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## Recent development of graphene materials applied in polymer solar cell



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#### ABSTRACT

Graphene, a rapidly rising star on the horizon of material science, has a unique two-dimensional nanostructure as well as exceptional mechanical, optical and electronic properties. Despite its short history, graphene has exhibited great potential applications in counter electrodes, acceptor and hole transporting layers of polymer solar cells. This astonishing achievement is the subject of this article, which reviews the developments in counter electrodes, hole transporting layers and acceptor of polymer solar cells based on graphene materials in the past five years. Polymer solar cells evolve in an exponential manner in the three key areas of power conversion efficiency, stability and flexibility. In this review, we summarized briefly the effect of functional graphene sheets on power conversion efficiency and stability, and also pointed out the prospective development trends of functional graphene sheets to be applied in polymer solar cells in the future.

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#### 1. Introduction

Currently, silicon solar cells cover more than 85% of the market of photovoltaics. However, silicon cells technology is still not cheap enough to allow a wide diffusion of photovoltaic energy conversion in the absence of government incentives [1]. For this reason huge

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http://dx.doi.org/10.1016/j.rser.2014.11.040 1364-0321/© 2014 Elsevier Ltd. All rights reserved. efforts of research and development have been spent in the past years to find alternative and improved solutions in this field. PSCs represent a new technology that in the midlong term could lead to affordable energy [2]. In addition to this, PSCs (meaning of the abbreviations was shown in Table 1.) are lightweight and can be made flexible, opening the possibility for a range of new applications. And large-area PSCs can be fabricated easily and inexpensively by employing cost-effective techniques, for instance, inkjet, screen printing, slot-die, gravure and spray coating [3].

The PCE, stability and flexibility have played a key role in the potential application of PSCs [4]. Many research activities have been carried out to improve PCE, stability and flexibility, such as optimizing chemical structure and preparation conditions, and so on [2-10]. In normal geometry, the layer stack was usually built on top of an ITO electrode followed by a hole transporting layer PEDOT:PSS, the active laver (PBDTTT and PCBM), an electron transport layer (e.g., LiF) and finally a low work function metal electrode (e.g., Al). And then the high PCE (ca. 7.7%) has been obtained [3]. However, the PCE of PSCs was difficult to be further improved and the flexible PSCs were difficult to be prepared with these materials. In addition to this, the PSCs exhibited instability [4]. These results were attributed to the following reasons. Firstly, ITO was not compatible with large scale manufacturing methods in contrast with today's market requirements for a flexible electronics and it failed under bending [11]. Furthermore, device issues liked susceptible ion diffusion into the polymer layers and mechanical brittleness limited the applicability of ITO in flexible PSCs [11]. Secondly, PEDOT:PSS had several problems including high acidity, hygroscopic properties and inhomogeneous electrical properties, resulting in poor long-term stability [12]. Therefore, a substitute for ITO and PEDOT:PSS with a similar performance but lower cost was clearly needed.

In recent years, graphene materials have been applied widely in PSC technology due to high transparency, conductivity, chemical stabilization and flexibility [13]. Pioneering experiments about graphene materials as transparent electrodes, acceptor and hole transporting layers were done by Loh et al., Chen et al. and Chhowalla et al., respectively [14–16]. And then, a number of works have reported the effects of graphene materials on PCE, stability and flexibility of PSCs [15–25]. But the PCE ( < 3.6%) of PSCs based on graphene materials was far lower than the value (ca. 8.5%) of traditional PSCs in these works [14–25].

Despite all the studies done so far in this field, and the graphenebased functional materials for organic solar cells have been reviewed [26,27], up to now only very few and isolated facts were available on how graphene materials affect the performance of PSCs, i.e., how to work the mechanism might and the PCE was far lower than the value of 8.5% as discussion above. Several points have contributed to these lackings. Firstly, the high *T* and Rs of graphene thin film were difficult to be obtained according to the previous works [17–25]. Secondly, the donor polymeric structure of active layer was few discussions to improve PCE and stability of PSCs based on graphene electrodes, hole transporting layers and acceptor. Thirdly, the effect of graphene on the performance of PSC was a relatively new physical process first described about 5 years ago. So it was quite usual that most of the work done so far was devoted to the development and optimization of the effect than to a deeper understanding of the mechanism of the physical process [15-27].

The scope of this paper was to discuss some mechanistic aspects of the physical process between the properties of

The list of abbreviations in the paper.

graphene materials and the performance of PSCs, which were plausible and in line with earlier and new findings. The proposed discussion will give an explanation for the improvement of PCE, stability and flexibility of PSCs. Although some parts of the explanation were reported earlier [21,22], here the whole explanation will be introduced to a broader community for the first time. It may be a help for further discussions and for the development of better PSCs based on graphene materials.

## 2. Preparation of graphene counter electrodes, acceptor and hole transporting layers of PSCs

#### 2.1. Graphene counter electrodes of PSCs

The graphene counter electrodes of PSCs were prepared by the Hummers method as shown in Scheme 1. Firstly, the GO solution







**Scheme 2.** Preparation process of graphene counter electrode applied in PSCs by the CVD method.

Word	Abbreviation	Word	Abbreviation
Polymer solar cells	PSCs	Power conversion efficiency	PCE
poly(3,4-ethyleneuloxythiophene).poly(styrene sunonate)	FEDOI.F33	diyl-alt-4-substituted-thieno[3,4-b]thiophene-2,6-diyl]	FDDIII
Semitransparent indium tin oxide	ITO	Phenyl-C61-butyric acid methyl ester	PCBM
Graphene oxide	GO	Poly[3-(potassium-6-hexanoate) thiophene-2, 5-diyl]	P3KT
Poly(3-hexylthiophene)	P3HT	Poly(3-octylthiophene)	P3OT
Fullerene	C <sub>60</sub>	Chemical vapor deposition	CVD
Polyethylene terephthalate	PET	Polymethyl methacrylate	PMMA
Lowest unoccupied molecular orbital	LUMO	Highest occupied molecular orbital	HOMO
Sodium dodecyl sulfate	SDS	1, 2-dichlorobenzene	DCB
Optical transparency	Т	Sheet resistance	R <sub>s</sub>
Pyrene buanoic acid succidymidyl ester	PBASE	Electrical conductivity	σ

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