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⁵ 2D organic semiconductors, the future of green nanotechnology

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ARTICLE INFO	ABSTRACT	
Keywords: 2D organic semiconductor Green nanotechnology OTFTs OLEDs Photo-diodes Organic solar cell Optical wave guide	The discovery of 2D organic semiconductors of atomically thin structures has attracted great attention due to their emerging optical, electronic, optoelectronic and mechatronic properties. Recent progress in such organic nano- structures has opened new opportunities for engineering material properties in many ways, such as, 0D/1D/2D nanoparticles hybridization, strain engineering, atomic doping etc. Moreover, 2D organic nanostructures exhibit a unique feature of bio–functionality and are highly sensitive to bio-analytes. Such peculiar behavior in 2D organics can be utilized to design highly-efficient bio-sensors. Also, a bio-molecular integrated electronic/optoelectronic device with enhanced performance can be attained. Furthermore, the bio-degradable, biocompatible, bio- metabolizable, non-toxic behaviour and natural origin of organic nanomaterials can address the current ecolog- ical concerns of increasing inorganic material based electronic waste. This review highlights the benefits of 2D organic semiconductors. Considering the importance of strategic techniques for growing thin 2D organic layers, this review summarizes progress towards this direction. The possible challenges for long-time stability and future research directions in 2D organic nano electronics/optoelectronics are also discussed. We believe that this review article provides immense research interests in organic 2D nanotechnology for exploiting green technologies in the future.	

1. Introduction

The discovery of organic semiconductors has opened up new opportunities for low cost, bendable and green electronics. Nature has preserved an infinite variety of organic materials, and these materials have much better scopes for easy manufacture, shaping and tuning of materials properties compared to inorganic materials [1–3]. π – π stacking in organic materials reveals good conductivity of charge carriers [1,2]. Towards optical performance, organic nano/sub-micro structures have shown near unity photoluminescence quantum yield [3]. Hence, organic materials are emerging in electronic, optoelectronic and photonic device applications. However, since the introduction of organic materials, research progress using these materials was very slow; even though many believed that organic materials will be the ultimate alternative for conventional inorganic materials. But after the demonstration of a two-layer 2D organic thin film based photovoltaic cell and electroluminescent diodes by Tang et al. in the mid 1980's [4,5], research activities towards organic semiconductors were dramatically tuned in rapid progress. Since then, numerous useful applications including organic thin film transistors

(OTFTs), organic light emitting devices (OLEDs), organic solar cells, organic lasers, bio/chemical sensors and memories have been fabricated [6-10].

Observation of fascinating characteristics of 2D inorganic nanomaterials [11-16], but also, realization of certain limitations with them have hugely increased attention to studying 2D structure of organic materials [17-19]. In particular, 2D nanomaterials are ultrathin sheet of single crystals with atomic or molecular level thickness. Due to large surface area to volume ratio and decreased dielectric screening in ultrathin 2D feature, they can show a unique physical, chemical, mechanical, electrical, optical, optoelectronic, magnetic properties those cannot be achieved by bulk form of materials. The atomic-level thickness is generally smaller than most particles' transport mean free path, hence, particles follow ballistic transportation rather than scattering or diffusion [20,21]. This quantum confinement effect fundamentally changes the electronic behavior of these materials. Also, excellent optical transparency and a fast response have made them ideal for both fundamental study and novel electronic applications. Towards the 2D organic materials, the capability of designing and tuning structures greatly enriches

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Fig. 1. Structural differentiation of a large family of organic molecules; (a) small molecules (Rubrene, Pentacene, Alq3 etc.) and (b) polymers (Poly(3-hexylthiophene), Poly(3-hexylthiophene-2,5-diyl) etc.).

2D organic nanomaterials and endows them with promising electronic/optoelectronic properties. They have excellent flexibility, molecular diversity, bendability and solution-processability of large area synthesis [22,23]. Hence, they can hugely boost up numerous new materials into 2D nanomaterials family. 2D organics can effectively address the challenges of high cost issues, and the ecological concerns of electronic waste constrained in 2D inorganics. Also, 2D organics possess interesting layer-dependent properties, and AFM imaging can clearly illustrate layers numbers; however, it has to be more explored. Recently, layer dependent carrier transport properties in thin 2D organics [24,25], and organic layer dependent exciton pumping across organic-inorganic interface have been exhibited [26]. Hence, combining the advantages of 2D architecture of organic materials is of great importance for the development of next-generation electronics and optoelectronics.

Organic materials are generally categorized into two groups; i.e., small molecules such as pentacene, rubrene, Alq3, nucleic acid, etc., and polymers such as poly(3-hexylthiophene), polypropylene, polystyrene, proteins, etc. Small molecules are low molecular weight organic materials (<900 unit mass), while polymers consist of high molecular weight species that are made up of many repeat unit monomers. Fig. 1 shows the structural differentiation of small molecules and polymers. Fig. 1a demonstrates the molecular structures of small molecules; rubrene, pentacene, Alq3; whilst, the organic polymer poly(3-hexylthiophene) and poly(3-hexylthiophene-2,5-diyl) are shown in Fig. 1b. Towards 2D organic architecture, there are several synthesis methods available, which are also discussed in following sections. This review will firstly overview the current synthesis protocols of thin 2D organic materials. Then, it will highlight the importance of 2D organic thin films for incipient nanotechnological applications and their ecological concerns. Finally, this review will attempt to discuss challenges and the future prospects of 2D organic semiconductors in various fields such as OTFTS, OLEDs, Solar cells, photo detectors, chemical/bio sensors and optical

wave guides.

2. Synthesis protocols for 2D organic thin films

Fabrication of large area and uniform film 2D organic single crystals require some particular techniques. This chapter demonstrates some specific methods as shown in Fig. 2 which have been employed to synthesize thin and good crystalline 2D organic single crystals. Current synthesis protocols have achieved film thickness' from few molecular layers down to a single molecular layer.

2.1. Floating-coffee-ring driven assembly

The phenomenon coffee-ring is named for the characteristic of ringlike deposit along the perimeter of a spill of coffee [27], and the mechanism behind the formation of similar rings from any liquid is commonly known as the coffee ring effect. The coffee-ring pattern generally originates from the capillary flow of liquid induced by the differential evaporation rates across the drop, and the process initiates with the outward flow of dispersed liquid towards the edge of the substrate [28]. This coffee-ring method is widely employed to prepare thick organic film.

Towards the synthesis of ultrathin 2D organic crystal, a little modified, floating coffee-ring driven method has been introduced as a very effective method. The key characteristic of this technique is the mixing of organic molecules in a mixture of good solvent and antisolvent where the antisolvent with a relatively higher boiling point is selected [29,30]. Then, the mixture is drop casted onto the substrate and a mechanical pump generates a continuing airflow on the top of the droplet. This airflow drags the droplet to diffuse quickly across the substrate. Near the edge of droplets, the evaporation of the solvent occurs which leads to a floating-coffee-ring effect on the antisolvent. The high evaporation rate of a good solvent near the edge of a droplet results in the thin coating of

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Fig. 2. Various synthesis protocols for growing thin 2D organic layers.

organic layers over the substrate. Using single solvents with low boiling points, such as chloroform, toluene, *etc.*, these drop casted organic molecule solutions commonly form micro thickness films, and organic molecules are barely coated on the substrate when using high boiling point solvents such as chlorobenzene, anisole *etc.* Hence, the floating coffee ring method is an important technique to obtain ultra-thin and uniform 2D organic crystals. The mixing ratio of solvent and antisolvent is also crucial in this method; synthesis of large area mono and bi layers of dioctylbenzothienobenzothiophene (C8-BTBT) have been reported using 0.2 and 0.5 wt% of immiscible p -anisaldehyde and ~98.5 wt% of miscible anisole [31]. However, this method is still limited to make it applicable to many soluble organic materials, and also enabling the fabrication hetero bilayers or super lattice structures of 2D organic single crystals.

2.2. Layer-by-Layer deposition

Layer-by-Layer (LbL) deposition is also an effective technique for the fabrication of ultrathin organic molecular layers or tens of nanometre thick organic films. The usual concept of LBL is building of one layer below ~ 1 nm at a time. LbL is a pervasive method for coating substrates with organic molecules, it offers superior control and versatility compared to other thin film deposition techniques. Interestingly, LbL methods can be applied to both planar and particulate substrates. LbL assembly uses diffusion-driven kinetics to promote adsorption onto the substrate. This method is simply performed by immersing the planner substrate in a chemical solution, followed by rinsing steps to wash off the unbound material [32]. While for particulate substrates, more steps are involved and it requires dispersing the substrates in a chemical solution followed by pelleting using centrifugation for the washing steps [33,34]. In principle, LbL films are kinetically trapped structures due to the assembly methods, and can be post-treated to shrink, swell or reconfigure using stimuli such as pH [35], heat [36], mechanical forces [37], competitive binding of salts [38], etc.

Towards the synthesis ultrathin of 2D organic crystal, this method initially requires charged substrate and organic molecules consisting of opposite charges to be deposited via merging the substrate into an organic molecular solution [39]. Piranha solution, RCA treatment and oxygen plasma treatments are some easy and common techniques to

charge the substrates [40,41]. Immersion of the charged substrate into an organic solution of opposite charges (anionic or cationic), results in an organic monolayer on the substrate [39]. Since most ionic group of organic molecules remain exposed to the interface with the solution and surface charge is reversed, subsequent immersion of charged substrate in the solution containing the opposite ionic molecules and rinsing it with distilled water at each step can be utilized to obtain 2D organic hetero bi-layers or thicker organic films. This method has been used to deposit several layers of partially doped polyaniline, sulfonated polystyrenes on a substrate [42]. Most recently, an ultrafast LbL method was utilized to make a 58 layer assembly of divalent triazolinedione and a trivalent diene molecule [43]. This method is often useful to generate 2D organic molecular heterostructure or superlattice of structures consisting of opposite/alternate charges of each. Instead of the solution dipping LBL method, a spin-coating-assisted LbL method was developed to speed up the multilayer assembly [44]. However, this method is not an efficient method due to inefficient use of adsorbate solutions on the substrate.

2.3. Vapor deposition method

For a long time, the vapor deposition method has been employed for the fabrication of organic thin films. This method can be categorized into two primary classes; i) physical vapor deposition (PVD), and ii) chemical vapor deposition (CVD) techniques. Usually, thin films of insoluble small organic molecules are fabricated using these proficiencies.

PVD is fundamentally a vaporization-coating technique which transfers the resulting vapor phase of a material onto the target substrate via condensation. The most common PVD operations are thermal evaporation and sputtering. Thermal evaporation involves heating a material in a vacuum chamber until the atoms have enough energy to be released. After being vaporized, the atoms are channeled through a vacuum chamber to coat a target substrate. This deposition technique is likewise suitable for fabricating hetero-organic layers. It can produce extremely pure and high quality films. This method is most commonly used to generate inorganic nano films. Recently, many research groups have demonstrated this technique to fabricate pentacene monolayers [45,46].

In a CVD process, the substrate is exposed to one or more volatile precursors, which react and/or decompose on the substrate surface to produce the desired materials thin film deposit. The major difference

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between PVD and CVD is the process of depositing the source material onto the substrate. In a CVD process, the substrate is put into a vacuum chamber and a source material is put either inside the same chamber or in a neighboring chamber. Then, the source material is either heated or the atmospheric pressure is decreased until the source material vaporizes. Also, one or more precursors can selectively be used to react with the source material allowing for it to be deposited on the substrate. The vaporized material then reacts on the substrate to generate a uniform thin film. Modifying the temperature and time duration of the sequence helps to manage the thickness of the film. Polymer based thin films such as poly(tetrafluoroethylene) and fluoropolymer thin film have been demonstrated with this technique [47]. However, this technique has been more widely used for 2D inorganic compounds such as transition metal dichalcogenides, monochalcogenides and graphene [48,49].

2.4. Spin-coating

Spin coating is an easy and simple technique for the preparation of uniform thin solid film on flat substrates from sol-gel samples. In this process, solid particles dissolved in a solvent are spin-coated on the substrate surface using a spin coater. Subsequent removal of the solvent due to centrifugal forces solidifies the gel, resulting in a solid film. A solution of material is dispensed onto the centre of a wafer, which is then rotated at high speed. Rotation continues until the excess solution spins off the substrate and the desired thickness of the film is left on the substrate. The applied solvent is usually volatile and evaporates during deposition. The thickness of spin coated film is greatly affected by the spin speed and the viscosity of the solution; and some other parameters such as spin time, solution density, solvent, evaporation rate and surface wettability also pay role. This technique has also been employed for growing organic thin films on desired solid substrates.

A typical spin-coating process involves depositing an organic molecule fluid onto the center of a substrate and then spinning the substrate at high speed up to a few thousand revolutions per minute (rpm) [40,41]. The parameters chosen for the spin process strongly affect the thickness of the film. Slight variations in the parameters can result in drastic variations in the coated film thickness. The centrifugal acceleration results in spreading organic molecules on the substrate and develops a thin film of organic molecules on the surface of the substrate. In this process, organic film thickness also depends on the nature of the solvent used, especially viscosity, drying rate and surface tension of the solvent. Spin coated nano-films of poly-methyl-methacrylate (PMMA), 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8-BTBT) are the most commonly reported nano-films fabricated using this procedure [50,51].

2.5. Self-assembly

Self-assembly literally represents a phenomenon where the components of a system assemble themselves to form a larger functional unit. This is a promising strategy for creating organic nanostructure films with surface functionalization in a well-controlled manner. Hence, this method is suitable to fabricate 2D organic films with tailored properties and additional surface functionality to be used as sensing devices. A molecular self-assembly technique is a spontaneous association of molecules under equilibrium conditions into steady, structurally welldefined aggregates adhered by non-covalent bonds [52]. The subtle interplay between molecule-molecule and molecule-substrate interactions plays the role in developing molecular building blocks on the substrate [3]. Strong hydrogen bonding and π - π stacking between organic molecules are exploited to create self-assembled two-dimensional structures [53]. This technique is suitable to construct desired functional nano-architectures with the synthesis of molecular building blocks of predetermined intermolecular binding properties. There are basically two types of self-assembly; i) intra molecular and ii) inter molecular. Commonly, the term molecular self-assembly refers to intermolecular

self-assembly, while the intra molecular analogue is more commonly called folding. Self-assembled nanofilms of various molecules such as perylene-3,4,9,10-tetracarboxylic dianhydride (PTCDA), alkanethiols, $CH_3(CH_2)_{n-1}$ *etc.* have been demonstrated [54,55], and recently, borophene/PTCDA lateral heterostructure has also been synthesized with using this technique [56].

2.6. Langmuir-Blodgett technique

The Langmuir-Blodgett (LB) is also an effective technique to develop ultra-thin organic layers, monolayer organic molecules can be synthesized from this technique. Langmuir-Blodgett films are named after Irving Langmuir and Katharine B. Blodgett, who invented this technique [57]. In this technique, nano thickness of amphiphilic organic films are fabricated by immersing the solid substrate into a liquid. Initially, a selected organic material is dissolved in a volatile and water insoluble solvent such as chloroform, benzene etc. After that minute amount of the solution is spread onto air-water interface using a micro syringe. Water insoluble amphiphilic molecules form floating monolayer at air-water interface. And, an organic monolayer is adsorbed homogeneously with each immersion step, and after the evaporation of solvent almost one molecule thick film can be formed [58]. The monolaver is assembled vertically and is composed of amphiphilic molecules with a hydrophilic head and a hydrophobic tail. Usually, long chain fatty acid, lipid molecules etc are compatible molecules with this techniques. This technique is one of the most promising techniques for preparing organic thin films with precise control over monolayer thickness and homogeneous deposition of a monolayer over large areas. This technique also provides opportunities to make multilayer structures with varying layer composition. Till now, LB films of various organic polymer such as poly(vinylidene fluoride trifluoroethylene), naphthalene-tetracarboxylic-dianhydride (NTCDA), polypyrrole polythiophene, poly (3-hexylthiophene) polymer, etc, with their suitable applications have been demonstrated [59-61].

2.7. Patterned growth

Organic crystal film growth based on patterning is crucial for the demonstration of high-performance organic nano devices. This approach can be effective for introducing metallic electrodes with significantly less gate leakage current [62]. The widely applied top-down photolithog-raphy technique in 2D inorganic semiconducting films is not compatible with organic crystals. Organic molecules are vulnerable to high temperature and using multiple stages of organic solvent cleaning during this process can harshly degrade organic molecules. Hence, a different type of bottom-up patterning and growing of 2D organic molecules film into a designed ordered structure can be an effective approach for fabricating future organic semiconducting nanodevices.

Currently, templet assisted and substrate assisted bottom up approaches have been introduced to grow organic film with good contact metallic patterning. The template-assisted method utilizes a particular template to control the pattern and growth of organic crystal films. The template serves as a mold to have particular shapes and organic molecules are guided for self-assembly into crystal patterns according to confined geometries using a template. Elastomeric polydimethylsiloxane (PDMS), is commonly used as the template material [63,64]. While in substrate assisted patterning, the substrate wettability concept is utilized to locate and pattern organic crystal films. A substrate surface functionalized with phenyl groups exhibits a wetting property to facilitate the growth of organic materials. Usually, substrate surface modified alkyl or fluoroalkyl groups develop a de-wetting property with poor affinity to organic solvents. Combining these strategies is suitable for designing micropatterned organic single crystal thin films at desired locations [65]. The solution containing organic semiconductor, 3,9-bis(4-ethylphenyl)-peri-xanthenoxanthene (C2Ph-PXX) has been shown to spread out uniformly over pre-defined areas with a coating applicator [65].

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3. Advantages of 2D organic semiconductor

3.1. Eco-friendly materials

Nowadays, the electronic market is highly competitive, wellsophisticated and novel electronic device progress rapidly with the expansion of science and engineering. Such progress in electronics has not only benefited society but has also put a great challenge of proper disposal of electronics at their end-of-life. The improper disposal of such electronic wastes results in severe environmental impacts because they are usually toxic, flammable, corrosive, highly reactive, infectious or radioactive [66]. Such wastes also contribute to the formation of greenhouse gases, such gases are responsible for global warming and climate changes. Also, possible chlorination or bromination effects from many devices can cause the depletion of ozone layers which allows hazardous solar ultraviolet radiation to pass towards Earth's surface. Landfill and incineration are the main methods for the disposal of electronics [67]. For long-term sustainability, a waste management system should primarily consider the reduction, reuse, recycle, recovery of energy and reclamation of land [68]. Waste disposal issues with future nanotechnology markets can bring severe concerns. Nanostructures can easily enter the human body and cross various biological barriers. So far, nanoparticles with sizes less than 10 nm (more likely said as quantum dots) act similar to a gas and can enter human tissues easily during disposal or manufacturing processes. Usually, many inorganic nanostructures are found to be bio-compatible, however, they are not bio-degradable. In contrast, organic materials are biodegradable and most of them are also non-toxic [66]. Landfill disposing of organic electronics at their end-of-life will seriously address the concerns of electronic waste, greenhouse gas effects and ozone depletion. Hence, the development of organic electronics can sustain environmental ecology much better than their inorganic counterparts.

3.2. Flexible devices

Flexible electronic devices are considered to be bendable, stretchable and unbreakable. They can be wrapped around a matchstick thousands of times without damage. This type of electronic offers high potential for innovative products by rendering electronic hardware compatible with nonplanar surfaces. Flexible electronics are most suited for their potential in wearable electronics. Currently, available commercial electronics are silicon-based which are breakable during bending and stretching. Incorporating unbreakable plastic can also be a unique choice for safer use. Therefore, the bendable, stretchable and unbreakable nature of organic semiconductors can revolutionize the scope of flexible electronics for future robust devices in many applications. Furthermore, the development of large scale, low cost flexible organic electronic devices and their roll-to-roll production processes will allow their market implementation in numerous applications such as displays, photovoltaics, chemical sensors, exotic devices ete [69].

3.3. Bio molecular integration electronics

Organic semiconductors are uniquely suited for bio-integrated electronics as they have functionalities accessible for biomolecules. The soft nature of organic materials offers excellent mechanical compatibility with many biological tissues. The ability of organic semiconductors to conduct ions in addition to electrons and holes also opens up a new communication channel with biology; for example, an enzymatic reaction changes the current flowing through a polymer transistor [70]. Also, an organic electronic device can trigger a biochemical reaction [71]. Furthermore, nature has developed a tool kit of biomaterials that is capable for signature recognition; such as, enzymes that metabolize substrates, oligonucleotides that hybridize with complementary strands and antigens that pair with antibodies [70]. Organic nanodevices, such as OFETs can be very suitable platforms for the recognition of such

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biological molecular coupling due to their bio-compatible nature and highly sensitive electronic response. Such biomolecular recognition can be grounded on the optical performance of 2D organic films as they have high PL yield and highly sensitive potential with doping and de-doping, according to the adsorption and desorption of biomolecules. Moreover, bio-molecular integration in organic electronics can be helpful in enhancing device performance significantly due to probable doping or possible application of strain from bio-molecules.

3.4. Property tuning: surface functionalization, doping, heterostructures

Π-conjugation structural driving electrical conductivity of organic nanostructure depends on the concentration and mobility of charge carriers. Intermolecular spacing of neighboring molecules and the electronic structure of the conjugated system are important for electrical conductivity [72]. Other quantities such as ionization potential, electron affinity, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels difference of such a conjugated system are more likely to control the injection of holes and/or electrons in the material [73]. Higher surface to volume ratio in 2D organic materials and hydrophilicity nature provide higher chances of surface hybridization with many semiconducting and plasmonic nanostructures. Such hybridization can manipulate the two dimensionally confined exciton complexes, such as excitons, trions, biexcitons or higher order excitons in 2D organic semiconducting crystals [45,74,75]. Probable electron/hole transfer through the interphase or plasmonic nano particles inducing locally enhanced electric fields are responsible for such tailoring of 2D organic material characteristics. Fig. 3(a-c) shows the huge decrease in photoluminescence (PL) for mono and few-layer pentacene when it is hybridized with 1L-MoSe₂. Similarly, a huge tailoring in the transfer curve for the anti-ambipolar nature in a CuPc-MoS₂ (2L-MoS₂) heterostructure has also been experimentally demonstrated as shown in Fig. 3 (d, e). As observed in many 2D transition metal dichalcogenides strain inducing band gap engineering [76-78] effects can be more significant in 2D organic thin films due to their flexibility and bendability.

4. Applications of 2D organic nanotechnology

2D organic semiconductors demonstrate a solid potential for various nanotechnological applications. They have very strong opto-electromechanical performance, bendability, bio-functionality and echo friendly nature; in addition to cost effectiveness and enormous availability, which has attracted research interests and commercial applications. Fig. 4 displays the most potential applications of 2D organic semiconductors; particularly in OTFTs, OLEDs, photodetectors, optical waveguides, chemical/bio sensors and solar cells.

4.1. Electronics and optoelectronics applications

4.1.1. OTFTs

OTFTs are three-terminal devices with consisting of a source, drain and gate electrodes, and the channel conductivity can be modulated under electrical fields generated by the gate bias. Particularly, OTFTs are switching devices, and for an ideal p-type or n-type OTFTs, the off state is realized when $V_{GS} = 0$ V. At zero gate bias in OTFTs, nearly flat energy bands are realized, so channel resistance is dominated by their intrinsic conductivity, where most of the charges are trapped at the band tail. Hence, a very high channel resistance is occurred at zero gate bias. But in the on state, the amount of V_{GS} increases until the channel conductivity reaches saturation. In p-type OTFTs, negative gate bias induces band bending of both dielectric insulator as well as the organic semiconductors in the active layer, while in n-type OTFTs, positive gate bias causes band bending. At the dielectric/organic semiconductor interface, the HOMO or LUMO of the organic semiconductor moves toward the bulk fermi level which result in increase of carrier density exponentially in the

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Fig. 3. An advantageous property of 2D organic semiconducting materials; their hybridization with 2D inorganic semiconductors can significantly tailor the intrinsic optical, electronic and optoelectronic properties. (a) Schematic diagram of the heterostructure of monolayer MoSe₂ and 2D pentacene (PEN). (b) Optical images showing before and after formation of heterostructure, on where wetting layer (WL), one-layer (1L), and two-layer (2L) pentacene regions are in upper panel and their heterostructure with 1L-MoSe₂ is in lower panel. Scale bar: 10 μ m. (c) (I) PL spectra of 1L MoSe₂ + WL PEN heterostructure, 1L MoSe₂, and WL PEN and (II) PL spectra from 1L MoSe₂ + 1L PEN heterostructure, 1L MoSe₂, and 1L PEN. (d) Schematic of p-n 2D organic/inorganic (CuPc-MoS₂) heterostructure device with its electrical connection. (e) Comparison of transfer curves for CuPc-MoS₂ heterostructure, CuPc only and MoS₂ only. This shows the tailored diode behaviour, anti-ambipolar nature in CuPc-MoS₂. Images a-c adopted with permission from Wiley-VCH verlag [45], and Images d,e adopted with permission from Royal Society of Chemistry [75].



Fig. 4. Wide scopes of 2D organic semiconductors for emerging nanotechnological device applications.

channel region. The source-drain channel current is orders of magnitude larger than it is in the off-state. However, when the channel thickness is down to a few nanometers, or even to a monolayer, the carriers are confined within that layer thickness, and can achieve uniform gate modulation [79]. In a 2D semiconductor with the layer thickness d_{sc}, a

constant carrier density is expected because of the confinement effect [79]. By adopting the Vissenberg–Matters model [80], the bulk (3D) channel drain–source current of OFET at high gate bias was estimated as

$$I_D^{3D} \alpha (V_{th} - V_G)^{2T0/T-1}$$

where $V_{\rm th}$ is threshold voltage, V_G is gate bias, and T is temperature. However, it shows weaker temperature dependence for 2D material, and drain-source current be

$$I_{D}^{2D} \alpha (V_{th} - V_{G})^{2T0/T-1}$$

Various studies have shown that the 2D carrier density distribution is valid rather than the 3D distribution for various top gate configuration polymer FETs even if the polymer film thickness is 60–100 nm [81].

2D OTFTs have been extensively investigated due to their peculiar advantages as explained earlier. Various polycyclic aromatic compounds such as oligoarenes, oligoacenes, tetrathiafulvalenes, and polycyclic aromatic hydrocarbons are qualified as good candidates for OTFTs [75]. However, current results in OTFTs have shown relatively slow carrier mobility and this was found to decrease at elevated temperature. The increase in lattice vibrations and phonon scattering are responsible for the decrease in carrier mobility at higher temperatures [82,83]. Zhang et al. observed the mobility of a 1.58 nm pentacene film (two layers thickness) grown on hexagonal-BN to be 3.0 cm²V⁻¹s⁻¹ at room temperature [84]. Until this date, He et al. demonstrated the highest field effect mobility of 30 cm²V⁻¹s⁻¹ in a C₈- -BTBT monolayer grown on a h-BN substrate [85]. Based on recent progress, carrier mobility of several organic thin film devices are also listed in Table 1.

2D organic electronic devices with much improved carrier transport mobility are in high demands. Such electronic devices of high mobility, high on-to-off current ratios, and high stability can be achieved from the

Growth method

Self assembly

Spin coating

Self assembly

Self assembly

Dip coating

Self assembly

Self assembly

Self assembly

PVD

Langmuir-Blodgett

Dual solution-shearing (DSS)

PVD

PVD

CVD

Organic materials

HNPNA-1, HNPNA-2

Pentacene

C₆-DBTDT

C₁₀-DNTT

C₆-DPA

HBPs

HTEB

POBOC8

C_o-BTBT

BTBT-C12-PA

Glycol-C60-C6-PA

CuPC

C60

C_oBTBT

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Comparison of electrical performance of various 2D organic crystals.

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Thickness (nm)

1.64, 3.22

8.26 and 2.7

Ref.

[84]

[86]

[87]

[88]

[89]

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Carrier types

p-type

p-type

p-type

n-type

p-type

n-type

P-type

P-type

P-type

P-type

n-type

p-type

p-type

p-type

6	7
6	8
6	9
7	0
7	1
7	2
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7	6
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improvement of materials synthesis. Residue free, high crystalline, uniform and ultrathin 2D organic crystals can improve the OTFTs performance. Accurate chemical tailoring of organic molecules can be a strategic way to enhance field effect mobility in OTFTs. Chemically modified thiophenes with enhanced carrier transport have been demonstrated [97]. Mild doping in such 2D organic semiconductors can also boost conductivity. Luo et al. reported enhanced charge carrier mobility in DPP-thieno[3,2-b]thiophene–conjugated polymer by 24 times by incorporating the ionic additive, tetramethylammonium iodide [98]. High performing OTFTs can be employed to pave the way for next-generation electronics with energy-efficient security systems, sensors, photonics, and spintronic memories.

4.1.2. OLEDs

2D OLEDs are the most promising scope of organic technologies. OLEDs are highly energy-efficient, eco-friendly lighting source and are the best alternative to existing lighting technology. Significant effort has also been made in both academia and industry to investigate π -conjugated molecules for their unique electrical or opto-electrical properties over the last three decades. Both organic small molecules and polymer thin films have shown highly performing OLED applications [86,90,99]. An OLED consists of semiconducting organic layers sandwiched between two electrodes where light is produced by the process of electroluminescence through the injecting electrons and holes in the emissive layer. So, efficient injection of electrons and holes result in better performance for OLEDs. Usually electrodes with a lower work function compared to the active organic layer are used for better injection of electrons, and higher work function electrodes are used for better hole injection. In OLEDs, light escaping from the OLED is also an important concern. So, one of the electrodes used in OLEDs must be transparent. In this regard, high work function ITO has been a widely used electrode which is transparent to visible light and also promotes injection of holes into the emissive laver.

OLEDs works on the singlet and triplet exciton radiation mechanisms as shown in Fig. 5a. Picking triplet excitons is one of the successful ways to fabricate an efficient OLED. Usually, phosphorescent materials produce light from both of singlet and triplet excitons, have found about four times more efficient than fluorescent materials [100,101]. Also, a larger device thickness gives rise to a larger impediment against carrier injection in, which results in increased turn-on voltage and luminous intensity and power efficiency are reduced. But using of thinner 2D enables high electric field even at low applied voltage [102], which may increase the carrier density, improves the carrier mobility and electroluminescence output as shown in Fig. 5b. Zhu et al. has reported a warm white OLED (CIExy value: (0.41, 0.44)), an efficacy of 63.2 lm W⁻¹ by employing an ultra-thin layer of bis(2-phenylbenzothiozolato- N,C20)iridium(acetylacetonate) (Ir(bt)2(acac) between two 5 nm layers of the 20 wt% FIrpic doped 2,6-bis- (3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy) host [103].

Nowadays, OLEDs have become a mainstream display technology. Also, great advances in improving the internal quantum efficiency of an OLED have been reported to nearly 100%, which was realized using phosphorescent and thermally activated delayed fluorescent emitters [104,105]. Fig. 5(c-e) show the characteristics of high-performance OLEDs with nearly 100% exciton on CBP-BP-PXZ-based intrinsic OLED device. But, the external quantum efficiency of OLEDs is still lacking, which has been estimated to be 20%–25%, due to a large amount of light trapped inside the device [106]. Hence, recovering the optical losses and improving the external efficiency is an important concern to be addressed for future devices. Also materials and device design are essential for improving external efficiency of OLEDs [107,108].

Mobility $(cm^2V^{-1}s^{-1})$

1.6 and 3

1.84 and 0.23

0.01

10

29

2.8

10.4

0.02

4.0

0.076

 10^{-4}

1.0

0.13

30

4.1.3. Photo-diodes

Substrate

h-BN

SiO₂

h-BN

h-BN

SiO₂

SiO₂

SiO

AlO₃

SiO₂

SiO

AlO.

SiO

 SiO_2

h-BN

A photodiode is a semiconductor device which converts light into an electrical current. In contrast to OLEDs, organic photodiodes (OPDs) utilize the organic semiconductor to absorb incident light and convert it to an electric current. In a photodiode, a built-in electric field is formed via a p–n junction or a Schottky junction between metal and semiconductors. The incoming photon excites semiconducting layers, and generates electrons and holes which moves to opposite contact electrodes driven by that built-in potential. Their photo response speed is determined by the transit time of excess charge carriers defined by

$t_{transit} = L^2 / \mu V_{bi},$

where L is the channel length, μ is the carrier mobility, and V_{bi} is the built-in potential across junction [109].

Similar to OLEDs, the ability to fabricate OPDs with low cost easily paves the way for mass production and has attracted great attention for commercial applications. The flexibility and large-area processability of the OPD in particular have opened the scope in X-ray digital imaging applications [110]. Color image sensing technology can also be revolutionized with the development of OPDs. In particular, currently used commercial photographic technique based on metal-oxide semiconductors [111,112], camera phones have already been significantly improved. However, more sensitive and efficient camera applications will be in large demand in medical, scientific, automotive and industrial applications in the coming years. The emphasis will be focused on making detectors smaller, packed with more analog-to-digital functions with a wider range of optical formats, as well as having different dynamic range specifications and signal-to-noise ratios, a lower power consumption, wireless communication, the ability to interact with a variety of interfaces and full color detection [110,113].

In spite of the good electrical properties of inorganic semiconductor based photodiodes, they have a number of limitations that can restrict their application [113,114]. OPDs can overcome many of the limitations in traditional inorganic materials. Using an ultrathin organic active layer can yield high level photon detectivity [1]. Observation of a photo responsivity 0.37 mA W^{-1} in perylene-3,4,9,10-tetracarboxylic G.P. Neupane et al.

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Fig. 5. Schematic illustration of working principle and experimental achievement of high-performance OLEDs with nearly 100% exciton. (a) Singlet and triplet excitons radiation phenomena in OLEDs. (b) Charge distribution in thin device layer architecture. (c) Luminance–voltage–current density and (b) current efficiency–luminance–power efficiency curves of non-doped OLEDs. Inset in plane (d): EL spectra at luminance of 5000 cdm⁻². (e) Relative luminance–time curves of CBP-BP-PXZ-based non-doped OLED device at different initial luminance. Image a adopted with the permission from Royal Society of Chemistry [101], and c-e adopted with permission from Wiley-VCH Verlag [105].



Fig. 6. Organic thin film photo-transistor. (a) Schematic illustration of a typical phototransistor based on dioctylbenzothienobenzothiophene (C_8 -BTBT) thin film/ graphene FET. (b) Transfer characteristics of the graphene device, before (black) and after (red) deposition of C_8 -BTBT. Photo response of C_8 -BTBT/graphene phototransistors and the evolution with increasing C_8 -BTBT thickness. (c) The dynamic photocurrent response of the devices under the same experimental conditions: laser power density: 7000 µW cm⁻², $V_{ds} = 0.1$ V, $V_g - V_o = 10$ V. The top panel shows the photocurrent response of pure graphene device without C_8 -BTBT. (d) EQE (red triangles) and IQE (blue squares) as a function of average number of C_8 -BTBT layers for all the measured devices (laser power, 100 µW cm⁻²). The red dashed line reflects the linear increase of EQE. (e) Photoconductive gain (Gph) as a function of average number of C_8 -BTBT layers for all the measured devices for all the measured devices (laser power, 100 µW cm⁻²). The red dashed line reflects the exponential increase of Gph. Images adopted with permission from Wiley-VCH Verlag [116].

dianhydride (PTCDA) monolayer/C₈-BTBT bilayer p-n diode [115], motivated for more detail study. Liu et al. demonstrated a C₈-BTBT/graphene phototransistor with a photo responsivity of 1.57×10^4 A W⁻¹, a photoconductive gain larger than 10^8 , an external quantum efficiency (EQE) on the order of 10^4 , and a response time of 25 ms with a monolayer thickness of C₈-BTBT as shown in Fig. 6 [116].

4.2. Mechatronics applications

2D organic semiconductors have great potential to be used in nano electro-mechanical system (NEMS). They can be an effective alternative to currently existing Si-based micro electro-mechanical systems (MEMS). MEMS technology is more often used for sensing of bio or chemical molecules. MEMS can recognize the target analyte according to a mechanical deflection of active surface caused by an analyte. The mismatch in the surface stress between the top and bottom surfaces of free standing active layers due to the binding of the target analyte causes the deflection of active layers. This deflection is monitored to recognize the analyte. Compared to Si and/or 2D inorganic based MEMS devices, ultrathin organic active layer based NEMS devices can be more effective for bio/ chemical sensing due to their soft mechanical properties. 2D organic NEMS allows a much larger mechanical deflection compared to inorganic ones and enhanced sensitivity can be achieved.



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Towards organic NEMS, these are usually categorized into piezoresistive, capacitive and piezoelectric types [78–81]. Piezoresistive NEMS offers great mechanical stability [117]. Capacitive NEMS are relatively easy to fabricate and highly scalable [118]. Piezoelectric NEMS are highly sensitive strain sensors [119]. In this regard, Seena et al., reported a standard OTFT integrated epoxy resin SU-8 based micro-cantilever which showed the maximum relative variations of drain current and mobility over 250% for 0.1% of applied strain [120]. Hence, 2D organic NEMS can be effectively used in bio/chemical sensing. In which a mild analyte causing sensible strain can also be converted into a detectable electrical signal. 2D NEMS devices based on polyvinylidene fluoride (PVDF) and its derivative; copolymer poly(vinylidene fluoride/trifluoroethylene) P(VDF-TrFE) have also been demonstrated to be suitable for flexible ferroelectric memory devices [121,122].

For the commercial use of 2D organic NEMS, mechanical durability is an important concern. Fukuda et al. studied the durability of p-type and n-type OTFTs under tensile strain. The on-current of the device was decreased with the increase of tensile strain and it was found to be reproducible even with a bending radius of 4 mm [123]. Fig. 7 demonstrates the transfer characteristics of the p-type semiconducting pentacene based NEMS device and n-type semiconducting benzobis (thiadiazole) (BBT) derivative with trifluoromethylphenyl groups (FPTBBT) NEMS with a channel length of 10 mm before, during, and

> Fig. 7. Mechatronic performance of OTFT devices with printed electrodes. (a) Optical microscope image of a OTFT and magnified image for channel region. (b) Transfer characteristics of the p-type semiconducting pentacene based OTFT device with channel length of 10 mm before, during, and after application of 1.6% tensile strain. (c) Saturation source-drain current (IDS) monitored at VDS = VGS =-20 V as a function of measurement number. IDS is normalized from its initial value (without application of strain). (d) Transfer characteristics of the n-type semiconducting benzobis (thiadiazole) (BBT) derivative with trifluoromethylphenyl groups (FPTBBT). (e) Saturation source-drain current (IDS) monitored at VDS = VGS = +20 V as a function of measurement number. OTFT device with channel length of 10 mm before, during, and after application of 1.6% tensile strain. Images adopted with permission from Nature publishing group [123].

after application of 1.6% tensile strain.

4.3. Others applications

4.3.1. Solar cell

As a renewable, free source of energy and also a non-polluting source of energy, solar energy is being of very important energy. Based on the photoelectric effect, an electrical device solar cell converts the solar energy into DC electricity. Since the 1950s; the introduction of the first crystalline silicon solar cell, the efficiency has reached up to 24% for crystalline Si-solar cells, and a theoretically predicted upper limit of 30% [124]. However, Si-based solar cells have major obstacles for market implementation due to their large production costs [125]. Despite a great deal of effort to reduce costs, large scale production of Si-based solar cells will also be limited by the availability of raw materials, such as solar-grade Si. Therefore, to ensure a sustainable technology path for solar cells, the development of new materials and device structures is of utmost priority. In this regard, organic materials can be a promising alternative compared to their inorganic counterparts due to their low-cost, easy fabrication and large-area compatibility with a flexible substrate. An organic solar cell uses conductive organic polymers or small molecules for light absorption and charge transport to produce electricity from sunlight via the photovoltaic effect. To this date, large research progress on organic solar cells has been reported [124-132]. In an organic solar cell device, the charge carriers have to pass through the organic/electrode interfaces. Therefore, the electronic structures of the molecules are an important factor that affects the overall efficiency of the cell.

Considering the importance of organic solar cell operation, trace amounts of water and oxygen can degrade the crystallinity of the active layer and electrode materials. Hence, this type of solar cell must be protected from humidity. Even with proper protection there are several degradation processes that need to be eliminated to ensure stability. Apart from device integrity, the materials must be photochemically stable and the nanoscale uniformity of donor-acceptor blend in the active layer must be preserved. One of the crucial parameters for increasing the photocurrent of organic solar cells is the absorption of more photons. This can be achieved by increasing the layer thickness and by shifting the absorption spectrum of the active layer to longer wavelengths. But the layer thickness is presently limited by the charge carrier mobility and lifetime. When the mobility is too low or the layer is too thick, the transit time of photo-generated charges in the device becomes longer than the lifetime, resulting in charge recombination. The use of polymers such as P3HT that are known to have high charge carrier mobilities allows an increase in film thickness from the usual ~100 nm to well above 500 nm, without a loss of current [128,129]. Thus in the future, there is a probability that modifying the organic semiconductor material structure and varying their thickness in device preparation, can lead to further improvement in the performance of organic photovoltaic devices. In this regard, the tandem cell strategy can be an effective way to simultaneously address these issues [130-132]. The use of tandem cells can overcome the thickness constraint of single-junction cells due to the low mobility of organic materials because wide and efficient absorption can be achieved by stacking the active layers with complementary absorption in tandem cells. Meng et al. presented a 17.29% power conversion efficiency for a two-terminal tandem organic cell [124]. This experimental strategy was guided by a semiempirical model analysis using the tandem cell strategy to overcome such issues, and taking advantage of the high diversity and easily tunable band structure of organic materials. The detailed optical simulation model is shown in Fig. 8, which shows a maximum predicted efficiency value of 17.36%.

4.3.2. Optical wave guide

Optical wave guiding at the nanometre scale has attracted serious attention in nanophotonics. Such materials characteristics in the nanostructural regime are very important for minimizing photonic circuits and optical communications. Researchers have applied great effort into optical waveguide technologies based on 1D inorganic nanostructures [133–136], 1D organic nanostructures based on polymers [138] and small organic molecules [138–141]. But optical waveguides based on 2D



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Fig. 8. Optical simulation and photovoltaic performance of the tandem solar cells. (a) Device architecture of the tandem cell. (b) Energy level diagram of the tandem solar cell. (c) Simulated current density generated in a tandem cell as a function of the thicknesses of the active layers. (d) J-V curve. (e) EQE and 1-reflectance (1-R) of the optimized tandem solar cell and (f) J-V curve of the tandem cells under different light intensities, ranging from 4.97 to 112.68 mW/cm2. The dashed vertical line at 720 nm in (e) is the cross point of the EQE plots of the two sub cells, and the dashed vertical line at 985 nm indicates the effective absorption upposition (assuming 50% EQE) of the rear sub cell. Images adopted with permission from the American Association for the Advancement of Science (AAAS) [124].

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Fig. 9. Optical wave guiding in CN-TSDB micro plate. (a) Color CCD images of CN-TSDB microplates showing PL waveguides at various excitation positions. (b) Optical microscopy image of a microplate with the moving directions of an input laser emitted signals by yellow arrows. (c) LCM PL intensity versus propagation distance along the [A], [B], and [C] directions. (d-f) The LCM Raman intensity of the (d) C-Br mode (667 and 687 cm-1), (e) -CF3 mode (1216 cm-1), and (f) –C=C– aromatic (1580 and 1594 cm⁻¹) stretching modes versus propagation distance along the [A], [B], and [C] directions. Images adopted with permission from Royal Society of Chemistry [143].

structures are still in their infancy. In particular, 2D organic nano structures can be more suitable for the application of controlling directional waveguides due to their anisotropic nature. Recently, Heng et al. demonstrated photoluminescence (PL) guiding along the abscissa in hexaphenylsiloles (HPSs) microtiles [142]. Jo et al. further demonstrated multi-directional guiding of PL and Raman signals in a 1,4-bis(3,5-bis(trifluoromethyl) cvanostyryl)-2,5-dibromobenzene (CN-TSDB) microplate single crystal structures [143]. Fig. 9 demonstrates PL and Raman signal wave guiding in a CN-TSDB micro plate, which exhibits the propagation of both PL centered at 450 nm and Raman signals (various modes: C-Br mode at 667 and 687 cm^{-1} , $-\text{CF}_3$ mode at 1216 cm^{-1} , and -C=C- aromatic stretching modes at 1580 and 1594 cm⁻¹) up to $\sim 20 \,\mu m$ distance along the [A], [B], and [C] directions. PL signal propagation was clearly visualized in color CCD imaging (Fig. 9a). For the efficient propagation of PL signal over long distances, the proper input excitation wavelength should be selected such that the PL emission can have efficient overlap with the absorption of the materials. In contrast, a Raman signal waveguide can uniquely provide signal transport using specicmolecular interaction modes such as vibrational, rotational and other low-frequency modes in highly crystalline organic nano- or micro-structures [144,145]. In particular, Raman waveguides are very important for transporting multiple signals that correspond to various distinguishable molecular orientations and vibrations which suggests that significantly more information can be transmitted using Raman waveguiding. Extending the optical communication from the PL and Raman signals wave guides is an interesting subject for future research. Hence, future investigations into optical signal wave guiding through ultra-thin 2D organic materials may revolutionize currently existing 1D-inorganic nano-photonic technologies.

5. Conclusion and outlook

[A]

[B]

[C]

[A]

[B]

[C]

Promising optical, electrical and optoelectronics characteristics of thin film organic semiconductor hugely opened their scopes for use in OTFTS, OLEDs, OPDs, solar cells and optical wave guiding technologies. Moreover, peculiar properties of 2D organic structure such as lightweight, low cost to fabricate and ease of production, eco-friendly nature etc have enormously extended their scopes into many areas of 2D nano devices formerly occupied by inorganic 2D nanomaterials. Organic materials are uniquely suited to produce sustainable, bendable, and biodegradable ultrathin electronics. Good accessibility of bio/chemicalfunctionalities and material softness, 2D organics promise high potential for use in bio/chemical sensing applications. However, like to inorganic 2D materials, 2D organics materials and nanodevices are not muchstudied yet. Recently, synthesis of ultra-thin large area 2D organic materials are rapid in progress, which will direct to explore more detail fundamental characteristics of single molecular layer thickness and layer numbers thickness dependent behavior. Detail understanding of quantum confinement effect in 2D organic materials in molecular thickness will pave the way to design high performing suitable nanodevice. Same to in 2D inorganic nanomaterials, various engineering techniques such as nano structural hybridization, bio-molecules functionalization, applying strain or chemical treatments can hugely manipulate their intrinsic characteristics and in particular, device performances can be significantly enhanced. Despite the emerging potential with 2D organic technologies, they have shown low resistance to thermal and poor conductivity, which partially limit their use and need to be addressed for future technology. However, insightful fundamental study of ultrathin 2D organics nanomaterials has not well-studied, which is highly

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demanded to be explored. Also, degradation of organic nanomaterials with environmental stresses is a very serious concern to be addressed. Research progress towards 2D organics nanomaterials has been continuously building up a foundation that will undoubtedly offer emerging techniques to resolve the limitations too. In conclusion, 2D organics nanomaterials will herald an emerging green technological nextgeneration electronics base with high energy efficiency, inexpensive, and flexible nano devices.

Q3 Uncited reference

[137].

Declaration of competing interest

The authors herein declare no conflicts of interest.

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