Invited Paper

Prospects of graphene electrodes in photovoltaics

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ABSTRACT

Transparent conductors (TCs) are becoming extremely popular in many different electronic applications such as touch panels, displays, light emitting devices, light sensors and solar cells. The commonly used electrode in these applications is Indium Tin Oxide (ITO). However, the cost of ITO is increasing rapidly due to the limited supply of Indium. Other issues such as lack of flexibility and cost of the deposition process make ITO less favorable in transparent electrode applications. Graphene has been under exploration as an alternative material for TC applications in the recent years. Graphene based TCs have been shown experimentally to exhibit promising electrical and optical properties. In this paper, the prospects of graphene for transparent conductors in photovoltaics are discussed. The recent advancements in this field as well as the theoretical predictions and possible pathways for improvements are presented. In the process section, we discuss methods to synthesize few-layer graphene (FLG) with high quality in a controllable manner.

Keywords: Graphene, photovoltaics, transparent conductor

1. INTRODUCTION

Transparent conductors (TCs) are essential components of modern electronics and photonics commonly found in touch panels, displays, light emitting devices, light sensors and solar cells [1], [2]. The market for transparent electrode is growing rapidly, and is projected to surpass $11 billion in 2016. The technical requirements for TCs in solar cell applications are *sheet resistivity* (R_s) less than 10 Ω/square and *optical transparency* (T) larger than 90%.

The commonly used electrode in these applications is ITO. However, the cost of ITO is increasing rapidly due to the limited supply of Indium, lack of flexibility and cost of the fabrication process. Therefore, exploration of new materials for transparent electrode applications is necessary to achieve low cost and high efficiency. Potential replacement materials include metal grids [3], [4], metal oxides [5], [6], and thin film metals [7], [8]. Low sheet resistivity and high optical transmittance are the fundamental requirements for these electrodes. Currently, neither ITO nor the alternative electrodes satisfy the industry's future requirements.

High transmittance, high conductivity, high mechanical flexibility as well as impermeability to moisture (leading to improved reliability) make graphene a promising electrode material for a variety of photovoltaic applications. In particular, graphene synthesis by chemical vapor deposition (CVD) (which is the only way of producing large area graphene transferrable to any substrate) allows the use of graphene as electrode material in various applications. Recent work characterizing graphene under electrostatic discharge (ESD) conditions (a key reliability issue in the electronics industry) has also shown that graphene is highly resilient against ESD, which is crucial for various TC applications [9]. However, the sheet resistance of single-layer graphene (SLG) is not yet satisfactory for transparent electrode applications.

In [10], [11] sheet resistance down to 30 Ω/sq has been reported by use of individually stacked tetra-layer graphene and chemical doping. Moreover, a theoretical work indicates that it is possible to achieve a sheet resistance of 10 Ω/sq with 90% transmittance by chemical doping and stacking 4 layers of graphene [12]. However, this layer-to-layer stacking technique relies on the quality of the individual monolayer graphene sheets and a complex transfer process. Note that the sheet resistivity of SLG is generally larger than 100 Ω/sq, which is much higher than that of few layer graphene (FLG). Therefore, FLG poses as a much stronger alternative for TC applications compared to SLG.

In this paper, the prospects of FLG) for transparent conductor applications are reviewed. Various developments in graphene TCs as well as the trade-offs between the film thickness and the optical and electrical conductivities are discussed, which are required for optimization of these structures. In the process section, we discuss methods to synthesize FLG (less than 4 layers) with high quality in a controllable manner. Furthermore, doping of FLG via various methods is also discussed that presents new opportunities in improving the performance of FLG as a TC. The integration of FLG based TCs with semiconductors is another important challenge in the development of FLG

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based TCs. Hence, we discuss optimization of the interface between FLG and semiconductors including the interactions of FLG with semiconductors, which is necessary for achieving high performance photovoltaic devices.

2. APPLICATIONS OF GRAPHENE ELECTRODES

Graphene based electrodes can be used in a variety of applications as transparent conductive electrodes such as touch panels, displays, light emitting devices, light sensors and solar cells. The industry standards for the electrical sheet resistivity and optical transparency of TCs vary within a wide range in these applications. For example, R_s<10 Ω/square and T>90% is required for TCs in solar cells. However, for touch panels R_s of ~300-1500 Ω/square and T>85% is considered sufficient. While many of the reported graphene based TCs can satisfy the requirements for touch panel applications, the research continues into fabrication of TCs, which can meet the standards of solar cell applications. A fabricated graphene based touch panel is reported by researchers in [11], and shown in Figure 1. The fabricated touch panel exhibits high mechanical flexibility. They also showed that graphene can handle more than twice the strain that ITO can handle. Unlike ITO panels, which break at 2-3% strain, the graphene based touch panels were able to withstand 6% strain. The strain limit on the graphene based panel came from the silver printed electrodes and not from graphene's inherent limitation.

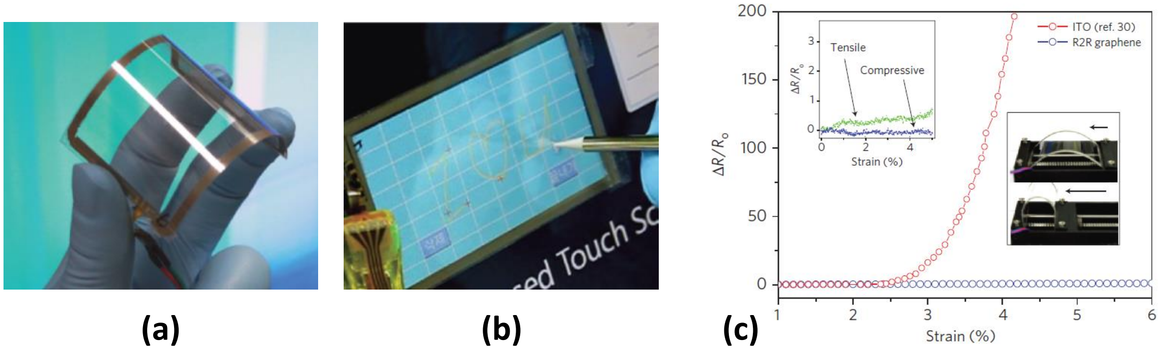

Figure 1. (a) A fabricated graphene/polyethylene terephthalate (PET) touch panel showing high mechanical flexibility. (b) Graphene/PET based touch panel working on a computer. (c) Comparison of the electromechanical properties of graphene/PET and ITO/PET electrodes under tensile strain. Graphene/PET electrode shows negligible sensitivity to the applied strain. The inset shows the resistance change with compressive and tensile strain applied to graphene/PET electrodes. Reproduced with permission from ref. [11]. © 2010 Nature Publishing Group.

3. ELECTRONIC AND OPTICAL PROPERTIES OF GRAPHENE

The electronic structure of single-layer graphene is described by a tight-binding Hamiltonian [13], [14]. In graphene, the separation of p_z bands and the σ bands near the Fermi energy is very large. So the p_z electrons play the dominant role in the electronic properties of graphite. Graphene has linear energy dispersion near the Dirac point with zero gap. The application of ultra-fast optical pulses to graphene leads to inter-band excitations and produces a non-equilibrium carrier population in the valence and conduction bands. Two relaxation time scales of ~100 fs (due to carrier-carrier intra-band collision and phonon emission) and ~1 ps (due to electron inter-band (*e-e*) relaxation and cooling of hot phonons) are observed [15]–[17].

SLG is a highly transparent material with a typical optical transparency of ~97.7% [18]. FLG can be considered as equivalent to a superposition of non-interacting SLGs [18], [19]. Therefore, the transparency of FLG becomes T=1-0.023N, with N being the number of layers. Experimental measurement results of the electrical conductivity and optical transparency of FLG exhibit a broad variation of these parameters as shown in Figure 2. Therefore, any improvement towards the sheet resistivity of FLG will improve the performance of FLG as a TC. A theoretical study [12] suggests that graphene has the potential to meet the future industry requirements for TC applications. Different methods of improving the sheet resistivity of graphene films will be discussed in this paper.

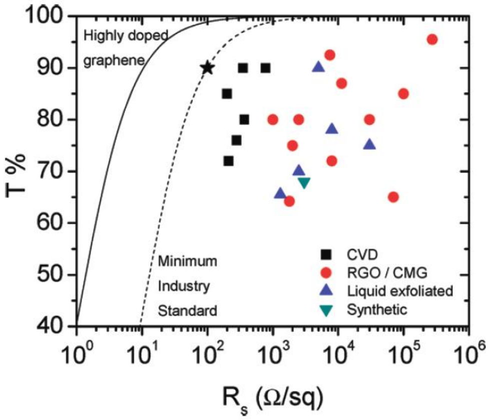

Figure 2. Optical transparency versus electrical sheet resistivity data for published articles. Reprinted with permission from ref. [12]. © 2010 American Chemical Society.

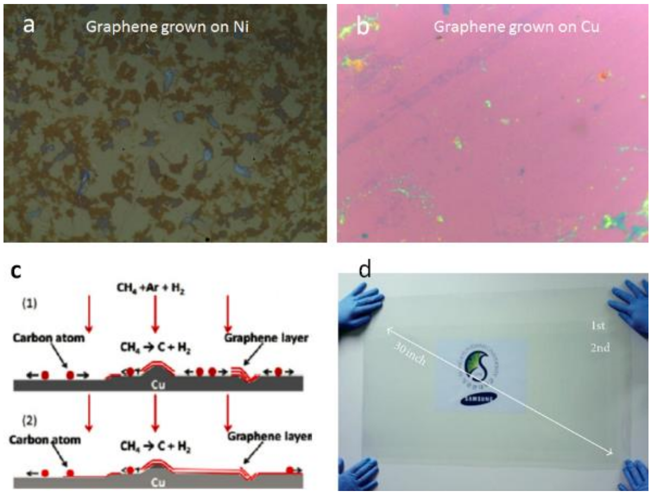

Figure 3. Optical microscope image of graphene grown on (a) Ni and (b) Cu. (c) Schematic of graphene growth process at low-pressure CVD condition: (1) Nucleation starts at the steps and grain boundaries, (2) Growth process after nucleation [29]. (d) A 30-inch graphene film transferred on a 35-inch PET sheet. Graphene was grown on Cu and transferred using thermal release tape and wet etching. Reproduced with permission from ref. [11]. © 2010 Nature Publishing Group.

4. GRAPHENE PREPARATION

The synthesis of large-area and high-quality graphene is important for the high-volume manufacturing of TCs. Recently, different methods have been demonstrated for the preparation of graphene, such as mechanical exfoliation of graphite [20], sublimation of epitaxial SiC [21], and catalyst-assisted CVD [11], [22]–[25]. CVD method, which uses Cu or Ni as catalyst, has been proposed as a promising approach allowing higher efficiency and scalability. Graphene forms on Ni surface through a carbon dissolution-precipitation process, which is originally from the carbon solubility in Ni at high temperatures. However, carbon exhibits different precipitation behaviors on the grain boundary and surface of Ni film, thereby resulting in a non-uniform growth of the synthesized graphene (Figure 3(a)). Compared with Ni, Cu has a better capability to grow high quality and uniform graphene (Figure 3(b)). Graphene growth on Cu mainly depends on a surface catalytic process due to the nearly zero carbon solubility below Cu's melting point (Figure 3(c)). It is worthwhile to note that a 30-inch graphene film (Figure 3(d)) has been demonstrated by a roll-to-roll method [11], indicating the possibility of producing graphene films at an industrial scale. Graphene transfer is another challenging task. It has been shown that the wet transfer technique will increase the density of residual moisture between graphene and substrate [2]. The residual molecules increase the contact resistance and degrade the performance of the devices. A dry transfer technique with use of PMMA layer in conjunction with thermal release tape lead to improved characteristics [2].

5. GRAPHENE ELECTRODE ENGINEERING

The electrical and optical properties of FLG can be varied by tuning the number of layers and doping density. While increasing the number of layers increases the available modes for conduction and improves the electrical sheet resistivity, it also reduces the optical transmittance. Intrinsic FLG has a very low electron density, which leads to low electrical conductivity. Doping of FLG is an effective way to increase the electron density, and improve the electrical conductivity. Several key methods are discussed here for improving the electrical properties of FLG and for optimizing its optical transparency.

5.1 Substitution doping

It has been experimentally demonstrated that the electrical properties of graphene can be tuned by substituting C atoms with boron (B) and nitrogen (N) atoms [26]–[28]. C atoms form stable chemical bonds with B or N atoms. BCN film and N-doped graphene films have been grown on Cu by CVD methods through a surface catalytic process. B-doped graphene has a *p-type* conducting behavior, while N-doped graphene is *n-type*.

5.2 Surface transfer doping

In contrast to the substitution doping in which the carbon atoms are replaced by the dopant atoms, the surface transfer doping method is achieved by electron exchange between FLG and adsorbed dopant atoms on the surface of FLG. This method is also called *adsorbate-induced doping*. In general, the surface transfer doping does not alter the band structure of FLG. Depending on the type of the molecule adsorbed on the surface, *n-type* or *p-type* doping is possible. For example, water vapor and ammonia lead to *p-* and *n-type* doping of graphene, respectively [20], [29]. The doping of SLG leads to a shift of the minimum conductivity point (Dirac point) in I_{DS}-V_{GS} measurement. *n-type* and *p-type* doping shift the Dirac point to more negative and more positive gate voltages, respectively. These experimental observations have also been interpreted by theory [30], [31]. Alternative dopants include Br_2, I_2 and K [32], [33].

5.3 Substrate engineering

Substrate engineering can be used to tune the doping density in FLG. Furthermore, in some photovoltaic applications, the interface between graphene and the substrate contributes to current transport. For example, in a solar cell where FLG is being used as a transparent conductor over a semiconducting active region, the contact resistance between the FLG and the active region should be kept as low as possible. Substrate engineering can be done by use of different substrates with different work functions and by doping the substrate. In addition, a clean substrate, which is free of dangling bonds is crucial because the dangling bonds and surface dipoles act as scattering centers.

5.4 Intercalation doping

Graphite intercalation compounds (GICs) have been used for more than 100 years to improve the characteristics of graphite. The first paper on GICs dates back to 1841, when Schafhautl published his first paper on H_2SO_4 intercalated graphite. Since then, numerous works have been published on improving the electrical sheet resistivity of graphite and graphene layers [34]–[47]. Intercalation of graphite is accomplished by introduction of a different atom or molecule in between the graphene layers. This can be done through introduction of a vapor phase gas to graphite in a controlled environment. Common intercalate materials include but are not limited to HNO_3, SbF_5, AsF_5, $CuCl_2$, $FeCl_3$, F_2, K-Bi and Br_2. GICs have improved the in-plane sheet resistivity of graphite by several orders of magnitude [37], [46], [48]. Similar to graphite, the intercalation of FLG leads to an improvement of the sheet resistivity [32], [35], [38]–[40], [49]–[55]. $FeCl_3$ doping of FLG has shown promise as shown in Figure 4. The temperature dependent sheet resistivity of $FeCl_3$ intercalated FLG is shown in Figure 4(a), and can be compared to the sheet resistivity of pristine FLG in Figure 4(b). As expected, the sheet resistivity reduces as the number of layers increases.

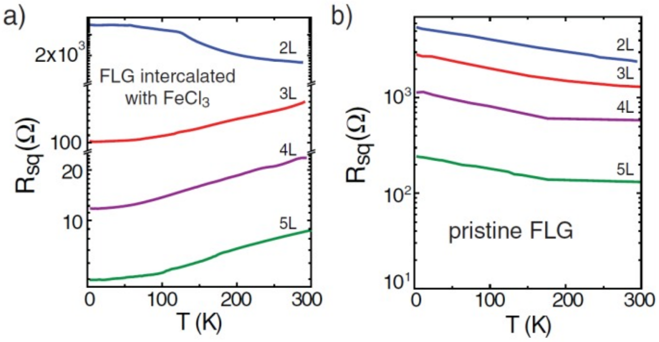

Figure 4. Temperature dependence of the sheet resistivity for (a) $FeCl_3$-FLG and (b) pristine FLG of different thicknesses. Reprinted with permission from ref. [56]. © 2012 Wiley Publishing Inc.

6. CONCLUSION

The recent advances in graphene based TC applications were discussed. FLG doped with intercalation materials such as HNO_3 and $FeCl_3$ has already shown promise for high electrical conductivity and high optical transparency. The theoretical predictions were discussed, which suggest that the electrical conductivity of FLG can be further improved by use of different doping mechanisms. With the projected predictions, the FLG based TC can be expected to pass the industry requirement for the next generation of TCs, including application in solar cells. A large scale fabrication process can potentially bring down the cost of fabrication of FLG based TCs, at which point the superb characteristics of FLG based TCs as well as their mechanical flexibility and impermeability to moisture will make FLG based TCs a cost effective and strong alternative to ITO and other TCs.

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