solution demonstrates the possibility of low-cost synthesis and the fabrication of large-scale graphene films. However, the assembled graphene films have relatively poor electrical conductivity owing to the high interlayer junction contact resistance and the structural defects formed during the oxidation and reduction processes. Thus, we can conclude that synthesis by chemical vapor deposition (CVD) methods is expected to be the most suitable for large-scale and high-quality graphene film production to be used as transparent electrodes.

The CVD-grown graphene films can be successfully transferred onto arbitrary substrates with various methods as shown in figure 2 [25–29]. Lee *et al* [29] recently reported a wafer-scale synthesis and transfer method that utilizes poly-di-methyl-siloxane (PDMS) or thermal-release tape as polymer supports to transfer graphene from on-metal layers to target substrates. The CVD-grown graphene can also be transferred by using spin-coated poly-methyl-methacrylate (PMMA) polymer films [26–28]. Li *et al* [28] found that the graphene transfer yield can be improved by introducing a second PMMA coating on top of the first PMMA/graphene layer because of the enhanced mechanical strength. However, the scale is also limited below the size of wafers because the PMMA layer has to be spin-coated on flat and rigid substrates.

In order to overcome the limitation of previous methods, Bae *et al* [30] developed a roll-to-roll transfer method utilizing thermal release tapes as shown in figure 3. The roll-to-roll method is easy to scale-up and also allows simple layer-by-layer (LBL) multiple transfers combined with the doping of individual layers that can enhance the sheet resistance of graphene.

3. Enhancement of sheet resistance

The versatility of graphene applications can be enhanced by adjusting the sheet resistance of graphene. Intrinsic

Table 1. Four representative methods for obtaining graphene films.

Method	Mechanical exfoliation	Chemical reduction from graphene oxide	Epitaxial growth on SiC	CVD based graphene growth on Ni, Cu, Fe, Co
Size	10~100 μm	> 6 inch	< 4 inch	> 6 inch
Mobility	best	bad	high	high
Transfer	yes	yes	no	yes
Applications	no	yes	little	most
Scalable	no	yes	not yet	yes

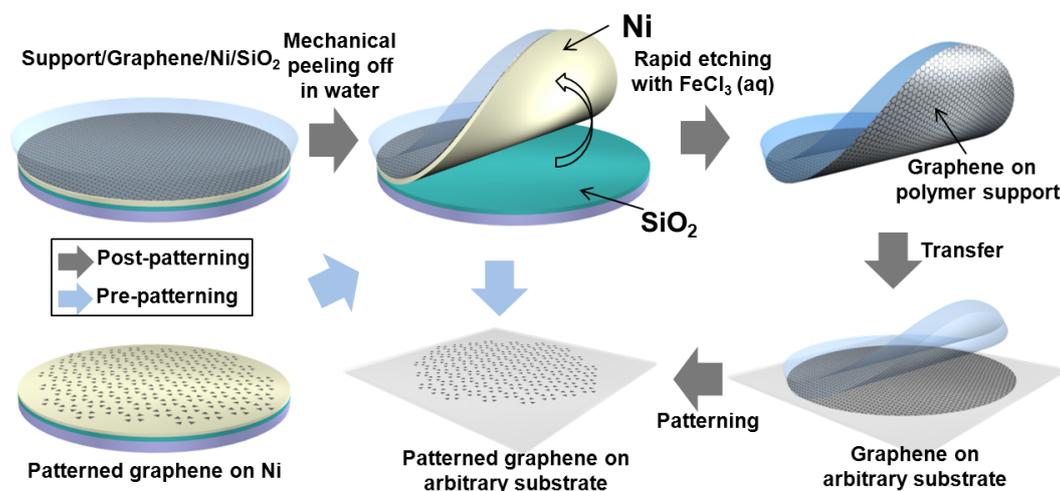


Figure 2. Schematic diagram of the polymer-based transfer method of graphene films grown on Ni or Cu/SiO₂/Si. Polymer-based graphene/metal layers are mechanically detached from a SiO₂/Si wafer. After etching of the metal layer, the graphene films can be easily transferred to target substrates.

single-layer graphene typically has a sheet resistance of $\sim 500 \Omega \text{sq}^{-1}$. While this sheet resistance value is good enough for touchscreen applications, the lower value is essential for solar cells, LED, LCD and electromagnetic interference shielding (electromagnetic interference, EMI) applications. There are two representative methods to decrease the sheet resistance: LBL transfer and chemical/electrical doping [27, 30, 31]. The LBL method is the most useful to decrease the sheet resistance by more than 50%, but after four-layer transfer, the decrease tends to saturate [28, 30].

Chemical doping methods can further enhance the conductivity of graphene [30, 32–34]. The decrease of sheet resistance for various chemical dopants is shown in figure 4(b). Combining these two methods, sheet resistance as low as $\sim 30 \Omega \text{sq}^{-1}$ at 90% transparency can be achieved, which is similar to that of commercial transparent electrodes such as ITO. However, the sheet resistance of the chemically doped graphene decreases with time due to the instability of doping agents. Therefore, recent research efforts have been devoted to stabilizing the doping effect by using protective barrier films.

4. Work-function engineering

A dominant transparent electrode used in current industrial applications, ITO, makes up n-type semiconductors along with In₂O₃ and SnO₂. ITO films are produced by various techniques such as evaporation in a reactive or vacuum atmosphere, sputtering, reactive ion plating, glow discharge deposition and different varieties of CVD [1]. For the deposition method of ITO, the properties of ITO can be adjusted according to the ratio of the deposited materials. Once the deposition process is completed, the work function of ITO remains fixed. However, this poses a practical problem for using ITO as an electrode since the work function plays a critical role. However, when a work function difference exists between ITO and the active graphene layer, the overall performance can be downgraded despite the superior optical and electrical properties of the used graphene layer. For best device performance, the active layer and the electrode should form ohmic contact to decrease the energy barrier of the contacting layers and achieve high carrier mobility. Thus, graphene promises a more favorable electrode, since its work function can be adjusted by LBL transfer stacking, chemical doping, self-assembled monolayer (SAM) treatment on the

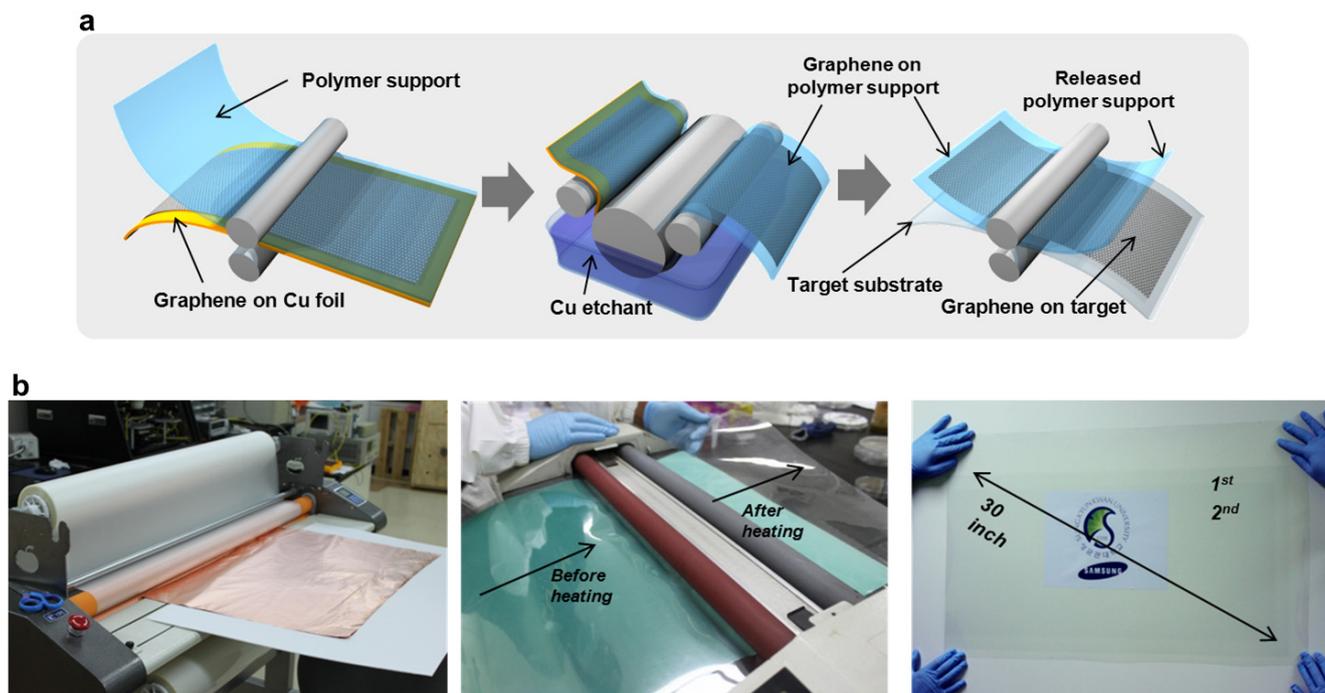


Figure 3. (a), Schematic diagram of the roll-based production of graphene films grown on a copper foil. The processes are divided into three steps: (1) the adhesion of polymer supports, (2) copper etching and (3) transfer to the target substrate. (b) Photographs of the roll-based production of graphene films on copper foil produced by the process shown in panel (a).

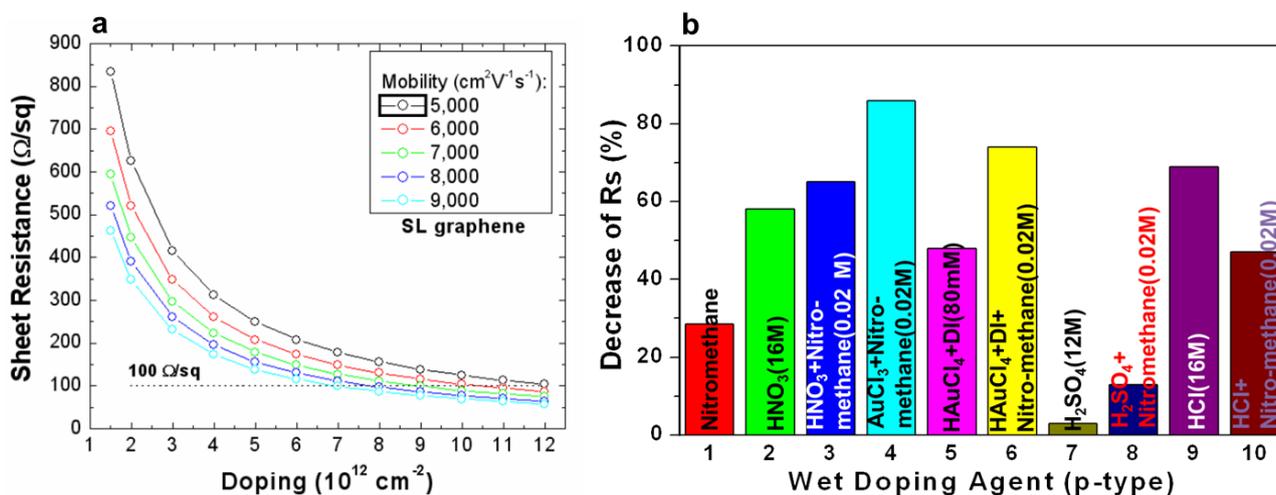


Figure 4. (a) Sheet resistance versus doping with different charge carrier mobilities. The sheet resistance is calculated by $\rho = 1/\sigma = 1/ne\mu$. (b) Decrease of sheet resistance versus various wet doping agents.

substrate or induced strain (figures 5(a)–(d)) [30, 35–38]. Figures 5(a) and (b) show the effectiveness of chemical doping in controlling the work function of graphene.

When nitric acid (HNO₃) is used for doping, the G and two-dimensional (2D) peaks of graphene are blue shifted and the carbon 1s peak of graphene in x-ray photoelectron spectroscopy (XPS) is broadened and red shifted upon p-doping (figure 5(a)). The work functions of each doped graphene layer measured by ultraviolet photoemission spectra (UPS) are shifted by ~130 meV as the doping time is increased (figure 5(b), inset). For SAM material-treated graphene films on SiO₂ substrates, the work functions obtained were 3.9, 4.25 and 4.5 eV, respectively (figure 5(c)). The surface treatment method

is a destructive doping technique, similar to substitutional doping or covalent functionalization of graphene that were previously reported [39]. In contrast to the chemical doping process, this technique allows lasting stability of the doping condition because the robust self-assembled ultra-thin layer forms a strong covalent bond with the SiO₂ surface. This confirms the effectiveness of SAM treatment in controlling the graphene work function, making it suitable for optoelectronic devices [40, 41]. The work function in uniaxially strained graphene is increased significantly with increasing strain. The theoretical calculation [42] and experimental result [43] of the graphene work function without strain is 4.5 eV. Depending on the direction of the strain, the change in the rate of the graphene work function is quite different and

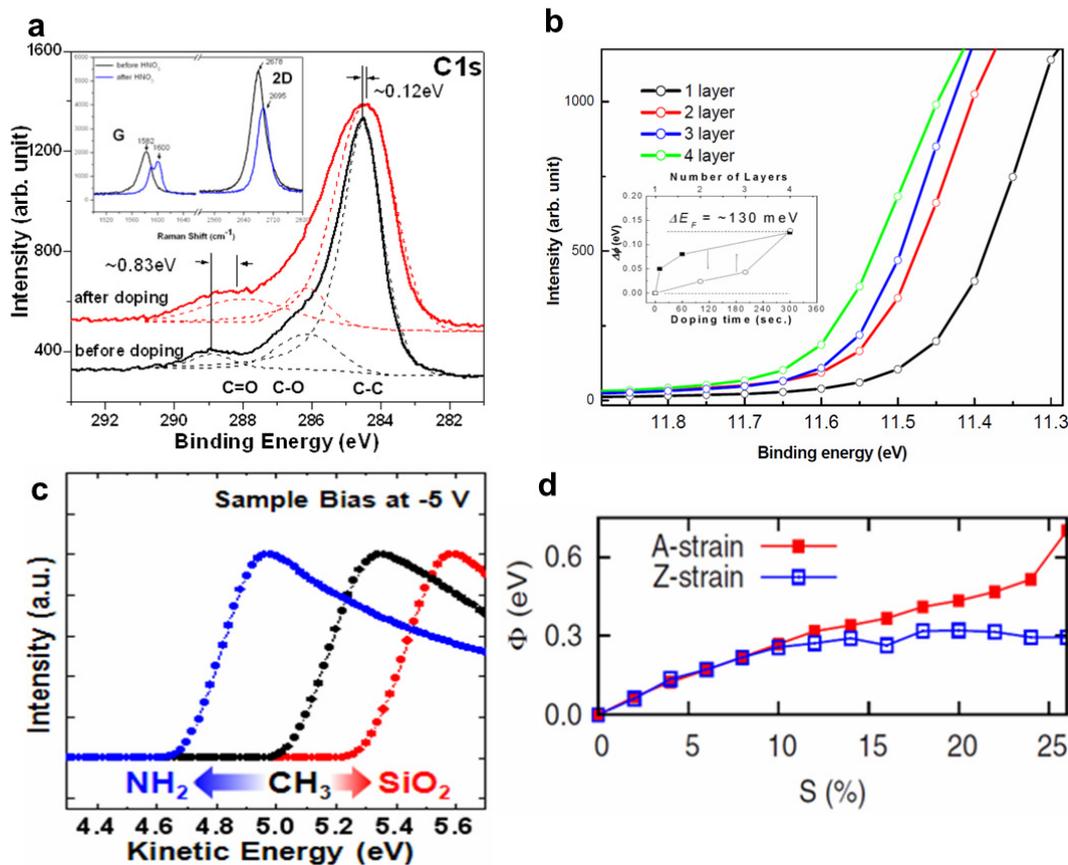


Figure 5. (a) XPS peaks of monolayer graphene films transferred to SiO₂/Si substrates, showing typical redshift and broadening of carbon 1s peaks (C1s) caused by p-doping. The inset shows Raman spectra of doped and undoped graphene films; both G and 2D peaks show ~ 18 cm⁻¹ blueshift. (b) UPS of HNO₃-doped graphene films with different numbers of stacked layers. The binding energy increases with the number of stacked graphene layers. The inset shows work-function changes ($\Delta\Phi$) with respect to doping time (the lower x-axis) and the number of stacked layers (the upper x-axis), measured by UPS. (c) UPS of the graphene films on SAM-modified SiO₂/Si substrates. (d) Calculated work functions (Φ) of graphene with the A and Z strains.

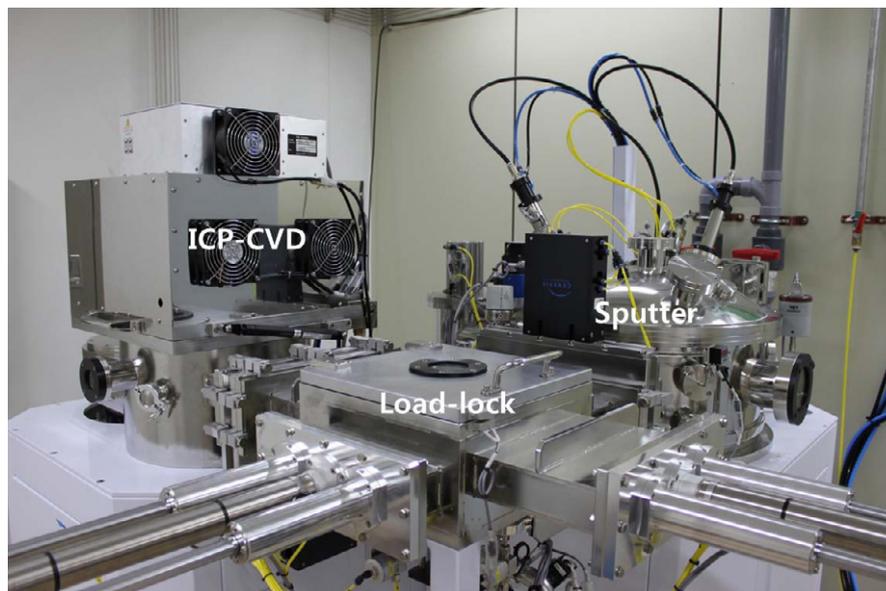


Figure 6. ICP-CVD system combined with a metal sputtering chamber connected through an automated load-lock module.

such a strain-induced work-function change can alter device characteristics. Calculation shows that the graphene work function increases linearly as the strain increases. With about 26% A/Z strain, the work function saturated at 5.2 and 4.8 eV in figure 5(d).

As seen from the aforementioned methods, many research efforts to adjust the graphene work function are being actively carried out and consistent studies on manipulating the work functions to better apply graphene for various electronic devices need to be carried out in the future.

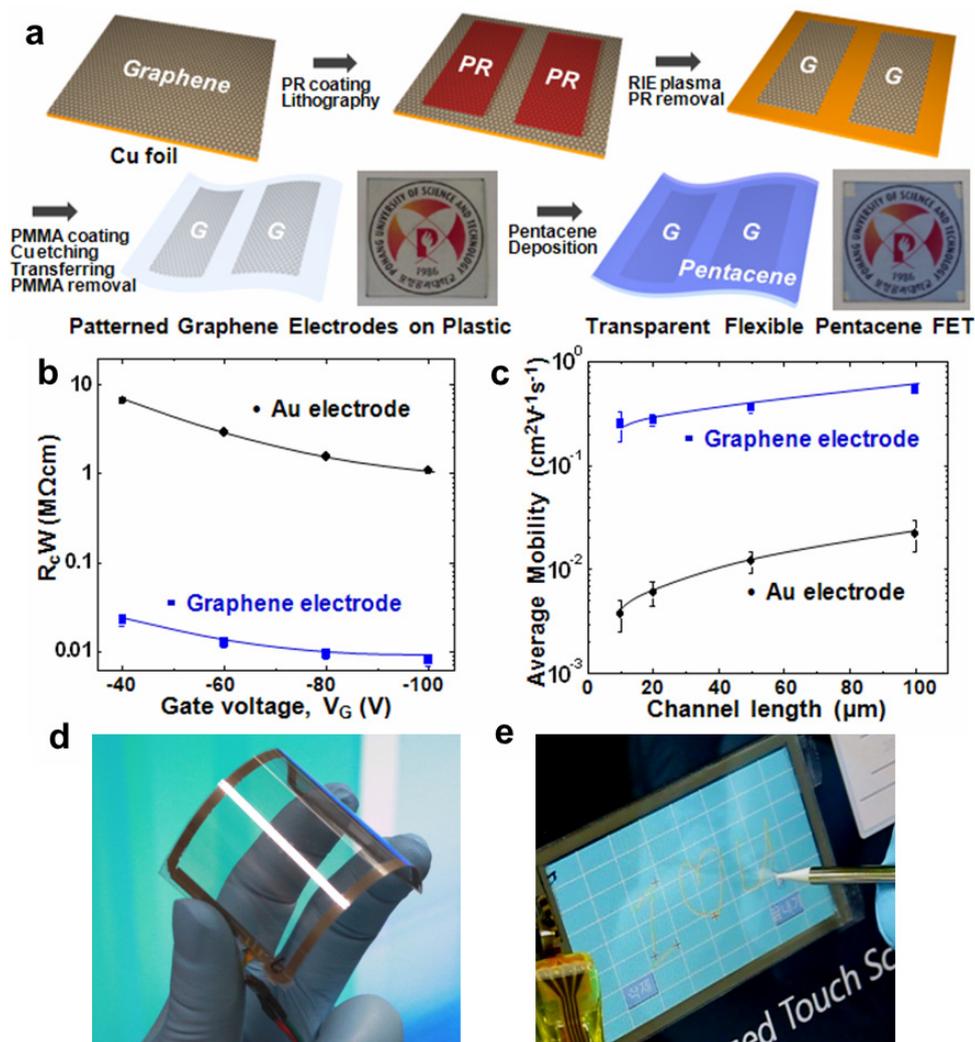


Figure 7. (a) Schematic representation of the procedure for fabricating patterned monolayer graphene electrodes on a plastic substrate. Digital camera images of flexible/transparent FETs with monolayer graphene source/drain electrodes before and after pentacene deposition are also shown. In $20 \times 20 \text{ mm}^2$ there are 120 FETs. (b) Calculation of the contact resistance values. Contact resistances of FETs with Au electrodes are shown for comparison. (c) Channel length-dependent field-effect mobilities of FETs with Au electrodes are shown for comparison. (d) Graphene/PET-based touch panel showing outstanding flexibility. (e) A graphene-based touchscreen panel connected to a computer with control software.

5. Low-temperature and low-cost synthesis

According to the most up to date literature, the growth of graphene films by CVD is more suitable for transparent electrode application than chemically reduced graphene (RGO) because of RGO's low electrical property resulting from poor interlayer junction contact resistance and structural defects formed during vigorous exfoliation and reduction processes. The conventional thermal CVD process requires high-temperature annealing and synthesis steps and long heating and cooling times, and accordingly requires a growth substrate that can withstand heating and cooling. Plasma CVD methods such as microwave plasma-enhanced CVD [44], surface wave plasma [45] and inductively coupled plasma (ICP)-CVD [46] are useful methods for obtaining graphene films at low temperature below 500°C . Recently, a successful application of plasma-CVD grown graphene as transparent electrodes has been reported [47]. Figure 6 shows ICP-CVD and sputter systems connected with a one load lock chamber, which prevents the oxidation of metal catalyst

to synthesize graphene with good quality. Graphene films can be synthesized on various metals on plastic substrates below 400°C by ICP-CVD and directly transferred to plastic substrates during the etching process. But the electrical and optical properties of graphene grown by ICP-CVD at low temperature need to be further enhanced for practical applications of future electronics. Nevertheless, within only six years since the discovery of graphene, numerous reports of synthesis, etching, transfer and fabrication have been published and developments have rapidly improved graphene properties for applications in physics, chemistry, mechanical engineering, biomedical engineering and the material science field. These indicate that the use of the low temperature growth technique for continuous graphene production will soon be accomplished for practical applications.

6. Future applications

In recent years, many researchers have been developing graphene-based applications for transparent electrodes such as

organic thin-film transistors (OTFT) [48], touchscreen [30], LEDs [49, 50], solar cells [51] and smart windows [5]. Figure 7(a) shows schematically the process of patterning graphene using photo-lithography and ultimately making transparent pentacene-graphene field effect transistor devices. When the graphene layer is used as the electrode and an organic material such as pentacene is used as the active layer, it shows about 100 times lower contact resistance and higher electron mobility than when Au is used as the electrode (figures 7(b) and (c)). As presented, low contact resistance between graphene electrodes and organic materials allows the production of transparent devices with improved performance. Figures 7(d) and (e) show the first successful application of graphene in touchscreens as a transparent electrode component. This shows excellent potential for applications to flexible transparent electronic devices. Although the practical device application is still at lab-bench scale, commercial scale adaptation of graphene as a transparent electrode does not seem far way.

7. Summary

Flexible and transparent electrodes, such as conductive polymer, CNTs, graphene and multi-layer graphene electrodes without indium, have been developed for use in the field of flexible stretchable electronic devices. Even though there are negligible markets for the flexible display, solar cell or touch panel, flexible and transparent electrodes will become important as the market grows. Flexible transparent electrodes with superior properties are expected as various materials have been studied for industrial application. Graphene is expected to be an alternative material to ITO, which is commonly used as transparent electrodes in touch sensors and solar cells. Additionally, because of its superior mechanical properties, it has great potential for flexible electronics such as OLED displays and solar cells. Even with only six years of research on graphene since the discovery in 2004, robust research efforts have allowed the development of various technologies for industrial applications, such as transparent electrodes and semiconductor devices. There is no doubt these technologies will quickly become more efficient in the near future.

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