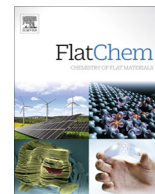




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## Recent advances in the application of two-dimensional materials as charge transport layers in organic and perovskite solar cells

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### ABSTRACT

Two-dimensional (2D) materials such as graphene and transition metal chalcogenides (MX<sub>2</sub>, M: transition metal, X: S, Se, Te) have emerged as a new class of materials due to their high carrier mobility, high transparency, tunable band gap, low cost, and solution-processable properties. These materials can be fabricated into single layers or few layers through facile processes such as chemical vapor deposition or mechanical exfoliation to unlock their superior electrical and optical properties. The ability to tune the work function enables their application as hole transport layers and electron transport layers in optoelectronic devices. In this review, we focus on recent progress in the application of 2D materials as hole transport layers and electron transport layers in organic solar cells and perovskite solar cells.

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**Abbreviations:** 2D, Two dimension; OSC, Organic solar cell; PSC, Perovskite solar cell; HTL, Hole transport layer; ETL, Electron transport layer; PCE, Power conversion efficiency; J<sub>sc</sub>, Current density; V<sub>oc</sub>, Open circuit voltage; PEDOT, PSS, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; P3HT, Poly(3-hexylthiophene-2,5-diyl); PBDDTPD, Poly[[5-(2-ethylhexyl)-5,6-dihydro-4,6-dioxo-4H-thieno[3,4-c]pyrrole-1,3-diyl][4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl]]; PCDTBT, Poly[N-9'-heptadecanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)]; PTB7, Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]]; PCBM, [6,6]-Phenyl C61 butyric acid methyl ester; SAM, Self-assembly material; ITO, Indium tin oxide; GO, Graphene oxide; rGO, Reduced graphene oxide; pr-GO, p-TosNHNH<sub>2</sub> reduced graphene oxide; SWCN, Single wall carbon nanotube; KMnO<sub>4</sub>, Potassium permanganate; K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>, Potassium persulfate; P<sub>2</sub>O<sub>5</sub>, Phosphorus pentoxide; H<sub>2</sub>O<sub>2</sub>, Hydrogen peroxide; TCNQ, Tetracyanoquinodimethane; VO<sub>x</sub>, Vanadium oxide; MoO<sub>x</sub>, Molybdenum oxide; e-MoO<sub>3</sub>, evaporated molybdenum oxide; TMD, transition metal dichalcogenide; MoS<sub>2</sub>, Molybdenum disulfide; WS<sub>2</sub>, Tungsten disulfide; TaS<sub>2</sub>, Tantalum disulfide; NbSe<sub>2</sub>, Niobium diselenide; Bi<sub>2</sub>Se<sub>3</sub>, Bismuth selenide; Al, Aluminum; Ag, Silver; LiF, Lithium fluoride; Ca, Calcium; ZnO, Zinc oxide; Ar, Argon; H<sub>2</sub>, Hydrogen; Cs, Cesium; Li, Lithium; TiO<sub>x</sub>, Titanium oxide; TFT, Thin film transistor; SiO<sub>2</sub>, Silicon dioxide; FTIR, Fourier transform infrared spectroscopy; AFM, Atomic force microscopy; SEM, Scanning electron microscopy; TEM, Transmission electron microscopy.

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## Introduction

In recent years, the need for renewable and sustainable energy resources has dramatically increased due to depleting resources of fossil fuels such as coal and oil. Among methods that produce clean and sustainable energy such as wind energy, hydraulic energy, and bio energy, turning solar energy to electric energy is most promising due to the unlimited energy provided by the Sun. Specifically, Si-based solar cells have been developed and applied in practice since 1970 [1]. However, the expensive fabrication method of Si-based solar cells has prohibited their wide usage. Thus, the replacement of Si-based solar cells with other types including dye-sensitized solar cells, organic solar cells (OSCs), quantum dot solar cells, and perovskite solar cells (PSCs) has been intensively investigated [2–5]. Emerging as a new class of solar cells, OSC and PSC have received tremendous attention due to their high power conversion efficiency, flexibility, and possibility of low-cost fabrication via a roll-to-roll process [6–8]. The power conversion efficiency (PCE) has been reported to be over 10% for OSCs and over 20% for PSCs [9–11]. PSCs have been predicted to beat commercial Si-based solar cells in the market due to their ease of fabrication and extremely high PCE, which significantly reduces the production cost.

Advancements in OSCs and PSCs are still underway through investigations on the active layer, electrodes, and interlayers, to boost their efficiency and to realize the stability needed for commercial use. One of the strategies to improve device performance is the insertion of interlayers between the active layers and electrodes to facilitate charge collection [12]. Generally, high-work-function polymers and metal oxide have been utilized as hole transport layers (HTLs) [13–15], whereas low-work-function polymers and metal complexes have been employed as electron trans-

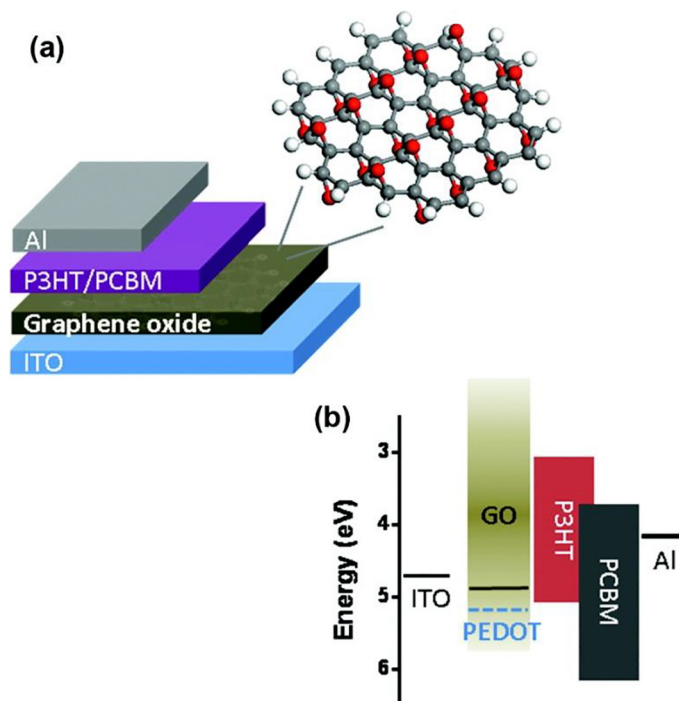
port layers (ETL) [16,17]. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is widely used as an HTL, but it has several problems including high acidity of suspension ( $\text{pH} \sim 1$ ), hygroscopic properties, and inhomogeneous electrical properties, resulting in poor long-term stability [18–20]. In addition, the metal oxide compounds for HTL or ETL are normally fabricated via low vacuum deposition, which increases the fabrication cost [14,15]. Therefore, abundant and solution-processable materials with low cost and high stability for HTL and ETL in OPVs and PSCs need to be identified.

Since a single layer of graphene was successfully fabricated by Novoselov et al. in 2004, the electrical and optical properties of graphene have been significantly enhanced [21,22]. The two-dimensional (2D) structures of transition metal dichalcogenides have also been explored, opening a universe of applications for 2D materials in the field of optoelectronic devices [23–25]. It is reported that the work functions of 2D materials are tunable through functionalization or specific treatment methods [26–28]. Further, 2D materials can be fabricated easily via solution-processable techniques [29,30]. Thus, 2D materials are highly promising alternatives to the traditional HTL and ETL materials in OSCs and PSCs. The recent progress in applying 2D materials to OPVs and PSCs are summarized and discussed in this review.

## The application of 2D materials as buffer layers in organic solar cells

### Graphene oxide (GO) and its derivatives

GO was used as an HTL in 2010 by Li et al. GO showed PCE comparable to that of PEDOT:PSS-based OSCs, demonstrating its potential in the field of optoelectronic devices, as shown in Figs. 1 and 2



**Fig. 1.** (a) Schematic of the photovoltaic device structure consisting of the following: ITO/GO/P3HT:PCBM/Al. (b) Energy level diagrams of the bottom electrode (ITO); interlayer materials (PEDOT:PSS, GO), P3HT (donor), and PCBM (acceptor); and the top electrode Al. Adapted with permission from Ref. [31].

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