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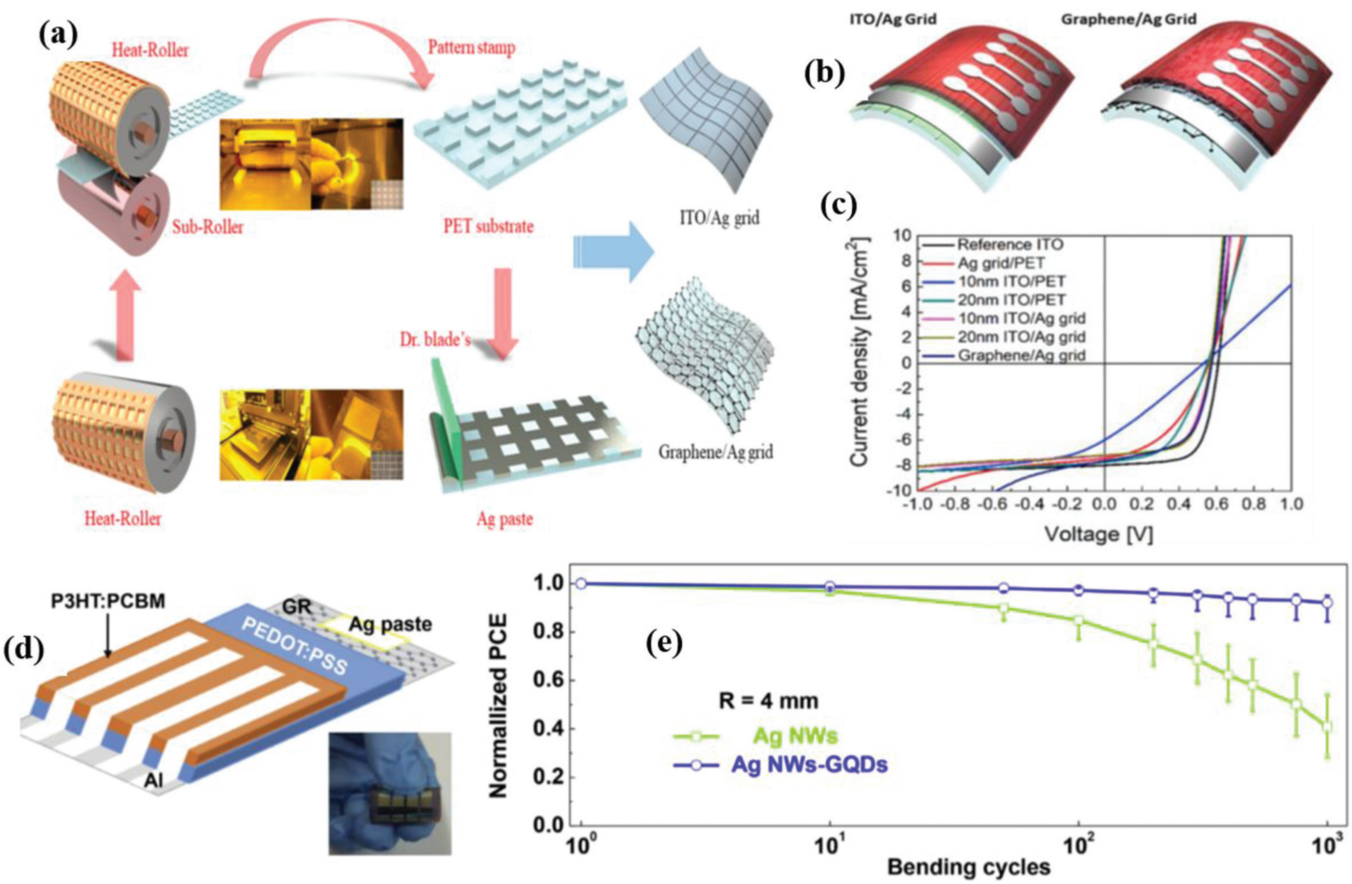

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MATERIALS ENGINEERING | REVIEW ARTICLE

Nanomaterials in 2-dimensions for flexible solar cell applications – a review

Benjamin Agyei-Tuffour[1,2,3]*, Kwadwo Mensah-Darkwa[4], Daniel Nframah Ampong[4], Elizabeth Adzo Addae[1], Gerald Selasie Gbadam[1], Clarisa Naa Shormeh Darko[1], Afia Owusua Akyaw[1], John Adjah[1], Joseph Asare[5], Guixiang Li[2] and Neill J. Goosen[3]

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Abstract: This review presents the progress, challenges and prospects of ultrathin flexible photovoltaic devices based on 2-dimensional (2D) nanomaterials. These devices have shown very high performance in bending stabilities for up to ~90% of their power conversion efficiencies (PCEs) after multiple bending deformations. They are thin film PVs with lightweight and mechanically robust structures that allow use in the continual advancing solar cell applications. In this paper, comprehensive assessments of 2D nanomaterials, their syntheses methods, performance, degradation, mechanical and opto-electronic characterization in flexible photovoltaic (PV) cells are highlighted. Semi-conductor materials such as conjugated donor and acceptor polymers, small donor/acceptor molecules and organometal halide perovskites for use as active layers in such flexible solar cell structures are reviewed. The challenges and prospects associated with the adoption of 2D nanomaterials in flexible solar cells are presented. The review highlights the need to transition laboratory results on 2D nanomaterials based flexible solar cells into scale up and

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PUBLIC INTEREST STATEMENT

The quest for solar powered wearable electronic devices for health, textiles, sports event monitoring, etc., has seen increased interest in two-dimensional (2-D) nanomaterials in recent times. This has led to numerous experimental techniques and copious results published in different journals. This review article in a summarized form presents the advantages, breakthroughs, limitations, current developments and future applications of 2-D nanomaterials in different flexible solar cells using inorganic, organic and perovskite materials. The article shows examples of 2-D nanomaterials, their syntheses techniques and the role they play in the flexible device. Graphene being the common 2-D nanomaterial is highlighted for its unique role in the performance of flexible oganic solar cells, flexible dye-sensitized solar cells and flexible perovskite solar cells. The versatility of 2-D nanomaterials has been highlighted with its limitations and would give the public a complete overview of nanomaterials in 2-dimensions in the flexible optoelectronic devices industry.

commercialized products despite the existing and also opens research areas for researchers to explore and achieve robust and high-efficient solar devices.

Subjects: Renewable Energy; Materials Science; Coatings & Thin Films-Materials Science; Nanoscience & Nanotechnology; Electronic Devices & Materials

Keywords: 2-D nanomaterials; syntheses techniques; device stability; flexible solar cells; power conversion efficiency

1. Introduction

Solar cell research has been a hot topic for decades because it is at the heart of the solar energy to electricity conversion system (Hecht, 2021). Solar energy, being the most abundant energy source on the planet, is seen as the most cost-effective substitute to fossil fuels, which have caused major pollution and climate change effects. Solar cells thus have been considered as the optimum device to this huge volume of solar energy to electricity. Solar cell technology has been classified into three generations. Solar cells made of monocrystalline silicon and polycrystalline silicon are "first-generation solar cells", and they account for the majority of the photovoltaic (PV) devices on the market (International Renewable Energy Agency, (IRENA, 2019)), National Renewable Energy Laboratory (NREL)). Thin film solar cells, such as copper indium gallium selenide (CIGS), copper indium diselenide (CIS), cadmium sulphide (CdS), cadmium telluride (CdTe), amorphous silicon solar cells, etc., are among the "second-generation solar cells". All other revolutionary PV technologies, such as perovskite solar cells (PSCs), dye sensitized solar cells (DSSCs), quantum dot sensitized solar cells (QDSCs), polymer solar cells, are classified as third-generation solar cells (Wu et al., 20192004; Nassiri Nazif et al., 2021; Koo et al., 2020). The rising global demand for flexible, portable and wearable solar powered gadgets has necessitated the development of flexible, lightweight, and efficient power production resources. Two-dimensional nanomaterials therefore play crucial roles in the fabrication and performance of flexible solar cells. The ultrathin 2D nanomaterials have over the past decade recorded increased research progress. Since the discovery of graphene by Novoselov et al., 2004, 2013 using the micromechanical exfoliation method, several other 2D nanomaterials such as conducting polymers, carbon nanotubes, metal nanowires and transition metal dichalcogenides (TMDs) have been investigated in quest for low-cost, mechanical stability and high-performing flexible transparent conducting electrodes (TCEs). These 2D nanomaterials have shown distinctly large surface area and tunable chemical properties compared with their bulk versions. The absence of an interlayer in their single layer nanosheets is responsible for their excellent electronic properties. Most importantly is the atomic thickness and in-plane covalent bonding which gives them high flexibility and optical transparency to enable their use in the energy industry. The unique physical and chemical properties of 2D nanomaterials render them suitable for applications in flexible solar cells and in other areas including electronics, optoelectronics, sensors, catalyses, biomedicine and biotechnology (Hou et al., 2018; C. Li et al., 2019; Y. Wang et al., 2022).

Graphene, a popular and actively studied 2D nanomaterial, is reported to be extra thin, low weight, has excellent optical, mechanical, electrical and conductivity properties (Novoselov et al., 2004). Extensive research has leveraged on its electronic properties and morphology in various applications including flexible solar cells. In comparing with graphene, 2D conducting polymers including polyaniline (PANI), Polypyrrole (PPy) and poly-(3,4-ethylenedioxythiophene) (PEDOT) have atomic/monomer units of ultrathin layer characteristics with spatially confined framework as a result of in-plane interactions than out-of-plane. 2D conducting polymers have self-supporting integrity, superior topological planarity, conformational crystallinity (periodicity), etc. This improved in-plane periodicity and minimal defects in 2D nanosheet-like conducting polymers are advantageous in the manipulation of charge carriers when adopted in optoelectronic devices (Barpuzary et al., 2019). Carbon nanotubes (CNT) are seamless cylinder-like fullerene structures of rolled up graphite sheets. They have high yield strength, high aspect ratio (l/d > 1000), high stability at increased currents, superior electron transport and high Youngs modulus. There are two main types of CNTs: the single walled and the multi-walled CNTs. The SWCNT has only one layer of graphene sheet rolled from edge to edge into a seamless cylindrical tube with ~1 nm diameter and ~ 1 mm length. The MWCNT has a concentric tube of graphene sheets with varying diameters and the interplanar distances between the concentric tubes range from ~ 0.32–0.35 nm. Due to improved adhesion energies between MWCNTs and other substrates compared with the SWCNTs, the former is often adopted in optoelectronic applications. The MWCNT has more active sites on the edges that can easily interact and adhere to substrates in various devices, (Bichoutskaia et al., 2008; Maheswaran & Shanmugavel, 2022). A 2D transition metal dichalcogenide (TMDC) is material with three atomic planes in which a transition metal atom is sandwiched between two planes of selenium, Sulphur or tellurium atoms (commonly called chalcogens). The electrons are confined in one-dimension and free to move in two-dimensions. 2D TMDCs have excellent optical absorption coefficients, narrow and varying band gaps and self-passivated surfaces which make them excellent solar cell materials. In the ultrathin state of 2D TMDC, they have an enhanced broadband and omnidirectional absorption in the visible spectrum. They therefore find applications in single, double junction and tandem solar cells, (Critchey, 2022). Flexible solar cells (FSCs) have thus been hailed as a potential PV technology due to inherent benefits such as lightness and bendability, which make them ideal for installation and integration with architectural and wearable electricity-generating devices. Figure Figure 1 and Table 1 present the current efficiencies of the best solar cells produced from different materials (National Renewable Energy Laboratory (NREL), Hasan et al., 2021). For instance, flexible perovskite solar cells (FPSCs) have made significant developments with ~26.1% and 29.5% power conversion efficiencies in perovskite and perovskite-tandem cells (National Renewable Energy Laboratory (NREL)). This PV technology is compatible with the roll-to-roll production due to the improved photoelectronic characteristics of the halide perovskite absorber and the device ease of fabrications. Organic solar cells (OSCs) and dye-sensitized solar cells (DSSCs) have achieved high mechanical flexibility, as well as their high

Figure 1. Solar Cell Efficiencies compiled from 1976 to 2020. Adapted from National Renewable Energy Laboratory (Maronchuk et al., 2019, NREL 2020).

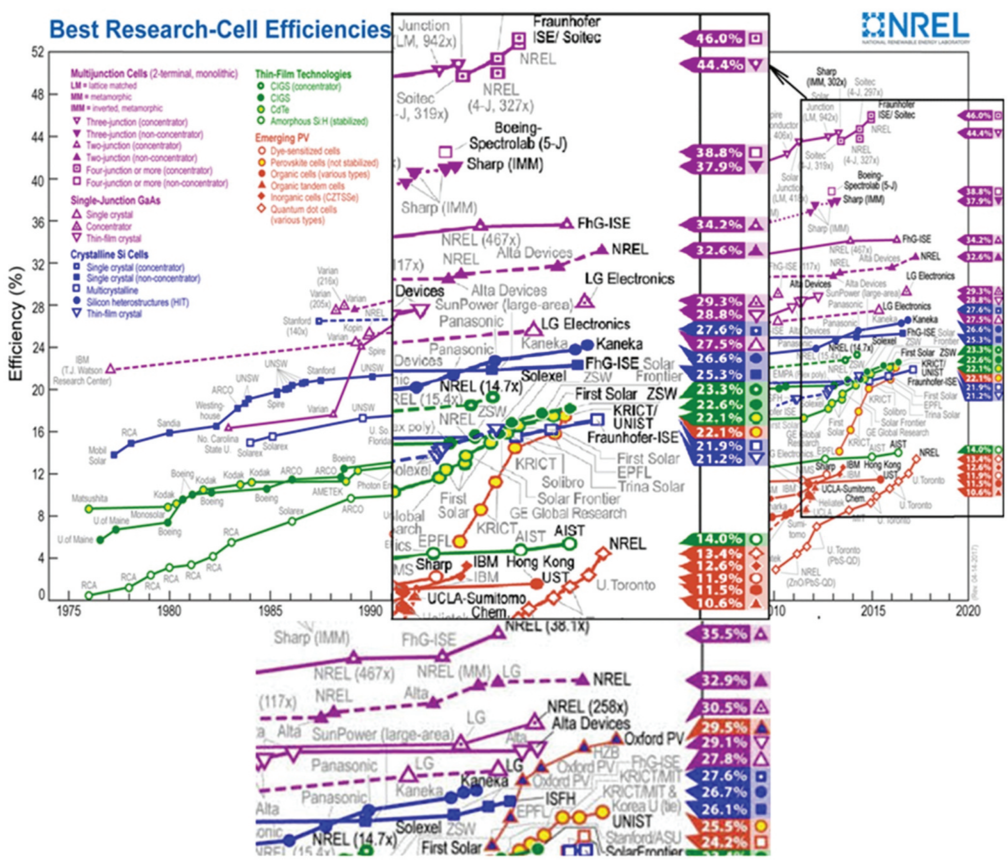
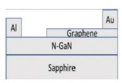
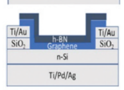
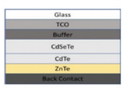
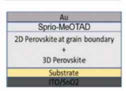
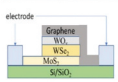
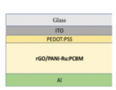


Table 1. Comparison of the different 2D nanomaterials-based solar cells and their device architectures, performance characteristics as well as methods used in their fabrication. (Hasan et al., 2021)

Device structure	Functional materials	Fabrication techniques	Device outputs	Ref.
	Graphene, Gallium nitride (GaN)	Metal–organic chemical vapor deposition (MOCVD)	V_{oc} = 0.225 V J_{sc} = 0.0257 mA cm^{-2} FF = 23%, PCE = 0.0013%	(Kalita et al., 2017)
	Graphene, Hexagonal boron nitride (h-BN)	Photolithography chemical vapor deposition (CVD)	V_{oc} = 0.547 V J_{sc} = 32.89 mA cm^{-2} FF = 54.2%, PCE = 10.93%	(Meng et al., 2016)
	CdSeTe CdTe	MOCVD, Vapor-transport deposition (VTD)	V_{oc} = 0.921 V J_{sc} = 30 mA cm^{-2} FF = 80%, PCE = 20.8%	(Metzger et al., 2019)
	Phenylethylammonium lead iodide ($PEA2PbI_4$)	Spin coating	V_{oc} = 1.13 V J_{sc} = 24 mA cm^{-2} PCE = 20.64%	(J.W. Lee et al., 2018) L. Li et al., 2022
	Graphene, WOx WSe_2, MoS_2	Beam lithography E-beam deposition	V_{oc} = 0.48 V I_{sc} = 0.69 μA FF = 45%, PCE = 5%	(Yang et al., 2020)
	rGO/PANI-Ru hybrid nanocomposite	Spin coating	V_{oc} = 0.732 V J_{sc} = 14.59 mA cm^{-2} FF = 63.6%, PCE = 6.8%	(Vinoth et al., 2017)

power conversion efficiency (PCE ~ 18.2%) (Koo et al., 2020). The architectural design of flexible electronics is generally based on a flexible plastic substrate coated with indium tin oxide (ITO) or fluorine doped tin oxide (FTO) (Du et al., 2021; Shin et al., 2019). Due to the development of cracks on the fragile ITO/FTO, it tends to degrade drastically under hard bending conditions with approximately 4 mm bending radius which reduces the power conversion efficiency (PCE). Therefore, highly flexible electrode materials with excellent electrical conductivity and optically transparent features are necessary to replace the brittle ITO/FTO electrodes in FSCs to achieve high mechanical flexibility and efficiency (Yoon et al., 2017). In this review therefore, graphene, chalcogenide, and nanocomposites of 2D materials and their use in flexible solar cells are presented. Their properties and syntheses techniques have been analyzed and summarized as well as their flexibility, stability and opto-electronic properties in flexible solar cells. The organic solar cells (OSCs) and dye-sensitized solar cells (DSSCs) based on graphene and chalcogenide as well as carbon nanomaterials such as carbon nanotubes and fullerenes have also been presented. Their flexibility, stability, opto-electronic performance, and other features as well the challenges and prospects associated with the adoption of 2D nanomaterials in flexible solar cells are presented.

2. Synthesis of 2D nanomaterials

2.1. Preparation methods of 2D nanomaterials

Many reliable synthetic strategies have been developed to explore and fabricate the several potential 2D ultrathin nanomaterials. Most of these discovered nanomaterials are derived from layered compounds with strong intra-plane covalent bonding. One of the two general approaches, top-down method utilizes exfoliation to separate such as these bulky crystals into nanosheets. The second approach referred to as the bottom-up employs direct synthesis from chemical reactions under specific conditions to form ultrathin 2D nanomaterials. There exist a lot more approaches under these two main methods for the fabrication of 2D nanomaterials.

2.1.1. Top-down approach

While natural processes are used to execute chemical processes, textural intermolecular forces in solids are exfoliated with the help of mechanical forces. After various layers of exfoliation, the intermolecular forces connecting the moderate layers are set to reduce. Liquid-liquid interfaces, polymers, or surfactants are used in order to capture and balance exfoliated sheets. Other top-down techniques includes ball milling, thermal evaporation, laser ablation and sputtering (Nicolosi et al., 2013)

2.1.1.1. Mechanical Exfoliation. The fabrication and general study of promising applications of 2D nanomaterials, for example the opto-electro-mechanical devices and functional nanostructured membranes, are as a result of the discovery of 2D materials including graphene and its derivatives and also because of their unique structural, mechanical and optoelectronic characteristics (Buehler et al., 2013). While keeping the crystal and structural properties, mechanical exfoliation is a straightforward technique to create nanosheets with a single or few layers (Alam et al., 2021). The separation of single and few layers using mechanical exfoliation remains the most utilized approach for researching their properties since it is relatively less destructive (Buehler et al., 2013). This strategy for partitioning of graphite utilizing sticky tape to pries separated graphene layers was the primary technique used to create graphene (Novoselov et al., 2004). The glue substrate gives a means to straightforwardly apply power to individual graphene layers to pries them separated, which is fit to produce extremely great huge graphene sheets (Chen et al., 2011).

However, due to its inability to control the quantity of size and layers as well as the contamination the surfaces of monolayers with sticky polymers, mechanical exfoliation is not advised for mass production (Radisavljevic et al., 2011). The exfoliation of multilayer motif conducting polymers in liquid phases results in individual layers. However, their isolation into stand-alone unit is quite challenging due the strong pi-pi stacking, covalent and H-bonding, or van der Waals type of interactions. Figure Figure 2 depicts the surface modification cycle in mechanical exfoliation.

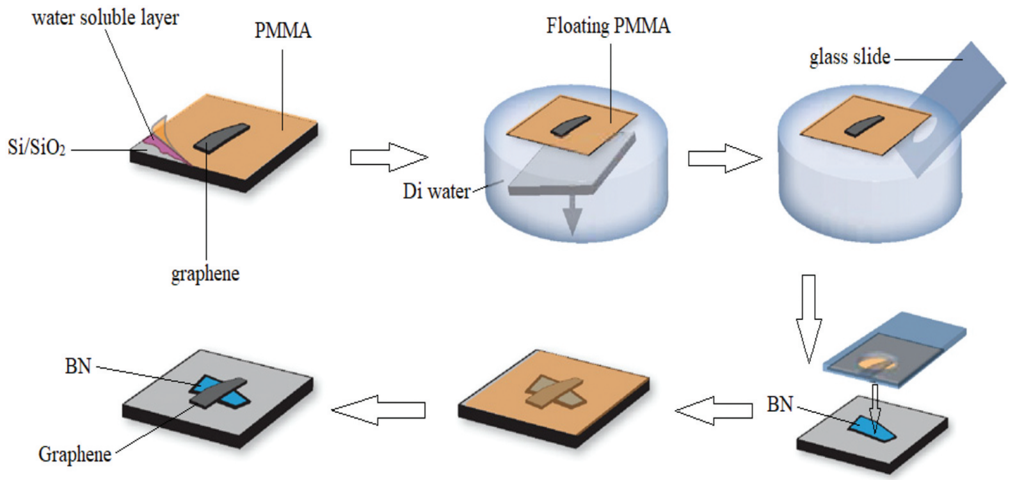


Figure 2. Mechanical Exfoliation process (Huang et al., 2020).

2.1.1.2. Liquid Exfoliation. To directly exfoliate single layer or multilayer flakes from the bulk material with use of sonication, intercalation, two-step expansion, etc., liquid exfoliation can separate bulk crystals in a particular solvent or surfactant and as a result, liquid exfoliation can overcome the confines of mechanical exfoliation (Baijua et al., 2021) Figure 3 describes the various liquid exfoliation mechanisms.

2.1.1.3. Thermal Evaporation. The use of evaporation technique to produce thin films of 2D nanomaterials is a common route to fabricate flexible solar cell layers. The material is placed in resistive boat and heated at high temperatures (and in vacuum) to cause the vaporization of the bulk sample and transport it to solidify on a substrate (Figure 4a). Usually the coil/boat containing the solid bar is heated with high DC current and high vacuum ($<10^{-4}$ Pa) which facilitates the evaporation and the transport of the nanoparticle to the target. In many instances, the substrate temperature can be modified depending on the nature of the nanomaterials to be produced. Prior work by S. Kim et al., (2018) reported on the relationship between the substrate temperature, film uniformity, morphology and thickness. 2D nanomaterials including C60, MoS_2, graphene,

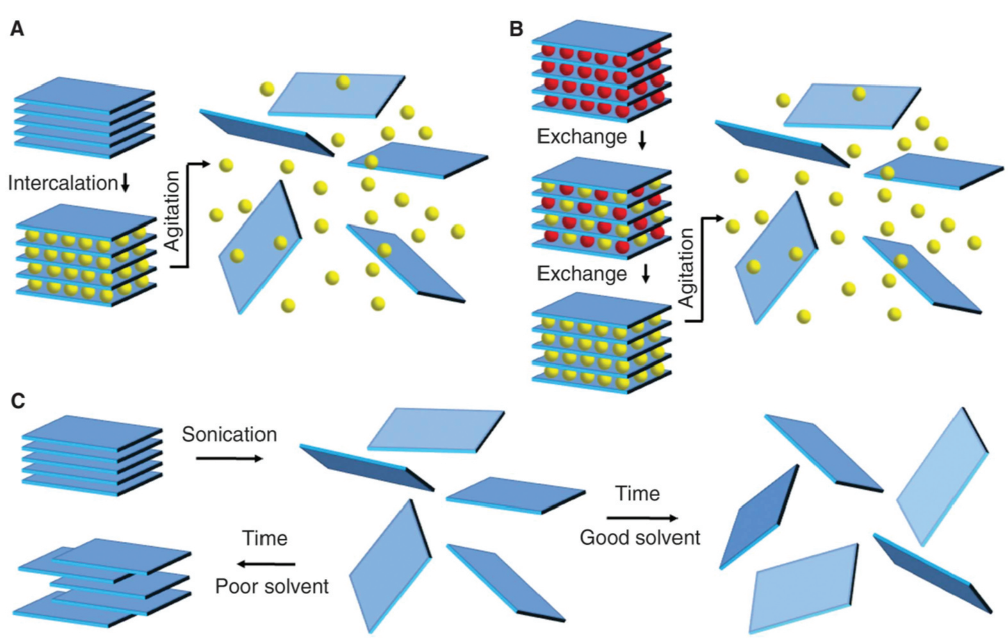
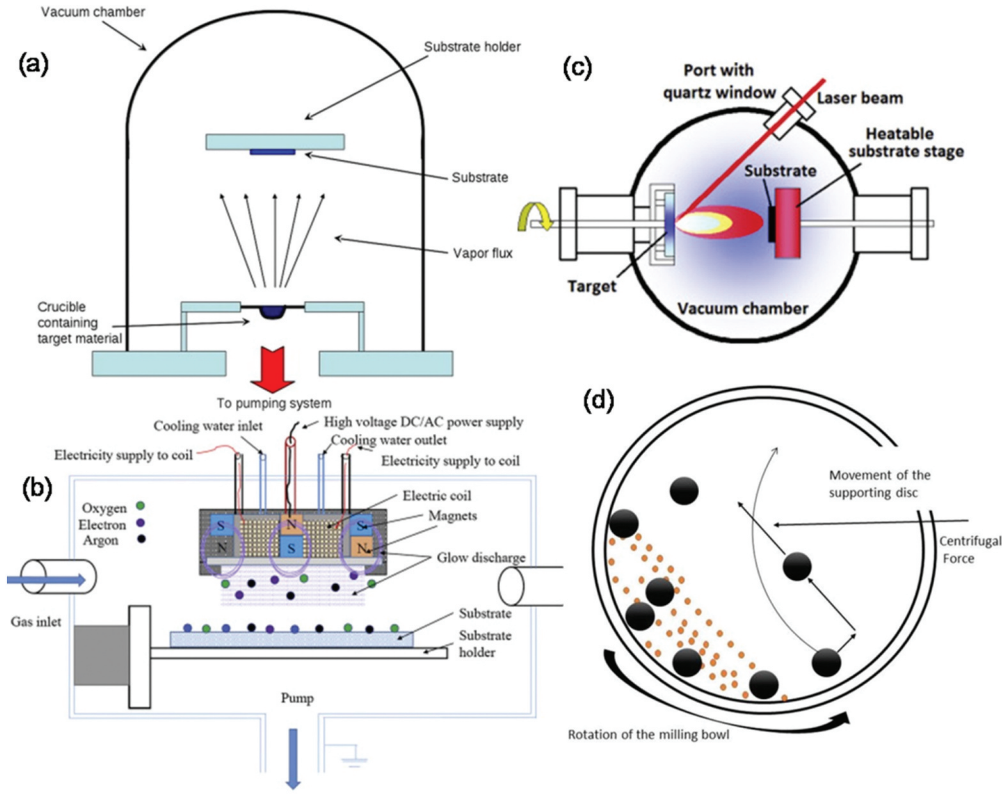


Figure 3. Schematic description of the main liquid exfoliation mechanisms. A. Ion intercalation. B. Ion exchange. C. Sonication-assisted exfoliation (Nicolosi et al., 2013).

Figure 4. Schematic representation of the top-down approach (techniques) to 2D materials syntheses: (a) thermal evaporation (Martin-Palma & Lakhtakia, 2013), (b) sputtering (Hasan, 2018), (c) Pulsed laser deposition (Varghese et al., 2019) and (d) ball milling (Singh et al., 2020).



molybdenum trioxide, tungsten trioxide, etc., have shown to possess high uniformity, variable thickness and the control of the growth dynamics of organic framework greatly influences the optoelectronic behavior of the 2D nanomaterials (Nguyen et al. 2020).

2.1.1.4. Sputtering. The sputtering process is based on the bombardment of target materials by ions generated by plasmas generated in DC and RF gas discharges (Figure 4b). When high-energy ions collide with the target material, the ions are ejected from the target material by the moving momentum of the Ar ions in the plasma, causing erosion of the target surface. The higher the RF or DC power applied to the target, the higher the energy of the particles emitted from the target. It must be noted that the energy of the ions ejected from the target depends on the momentum imparted by the ions in the plasma. Thus, the higher the density, the higher the sputtering rate from the target according to Bräuer et al., (2010). This high energy of ejected ions can be problematic due to the creation of defects on the substrate. Some of the recently studied 2D materials includes, graphene (semimetal), MoS_2, WS_2, $MoSe_2$, (semiconductor) so called transition metal dichalcogenides (TMDCs), elemental 2D materials such as phosphorene, silicene, germanene, borophene (semiconductor and metal), Mxene 2D materials (most of them metallic), NbS_2 (superconductor).

2.1.1.5. Laser ablation. The use of pulsed laser deposition technique to synthesize different two-dimensional nanomaterials is a common phenomenon. It is a physical vapor deposition process where a high power pulse laser beam is focused to strike a target of the desired composition. The material is then vaporized and deposited as a thin film on a substrate facing the target as shown in Figure 4c. The technique is very simple as the surface of a target is heated into a vapor phase using a laser beam. For example, Smalley, Kroto and Curl who won the Nobel Prize when they adopted the Nd-YAG laser to vaporize graphite and synthesized spherical fullerenes. The ultimate film produced by this technique will have a composition similar to the target material. Hence, it is often used to synthesize different materials by varying the pressures over a wide range. Carbon nanotubes (CNTs) and fullerenes (C60) are common materials produced by pulsed laser deposition (Varghese et al., 2019; Hong et al., 2019; Maheswaran & Shanmugavel, 2022).

2.1.1.6. Ball milling. Ball milling is an eco-friendly route to synthesize nanocomposites of two dimensions. It is one of the methods which take advantage of the mechanical shock forces to exfoliate bulk materials into layered forms of nano-dimensional thicknesses. The working principle is simple; impact and attrition size reduction take place as the ball drops from near the top of a rotating hollow cylindrical shell as shown in Figure 4d. Prior work by Singh et al., (2020) on the possibilities of fabricating reduce graphene oxide sheets, platelets of graphene as well as functionalized graphene achieved effective outcomes. For example, Krishnamoorthy et al., 2016 using N-methyl-pyrrolidone (NMP) both as solvent and agent ball milled to exfoliate molybdenum disulphide nanosheets adopted in the solar cell and other optoelectronic device fabrications. The merit of this technique is the fact that neither an inert atmosphere nor high vacuum is required.

2.1.2. Bottom-up approach

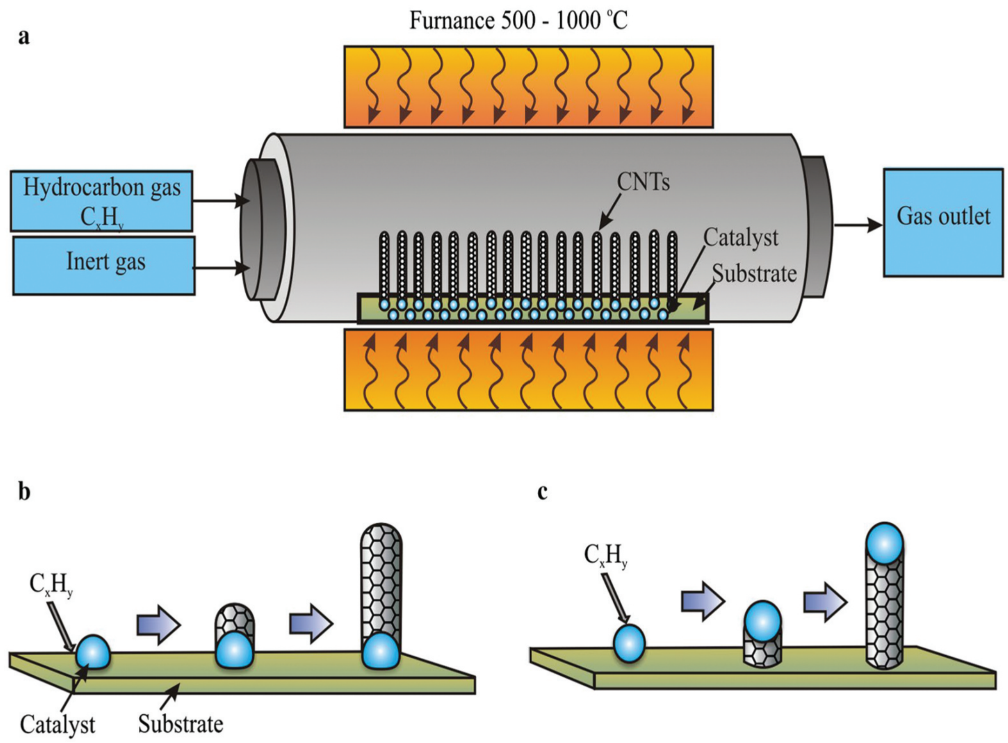
For making ultrathin and excellent nanocomposites with large parallel dimensions, the bottom-up approach is extremely important. From atomic to molecular sources, nanoscale materials are created using bottom-up technique which includes chemical vapor deposition (CVD), hydrothermal synthesis, co-precipitation and sol-gel. (Alam et al., 2021)

2.1.2.1. Chemical vapor deposition. Chemical Vapor Deposition (CVD) method involves the deposition of vapor created when the source material is heated at high temperature on a substrate at a relatively low temperature (Zhou et al., 2020). CVD approach was successfully applied in 2009 in a mixture of methane and hydrogen gas under 1000 °C to prepare large-area and high-quality graphene on Cu foil substrate (C. Li et al., 2019). 2D materials are made by separating gas, liquid, or solid predecessors in specially arranged environment (J. H. Lee et al., 2014). (Kim et al., 2012) arranged the monolayer and large-area h-BN through low-pressure CVD on the Pt foil by utilizing ammonia borane as the forerunner. Lee et al. arranged the single-layer and high-glasslike MoS_{22014} on the SiO_2 substrate by utilizing MoO_3 and S powders as the reactants (Y. H. S. Lee et al., 2012).

CVD is a favored strategy to plan large area, high-quality also, monolayer 2D nanomaterials, while the exploratory condition for CVD is moderately severe and the CVD strategy has not yet been created for setting up some other 2D nanomaterials, for example, phosphorene (Zhou et al., 2020). The CVD technique is able to produce 2D nanosheets of conducting polymers directly from their atom, ion or molecular structures. Figure Figure 5 illustrates the various procedures in synthesizing 2D materials using chemical vapor deposition.

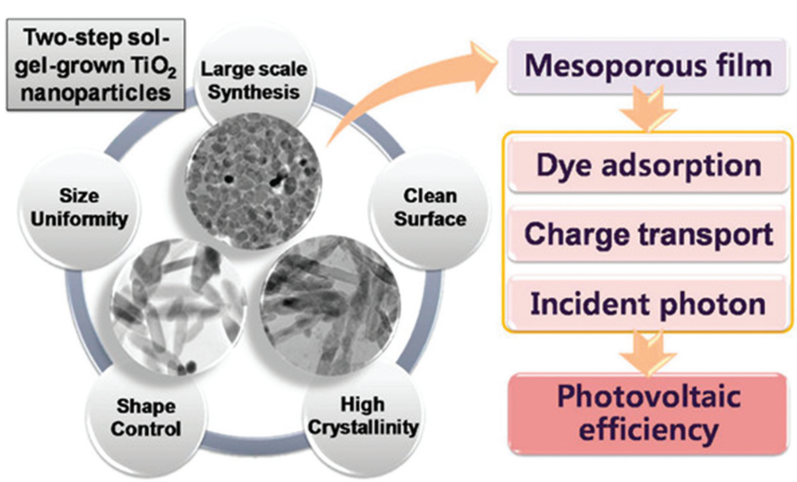
2.1.2.2. Solution-based chemical synthesis. Despite the less popularity of this method as compared to CVD, solution-based synthesis methods are manifesting as one of the preferences because of their simplicity account for mass production in a short time. Other than that, solution-based synthesis methods present new approaches to achieve things that cannot be done by using the CVD method, for example synthesis inside the optical fiber capillaries, and can be used for several kinds of flexible substrates, even polymer substrates. For Transition Metal Dichalcogenide (TMDC) thin film synthesis, chalcometalates are the most utilized precursors by thermal decomposition. The cation placed outside on the metal-chalcogen framework must decompose within the pyrolysis reaction and its molecular weight must not be too high to prevent weight loss during the thermal treatment. Oxidation can also be prevented by performing the thermal treatment under an inert surroundings. (Hoang et al., 2020)

Figure 5. Schematic representation of chemical vapor deposition (CVD) process. (a). Simplified scheme of a CVD reactor for CNTs synthesis; (b). base growth (Zaytseva & Neumann, 2016).



2.1.2.3. Sol-gel synthesis technique. Sol-gel method has gained popularity because it produces extremely stoichiometric, as well as multicomponent oxide nanocrystalline materials. This technique in combination with template-assisted, electrophoretic deposition, etc., has achieved successful outcomes of nanowires of multicomponent nature in a repeated order as represented in Figure Figure 6. The hydrolysis of a solution of precursor molecules to produce a suspension of colloidal particles (the sol), followed by the condensation of sol particles to produce a gel, is the foundation of sol–gel processing. Whereas in aqueous environments, precursors can be inorganic salts, they are metal alkoxides in organic solvents. The capacity to handle multicomponent complex oxides is the largest benefit of sol–gel processing.

Figure 6. Shows the two-step sol-gel synthesis technique adopted in nanoparticles for photovoltaic applications, (H.-F Lee et al., 2013).



3. 2-D Nanomaterials in Flexible solar cells (FSCs)

Flexible solar cells (FSCs) have recently received a lot of attention by researchers. The goal of this technology was to develop a portable power source that is also efficient. The architectural design of flexible solar cell is generally based on a flexible plastic substrate coated with indium tin oxide (ITO). Due to the development of cracks on the fragile ITO, it tends to degrade drastically under hard bending conditions with approximately 4 mm bending radius which reduces the power conversion efficiency (PCE). Another problem of the FSC in perovskite solar cell (PSC) application is the challenge of using fully low temperature during its preparation on a substrate. High flexible electrode material with excellent electrical conductivity and optically transparent features are necessary to achieve high mechanical flexibility and efficiency of a perovskite solar cell (Yoon et al., 2017). Graphene, chalcogenide, and nanocomposites of some 2D materials possess the above features and are even lighter and easy to obtain. These three materials are discussed in the subsequent sections.

3.1. Flexible solar cells based on graphene

Graphene has demonstrated to be an excellent electrode material for FSCs with good flexibility, high conductivity and excellent transparency. Using graphene as a transparent conductive electrode (TCE) helps to achieve large surface area and high-quality films on flexible polymer substrates. Usually, this is achieved using chemical vapor deposition (CVD). One major problem of using graphene is the low PCE due to the large resistance of graphene sheets which limits its application in devices. Despite its limitations, it has shown significant performance in flexible organic photovoltaics, perovskites, dye-sensitized and several solar cell architectures, owing to its high chemical stability (Du et al., 2021; Shin et al., 2019) and Table 1.

3.1.1. Flexible perovskite photovoltaic devices (PSCs)

The increase in demand for portable power applications has aroused great interests in perovskite solar cells (PSCs) due to the excellent flexible performance and PCE it possesses. The high efficiency of the PSCs stems from the bandgap tunability of the device which helps in achieving very high optoelectronic properties. Low temperature fabrication process creates wide window for high flexibility and transparency (Guo et al., 2015). Graphene electrodes help in achieving very high flexibility, low resistance of the graphene sheets, light weight, and many other promising features according to (Liu et al., 2016). They fabricated ultrathin PSCs using transparent graphene electrode. They employed low-temperature solution process to first prepare the PSCs on an indium tin oxide electrode and subsequently fabricated the FPSCs with the graphene electrodes by coating a two-layer graphene (2 L-G) with different polymer substrate film such as P3HT which has sufficient energy levels (see Figure 7a) to serve as hole transport layer (HTL). Under light illumination, the

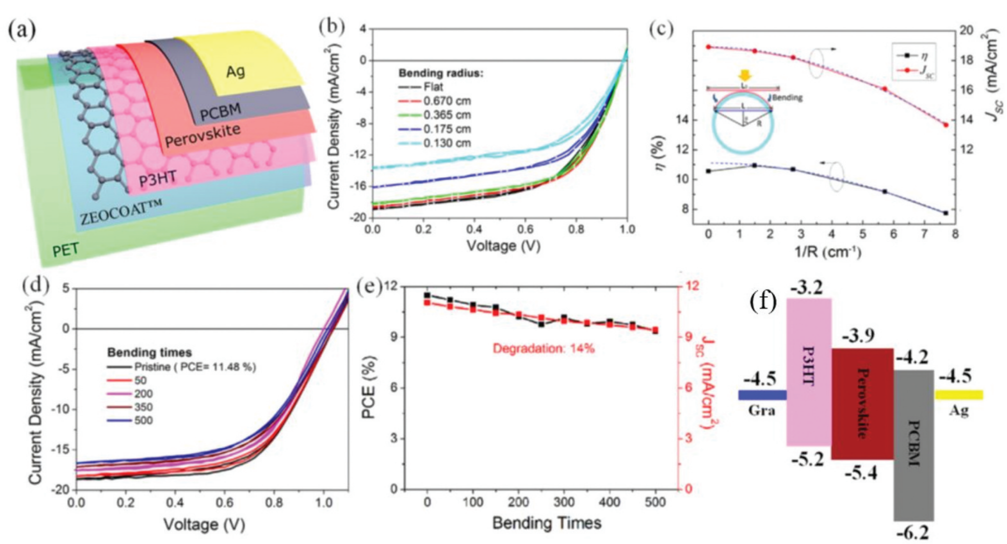


Figure 7. (a) Schematic representation of flexible PSC. (b) current-voltage behavior of PSCs with varying bending radius, (c) Flexing radius and efficiency of PSCs. (d) Bending cycle of PSCs on (d) *J-V* and (e) PCE and J_{sc}. (f) Energy diagram of the flexible PSC. (Z. Liu et al., 2016).

flexible device showed little hysteresis under various bending conditions (Figure 7b-d). The flexible PSC recorded a PCE of 11.5% (Figure 7e) with very high durability during various bending tests. They also compared the specific weight (the power output per weight) of the fabricated device with the conventional inorganic solar cells and obtained about 5 W/g which shows a better performance. The performance of this flexible device was attributed to the usage of the P3HT as hole transport layer which enables uniform coating on the graphene electrode with high band structure which paves way for its application in highly efficient flexible PSCs.

A year after, Yoon *et al*. (Yoon et al., 2017) recorded a maximum PCE of 16.8% with no hysteresis of a graphene-based flexible perovskite solar cell as a transparent electrode. They reported that after 5000 bending cycles using a bending radius of 2 and 4 mm, over 85% of the recorded PCE was maintained by the graphene-based devices. They also compared the resistance of their flexible device with the conventional ITO devices and realized it was higher in the graphene-based flexible PSC, but their device yielded a high open circuit voltage (*Voc*) and short circuit current (*Jsc*). This was achieved by the excellent energy level alignment and transmittance with the homogeneous HTL. After repeated bending tests, there was neither resistance increase in the graphene sheet nor any observable cracks from the flexible device as compared to the conventional ITO device. Here, a single layer graphene synthesized by CVD was wet transferred on a thin polyethylene naphthalate (PEN) film to enhance transmittance and flexibility. To improve the electrical conductivity of this single layer device, few nanometers of thick molybdenum trioxide layer (MoO_3) was thermally deposited to cause graphene hole doping (see Figure 8a-e). Recently, Jin and his colleagues have reported the performance of a an efficient and stable flexible PSCs without adopting a separate hole transport material. However they used carbon conductors and a substrate made from silver nanowires and graphene (Jin et al., 2021). A complete carbon-based PSC fabricated using low-temperature solution process was successfully achieved and the substrates involved were the graphene-AgNWs-polyethylene terephthalate and other substrate structures as shown in Figure 8f. An optimal efficiency of ~9.73% was obtained with enhanced mechanical stability with long term durability for the flexible devices (Figure 8g). After 1000 bending cycles using 10 mm radius, the Gr-AgNWs/PET flexible C-PSCs maintained 89% of its original PCEs which was higher than the ITO-based substrate as shown in Figure 8h with minimal degradation.

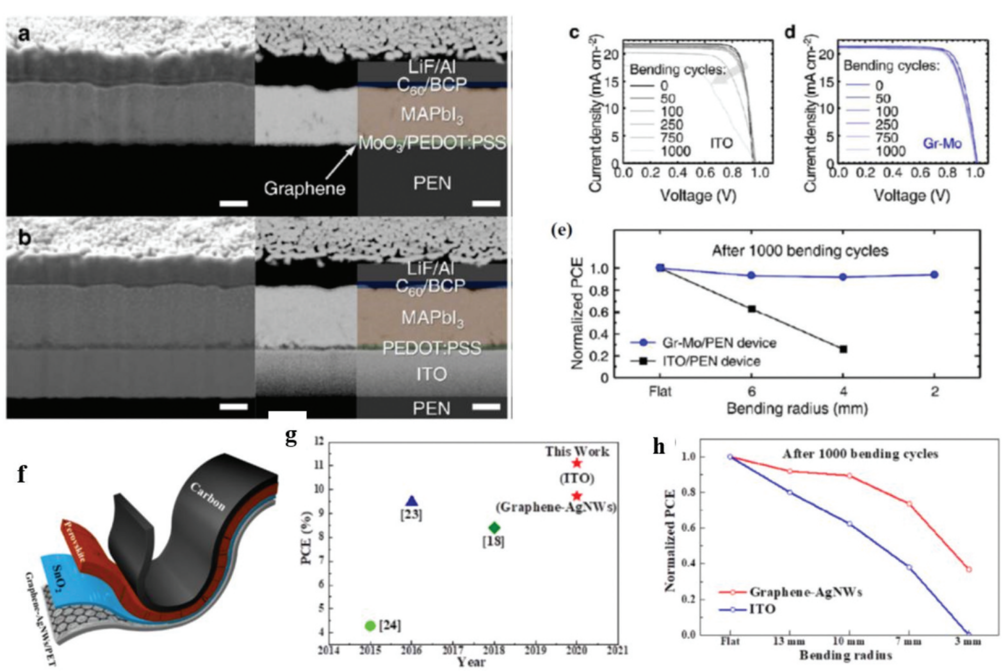


Figure 8. (a-b) Focused ion-beam assisted SEM images of complete perovskite devices fabricated on (a) Gr-Mo electrode/PEN substrate and (b) ITO electrode/PEN substrate. (c-d) J −V curves of the (c) ITO/PEN and (d) Gr-Mo/PEN devices throughout 1000 bending cycles with R = 4 mm. (e) Normalized PCEs as a function of bending radius after 1000 cycles for the Gr-Mo/PEN and ITO/PEN devices (Yoon et al., 2017). (f) Schematics of C-based FPSCs. (g) Comparing PCE of C-based FPSCs without hole transport materials over recent years. (h) Normalized PCE after 1000 bending cycles at different bending radii. (Jin et al., 2021).

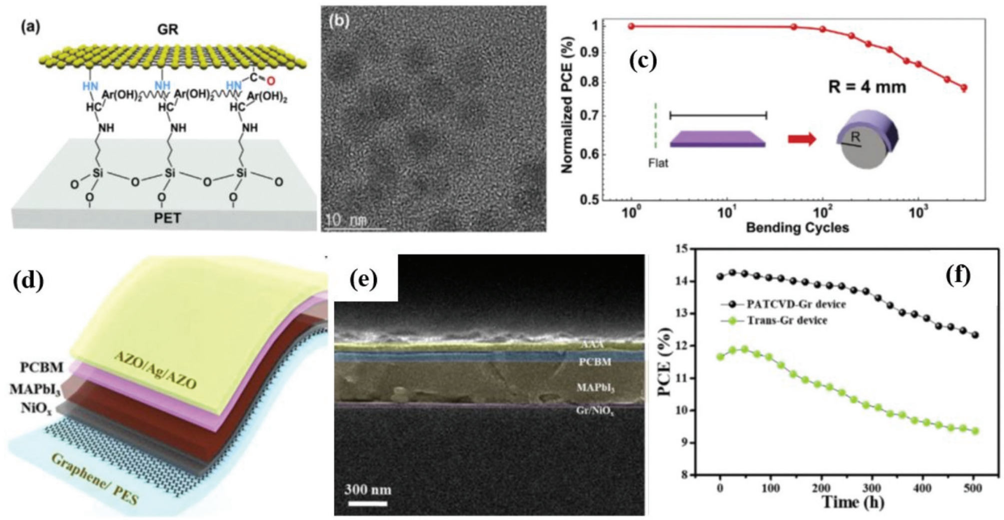


Figure 9. (a) Schematics of PET substrate and graphene sheet chemical interaction by an interlayer of APTES. (b) High TEM image of GQDs. (c) Normalized PCE degradation under repeated bending for 1000 cycles using a radius of 4 mm. (Shin et al., 2019). (d) Schematics of the semi-transparent flexible graphene-based PSCs. (e) SEM image of the full solar cell structure. (f) PCE variation of the devices under continuous exposure of solar light AM 1.5 G during 500 hrs. (Tran et al., 2019).

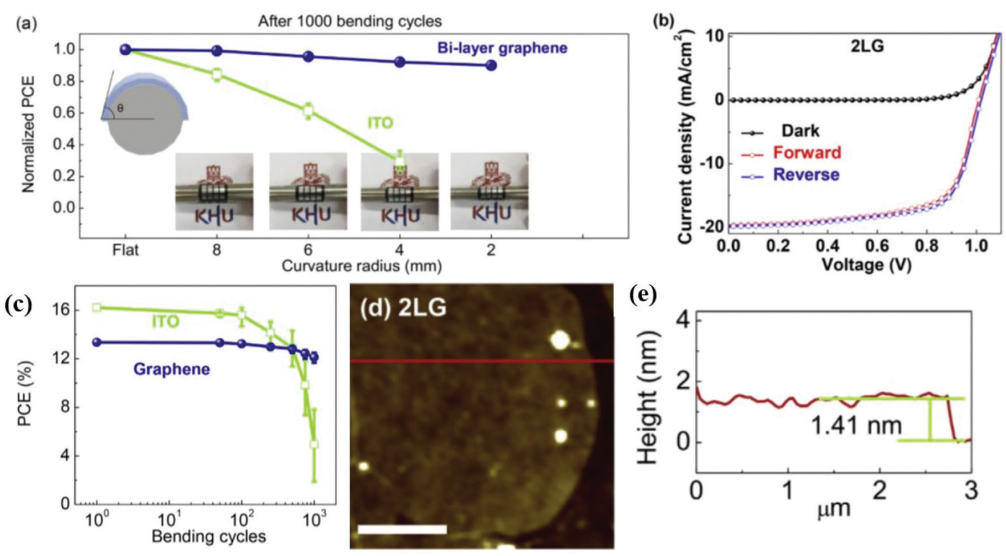
Prior study by Shin et al., (2019) reported a flexible PSCs which exhibited an excellent stability upon bending and retained about ~80% of its initial PCE recorded following a flexural cycle of 3000 and at bending radius of ~ 4 mm. In their report, a graphene-treated 3-aminopropyltriethoxysilane was first used as a transparent conductive electrode (TCE) and then quantum dots (GQDs) were doped with phenyl C61 butyric acid methyl ester (PCBM) to serve as electron transfer layer (ETL) for the application of FPSCs. The PCE of the fabricated PSCs increased up to 15.0% on a flexible substrate with the GR/APTES TCE as a result of increasing GQDs concentration (see Figure 9a-c). Tran *et al*. (Tran et al., 2019) first reported an ambient air fabrication of transfer-free graphene electrode for the application of super-flexible and semi-transparent PSCs. The device was synthesized on a polymer substrate using a CVD technique coupled with plasma and the PCE of ~14.2% was recorded and flowing a flexural cycle of 1000, efficiency of more than 90% was retained for the graphene-based devices under a tensile strain of 1.5% with high flexibility (see Figure 9d-f).

Semi-transparent FPSCs with an anode/cathode have been fabricated by graphene doping with bis (trifluoromethanesulfonyl)-amide (TFSA) and a high aperture FPSCs with triethylenetetramine (TETA) (Jang et al., 2019). The PCE was increased by the addition of Ag reflectors at the bottom/top of the FPSCs and retained 70% of its initial value at a bending radius of 8 mm after 1000 bending cycles. Kim et al. (S. S. Kim et al., 2018) also employed multiple graphene layers as TCEs and observed the layer number (L_n) effect on a FPSCs. For L_n = 2, the PCE of 13.35 and 13.94% was recorded for forward and reverse scans, respectively, with nearly no *J-V* hysteresis. The solar cells based on graphene displayed high-performance bending stabilities maintaining about 90% of the original PCEs using radius of 8–2 mm after 1000 bending cycles (see Figure 10a-e). These promising performances make graphene-based PSCs applicable in flexible portable power sources.

3.1.2. Flexible organic solar cells (FOSCs)

Organic solar cells (OSCs) have been on the radar of most researchers due to its prospects of mass production, light weight, and eco-friendly components. ITO are usually used as TCE, but there is the need to replace the poor transparent-brittle ITO with much more flexible OSCs. Graphene has shown good chemical stability, high flexibility, and hence, can be used as TCEs for flexible OSCs (Kumar & Chand, 2012). Recently, (Du et al., 2021) fabricated flexible OSCs with graphene-based anode architectures with different graphene layers and doping. They recorded a better performance with the 0.2 cm^2 3-layer graphene (BI-doped or pristine) TCE. A PCE of 6.85% was obtained for the BI-doped graphene anode with high flexibility, mechanical robustness and exhibited low degradation during 250 flexing cycles. Increasing the thickness of the BI-doped TCE with photoactive thick layer to 1.6 cm^2, a PCE of 1.8% was recorded, illustrating graphene TCE as a promising

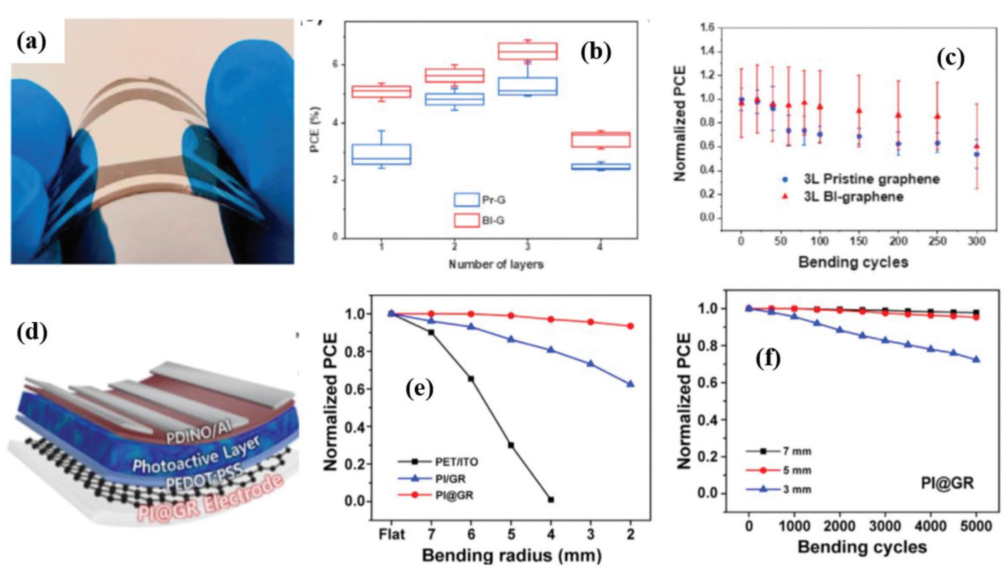
Figure 10. (a) Normalized PCEs of the FPSCs with ITO and 2-layer graphene vs bending radius. (b) Dark and photo *J-V* curves of FPSCs with 2-layer graphene in forward and reverse scans. (c) Absolute PCEs of the fabricated solar cells vs bending cycles at a radius of 4 mm. (d, e) AFM image and height profile of 2-layer graphene. (S. S. Kim et al., 2018).



candidate for flexible OSCs (see Figure 11a-c). By adopting polyimide-integrated graphene (PI@Gr) electrode for flexible OSCs, Koo research team (Koo et al., 2020) recorded 15.2% power conversion efficiency. The PI served as a bifunctional material for both substrate and carrier film for the graphene which enhanced the flexibility, thermal stability, and mechanical robustness of the devices during up to 10,000 bending cycles as shown in Figure 11d-f. This performance was highly comparable to the rigid ITO-based reference device, which serves as a promising candidate material for highly efficient and flexible optoelectronic devices.

Low-resistance transparent conductive film (TCF) with high flexibility has been developed by thermal roll imprinting which was embedded in Ag grid for applications in flexible OSCs (Raman et al., 2020). Excellent electrical and optical performances were shown when graphene and ITO second layer was coated onto the flexible Ag-embedded substrate as shown in Figure 12a-b. The high performing TCF was used to fabricate an OSC and recorded a power conversion efficiency of 2.551% and 2.649% for the graphene-embedded Ag grid and the ITO-Ag grid, respectively. The

Figure 11. (a) Picture of a 1.6 cm² flexible BI-doped graphene anode-based OSC. (b) PCE as a function of number of the fabricated OSCs layers. (c) PCE of the OSCs as a function of bending cycles. (Du et al., 2021). (d) Schematics of the fabricated device. (e,f) Normalized PCE of flexible the devices at various radii of curvature after 1,000 cycles of the bending test and under various bending cycles with different bending radii, respectively. (Koo et al., 2020).



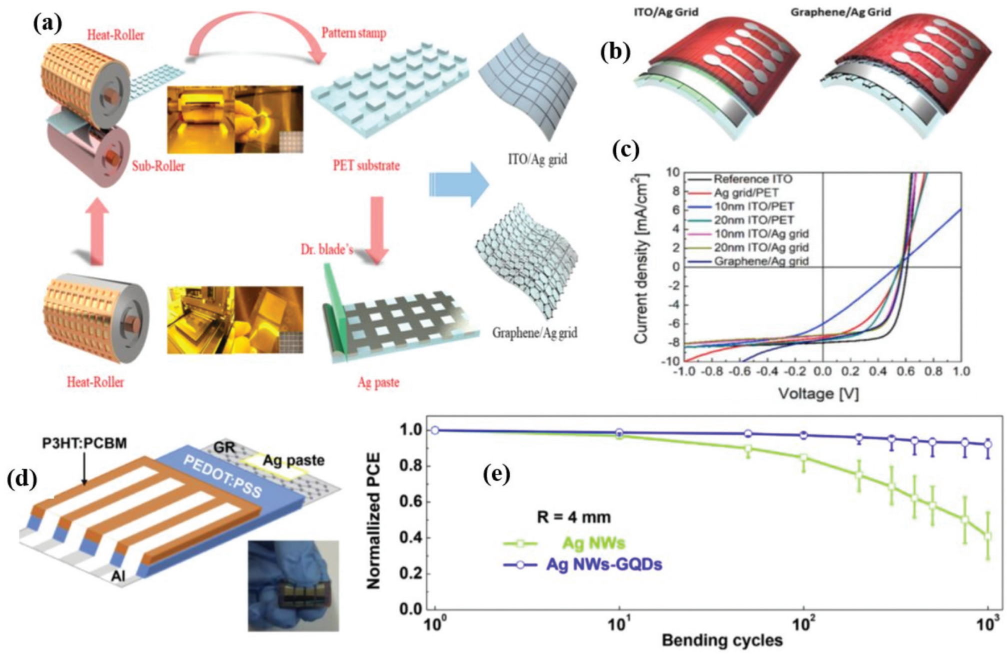


Figure 12. (a) Schematics of the embedded Ag grid electrode via thermal roll imprinted metal grid pattern process and inked Ag paste in grid pattern process. (b) Picture of the fabricated FOSCs using 20-nm-thick ITO/Ag grid and graphene/Ag grid electrodes. (c) *J-V* curves of the fabricated FOSCs on the hybrid Ag grid electrodes. (Raman et al., 2020). (d) Schematics of the fabricated FOSCs. (e) Normalized PCEs of the Ag NWs/GR TCE/FOSCs vs bending cycles at R = 4 mm for n_G= 0 and 0.02 g/L (Shin et al., 2018).

intaglio method used in the hybrid electrode fabrication (20 nm thickness) resulted in a moderately high performance with short-circuit current and open-circuit voltage of 7.174 mA/cm^2 and 0.565 V, respectively (Figure 12c). (Jayenta Singh et al., 2019) in an earlier work used computational studies (Silvaco TCAD Atlas tool) for graphene/PEDOT:PSS Schottky junction on a PET substrates used for flexible OSCs applications. They designed and simulated the PEDOT:PSS layer varying its thickness from 50–90 nm with the optimized device showing excellent photovoltaic features under AM1.5 G illumination. In this work, they proposed a band diagram to be used in explaining the phenomenon of carrier transport. Energy conversion efficiency, short-circuit current density and an open circuit voltage of 2.87%, 0.68 mAcm^{-2}, and 0.68 V, respectively, were reported. Graphene TCEs was doped with graphene quantum dots (GQDs) which has been mixed with silver nanowires (Ag NWs) and used for highly FOSC (Shin et al., 2018). With this work, GR TCEs doped with GQDs-mixed Ag NWs were first fabricated on a substrate of polyethylene terephthalate with varying doping concentrations of GQDs as shown schematically in Figure 12d. A power conversion efficiency of 3.66% was recorded for the Ag NWs/GR TCE as a result of the doping concentration of GQDs and consequently, resulted in decreasing resistance in the composite sheet. The bending flexibility was also enhanced and over 90% of the initial PCE was maintained for the OSCs after 1000 bending cycles at a radius of 4 mm (see Figure 12e).

3.1.3. Flexible dye-sensitized solar cells (DSSCs)

The architecture of dye sensitized solar cell (DSSC) consists of a platinum counter electrode, titanium dioxide nanocrystalline which is dye sensitized and a liquid electrolyte. To lower the tri-iodide (I_3^-) to I^- in the redox electrolytes, deposition techniques such as thermal annealing, vacuum coating, etc., are chosen to coat the precursors of platinum on a substrate of a transparent conducting oxide support. Due to the expensive nature of Pt even though it has excellent conductivity and enhanced stability, there is the need for alternative materials to achieve Pt-free as well as TCO-free devices to make them affordable (Regan & Grätzel, 1991). Graphene has demonstrated to possess these features. Textile fabric-based electrodes have recently received a lot of attention by researchers as wearable energy sources due to their flexibility, lightweight, and cost effectiveness, as well as their ease of production. (Sahito et al., 2016) fabricated a Pt-free highly flexible and conductive graphene nanosheets, coated on a cotton fabric (HC-GCF) for use as an efficient counter electrode (CE) in dye-sensitized solar cells. The fabricated HC-GCF counter electrode with polymer electrolyte recorded a high photovoltaic conversion efficiency of 6.93%.

The device also showed a negligible variation at various bending angles with enhanced electrocatalytic activity (ECA), making it a promising candidate material for DSSCs. Graphene with graphitic properties was also fabricated and compared to pristine graphite by utilizing metal titanium in a hydrochloride acid solution (Hung et al., 2014). The synthesized TCO-free graphene nanosheet was highly conductive and flexible which served as a counter electrode material in a DSSCs. Figure 13a-b presents an FESEM micrographs of graphene oxide (GO) as well as reduced graphene in titanium metallic powders (rGO-Ti(III)) with tiny white particle features resulting from the chemical reduction process. When utilized as a flexible counter electrode in dye-sensitized solar cells, this high-quality graphene film performs better than the standard sputtering Pt counter electrodes as shown in the *J-V* curves in Figure 13c. A two-step hydrothermal reaction process was employed to develop a mechanical stable 3-D composite of nitrogen-doped graphene/reduced hydroxylated carbon nanotube aerogel (NG/CNT-OH) (Jindan Zhang et al., 2016). This TCO-free composite material showed a distinct hierarchical porosity with a lot of exposed active sites for ion transport as shown in the SEM image in Figure 13d. The NG/CNT-OH was later compressed and achieved excellent flexibility and electrical conductivity making them suitable for flexible DSSC counter electrodes. After optimization, the DSSC TCO-free counter electrodes (CE) recorded a cell efficiency of 6.36% with a higher short-circuit current density of 13.62 $mAcm^{-2}$ as compared to the DSSC Pt CE (5.74% and 12.81 $mAcm^{-2}$, respectively) (see Figure 13e).

Again, CVD method has also been employed to synthesize flexible CEs which is Pt-free for applications in DSCCs (Lukaszkowicz et al., 2017). Here, the graphene films synthesized by CVD were used to produce ITO/PET counter electrodes with addition of other polymer substrates. The flexible electrodes used for the DSSCs application delivered a PCE of up to 3.95% as compared to the Pt-based DSSC CEs (4.39%).

The photoluminescence properties of graphene quantum dots (GQDs), which are produced from graphene domains, have been studied for DSSCs applications. GQDs are biocompatible and have high luminescence which indicate its possible integration into photovoltaic systems (Mustafa & Sulaiman, 2020). An electrochemical cyclic voltammetry technique was used to synthesized GQDs from reduced graphene oxide (rGO) and incorporated into TiO_2 paste (a binder free substrate). As a result of the binder-free TiO_2 incorporation, an increase in charge extraction was observed which accounted for an increase in PCE of the device by 5.48% as compared to the TiO_2 photoanode (Kumar et al., 2019). In

Figure 13. FESEM of (a) GO, (b) rGO-Ti(III). (c) *J-V* curve for DSSCs with Pt electrode, graphene (rGO-water) and Pt nanoparticle incorporated graphene (Pt-rGO-water). (Hung et al., 2014). (d) SEM image of interior microstructure of NG/CNT-OH composite aerogel. (e) *J-V* curves of DSSCs with different CE (The curves were obtained under AM 1.5 G simulated sunlight with a power density of 100 $mWcm^{-2}$, photoanode active area = 0.16 cm^2. (Jindan Zhang et al., 2016).

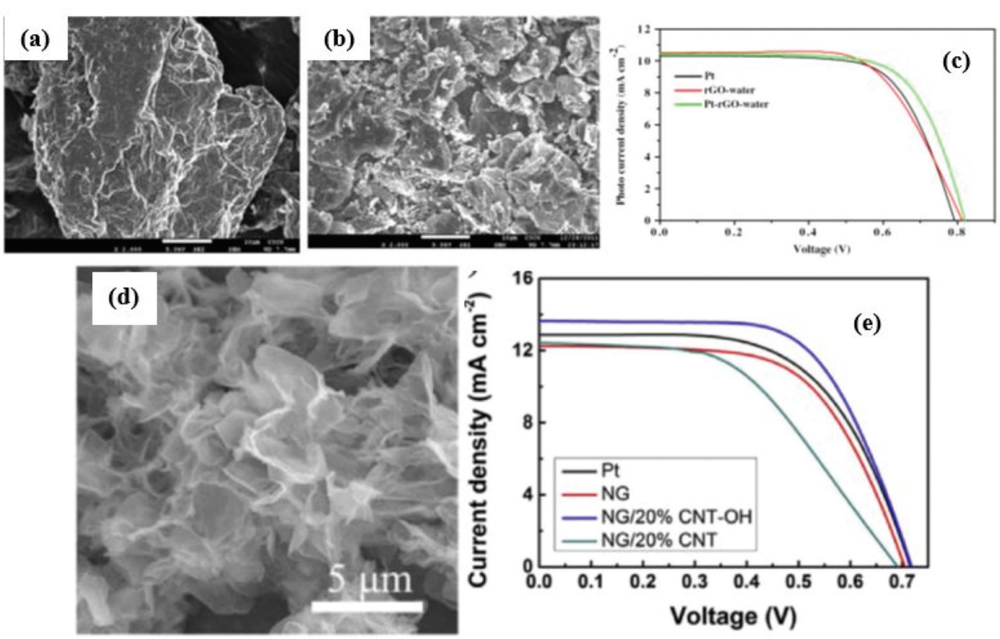


Table 2. Performance comparison of flexible organic, perovskite and dye-sensitized solar cell devices based on composite materials and fabrication methods

Composite material	Fabrication Method	PCE (%)	Bending radius (mm)	Bending cycles	Efficiency retention (%)	Young modulus (GPa)	Poisson ratio	Adhesion Force/energy (nN)	Reference
P3HT:PCBM	CVD	2.5–2.6	5.2	-	Fairly constant	6.02	0.3	2.9 J/m^2	(S. S. Lee et al., 2012), Agyei-Tuffour, Doumon et al., 2017
TETA/GR TCEs	CVD	3.30	12–6	-	99.0	3.2	0.3	1.24 J/m^2	(Shin et al., 2018)
Graphene oxide/ polyimide (PI) substrates	CVD	3.20	3.0	1000	92.0	20.5	0.3	2.3 J/m^2	Yang, 2011
Ag NW-graphene/PET	Simple brush painting	2.68	10.0	5000	Fairly constant	207	0.3	4.4 J/m^2	(Seo et al., 2014) Kang et al., 2016
$FAPbI_3$	LASP	19.38	.org/10.0	500	92.0	12.8	0.31	354.30 nN	(Wu et al., 2019) Ma et al., 2021 Ichwani et al., 2022
Amino-functionalized Gr-QDs (AGQDs)/NiOx	Low temp. solution process	18.10	10.0	1000	88.0	34	0.3	6.78 J/m^2	(Z. Z. Wang et al., 2020)
Ti/Al/Cu foil nanotubes (TNT)/CNT	One-step ionizat + CVD	6.01	7.5	100	84.2	92	03	1.53 J/M^2	(X. X. Wang et al., 2015) (Tsukamoto, 2020)
graphene/CNTs/Spiro-OMeTAD	CVD + Low temp. non-aq Rt	11.90	4.0	2000	86.0	32	0.3	105.92 nN	(Luo et al., 2018) Ichwani et al., 2022

(*Continued*)

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Table 2. (*Continued*)

Composite material	Fabrication Method	PCE (%)	Bending radius (mm)	Bending cycles	Efficiency retention (%)	Young modulus (GPa)	Poisson ratio	Adhesion Force/energy (nN)	Reference
Paper-based GD/PEDOT: PSS	"Soak and dry"	4.91	-	150	Fairly constant	1.42	0.3	0.5 J/m^2	(C.-P. C.-P. Lee et al., 2017), B. Agyei-Tuffour et al., 2016
CF@PANI20@CoSe	In situ polym. + HT reaction	10.28	5.0	600	78.6*	5.0	0.3	0.11 J/m^2	(Junxiang Zhang et al., 2019)
Tandem PET/ITO/MB-NiO /WBG **PVK/C60**/ SnO_2/Au/ PEDOT :PSS/NBG PVK/ C60 /B C P/Cu	ALD + Spin coating	24.7	15	10,000	95.1	20	0.3	24.45 nN	L. Li et al., 2022 Ichwani et al., 2022

CVD: Chemical vapor deposition

LASP: Ligand and additive synergetic process

*Under a deformation state of 90° bending angle

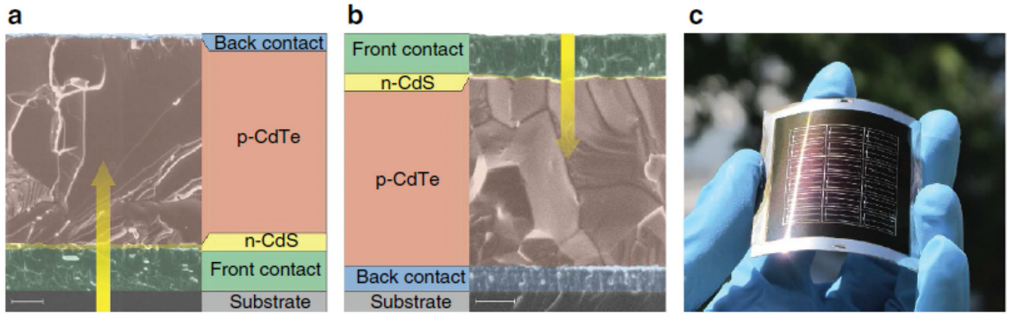


Figure 14. SEM micrograph and schematic cross-section of CdTe solar cell in (a) superstrate and (b) substrate configurations. (c) Photograph of a flexible CdTe solar cells on a flexible metal foil. (Kranz et al., 2013).

summary, graphene combines high inherent carrier mobility and outstanding flexibility with excellent mechanical strength and chemical inertness, making it an excellent choice for optoelectronic applications such as flexible PSCs, OSCs and DSSCs. Depending on the flexible substrate material and fabrication method employed, different device performance could be obtained; a summary performance comparison has been presented in Table 2.

3.2. Flexible solar cells based on chalcogenide

Cadmium telluride (CdTe) photovoltaics (PVs) are efficient and economical power systems which yield a high energy return on investment with a short payback time. A typical CdTe cell consists of a CdTe absorber layer; a cadmium sulfide (CdS) window layer; back and front contact layers; and a substrate, arranged in either the superstrate (Figure 14a) or the substrate (Figure 14b) configurations. The performance of a CdTe cell is strongly dependent on the morphology of the absorber layer, which is usually affected by the type of substrate used. In superstrate configuration, transparent substrates (glass and polymers) are usually employed to facilitate the transmission of light through the cell, whereas flexible metal foils are used for the substrate configuration. Because the substrate configuration usually yields lower efficiencies, its back contact has been doped with Cu to form a degenerate semiconductor layer in the form of Cu_xTe, Cu-doped ZnTe, HgTe:CuTe-doped graphite paste or As_2Te_3:Cu_5. The high processing temperatures during manufacturing of CdTe cells, however, result in the diffusion of Cu atoms into the adjacent layers, thus, deteriorating their electronic properties (Kranz et al., 2013).

Currently, efforts have been invested in the assessment of the performance of CdTe PVs based on ultra-thin glass (UTG) substrates as a substitute to the traditional metal and polymeric foils. Apart from the high reproducibility of UTG-based CdTe cells, these substrates are highly transparent and resistant to high processing temperatures. UTG substrates offer other advantages such as high mechanical flexibility, lightweight, high surface roughness, and high radiation resistance. Comparatively, polyimides, which may also offer the required optical properties, are limited by the poor thermal characteristics. The use of UTG in CdTe applications is very recent and the first CdTe solar cell on flexible glass was reported by Salavei *et al.* (Salavei et al., 2016). Ever since, there have been numerous attempts to improve the performance of these cells even in flexed conditions. However, the open voltage circuit V_{oc} has been found to drop when the device is flexed with a near-full recovery after release. On the other hand, the closed-circuit current (J_{sc}) and corresponding fill factor (FF) are both enhanced under these conditions leading to an overall boost in efficiency in both compression and tension as shown in Figures 15a, 15, respectively. Again, a further reduction of the bending radius results in the drop of V_{oc} and J_{sc} and the overall efficiency of the cell (Rance et al., 2014). Teloeken *et al.* (Teloeken et al., 2020) suggested that the degradation of photoelectric properties could be attributed to the Cu in the CdTe cells when doped. Specifically, Cu diffuses at a fast rate, thus, promoting charge recombination and degradation of the PV characteristics. On the other hand, doping the cells with elements such as As improves the performance even of very flexed cells because As is a bigger element and, hence, slower diffuser.

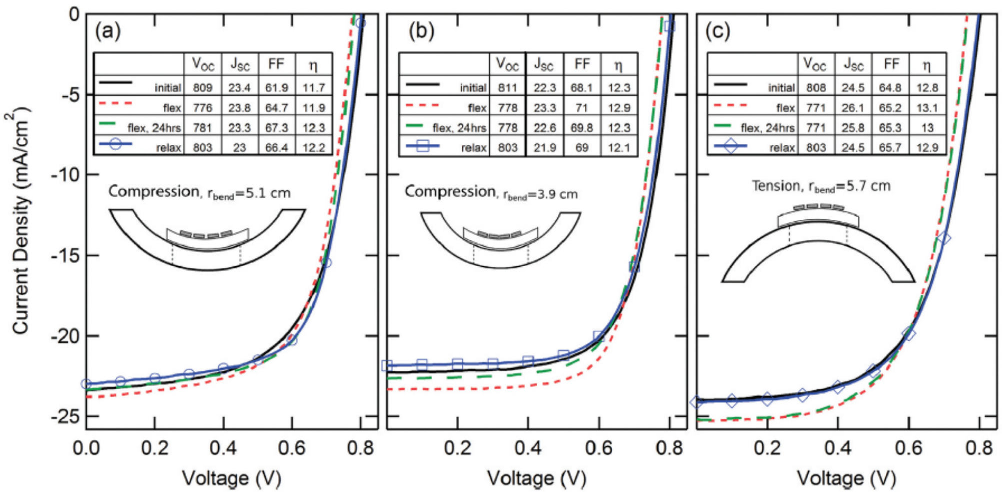


Figure 15. Photovoltaic performance of CdTe devices flexed at (a) 5.1, (b) 3.9, and (c) 5.9 cm bending radii. (Rance et al., 2014).

Another fast-growing poly-crystalline chalcopyrite solar technology is the Copper Indium Gallium Selenide (Cu (In, Ga) Se_2 (CIGS)) thin-film solar PV, a very promising alternative to the well-established crystalline silicon solar cells. CIGS solar cells possess a high optical absorption coefficient, high thermal stability, and an excellent photoelectric conversion efficiency which has reached 21.7% in the last few years (Sim et al., 2018). Like CdTe cells, CIGSs can also be deposited on flexible polyimides and metal foils. Stainless steel (SS) foil has the higher temperature stability, an ideal coefficient of thermal expansion, and is cost-effective. Its high thermal stability facilitates the growth of CIGS absorption layers during processing (W. W.-S Liu et al., 2015). However, the common diffusion of Fe, Ni, Al, and Mn from the substrate foil into the absorber film reduces the conversion efficiency of the cells to 17.7%. Fe, especially, reduces the FF, V_{oc} and J_{sc} by enhancing the high rate of charge recombination.

Cr, SiO_x, Al_2O_3 and the W–Ti used by (Gao et al., 2015) have been used as diffusion barrier elements to restrain Fe diffusion, whereas TiN-barriers are known to prevent tungsten and copper diffusion with Ti also acting as an effective adhesive layer that effectively counters the compressive stresses measured at the SS interface (W. W.-S Liu et al., 2015). These intrinsic compressive stresses between the SS substrate and the back contact usually result in peeling of the layers and the formation of defects due to the stress mismatch (W. W.-S Liu et al., 2015). The introduction of alkali elements such as Na, K, Rb, and Cs as well as Sb and Bi into the CIGS absorber films also enhances the photovoltaic performance of CIGSs (Kawano et al., 2020a). With Bi-doped CIGSs, the CIGS grain size and carrier lifetimes are optimized when the Bi thickness ranges from 20 to 65 nm. However, during high temperature processes (543–572 °C), a high Bi-diffusion is observed leading to the degradation of the photovoltaic properties. Figure Figure 16 shows the variation in the morphology of the CIGS layers as well as the photovoltaic behavior of the cells as a function of the Bi thickness.

Just like with CdTe solar cells, CISG substrates have also been replaced with glasses such as borosilicate and soda-lime glasses (SLG). SLG glasses are preferred to borosilicate glasses due to the suitable coefficient of thermal expansion as well as the favorable CIGS growth which is enhanced by the good Na supply. Unfortunately, SLG is not available in ultra-thin shape, but fairly high efficiencies have been achieved even with the thicker SLG glasses (280 μm) of 19.8%. Yet still, UTG glasses are preferred for the aforementioned advantages. Gerthoffer et al. (2015) demonstrated that CIGS on UTG has improved performance compared with rigid glasses. For a better performance, however, the Na content of the cell setup must be adjusted by doping to account for the lower levels of Na in UTG-based CIGS cells. It is also noteworthy that in this case, the curvature

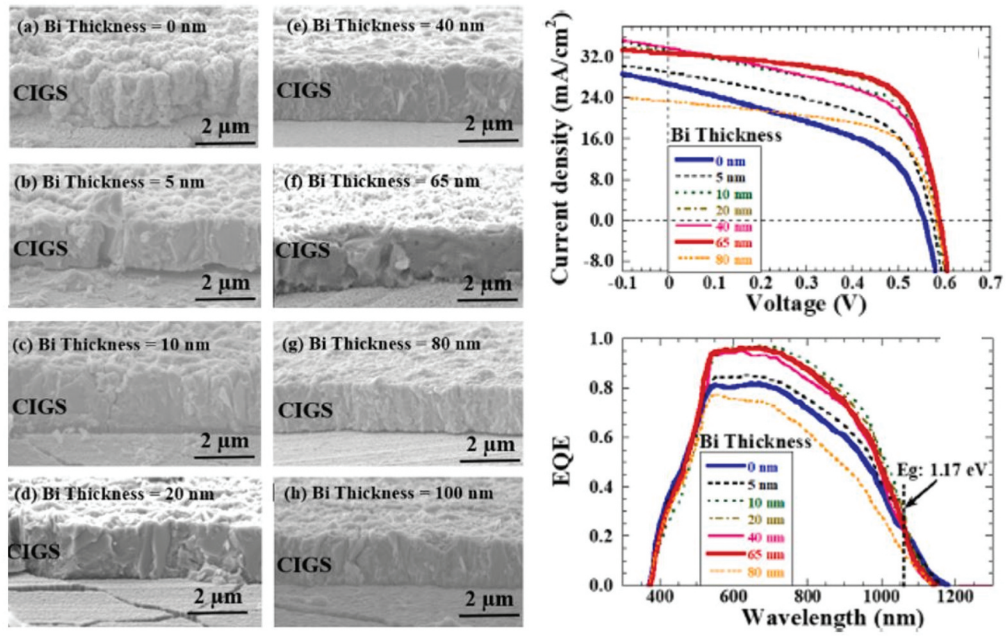


Figure 16. SEM cross-sectional micrographs of the Bi-doped CIGS films on SS substrates (on the left) and the photovoltaic response of the cells (on the right) (Kawano et al., 2020a).

of the cell must not be too pronounced, and the shape of the modules must remain unchanged for maximum performance.

The Ga content in the CIGSs can also be optimized to improve the overall performance of the cells and recent work has focused on the optimization of the CIGS absorber thickness: a 2 to 3 μm-thick CIGS absorber, in fact, increases the overall bandgap of CIGS cell and improves the luminous absorption range even in the long wavelength regions (over 700 nm), but reducing the thickness to less than 2 μm minimizes the raw material usage and, therefore, cost (Kawano et al., 2020b). Another proposed way of enhancing the performance of CIGSs involves the use of graphene and its derivatives as conductive films due to the superior electrical and optical properties, as well as mechanical flexibility and bending durability. The drawback, however, is that this method requires that the graphene films be grown on a copper foil and then transferred onto the desired substrates (as shown in Figure Figure 17) which often results in the formation of defects and imperfections, thus, active sites for recombination (Sim et al., 2018). The CIGS solar cells fabricated with graphene/Cu foil have yielded promising results, a power conversion efficiency of 9.91% with J_{sc} of 28.84 mA/cm^2, V_{oc} of 0.531 V and FF of 64.75%, and more work is recommended for possible consideration in future applications.

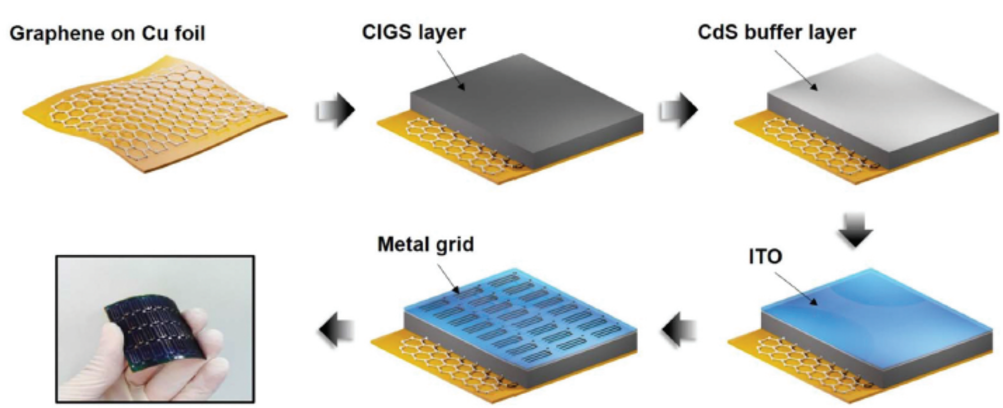


Figure 17. Schematic representation of the fabrication of a flexible CIGS cell on a graphene/Cu substrate. (Sim et al., 2018).

3.3. Flexible solar cells based on nanocomposites of 2D materials

Nanotechnology offers a lot of promise when it comes to harvesting solar energy efficiently with photovoltaic cells. Furthermore, nanotechnology has arisen as a multidisciplinary field with rising importance in other technical fields. When doped with silicon for inorganic cells and polymer for organic cells, nano particles operate as semi-conductive constituents of PV cells (S. A. Khan & Rahman, 2019). Several different conductive materials have been investigated based on their economic, flexural reliability and enhanced performance as transparent conducting conductor. Some include metal-NWs and grids, conjugated polymers, CNTs and graphene, etc. The common hurdle with these materials is the low intrinsic electrical conductivity which impedes their optoelectronic performances. Nanowires, unlike their mesh siblings, have nanometer-scale conducting networks that completely remove most of this interference. Another class of 2D materials, MXene, commonly made of carbides and nitrides, has attracted attention recently, credit to the exceptional characteristics similar to graphene (H. Wang et al., 2018). This material applications include energy storage and solar cells. Tang *et al.* (Tang et al., 2019) released a work on the fabrication of conductive, transparent, and flexible MXene/silver nanowire (AgNW) hybrid films, which received the highest figure of merit (162.49) in literature to date. The hybrid films recorded a low roughness, excellent electrical conductance and optical transmittance as well as superior flexing performance, thanks to a simple and scalable solution-processed approach as shown in Figure 18a. Following the film production, the hybrid electrodes were shown to work as transparent electrodes in organic photovoltaics (OPVs) using the fullerene molecule PTB7-Th:PC71BM and the nonfullerene molecule PBDB- T:ITIC. A ternary structure of PBDB-T:ITIC:PC71BM was exhibited in an effort to increase the performance of flexible OPVs, which resulted in a PCE of 8.30%. Following a bending radius of ~ 5 mm for the ternary flexible organic PV structure, the power conversion efficiency was only 2% less retaining ~ 98% of the original PCE (Figure 18b). The flexible ternary OSCs were capable of keeping 84.6% (see Figure 18c) of the initial PCE after 1000 bending and unbending cycles using a bending radius of 5 mm, according to mechanical characteristic tests.

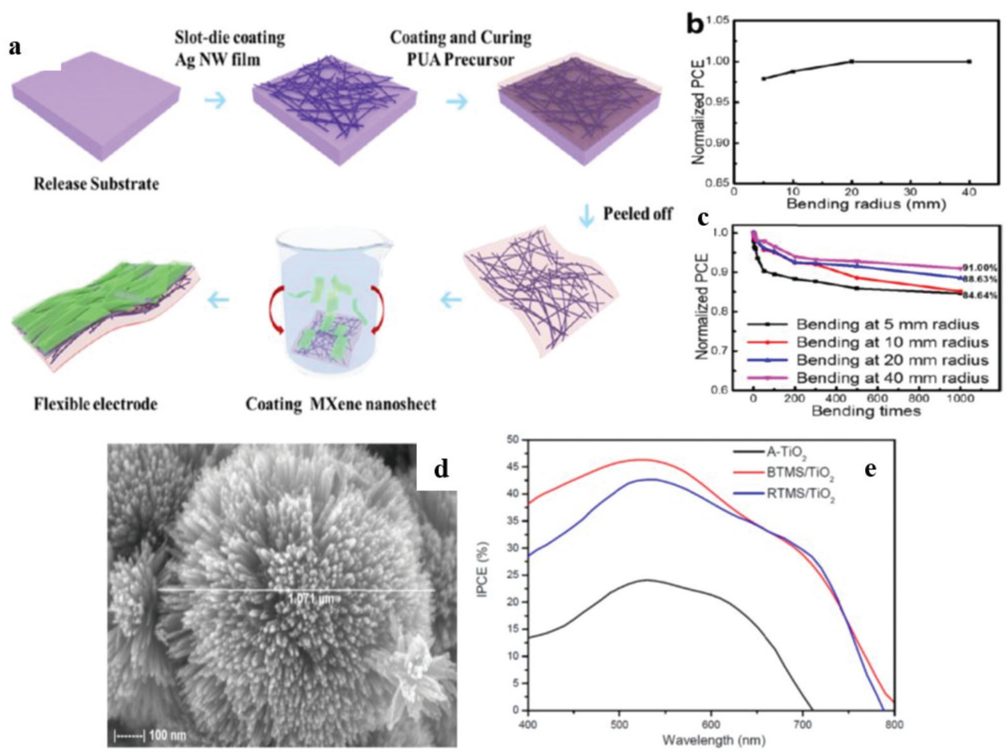


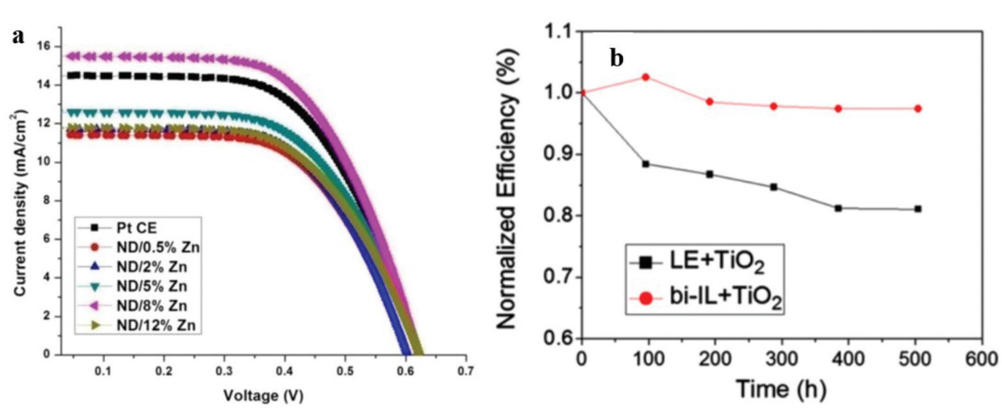
Figure 18. (a) Fabrication Process of MXene-Based Flexible Transparent Electrode. (b) Bending radius PCEs of flexible PSCs, (c) Bending radius-Bending cycle PCE of flexible PSCs. (Tang et al., 2019). (d) FESEM image of $BaTiO_3$ microspheres. (e) IPCE of anatase TiO_2 nanoparticles BTMS/TiO_2, RTMS/TiO_2 nanocomposites based DSSCs. (Baijua et al., 2021).

There has been a lot of effort on developing low-cost Pt-free dye-sensitized solar cells (DSSCs). Gurulakshmi and his colleagues (Gurulakshmi et al., 2019) presented a Pt-free carbon nanocomposite that could be used as a counter electrode catalyst for DSSCs. Graphene oxide (GO) was irradiated with Xe light source to produce reduced graphene oxide (SSrGO). The effectiveness of SSrGO and single walled carbon nanohorns (SWCNH) coated FTO glass substrate as a Pt-free composite CE for DSSC were examined, and it demonstrated equivalent catalytic activity to Pt. The recorded PCE of the test cell and the DSSC module was ~8.27% and 5.18%, respectively, (Baijua et al., 2021). During a hydrothermal process, a rutile phase TiO_2 intermediate was changed into tetragonal Barium titanate microspheres (BTMS), according to crystal structure and morphological investigations (Figure 18d). BTMS-TiO_2 nanocomposite based flexible DSSC photoanode recorded a PCE of 4.53% compared to pure anatase TiO_2 nanoparticles based flexible DSSC photoanode (PCE = 2.66%). This performance was attributed to the reduced charge recombination and superior light scattering ability at the interface of film/electrolyte by the ferroelectric dipoles of $BaTi_3$. BTMS composite has demonstrated an enhanced performance as shown in the incident-photon-to-current efficiency (IPCE) plot in Figure 18e, making it a promising material for flexible DSSC applications.

To improve the energy conversion efficiency of OSCs, Khan *et al*. developed and integrated a film of ZnO/PVA nanocomposite (A. S. Khan et al., 2021) for its application. The nano films were synthesized with different concentrations of ZnO nanoparticles in a PVA matrix using a solution casting process. The OSC was used to examine each nanocomposite material independently. The efficiency of OSCs changes significantly before and after the ZnO/PVA nanocomposite coating is applied. Before and after applying the nanocomposite layer, the solar cell recorded an optimum efficiency of 10.07% and 13.57%, respectively. The increased efficiency of OSCs demonstrated that the ZnO/PVA nanocomposite film has tremendous promise for improving the efficiency of organic solar cells. Chen research team (Chen et al., 2011) fabricated a photoanode consisting of an array of TiO_2 nanotubes (TNT) filled with a nanocomposite of TiO_2 (P90) and nanographite and used to build a flexible DSSC. TNT arrays were created via anodic oxidation of Ti foil, and this Ti foil incorporating TNT served as the DSSC photoanode. The average diameter of each tube in the array is 100 nm. The light-to-electricity conversion efficiency of the DSSC with the nanocomposite photoanode (photoanode labelled as Graphite/P90-TNT) was 5.75%. Cells using simple TNT photoanodes (photoanode labelled as TNT) and TNT filled with P90-TiO_2 photoanodes (photoanode identified as P90-TNT) recorded efficiencies of 4.44% and 5.14%, respectively. The improved efficiencies in favor of the cells containing P90-TNT and Graphite/P90-TNT were due to the filled P90 and nanocomposite, respectively. More conductive electron transfer pathways and a longer lifetime for electrons in the TNT film were anticipated to be provided by the filled particles.

As a potential substitute for platinum, a new nanodiamonds (NDs)/zinc (Zn) nanocomposite has been studied by Fayaz *et al* as CE in a flexible DSSC system (Fayaz et al., 2019). The CE was made using a simple drop casting procedure on an indium tin oxide (ITO) covered Polyethylene

Figure 19. (a) *I–V* performance of ND/Zn nanocomposite-based CE (Fayaz et al., 2019). (b) Normalized efficiency of full-plastic DSCs based on MPN-based and bi-IL based liquid (H. H.-F Lee et al., 2013).



terephthalate (PET) substrate. The effects of different Zn concentrations in the optimized NDs layer were studied. When compared to Pt-based CE at optimal layer thickness, the optimized layer of NDs showed 78.52%, which was a similar performance with Pt-based CE. The addition of Zn nanoparticles increased the catalytic activity even more, making it comparable to or even better than the performance of the Pt-based reference CE. When compared to a Pt-based reference system, a 6.23% improvement in performance was recorded at an 8% Zn concentration as shown in the *I–V* curves in Figure 19a. Due to agglomeration of nanoparticles and charge trapping sites, increasing the concentration reduces performance. Based on nanocomposite gel electrolytes, Lee and his group (H. H.-F Lee et al., 2013) created efficient and reliable full-plastic DSCs. To create TiO_2 photoanodes on indium-doped tin oxide coated polyethylene naphthalate, several compositions of nanocrystalline TiO_2 pastes with no binder were combined with a mechanical compression approach. The compressed photoelectrode, which was made up of commercial titanium powders P25 and 100 nm TiO_2 powders together with a TiO_2 solidified MPN-based nanocomposite gel electrolyte, had an energy conversion efficiency of 6.49%, according to the findings. Furthermore, DSCs that was based on TiO_2 solidified binary ionic liquid (bi-IL) electrolyte were shown to have greater endurance than MPN-based nanocomposite gel electrolyte, retaining >97% of the initial efficiency (see Figure 19b) after 500 hours of aging testing at 60 °C under continuous light irradiation (100 $mWcm^{-2}$).

3.4. Mechanical properties of 2D nanomaterials on flexible solar cells

The mechanical properties of 2D nanomaterials adopted in flexible solar cell fabrications greatly influence the bending performances and power conversion efficiencies of the devices fabricated from them. These properties include the Young modulus, Poisson ratio, adhesion energy as well as bending strain cycles. Prior work by (Ichwani et al., 2022; L. Li et al., 2022; Ma et al., 2021)have reported improved bending cycles, high efficiency, Young modulus and stable Poisson ratios (~0.3). Whereas conducting polymers and flexible substrates including polyethylene terephthalate (PET), polydimethysiloxane (PDMS) (Asare, Agyei-Tuffour, et al., 2020), poly-3-hexylthiophene (P3HT), poly-3,4-ethylenedioxythiophene-polystyrene-sulphonate (PEDOT.PSS) as well perovskite absorber layers, methyl ammonium lead iodide ($MAPbI_3$) and formamidinium lead iodide ($FAPbI_3$) as presented in Table 2 have recorded lower Young modulus of ~6.02, ~1.42 GPa and ~17.8 GPa and ~0.3 poisson ratio, the 2D graphene and TMDCs such as MoS_2 and WS_2 have relatively high Young moduli; ~1 TPa and ~270 GPa respectively, (Agyei-Tuffour et al., 2017, Jiang et al., 2022) and could be susceptible to easy fracture however they also possess increased interfacial toughness to withstand the multiple bending cycles (Souza et al., 2021, Ichwani et al., 2022). In their few layer thicknesses, ~5–25, they even possess much higher modulus of ~ 330 GPa with low poisson ratio of ~0.25–0.27. This is at variance with the conducting polymers which have low moduli and can allow deformation under different loading angles. The films are thinner and even in the presence of inter-locking hard particles (dust with E = ~70 GPa), they can deform around them to achieve superior interfacial surface contact that facilitates charge transport (Agyei-Tuffour et al., 2016, 2017). The bending cycles also vary significantly from ~100–10,000 cycles for Ti/Al foil nanotubes (TNT)/CNT to tandem PET/ITO/MB-NiO/WBG perovskite/C60/ALD-SnO_2/Au/PEDOT:PSS/NBG perovskite/C60/BCP/Cu devices, (L. Li et al., 2022). The graphene-based device recorded 2000 bending cycles whereas the AgNWs based device recorded ~5000 bending cycles (Table 2). This synergetic relationship of the optoelectronic, mechanical and electrical properties that exist in 2D nanomaterials allow their flexibility with great tolerance and with the deformations at the inner and outer surfaces which are considered as tiny compression and tension deformations (Jiang et al., 2022). By varying the thicknesses, direct and indirect bandgaps can be generated in the 2D materials leading to enhanced optical absorption phenomenon that is requirement in solar cell performance. These tunable properties, favorable mechanical strength make 2D nanomaterials reliable candidates for flexible solar cell devices.

4. Challenges and future prospects

It is noteworthy that whereas 2D nanomaterials for flexible solar cells using inorganic materials such as CdTe, CIGS, CIS have already found industry applications, the others based on organic and conjugated polymers, perovskites and dye sensitize solar cells will require further improvements in their long-term stabilities and efficiencies to make industrial impacts. These are clearly areas that require much attention with novel synthesis and fabrication techniques if flexible perovskite and organic solar cells with 2D nanomaterial components will move from the laboratory to the industrial scale. The synthesis methods of 2D nanomaterials will require the development of techniques that are sufficiently simple to achieve economic value and also preserve the properties of the parent materials. Future developments to realize the potential of these nanomaterials in flexible solar cell fabrications will require sophisticated materials characterization to establish their structure-property relationships. The prominent amongst the hurdles include the high precipitation rate of graphene and carbon nanotubes when good dispersion is usually required, their biodegradability, toxicity and biocompatibility require attention. Though graphene dominates the 2d nanomaterials for flexible solar cells, other materials such as the Mxenes hold future prospects.

Secondly, the Transition metal dichalcogenides (TMDs) though have great potential in flexible solar cells. They are difficult to synthesize in large scales, which means they will take much more time and cost compared to commercial materials. Compared to other 2D materials, TMDs have various compositions with versatile properties that are suitable for different fields. Nevertheless, there remain several hurdles to be addressed including material synthesis and device fabrication. The realization of flexible solar cells requires flexible substrates such as polymers, and the device thickness is usually at the micrometer scale. This brings challenges for the extensive production of TMD thin films with high quality and the maintenance of device reliability. Again, even though researchers have explored alternative approaches to directly grow TMDs on flexible substrates by plasma, magnetron sputtering, and laser beam methods. These techniques can achieve the crystallization of TMDs on flexible substrates with low damage fabrication. Nevertheless, they are not appropriate for extensive manufacturing. With the numerous studies having been published on TMD flexible solar cells, majority are on MoS_2-based. Hence, the exploration of other TMDs (beyond MoS_2) will contribute greatly to the field. For instance, the Janus TMDs have layered structures with similar electrical and mechanical properties as the normal TMDs. Though TMDs have proved their excellent feasibility in various flexible solar cell applications, the exploration of other immature and new areas should be explored including flexible displays, flexible antennas, solar wearable thermoelectric generators, etc. Lastly, though the challenges in practical use and industrial production of flexible devices based on TMDs still exist, it should be highlighted that more achievements will be obtained with continuous research.

Thirdly, the most efficient flexible organic, perovskite solar cells (PSCs) employ Spiro- OMeTAD as a hole-transport layer (HTL). However, it does not ensure device reliability and long-term stability under a damp-heat condition. Despite much effort, there are only a few candidates that meet the rigorous long-term stability requirements at the cost of lower PCE. Thus, developing a HTL that can meet both efficiency and stability standards by considering the following requirements is necessary for future works dopant-free HTMs with high mobilities, enhanced interfacial interaction between the HTM and the perovskite surface, improved thermal stability of the HTM itself, and modification of the HTL architecture would go a long to improve reliability.

5. Conclusions and summary remarks

The review article has presented a comprehensive thin film photovoltaic devices based on 2-dimensional (2D) nanomaterials. The 2D materials including graphene, metal nanowires, conducting polymers, carbon nanotubes, and transition metal dichalcogenides have shown very high performances in bending stabilities with ~90% of its power conversion efficiencies (PCEs) when used in different solar cells. The power conversion efficiency (PCE) of ~8.31%, current density of ~ 9.56–9.37 $mAcm^{-2}$, fill factor range of ~57–64% after a 100 bending cycles with an ~0.75 cm bending radius. Devices made from tandem 2D perovskite layers have achieved ~ 24.7% efficiency and specifically large devices of ~1.05 cm^2 has recorded 23.5% after 10,000 bending cycles and 15 mm radius. Graphene for example is a lightweight and mechanically robust 2D nanomaterial often adopted in flexible solar cells. The degradation mechanisms, mechanical and opto-electronic characterization of 2D-based flexible photovoltaic (PV) cells has been highlighted for inorganic, organic and perovskite solar cells. The electrical characterization, robustness and mechanical flexing of flexible PV cell structures fabricated specifically using 2-D nanocomposites and graphene sheets are analyzed and presented.

TMD-based flexible electronics have been reviewed. Starting from the material, the structure, physical properties, and synthetic routes of TMDs have been introduced, and the feasibility of TMDs as promising candidates for flexible electronics has been discussed. Compared to bulk materials, monolayer and few-layer TMDs present different photoelectric properties due to the different charge transport mechanisms, including the opening of the bandgap and the indirect/direct bandgap transition. Besides, 2D materials also exhibit better flexibility and are suitable for printing, casting, screening, or even constructing 3D configurations.

The review has highlighted the need to transition current experiments on 2D nanomaterials based flexible solar cell fabrications into scale up and commercialization despite the challenges identified for industry players and researcher to explore solutions to the challenges to achieve robust and highly efficient solar devices.

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