



A review of graphene based transparent conducting films for use in solar photovoltaic applications



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ABSTRACT

Graphene has been recognised for its various excellent properties and its potential to be applied in various applications such as transparent conducting films (TCF), optoelectronic devices and energy storage. Graphene has been successfully synthesised by various methods including chemical vapour deposition (CVD), exfoliation and solution-processed. As-synthesised graphene has been used in various applications and its potentials have been realised. However, graphene performance has been compromised due to various factors. In this review, we present the methods to improve graphene performance by doping, enhancement of transfer methods and hybrid films of graphene with other materials. In addition, graphene as TCF for photovoltaic applications has also been described. We also include a discussion on various Figure of Merit (FOM) determinations, to evaluate the performance of graphene as transparent conducting films.

1. Introduction

Transparent conducting films (TCF) are highly conductive and highly transparent materials used in various applications such as solar cells, touch screens, light emitting devices, etc. Indium tin oxide (ITO) as TCF is a commonly used material in various applications as it exhibits excellent properties of low sheet resistance (R_s) in the range of 10 – 25 Ω /sq and optical transmittance (T) greater than 90%. However, the shortcomings of ITO such as the high cost of raw material (scarcity of indium), brittleness when applied in flexible devices and expensive fabrication process (high temperature processing), have led to the study

of alternative materials. In addition, the problem of instability in acids or bases, vulnerability to ion diffusion into the polymer layer and poor electrical contact with organic materials, have reduced the possibility of ITO for further use in organic devices. On the other hand, the other widely used TCF for its comparable performance with ITO, fluorine tin oxide (FTO), is associated with problems such as degradable performance at high temperatures due to an increase in sheet resistance and defects of FTO, causing current leakage.

As TCF, the materials must fulfil the requirements of high electrical conductivity and high optical transparency for use in various device applications. On the other hand, other factors to be considered in

Abbreviation: a-C, amorphous Carbon; a-Si, amorphous silicon; a-Si:H, hydrogenated amorphous silicon; AgNC, silver nanocomposite; AgNW, silver nanowires; APTES, 3-Aminopropyltriethoxysilane; AZO, aluminium zinc oxide; BLG, bilayer graphene; BIPV, building integrated photovoltaics; C-CVD, Catalytic-Chemical Vapour Deposition; CdTe, cadmium telluride; CFG, crack-filled graphene; CIGS, copper indium gallium selenide; CNT, carbon nanotubes; CO, carbon monoxide; CuNF, copper nanofibers; CuNW, copper nanowires; CuNT, copper nanotrough; CVD, chemical vapour deposition; CZTS, copper zinc tin sulfide; DC, direct current; DSSC, dye-sensitised solar cells; EPDM, ethylene-propylene-diene monomer; ETL, electron transport layer; FLG, few layer graphene; FOM, Figure of Merit; FTO, fluorine tin oxide; GO, graphene oxide; hBN, hexagonal boron nitride; HFTCVD, hot filament thermal chemical vapour deposition; HJSC, heterojunction solar cells; HOPG, highly ordered pyrolytic graphite; HTL, hole transport layer; IPA, isopropyl alcohol; ITO, indium tin oxide; LB, Langmuir-Blodgett; LbL, layer-by-layer; LPCVD, low-pressure chemical vapour deposition; LT-PECVD, low-temperature plasma enhanced chemical vapour deposition; MIBK, methyl isobutyl ketone; MLG, multilayer graphene; MNWT, multi-walled nanotubes; OPV, polymer solar cells; OSC, organic solar cells; PAni, polyaniline; PCE, power conversion efficiency; PDMS, polydimethylsiloxane; PEDOT:PSS, poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate); PEI, polyethyleneimine; PET, polyethylene terephthalate; PMMA, polymethylmethacrylate; PSC, perovskite solar cells; PVA, polyvinyl alcohol; P3HT, poly(3,4-ethylenedioxythiophene); QD, quantum dots; rGO, reduced graphene oxide; SAM, self-assembled monolayers; SLG, single layer graphene; SWNT, single-walled nanotubes; TCF, transparent conducting films; TCL, transparent conducting layer; TCNQ, tetracyanoquinodimethane; TCO, transparent conducting oxides; TE, transparent electrode; TEM, transmission electron microscopy; TFSA, trifluoromethanesulfonyl-amide; TFSC, thin film solar cells; 0D, zero-dimensional; 2D, two-dimensional; 3D, three-dimensional; 6P, para-hexapheny

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Nomenclature			
Ar	argon	$\sigma_{DC,B}$	bulk DC conductivity
KMnO ₄	potassium permanganate	n	percolation exponent
CH ₃ NH ₃	methylammonium	t_{min}	critical thickness
Co	cobalt	R	reflectance
Cu	copper	ϵ_0	permittivity of free space
C ₁₀ H ₁₆ O	botanical derivative camphor	c	speed of light in a vacuum
HAuCl ₄	tetrachloroauric acid	n	refractive index of the film
HI	hydriodic acid	m^*	effective mass of the conduction electrons
H ₂	hydrogen	μ	mobility
H ₂ SO ₄	sulphuric acid	λ	visible wavelength of light
NaNO ₃	sodium nitrate	e	electronic charge
Ni	nickel	σ_{DC}	DC conductivity
SOCl ₂	thionyl chloride	σ_{OP}	optical conductivity
R_s	sheet resistance	n_{sub}	refractive index of the substrate
T	optical transmittance	Z_o	impedance in free space
σ	conductivity	ϵ_c	critical strain
ρ	resistivity	G_{sh}	sheet conductance
t	film thickness	A	absorption
α	optical absorption	K	coefficient of proportionality
		V_o	open circuit voltage
		J_{sc}	short circuit current density

developing TCF include low-cost materials, low-temperature processing methods for lower fabrication costs, and flexibility for flexible applications. Numerous materials for TCF alternatives have been studied, including transparent conducting oxides (TCO) [1], conductive polymer [2], metal nanowires [3–5], carbon nanotubes [6] and also graphene.

The semiconductor material of TCO was extensively applied as TCF owing to its properties of electrical conductivity ($\sim 10^{-4} \Omega \text{ cm}$) and high transmittance over the visible light range ($> 90\%$). Various metal oxides have been reported, including binary and multicomponent TCO as well as improvements to the TCO by proposing multilayers of TCO/metal/TCO [7,8]. Reviews on the status of TCO have been described in detail by Minami et al. [9,10], Stadler [11] and Ellmer [12]. Among TCO, aluminium zinc oxide (AZO) is known to have comparable performance to ITO and FTO including other advantages of less toxicity, inexpensive cost and simple fabrication methods. However, grain boundaries and electron scattering have reduced the carrier mobility of TCO, consequently affecting the performance of these materials.

Metal nanostructures consisting of nanowires, nanogrid, nanofibres etc. are also excellent candidate materials for TCF due to their exceptional properties including high conductivity and being highly transparent in visible wavelengths. The conductivity of these materials is dependent on the percolation network, of which the high aspect ratio of longer and thinner nanomaterial is preferable in order to obtain the best performance of TCF with low sheet resistance and high transmittance [13]. In addition, the mechanical flexibility of these materials makes them relevant for flexible applications. However, the shortcomings of metal nanostructures, i.e. the high junction resistance and poor adhesion to the substrate, require further improvement to these materials.

Carbon nanotubes (CNT) are one-dimensional carbon-based materials with high conductivity and high transparency, applicable as a substitute for ITO TCF. CNT have high mobility despite having much lower carrier concentration compared to ITO, i.e. $\sim 10^{17}/\text{cm}^3$ and $10^{20}/\text{cm}^3$, respectively. The properties of CNT differ according to the wall numbers such as single-walled nanotubes (SWNT) and multi-walled nanotubes (MNWT). Previously, CNT were applied as transparent electrodes for organic solar cells, showing a comparable performance to the solar cell with ITO as a transparent electrode [6,14]. However, the high sheet resistance of CNT compared to ITO is still the main concern.

Recently, graphene has been studied due to its exceptional properties such as high carrier mobility of $\sim 200,000 \text{ cm}^2/\text{Vs}$, high transparency in visible light and near infrared, and its being highly flexible.

Due to these excellent properties, graphene has been applied in various applications such as solar cells, sensors, flexible electronics and others. In application, graphene may be applied as charge carrier or as a transparent conductive electrode.

As graphene has been the centre of attention recently, many studies are conducted on the synthesis and application of graphene; as well as numerous reviews on graphene [15–17]. This review aims to give an update to the research progress in graphene, which includes graphene synthesis, the method to improve the performance of graphene as well as the performance of photovoltaic applications incorporating graphene-based transparent conducting films. We also include the performance evaluation of graphene-based TCF by using the Figure of Merit (FOM).

2. TCF characteristics properties and performance evaluation

2.1. Fundamentals of electrical and optical properties

The best way to describe the property of a material is by using the conductivity, σ (S/cm) and resistivity, ρ ($\Omega \text{ cm}$); the conductivity is a measurement of the ability for material to conduct electricity, whereas the resistivity is a measurement of the ability of material to oppose electricity flow. For a thin film, its resistance is measured by the sheet resistance, i.e. a resistance in regard to the thickness. The sheet resistance can be described by

$$R_s = \frac{\rho}{t} = \frac{1}{\sigma t} \quad (1)$$

where t is the film thickness (nm), $\rho = 1/\sigma$ is the resistivity and σ is the direct current (DC) conductivity. High conductivities (i.e. lower resistivity) can be obtained with a thicker film. However, the transparency of the thicker film may be affected due to an increase in the optical absorption of the film, as given by

$$T = e^{-\alpha t} \quad (2)$$

where α is the optical absorption [18]. The sheet resistance of bulk film may be obtained by combining Eqs. (1) and (2) to obtain

$$T = e^{-\alpha/\sigma_{DC,B} R_s} \quad (3)$$

where $\sigma_{DC,B}$ is the bulk DC conductivity.

The sheet resistance of nanostructure materials described by De et al. [19] is given by

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