

2D Materials for Wearable Energy Harvesting

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The growing demand for energy in wearable sensors and portable electronics necessitates the development of self-contained, sustainable, and mobile power sources capable of harvesting environmental energies. Researchers have made significant strides in implementing photovoltaics, thermoelectrics, piezoelectrics, and triboelectrics in 2D materials. This has resulted in significant advancements in wearable energy harvesting systems based on 2D materials. This review discusses the relationship between synthesis procedures, material structures/properties, and device performance in the context of 2D materials-based wearable energy harvesting technologies. Finally, challenges and future research opportunities are identified and discussed based on current progress.

1. Introduction

With the rapid development of wearable electronics, portable gadgets, and the Internet of Things (IoT), much work has gone into developing sustainable, mobile, and self-sufficient energy sources to fuel the operations of connected technology.^[1–7] Harvesting ambient energy and converting it to electricity has emerged as a promising strategy in this regard. Solar energy, thermal energy from body heat, and mechanical energy from human activities are all examples of energy acquired from the

environment.^[8–13] Leveraging the fundamental science and understanding of photovoltaic, thermoelectric, piezoelectric, triboelectric, and other effects, various technologies have been developed to target different types of energy harvesting.^[14–16] Innovations in materials design and energy harvesting have substantially increased the device's performance, thanks to recent advances in nanotechnology and nanomanufacturing.^[17–20] The first piezoelectric nanogenerator (PENG) and the first triboelectric nanogenerator (TENG) were developed in 2006 and 2012, respectively, ushering in a new era of low-frequency energy harvesting, while the

realization of 2D materials with controlled structures and properties allows photovoltaic cells and thermoelectric harvesting to be optimized.^[9,21–24] Other innovative technologies have been proposed to capture energy in various forms or via various methods (Figure 1).^[16,25–27]

The growing interest in 2D materials and substantial studies into their properties and applications have accelerated the growth of energy harvesting with 2D materials. Atomically thin, layered 2D materials with intriguing electrical, optical, thermal, and mechanical properties, such as graphene and graphene-based materials, 2D black phosphorus (BP), transitional metal dichalcogenides (TMDCs), and transitional metal carbide, nitride, or carbonitride (MXene), offer a broad spectrum of solutions in materials science and are promising candidates for energy application technology.^[28–34] For instance, the superior mechanical strength and flexibility of 2D materials perfectly fit the durability and adaptability requirement for wearable energy harvesting.^[35,36] The intriguing band structures and charged carrier transport features that result from the quantum confinement effect in 2D materials provide appealing electrical and optoelectrical performance.^[37–41] The structure- and chemistry-dependent features of 2D layered materials allow scientists to adjust the materials' structure and chemistry for precise control over the materials' properties and to maximize device performance.^[42–44] 2D materials have shown tremendous potential for wearable energy harvesting.^[45–47]

A few reviews were published summarizing the recent development of 2D materials for wearable energy harvesting.^[48–50] For example, Fan et al. discussed the recent progress of monolayered 2D materials-based energy harvesters.^[48] Yang and Lee summarized the application of layered 2D materials in piezoelectric and triboelectric energy harvesting.^[49] The use of layered 2D materials in PENGs and TENGs was also discussed by Han et al.^[50] However, the scope of the aforementioned reviews is limited to specific types of materials or energy harvesting mechanisms. This article summarizes and discusses the progress of 2D materials and their applications in wearable energy harvesting. To provide a foundation for understanding

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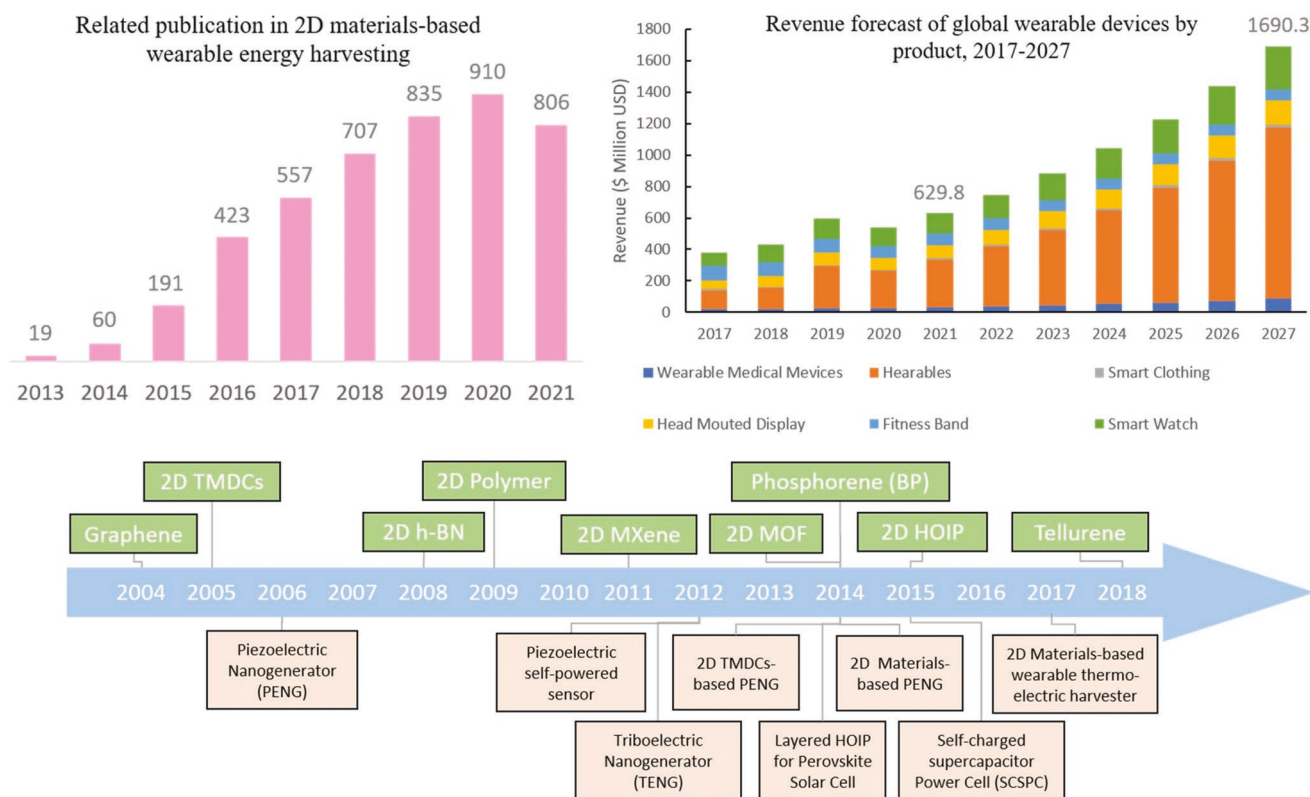


Figure 1. Top left: related publications count in 2D materials-based wearable energy harvesting. Source: Web of Science. Search index: TOPIC: 2D materials and TOPIC: wearable energy harvesting. Top right: global wearable device revenue forecast by product type from 2017 to 2027. Bottom: historical timeline of experimental realization of 2D materials and development of wearable energy harvesting techniques after the discovery of graphene.^[21,76,129,281,361,362,431,967,997,1168,1177–1182]

how the structure of 2D materials may affect their electrical, thermal, optical, and mechanical properties, we begin in Section 2 by examining their characteristics, biocompatibility, and cytotoxicity. Section 3 introduces the widely documented top-down and bottom-up approaches to the preparation of 2D materials, demonstrating the connection between the synthetic process and the structure of 2D materials. The review's fourth section examines a variety of energy harvesting technologies, including photovoltaic cells, thermoelectric harvesters, PENG, TENG, and other emerging approaches, with a particular emphasis on the integration of 2D materials-based wearable energy harvesters with wearable electronics and sensors. We are primarily interested in works that demonstrate wearability or have a great potential to do so. Finally, in the third section, we will highlight current research challenges, such as large-scale 2D material synthesis and harvester integration with energy storage devices, as well as potential future applications of 2D materials for wearable energy harvesting.

2. 2D Materials and Their Properties

The properties of 2D materials, which typically have a monolayer thickness to a few layers and a lateral size of several tens of microns, differ dramatically from those of bulk materials.^[51–53] Strong in-plane chemical bonds and weak out-of-plane

van der Waals interactions are common in 2D materials with layered structures. This opens up fascinating research in heterostructures generated by 2D materials stacking and quantum phenomena like this Dirac fermions and quantum spin Hall effect.^[38,54–59] In this section, we will review the qualities of 2D materials relevant to wearable energy devices and the relationship between structure and properties.

2.1. Graphene

Graphene consists of single-layer carbon atoms packed into 2D hexagonal lattices through sp^2 hybridized π bonds with an average bond length of 1.24 Å (Figure 2a).^[60,61] Graphene's strong in-plane elastic stiffness (340 N m⁻¹) and breaking strength (42 N m⁻¹) are due to the sp^2 bonding between two neighboring carbon atoms.^[62,63] Theoretical calculations and actual measurements indicated that graphene has a high Young's modulus of 1 TPa and inherent tensile strength of 130 GPa, the highest of any known material values.^[63–65] Such high stiffness and robustness make graphene a good candidate for wearable devices and sensors.^[66–70]

Graphene offers remarkable electrical properties due to its linear dispersed band structure near the Dirac point, and its Fermi level may be easily modified via electrostatic doping.^[62] Novoselov and co-workers initially reported high carrier

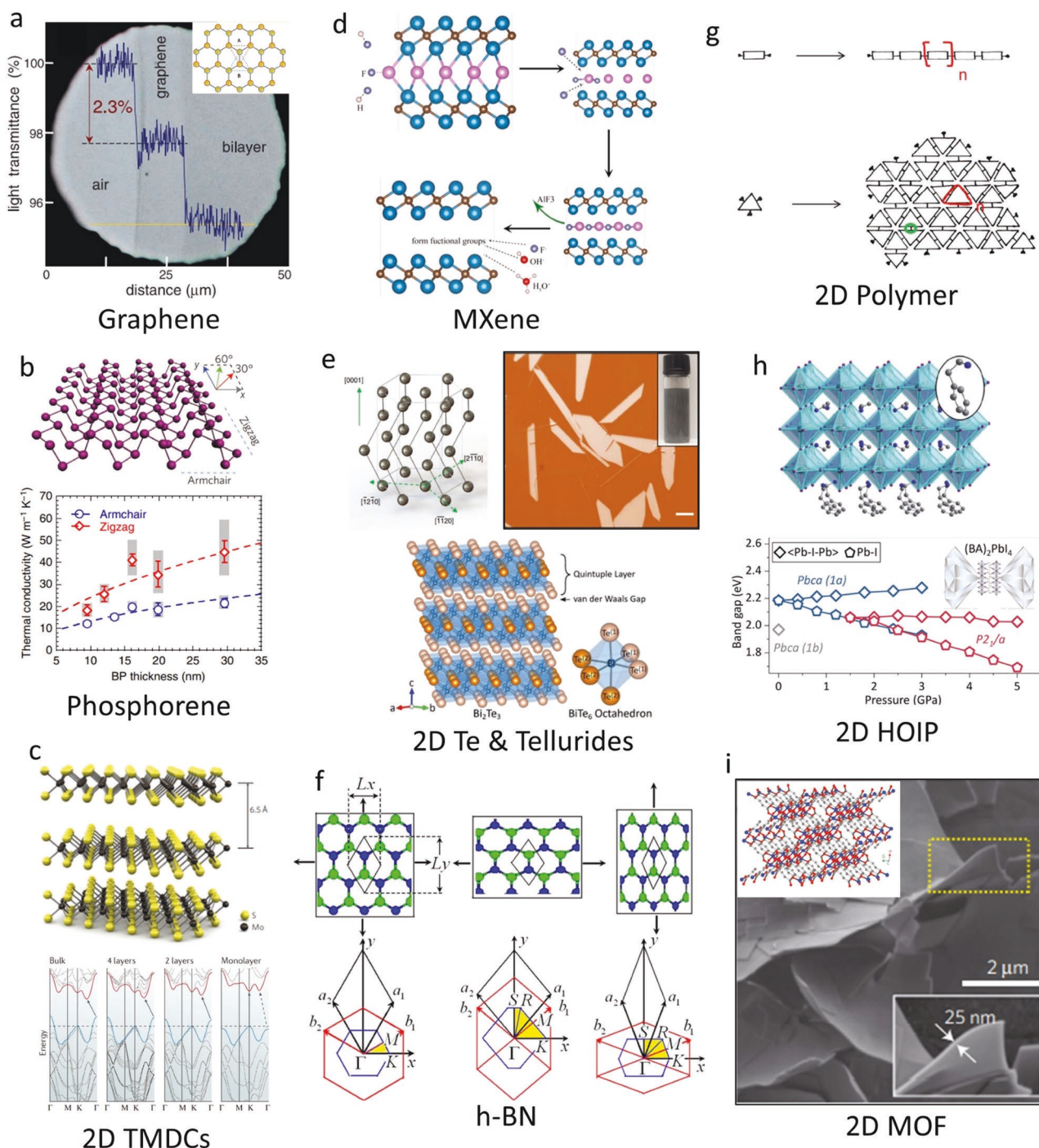


Figure 2. Structure and properties of 2D materials. a) Photo of a partially covered 50- μm aperture by monolayer and bi-layer graphene and the corresponding light transmittance along the yellow line. Reproduced with permission.^[93] Copyright 2008, American Association for the Advancement of Science. Inset: schematic of graphene lattices and sub-lattices showing interpenetrating graphene sub-lattice with the atomic sites of sub-lattice defined by the center of another sub-lattice. Reproduced with permission.^[62] Copyright 2010, Elsevier. b) Top: schematic showing atomic structure of monolayer 2D BP with crystal orientation. Reproduced with permission.^[123] Copyright 2015, Springer Nature. Bottom: in-plane thermal conductivity of 9.5 nm thick BP along the armchair and zigzag directions. The dash lines are linear fits while the grey rectangles represent the uncertainty of thermal conductivity of the substrate used in the measurement. Reproduced under terms of the CC-BY license.^[156] Copyright 2015, The Authors, published by Springer Nature. c) Top: schematic illustration of MoS_2 structure. Reproduced with permission.^[1183] Copyright 2011, Springer Nature. Bottom: calculated band structure of 2H- MoS_2 and transition of indirect to direct bandgap from bulk to monolayer. Reproduced with permission.^[159] Copyright 2017, Springer Nature. d) Schematically illustration of MXene fabrication by etching and exfoliation of MAX phases. Reproduced with permission.^[261] Copyright 2012, American Chemical Society. e) Top: schematic atomic structure of Te (left) and optical microscope image of solution-grown 2D Te nanoflakes (right). Inset is an image of Te nanoflakes dispersion. Scale bar indicates 20 μm . Reproduced with permission.^[281] Copyright 2018, Springer Nature. Bottom: schematical

mobility of $10\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for few-layer graphene, while May-rov et al. later claimed carrier mobility of $250\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for graphene enclosed in an h-BN substrate.^[71–76] Additionally, theoretical calculations revealed that the electrical conductivity of few-layer graphene is temperature and layer count dependent.^[77–79] Fang et al. demonstrated that graphene conductivity increases with increasing temperature and decreasing layer count, implying a negative temperature coefficient of resistance and increasing surface channel conductivity of multi-layer graphene, respectively.^[77] Graphene has been reported as a semimetal with zero bandgap due to overlapping the conduction band minimum (CBM) with the valence band maximum (VBM) at the K point of the Brillouin zone.^[76] However, it is necessary to create a bandgap in graphene to construct graphene-based electronic devices.^[62] The band structure of graphene can be modified by introducing defects (sp^3 hydrocarbon and dopant, for instance) into the honeycomb lattices,^[62,80–82] or by applying uniaxial strain through graphene–substrate heterojunction to distort graphene unit cells.^[62,83,84] While previous predictions indicated that a large global strain (>20 percent) is required to open a bandgap in graphene, recent calculations indicate that a tunable bandgap can be formed by forming a van der Waals heterojunction between graphene and substrates such as PtSe_2 , $\text{g-C}_3\text{N}_4$, and Ga_2SSe , and that a bandgap of 1.7–2.1 eV can be created by laser shock imprinting the e-beam fabricated graphene.^[60,85–88]

Single-layer graphene demonstrates a broad absorption spectrum extending from the visible to the near-infrared.^[89–91] Nair and co-workers were the first to report a constant absorption coefficient of 2.3% due to graphene's unique electrical structure and a negligible reflection of less than 0.1% (Figure 2a).^[92,93] Due to single-layer graphene's high absorption coefficient, the one-atomic layer thick nanosheet may be observed with the naked eye.^[93,94] Additional measurements demonstrated that the opacity is wavelength-independent and increases linearly with the number of graphene layers.^[62,93]

Due to the bonds produced by the unoccupied $2p$ orbitals, graphene exhibits remarkable thermal characteristics.^[64] Balandin et al. first reported a room temperature in-plane thermal conductivity of up to $5300\text{ W m}^{-1}\text{ K}^{-1}$ on suspended, single-layer graphene, and Ghosh and co-workers reported a long phonon mean free path of 775 nm at room temperature on Si/SiO₂ supported graphene.^[95,96] Although subsequent research indicated a considerable fall in graphene's thermal conductivity when supported on substrates, e.g., $600\text{ W m}^{-1}\text{ K}^{-1}$ when supported and encased on Si/SiO₂ substrate, supported graphene's thermal conductivity remains significantly better than that of Si ($145\text{ W m}^{-1}\text{ K}^{-1}$) and Cu ($400\text{ W m}^{-1}\text{ K}^{-1}$).^[97–101] On the other hand, a substantial study has been conducted on graphene's

size-dependent thermal conductivity.^[100,102–104] Ghosh et al. demonstrated that when the number of layers in graphene rose from two to four, the thermal conductivity decreased from 2800 to $1300\text{ W m}^{-1}\text{ K}^{-1}$.^[102] The decrease in κ value was attributed to the cross-plane coupling of the low-energy phonon and the growing phonon Umklapp scattering, both of which are quenched as graphene is thinned to monolayer thickness.^[97,102] Additionally, the κ value depends on the length of suspended graphene.^[100,102] Wang et al. also reported an 85% reduction in heat conductivity as the graphene length was decreased from 5.0 to 1.0 μm .^[100] Further research done by Xu et al. revealed that the κ value increases with the length (L) of the graphene nanosheet at a $\approx\log L$ scale.^[103] This result was further explained by the molecular dynamic simulation's mix of reduced dimensionality and phonon population displacement under nonequilibrium conditions.^[103]

In summary, graphene's fascinating features, such as its high stiffness, high carrier mobility at room temperature, high thermal conductivity near room temperature for both suspended and supported graphene, and broad adsorption spectrum, make it well-suited for use in wearables, nano-electronics,^[96,105] thermal interface materials,^[106] and solar cells.^[94,107]

2.2. Black Phosphorus

Black phosphorus was first discovered in 1914 when Bridgman transformed white phosphorus to bulk black phosphorus at elevated temperatures and pressures.^[108,109] However, the first 2D black phosphorus was not reported in the literature until early 2014.^[110–113] Black phosphorus (BP) in a single layer, often known as “phosphorene,” is made up of puckered hexagonal lattices (Figure 2b).^[111,113] Each phosphorus atom covalently bonds to three adjacent atoms with a bond length of 2.18 Å, an average bond angle of 102° , and an inter-layer P–P distance of 5 Å.^[111,113–115] This puckered hexagon lattice exhibits a high degree of intrinsic structure anisotropy, with two highly symmetric in-plane directions, commonly referred to as the armchair direction (along the a -axis, sometimes referred to as the “ x -direction”) and the zigzag direction (along the b -axis, sometimes referred to as the “ y -direction”), that can be identified.^[116–120] Additionally, the structural anisotropy results in anisotropy in the electrical, thermal, mechanical, and optical characteristics of BP, all of which display markedly different behavior in the armchair and zigzag orientations.^[121–124]

Black phosphorus is an intrinsic p-type semiconductor with a direct, thickness-dependent bandgap.^[111,125,126] Calculations projected a 2 eV direct bandgap at the Γ position of the first Brillouin zone,^[127–130] and as the thickness of phosphorene increases, the

illustration of quintuple-layer structured Bi_2Te_3 . Reproduced with permission.^[312] Copyright 2019, Wiley-VCH. f) 2D h-BN under different strain distribution and the corresponding primitive unit cell (black), reciprocal lattice (red) and the Brillouin zones (blue): symmetric along both perpendicular (L_x) and parallel (L_y) to the B–N bonds (left), asymmetric along the L_x direction (middle) and asymmetric along the L_y direction (right). The irreducible Brillouin zone is indicated in yellow. Reproduced with permission.^[340] Copyright 2008, American Institute of Physics. g) Schematic sketch of repeating units of 2 (top) and 3 binding sites (bottom) forming 1D linear polymer chain and 2D polymer, or “molecular fisherman's net” structure, respectively. Reproduced with permission.^[364] Copyright 2016, The Royal Society of Chemistry. h) Top: schematically showing the structure of 2D layered perovskite $(\text{PEA})_2(\text{MA})_2[\text{Pb}_3\text{I}_{10}]$. Reproduced with permission.^[1182] Copyright 2014, Wiley-VCH. Bottom: bandgap and phase transition of 2D perovskite $(\text{BA})_2\text{PbI}_4$ as a function of applied pressure. Reproduced with permission.^[402] Copyright 2018, American Chemical Society. i) SEM image of 2D CuBDC MOF nanosheet. Bottom right corner: showing a 25 nm thick CuBDC MOF nanosheet. Upper left corner: schematic 3D structure of CuBDC MOF. Blue, red and gray spheres represent the copper, oxygen, and carbon atoms. Reproduced with permission.^[432] Copyright 2014, Springer Nature.

bandgap lowers to 0.3 eV, while the direct bandgap shifts to the Z point.^[111,131,132] The bandgap of BP can be further tweaked by adding dopant, an electric field, and strain, effectively converting it to a semimetal.^[131,133–136] Deng et al. demonstrated the ability to tune the bandgap of BP thin films with varying thicknesses using an electric field.^[134] Due to the dielectric screening effect, the bandgap of a 10 nm thick BP film can be lowered to 50 meV when an external displacement field of 1.1 V nm⁻¹ is applied.^[134] On the other hand, Kim et al. reported in situ doping of potassium atoms as the charge donor in BP.^[133] BP can be converted to a Dirac semimetal via a band-inversion transition when strongly doped with potassium.^[133] Koenig and co-workers further reduced the bandgap of few-layer BP to 0.09 eV by doping Cu adatoms onto it and changing it from p-type to n-type (room temperature electron mobility of 380 cm² V⁻¹ s⁻¹).^[136] Due to the anisotropy of BP's in-plane structure, theoretical calculations projected that its carrier mobility along the armchair direction would be 1.5 to 3 times that along the zigzag direction, leading to a lower effective mass in the *x*-direction.^[119,137] The experimental results corroborated the calculation: high charged carrier mobilities of 1000 and 600 cm² V⁻¹ s⁻¹ were measured along the armchair and zigzag directions, respectively, on 15 nm thick BPs at 120 K, which corresponds to a conductance anisotropy of 63.7%.^[113,119,128] Additionally, when BP is supported on, sandwiched between, or encapsulated within h-BN substrates, more significant charge mobility (2000 to 4000 cm² V⁻¹ s⁻¹ at low temperatures) and an increased on/off ratio (above 10⁵ at ambient temperature) can be attained.^[138–140]

Due to the puckered hexagonal lattices, BP displayed high direction-dependent adsorption.^[141–143] Lan and co-workers determined and extracted the anisotropic optical characteristics of BP by using incident light polarized in the various directions of the material.^[143] A substantial increase in adsorption was detected along the armchair direction (*a*-axis), indicating a greater phonon-matter interaction along the *a*-axis.^[143] Mao et al. also reported few-layer BP with optical anisotropy in the visible range using polarized optical microscopy.^[141] The armchair direction exhibited greater adsorption, consistent with earlier findings.^[141] Additionally, the optical characteristics of BP are greatly dependent on the sample thickness.^[144–146] Theoretical calculations predicted that as the film thickness decreased, the adsorption edge would increase, and the optical conductivity would drop. Experimental results confirmed that few-layer BP shows tunable optical characteristics in the mid-infrared range.^[145,147,148] Thus, the adsorption window may be controlled by varying the number of layers of BP, making BP useful for optoelectronics and photodetectors.^[146,149]

Additionally, BP's mechanical properties, such as Young's modulus and breaking stress, are highly anisotropic.^[119,150–152] For both single- and few-layer BP, first-principles calculations projected an anisotropic Young's modulus ratio of 2.5.^[150] Tao et al. determined Young's modulus and breaking stress of a few-layer BP and found that the measured values were in close agreement with theoretical predictions.^[119] The Young's modulus of BP strip was determined to be 58.6 ± 11.7 GPa in the zigzag direction and 27.2 ± 4.2 GPa in the armchair direction, respectively. In contrast, the breaking stress that scaled linearly with Young's modulus was measured to be 4.79 ± 1.43 GPa and 2.31 ± 0.71 GPa, respectively.^[119] Further research done

by Wang et al. on thicker BP (14–34 nm) showed that the elastic modulus of BP nanosheet is layer-dependent.^[152] The maximum elastic modulus of the 14 nm thick BP nanosheet was 276 ± 32.4 GPa, and the elastic modulus declined to 89.7 ± 26.4 GPa as the thickness increased beyond 30 nm.^[152] Wei and Peng demonstrated that monolayer BP could bear up to 27% and 30% of tensile strain, respectively, in the zigzag and armchair directions.^[151] When tensile strains were applied in the zigzag and armchair directions, significantly different behaviors were predicted: 12.6% increase in the P–P bond length relative to the relaxed condition and stretched puckered unit cells in the armchair direction, respectively.^[151] BP's excellent mechanical qualities enable it to be used in wearable and deformable devices and electronics.

The anisotropy of BP's structure has also had a significant effect on its thermal qualities.^[109] Recent findings using first-principles calculations projected that the heat conductivity along the zigzag path is approximately two to three times that along the armchair direction.^[116,153–155] Luo et al. later confirmed the calculation results using micro-Raman spectroscopy (Figure 2b).^[156] The thermal conductivity of a mechanically exfoliated BP film with a thickness of 15 nm was measured to be 20 and 40 W m⁻¹ K⁻¹ in the armchair and zigzag directions, respectively, and was further lowered to 10 and 20 W m⁻¹ K⁻¹ in a 9.5 nm thick BP film.^[156] The decrease in thermal conductivity with the increasing thickness of BP film is due to significant surface phonon scattering, limiting phonons' mean free path.^[156,157] Another study conducted by Jang et al. on thicker BP films demonstrated thickness-dependent thermal conductivity (thickness ranging from 138 to 552 nm).^[116] Thermal conductivity values recorded for the armchair and zigzag directions are 26–34 and 62–86 W m⁻¹ K⁻¹, respectively, resulting in an anisotropic ratio of 2.4, which is consistent with both theoretical calculations and prior experimental results.^[116,153–156] It is worth noting that the authors state that as the thickness of BP films grows, the surface phonon scattering becomes less relevant, resulting in a thermal conductivity closer to the inherent value.^[116]

2.3. Transition Metal Dichalcogenide (TMDC)

Transition metal dichalcogenides (TMDCs), which have the general chemical formula MX₂, where M is a transition metal from group IVB, group VB, group VIB, or group VIIB, and X is a group VI element, can exist in a variety of structural phases due to the transition metal's different coordination, including the 1T-, 2H-, and 3R-phases.^[158–160] Generally, TMDCs containing a group IVB transition metal have thermodynamically stable 1T-phases with X-M-X stacking in the sequence of "ABA."^[158,159] However, TMDCs, including a group VIB transition metal, have more favorable 2H-phases with an "ABC" stacking.^[158,159] It was recently shown that further chemical exfoliation or lithium intercalation caused phase transition in the 2H-phased TMDCs, resulting in the formation of 1T'-phase, a deformed octahedral structure.^[161–166] This transition was attributed to structural relaxation caused by electron rearrangement in the transition metal's d-orbitals.^[167–169] The X-M-X layers are stacked and held together by weak van der Waals interaction, with an average monolayer

thickness of 6–7 Å (Figure 2c).^[160,162,170,171] The X–M–X layers of TMDCs are stacked and held together by weak van der Waal interaction, with an average monolayer thickness of 6–7 Å (Figure 2c).^[160,162,170,171] The bond length of M–X can range between 3.15 and 4.03 Å depending on the size of the transition metal and chalcogen ions.^[170]

Due to their excellent mechanical properties, TMDCs are ideal for flexible and stretchable electronics and optoelectronics.^[172] Bertolazzi et al. revealed that the suspended, free-standing monolayer MoS₂ had an average strength of 22 ± 4 GPa and an effective Young's modulus of 270 ± 100 GPa.^[173] Castellanos-Gomez et al. reported a similar result for suspended MoS₂ nanosheets with a few layers (5–25 layers).^[174] Tungsten-based TMDCs such as WS₂, WSe₂, and WTe₂ all have a high Young's modulus in the range of 149.1 ± 9.4 GPa to 302.4 ± 24.1 GPa and strength in the range of 6.4 ± 3.3 to 47.0 ± 8.6 GPa at monolayer thickness, with values decreasing as the thickness grows from monolayer to trilayer.^[175] Zhang et al. previously reported Young's modulus of 167.3 ± 6.7 GPa for multilayer WSe₂, a considerable decrease in elasticity compared to monolayer WSe₂ (258.6 ± 38.3 GPa), indicating that the mechanical properties of TMDCs are thickness dependent.^[175,176] Other TMDCs, including MoSe₂ and MoTe₂, displayed comparable mechanical properties, such as a high Young's modulus and strength.^[177–180]

Due to their diverse chemical compositions and structural phases, TMDCs exhibit a broad spectrum of electrical properties, ranging from metallic to semiconductor to insulator, as well as unique topological electronics phases.^[159,181–184] Specifically, the electronic properties of TMDCs are strongly dependent on the filling of the transition metal ions' nonbonding d orbitals: metallic behavior occurs when the nonbonding d orbitals are partially filled, while semiconducting behavior occurs when all nonbonding d orbitals are occupied.^[162] On the other hand, chalcogen ions have an effect on the bandgap of TMDC materials: the bandgap decreases with the increasing atomic size of the chalcogen elements.^[162,185–187] The bandgap of TMDCs is typically between 1 and 2 eV,^[183,185,187–189] and can be adjusted in a variety of ways: maintaining control over the thickness of TMDCs,^[186,189] applying strains,^[183,189,190] introducing dopants,^[172,191–193] alloying^[188] and forming heterostructure with other 2D materials.^[194]

The bandgap of semiconducting TMDCs, like graphene and phosphorene, is thickness-dependent.^[172,189,195–197] With decreasing sample thickness, the bandgap of semiconducting TMDCs grows and switches from indirect to direct bandgap. (Figure 2c).^[159,186,189,195,197–199] Metallic TMDCs, on the other hand, such as ReS₂ and NbS₂, were projected to exhibit thickness-independent metallic properties.^[197] Due to the reduced sample thickness, the transition metal d-orbitals contribute differently to the total density of states near the Fermi level. According to previous research, the direct bandgap is mainly produced from the xy/x^2-y^2 (VBM) and z^2 (CBM) orbitals, whereas the indirect bandgap is derived from the z^2 (VBM) and xz and yz orbitals (CBM).^[189,197,200–204] This indirect to direct gap transition has been shown to increase the photoluminescence quantum yield of TMDCs, hence enabling novel optoelectronic applications.^[204–207] Recently, interest in 1T'-phase TMDCs has increased because of their intriguing electrical and topological features.^[166,167,208–212] The zigzag chain structure of 1T'-phase

TMDCs reduces the transition metal's d-orbitals even further, resulting in band inversion and an inverted bandgap.^[167,59,213] Due to the strong spin-orbital coupling (SOC) and inversion symmetry of 1T'-phase TMDCs, quantum spin Hall states are generated, which were predicted theoretically and validated experimentally, making it a viable 2D topological insulator.^[59,210,212,214,215]

Strain engineering has been deemed an effective method for tailoring the band structure and tuning the electrical properties of TMDCs by utilizing their exceptional mechanical capabilities.^[183,216–218] Yun et al. calculated the bandgap variations in a single layer, 2H molybdenum, and tungsten-based TMDCs when applied strain or stress.^[189] While compressive strain increases the bandgap of monolayer 2H TMDCs, tensile strain and tensile stress decrease the bandgap, increasing the materials' metallicity.^[189] Johari and Shenoy further demonstrated that pure shear strains could be used to decrease the bandgap of some semiconducting TMDCs.^[183] Additional experimental observations on MoS₂ demonstrated excellent agreement with theoretical predictions, with a maximum bandgap change of 300 meV per 1% strain applied.^[219,220–222]

Additionally, the bandgap of TMDCs can be adjusted at the material level by alloying two distinct TMDCs or by inserting dopants.^[188,191,192,223,224] Komsa et al. demonstrated the feasibility of alloying molybdenum-based TMDCs (MoS₂, MoSe₂, and MoTe₂) and creating thermodynamically stable 2D compounds using theoretical calculations.^[223] Chen et al. showed experimentally that the sulfur-containing TMDCs (MoS₂ and WS₂) 2D alloy could be created with an adjustable bandgap depending on the tungsten concentration in the compound.^[224] The bandgap of Mo_{1-x}W_xS₂ can be adjusted between 1.82 and 1.99 eV when Mo_{0.8}W_{0.2}S₂ and WS₂ are used.^[224] Susarla et al. alloyed quaternary TMDCs using CVD and showed that by manipulating the CVD growth temperature, the bandgap of such quaternary TMDCs could be controlled between 1.61 and 1.85 eV.^[188] On the other hand, Neupane et al. described a simple postsynthesis treatment that immerses monolayer TMDCs in methanol to modify their electrical characteristics.^[192] Due to the n-doping, there is an increase in the field-effect mobility of monolayer MoS₂, WS₂, and WSe₂ and a decrease in the number of defects caused by the methanol treatment.^[192]

Utilizing heterostructures' different features and greater flexibility over pure 2D van der Waals layered materials, another emerging strategy for fine adjusting electrical properties is to build heterostructures with 2D TMDCs.^[194,225–229] Numerous atomically thin heterostructures include TMDCs, notably WS₂/WSe₂,^[194] MoS₂/graphene,^[226] WSe₂/SnS₂,^[227] and MoS₂/WS₂,^[230] have been demonstrated experimentally, and their electrical characteristics have been measured. Hill et al. analyzed the band alignment of the MoS₂/WS₂ heterostructure and discovered a type II band alignment with a 1.45 ± 0.06 eV heterostructure bandgap.^[230] Withers et al. created heterostructures using graphene, hexagonal boron nitride (h-BN), and a variety of 2D TMDCs, including MoS₂, WSe₂, and WS₂, to serve as quantum wells (QW) for light-emitting diodes (LED).^[228] Zhao et al. demonstrated that capillary force may turn a WSe₂/SnS₂ heterostructure into a 1D van der Waals superlattice. The originally semiconducting material becomes metallic as a result of type-III band alignment.^[227]

With a typical bandgap of 1–2 eV, TMDCs exhibit optical absorption in the infrared and visible regions.^[37] Due to the direct bandgap, the absorption in monolayer TMDCs is dominated by direct band-to-band transitions around the K and K' points in the Brillouin zone.^[37,160] Electrical properties profoundly influence the optical properties of TMDCs. Both absorption and photoluminescence behaviors of TMDCs can be tuned by forming heterostructure,^[228] alloying,^[224] thickness control,^[198] and postsynthesis chemical treatment.^[192,231] For example, MoS₂ demonstrated increasing photoluminescence intensity as the thickness decreased due to the transition from an indirect to a direct bandgap transition from bulk to monolayer.^[198] Additionally, the direct bandgap of monolayer MoS₂ results in a high absorption coefficient, which makes it appropriate for photodetector applications.^[28] Yamada et al. demonstrated the use of bis(trifluoromethane)sulfonimide (TFSI) as a post-synthesis chemical treatment on both mechanically exfoliated and chemical vapor deposition (CVD) generated monolayer MoS₂ to increase the intensity and lifespan of MoS₂ photoluminescence.^[231] Compared to pristine MoS₂, the treated MoS₂ had a 30-fold longer photoluminescence lifetime and a 230-fold greater photoluminescence intensity.^[231] The increased photoluminescence performance was attributed to OH radicals and a robust H⁺ donating environment on the MoS₂ surface following TFSI treatment.^[231] Additionally, an essential optoelectronic property of TMDCs is the presence of inequivalent degenerate valleys, which result from spin splitting caused by the high spin–orbital coupling and absence of inversion symmetry at the Brillouin zone's K and K' points.^[194,232–236] The spin-valley dynamics of TMDCs add a degree of freedom that can be manipulated electronically, opening up new opportunities in quantum photonic devices, as well as next-generation quantum electronics and computation.^[232,234,237,238]

2.4. MXene

MXene is a new class of 2D materials that comprise transition metal carbides, nitrides, and carbonitrides. It has the generic chemical formula $M_{n+1}X_n$ ($n = 1, 2, 3$), where M and X are transition metals (Ti, Nb, V, Cr, Ta, etc.) and carbon or nitrogen, respectively.^[239–243] MXene are commonly synthesized by selective etching and exfoliation from layered MAX phases with hexagonal geometries in which the A layers (group IIIA or IVA elements) alternately stack between the $M_{n+1}X_n$ layers (Figure 2d).^[240,244] Due to MXene's hydrophilic nature, the surface would be covered with functional groups whose proportions are unknown and change during the etching process.^[240,245,246] Thus, an alternative generic form of the MXene formula, $M_{n+1}X_nT_x$, is frequently reported in the literature, where T denotes the functional group adsorbed onto the surface.^[240] Additionally, the adsorbed functional groups may alter the mechanical characteristics of MXene.^[247,248] While 2D Ti₃C₂T_x theoretically has Young's modulus of 502 GPa, Lipatov et al. experimentally determined a value of 333 ± 30 GPa for monolayer Ti₃C₂T_x.^[248,249] The disparity in mechanical characteristics between theoretical and experimental values is due to the contribution of the functional groups populating the thin (0.98 nm) Ti₃C₂T_x layer.^[248,250] Additional first-principles calculations revealed that, whereas the

O-terminated MXenes displayed greater rigidity,^[240,247] MXenes with F- and OH-terminations have a lower elastic rigidity.^[247] The strong interaction between the O atoms and the Ti–C bonding is responsible for the greatest shift in Ti–C bonding between the three functional groups of interest.^[247] While functionalized MXenes have a lower Young's modulus, experimental results indicate that they can tolerate a more significant tensile strain by using the surface functional groups as a “buffer layer.”^[251,252] Further research has revealed an enhancement in the mechanical properties of MXenes when combined with carbon nanotubes (CNTs) or other polymers.^[253–256]

MXenes have a broad spectrum of electronic characteristics due to their diverse compositions, ranging from metallic to topological insulators.^[257–265] MXenes' surface functional groups are critical in determining their electrical characteristics.^[240,266,267] Calculations indicate that all pristine MXenes will be metallic without surface functionalization.^[240,268] Certain MXenes become semiconductors when functionalized, with bandgap values ranging from 0.25 to 2.0 eV.^[268] Experiments were conducted to determine the electrical characteristics of Ti₃C₂T_x and Ti₂CT_x.^[269,270] Miranda et al. revealed that Ti₃C₂T_x has a metallic character consistent with earlier theoretical predictions and low carrier mobility of $1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ when gated externally.^[269,271] Halim et al. also reported on Ti₂CT_x's metallic behavior.^[270] Further in situ measurements by Hart et al. revealed a fourfold improvement in conductivity after vacuum annealing to partially eliminate the surface functional group.^[267] Additionally, it is worth noting that due to the change in surface dipole moment following the adsorption of functional groups, the predicted work function of metallic MXenes can vary significantly between 1.8 and 8 eV, with a negative surface dipole moment resulting in a drop in work function.^[240,266] However, due to the mix termination created during the fabrication process, achieving an extreme value in the work function is challenging and requires precise control of functional group adsorption.^[240,272] Additionally, while it has been predicted that MXenes' electrical properties can be tuned using strain engineering and are layer-dependent, no experimental evidence has been found to support such band tuning procedures in MXenes.^[273,274]

Controlling the surface termination and composition of MXenes are two essential aspects affecting their optical characteristics.^[247,273,275–277] The computational results indicated that different terminations alter the adsorption spectrum of MXene.^[275,278] Berdiyev demonstrated that while O-terminated Ti₃C₂T₂ has a higher absorption in the visible band, functionalizing Ti₃C₂T₂ increases its absorption and reflectance in the UV region.^[275] Hantanasirisakul et al. demonstrated that Ti₃CNT_x and Ti₃C₂T_x have diverse visible-near infrared absorption spectra due to their various production processes.^[279] Ti₃C₂T_x had a local absorption maximum at 770 nm, whereas Ti₃CNT_x had a local absorption maximum at 670 nm and a plateau at 670 nm, depending on the synthesis solution utilized.^[279] Jiang et al. investigated the nonlinear optical response of few-layer Ti₃C₂T_x in the infrared region. They discovered a high effective nonlinear absorption coefficient and a nonlinear refractive index comparable to graphene.^[280] Although little was known about the MXene family's nonlinear optical properties, the work demonstrated that MXenes have significant promise for high-performance photonic devices.^[280]

2.5. Other 2D Materials

2.5.1. Other 2D Inorganic Materials

Beyond the materials discussed above, other 2D inorganic materials have also found applications in wearable energy harvesting. Amongst all, tellurene, Bi_2Te_3 and h-BN have been frequently reported for their usage in wearable energy harvesters. These materials are intriguing candidates for wearable energy harvesters due to their different electrical, thermal, and optical characteristics from the aforementioned 2D materials.

Tellurene, a 2D version of tellurium, is a promising 2D material for wearable energy harvesting. In contrast to other layered 2D materials, tellurium (Te) is formed of helical Te chains that spiral around the [0001] direction and are piled by weak interactions (Figure 2e).^[281,282] Due to the unique chain structure, Te exhibits structure anisotropy, which favors the formation of 1D Te nanostructures.^[283–288] 2D tellurium growth has been attempted using vapor-phase deposition techniques such as physical vapor deposition (PVD) and molecular beam epitaxy (MBE). However, the products were constrained by their thickness and epitaxial substrates.^[282,289] Wang et al. reported a solution-based, substrate-free synthesis of tellurene in 2018 (see Figure 2e).^[281] Tellurene grown in solution exhibits the same phase as its bulk analog (α -phase), which is projected to be the most stable phase at room temperature and pressure.^[281,290,291] Additionally, as the thickness of tellurene is lowered to a monolayer, a transition of the most stable phase from the trigonal α -Te to the tetragonal β -Te can be observed as a result of strain relaxation.^[291,292] The indirect bandgaps of α -Te and β -Te were calculated to be 0.75–1.15 and 1.47–1.79 eV, respectively.^[291,293] Experimental measurements of epitaxial Te on a graphene/6H-SiC(0001) substrate revealed a monolayer Te bandgap of up to 0.92 eV.^[282] Tellurene's bandgap is thickness dependent, rising from 0.33 eV in bulk Te to 0.92 eV in monolayer α -Te.^[281,282,293] This thickness dependence, caused by charge screening and interlayer coupling, was also observed in the field-effect mobility of tellurene field-effect transistors (2D Te FETs), which peaked at $750 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at a thickness of roughly 10 nm, as Wang et al. reported.^[294–296] Additionally, due to tellurene's high electrical conductivity and low thermal conductivity ($1.5 \text{ W m}^{-1} \text{ K}^{-1}$), 2D Te is an excellent thermoelectric material.^[297–299] The structural anisotropy also contributed to the anisotropic optical properties, as demonstrated by theoretical calculations and experimental measurements, which indicate a higher absorption along the chain direction.^[291,300] 2D Te has shown tremendous potential in photonics and optoelectronics due to its high carrier mobility, variable bandgap, and outstanding air stability.^[300–302]

Due to its high figure of merit in bulk materials (ZT 1.14 at room temperature) and 2D nanostructures around room temperature ($T < 250 \text{ }^\circ\text{C}$), bismuth telluride (Bi_2Te_3) has been widely researched in thermoelectric applications.^[303,304] Bi_2Te_3 is made of quintuple atomic layers with a repeated stacking pattern of Te-Bi-Te-Bi-Te along the c -axis, and each “monolayer” is stacked via weak van der Waals interaction (Figure 2e).^[305–308] It has been predicted and measured that by lowering the thickness of Bi_2Te_3 from bulk to a few layers, a 13-fold increase in figure of merit can be attained compared to the bulk counterpart due

to the lower thermal conductivity and increased electrical conductivity.^[306,307,309,310] Teweldebrhan et al. exfoliated stacked Bi_2Te_3 and noticed a considerable reduction in in-plane thermal conductivity ($1.1 \text{ W m}^{-1} \text{ K}^{-1}$) as compared to bulk materials ($1.5\text{--}2.0 \text{ W m}^{-1} \text{ K}^{-1}$).^[307] On the other hand, the semiconducting Bi_2Te_3 (bandgap 0.16 eV in bulk) exhibited an intriguing band evolution with thickness.^[311–313] Computational results indicated that as the thickness of Bi_2Te_3 was reduced from bulk to five quintuple layers, the bandgap closed, and the surface states became metallic. In contrast, bandgap values of 0.27 and 0.05 eV were calculated when the thickness was further reduced to one and two quintuple layers.^[313] Specifically, the interaction between the top and bottom topological surface states in a quintuple layer is critical for increasing the electronic contribution to ZT, which improves the thermoelectric properties of monolayer Bi_2Te_3 in conjunction with the lower thermal conductivity.^[313] Apart from the increased thermoelectric capabilities when the thickness of Bi_2Te_3 decreases to monolayer, the increase in topological surface states results in the formation of 2D Bi_2Te_3 topological properties, making it a preferred material for topological insulators and saturable absorbers.^[314–316]

Hexagonal boron nitride (h-BN), a structural counterpart of graphene, is composed of honeycomb lattices with alternating boron and nitrogen atoms connected via sp^2 bonds.^[317,318] The lattice parameters of h-BN were calculated to be 2.5 Å and 6.66 Å along the a - and c -axis, with an interlayer spacing of 3.33 Å.^[318,319] h-BN, like graphene, displayed an exceptional mechanical strength of 26.0 N m^{-1} and Young's modulus of 0.87 TPa, both of which are equivalent to graphene.^[320–323] In contrast to graphene, whose mechanical strength declines as the number of layers increases, the mechanical strength of h-BN has been demonstrated to be independent of layer thickness up to nine.^[324] Additionally, molecular dynamics simulations demonstrated that the mechanical characteristics of h-BN are temperature and strain-rate dependent. Using DFT calculations, Peng et al. predicted a nonlinear elastic deformation of 2D h-BN prior to strain softening failure.^[325,326] The results indicated that h-BN is a wide-bandgap semiconductor with an indirect bandgap of 5.95 eV and a direct bandgap of 6.47 eV, which is compatible with theoretical models.^[317,327–334] The direct bandgap was discovered to exist at the K point of the first Brillouin zone, whereas the CBM and VBM produced the indirect bandgap at the M and K points of the first Brillouin zone.^[333,335–339] In contrast to graphene, where applied strain has a negligible effect on its bandgap, calculations indicate that the bandgap of h-BN is strain-sensitive.^[340,341] When a mild symmetric compressive strain (1.5 percent) or an asymmetric tensile strain (1.2 percent) is applied to h-BN, Li et al. observed a direct-to-indirect bandgap transition (Figure 2f).^[340] Wu et al. further indicated the possibility of adjusting monolayer h-BN from insulator to semiconductor by applying biaxial tensile strains of 18% and 27% on monolayer h-BN.^[341] Additionally, further research found that the bandgap of h-BN varies at a rate of 5.9 meV K^{-1} with temperature.^[342] Because h-BN has a large bandgap, it exhibits photoluminescence in the deep ultraviolet range (215 nm). There has been renewed interest in single-photon emission (SPE) and nonlinear optical characteristics of h-BN.^[334,337,342–344] Originating from localized vacancy defects in h-BN monolayers, the h-BN-based SPE demonstrated

a high quantum efficiency and brightness, making it the only material capable of SPE in the deep ultraviolet region.^[337,344–346] Kumbhakar et al. also observed the non-linear optical characteristics of h-BN using IR excitation and Z-scan methods.^[342] h-BN exhibits exceptional thermal characteristics, with a monolayer h-BN thermal conductivity projected to exceed $600 \text{ W m}^{-1} \text{ K}^{-1}$.^[347] It was previously considered that the significant phonon-phonon scattering in h-BN contributed to its lower thermal conductivity than graphene.^[347] However, experimentally synthesized h-BN has a lower thermal conductivity than expected. Due to microstructure defects and polymer residue from the fabrication process, the thermal conductivity of few-layer h-BN was measured to range between $100 \pm 10 \text{ W m}^{-1} \text{ K}^{-1}$ in freestanding h-BN to about $484 \text{ W m}^{-1} \text{ K}^{-1}$ in suspended bilayer h-BN.^[348–350]

2.5.2. 2D Polymers and 2D Covalent Organic Framework (COF)

2D polymers, including synthetic 2D polymers and 2D covalent organic frameworks, are a rapidly growing class of 2D materials that have applications in gas storage, separation, and optoelectronics.^[351–360] Although the concept of a 2D polymer dates back to the 1930s when Gee et al. synthesized 2D macromolecules using air-water interfacial synthesis, it was not until 2009 that the concept of synthetic 2D polymers was reintroduced.^[361–363] Although the term “2D polymers” may have multiple definitions, the following five characteristics are generally recognized by chemists as the widely accepted definitions of 2D polymers:^[361,364–367]

- 1) The polymer must have planar topological sheets.
- 2) 2D polymers must contain at least one crystalline phase of their repeating units.
- 3) Covalent bonds must be used to connect the repeating units.
- 4) The thickness of 2D polymers should be comparable to the repeating units.
- 5) Each 2D polymer monolayer should be self-supporting under typical gravitational conditions.

It is worth noting that because synthetic 2D polymers are frequently referred to as those that can be synthesized at room temperature and pressure, polymers such as graphene, poly- C_{60} , and $(\text{BN})_x$ may occasionally be excluded from this group.^[366]

According to the definition, 2D polymers are composed of repeating units covalently bound together.^[361,364] These repeating units, each with three, four, or six binding sites, span the 2D plane and serve as the structure's skeleton and ordered porosity structure (Figure 2g).^[364,368–370] 2D polymers often use aromatic rings and their derivatives as building blocks to make rigid networks.^[371] Compared to a typical 1-D polymer chain, where the structure and molecular weight are directly tied to the monomer polymerization, a 2D polymer with at least three binding sites can retain structural integrity even when monomers are missing from part of the binding sites.^[361] Thus, with sheet sizes of several μm^2 , the typical average molecular weight of the 2D polymer can reach several Mda.^[361,372] Additionally, because a crystallinity-rich phase is required, the structure of 2D polymers can be studied using standard structure characterization techniques such as single-crystal XRD (SC-XRD).^[361,373–375]

The five prerequisites for 2D polymers govern their chemical structure and electrical properties, including Dirac cones, topological flat bands, and topological insulators.^[376–378] Gutzler calculated the band structure of 2D conjugated polymers and established a link between the band structure and structure parameters such as repeating unit length, in-plane dihedral angles, thickness, and isomerization.^[377] This mathematical finding paved the way for band engineering on 2D polymers and COF by screening for acceptable building blocks and motifs during the materials design and synthesis processes.^[377] Galeotti et al. synthesized a mesoscale-ordered 2D P^2TANG conjugated polymer featuring a Dirac cone and a semiconducting flat band.^[378] Additionally, it was proved that the Dirac cone and flat band of the P^2TANG might be changed by replacing the center atom or the bridging units, which opens up enormous opportunities for future research in the Hall effect and superconductivity.^[378] Springer et al. studied the topological properties of 2D polymers and shed light on the elements that contribute to their topologies, such as spin-orbital coupling (SOC), second-neighbor interaction, and the effect of the network on the polymer's electrical properties.^[376] Gámez-Valenzuela et al. recently explored the structural and electrical properties of C_3 -symmetric 2D polymers.^[379] The resulting bandgap was calculated to be between 1.17 and 3.13 eV, indicating that the pore size, geometry, bandgap, and band dispersion can be modified with the appropriate selection of repeating units and building blocks.^[379]

2.5.3. 2D Organic–Inorganic Hybrid Materials

Metal halide hybrid organic–inorganic perovskite (HOIPs), having the chemical formula AMX_3 , where A, M, and X are small organic cations, divalent Group 14 elements, and halogen, respectively, has been widely employed in solar cells due to its excellent defect tolerance and charge transport.^[380–390] By reducing 3D HOIPs crystals to a single-layer thickness along the (100) direction, 2D HOIPs are structural analogs to the Ruddlesden-Popper phase with the expression $\text{A}_2\text{A}'_{n-1}\text{M}_n\text{X}_{3n+1}$ and exhibit greater ambient stability than their 3D counterparts while retaining excellent optoelectronic properties.^[381,391–398] Thus, 2D HOIPs can be thought of as layers of $[\text{MX}_6]^{4-}$ octahedral material sandwiched between two layers of organic spacer cations (Figure 2h).^[398,399] The mechanical properties of 2D perovskite are thought to be dependent on the number of $[\text{MX}_6]^{4-}$ octahedral layers, the length of organic spacer molecules, and the strength of the M–X bond.^[381,400–402] While 2D HOIPs are mechanically weaker than their 3D counterparts, Tu et al. discovered that the out-of-plane mechanical hardness and Young's modulus of 5-layered 2D HOIPs are comparable to those of their 3D counterparts using nanoindentation.^[400] On the other hand, when the chain length of the organic spacer molecules increases, the out-of-plane Young's modulus declines initially before plateauing, showing that the softening effect in long-chain spacer cations has been saturated.^[400] Further calculation done by Gao et al. showed that the elastic moduli of 2D $(\text{C}_6\text{H}_5\text{CH}_2\text{NH}_3)_2\text{PbCl}_4$ are nearly isotropic across the *a* and *c* axis.^[401] Later, Kim et al. examined the in-plane Young's modulus of single-layered 2D HOIPs $(\text{R-NH}_3)_2\text{PbX}_4$, where R and X represent alkyl and halogen groups, respectively.^[381]

The bond strength of Pb–X was discovered to be a significant factor in determining the in-plane Young's modulus and had a minor effect on the out-of-plane Young's modulus, owing to the stronger Pb–X bonding resulting in a larger in-plane Young's modulus.^[381] Interestingly, the chain length of organic spacers can also affect the in-plane mechanical stiffness by distorting the $[\text{PbX}_6]^{4-}$ octahedral structure and altering the length of the Pb–X bond.^[381] Another study conducted by Yin et al. investigated the phase transition and optical characteristics of 2D $(\text{C}_4\text{H}_9\text{NH}_3)_2\text{PbI}_4$ flakes at various applied pressures.^[402] At 0.1 GPa, a phase transition from *Pbca* (1b) to *Pbca* (1a) was detected, followed by a phase transition to *P2₁/a* above 1.4 GPa.^[402] The increase in the Pb–I–Pb bond angle and the decrease in the Pb–I bond length due to external pressure results in a red shift of the excitonic bandgap, suggesting a narrower bandgap due to the materials' structural changes (Figure 2h).^[402]

Wang et al. also reported on the bandgap shift caused by the deformation of the 2D HOIPs structure.^[403] Strain engineering of 2D $(\text{BA})_2(\text{MA})_{n-1}\text{Pb}_{n+1}\text{I}_{3n+1}$ microplates revealed a decreasing rate of change in bandgap with increasing numbers of layers.^[403] External strain-induced distortion of the stacked octahedra was thought to impair the stability of the 2D HOIPs by facilitating moisture and oxygen penetration.^[403] Parveen et al. confirmed that the bandgap of 2D HOIPs is thickness dependent and increases linearly when the sample thickness is reduced from 14 layers to bilayer.^[393] The discovery was consistent with a prior demonstration by Guo et al. on solution synthesized $(\text{C}_6\text{H}_5\text{C}_2\text{H}_4\text{NH}_3)_2\text{PbBr}_4$ (PEPB), in which photoluminescence of PEPB nanocrystals of varying thickness displayed a spectrum of colors, indicating a thickness-dependent bandgap.^[404] Zhang et al. detected a 2D–3D transition in the temperature-dependent photoluminescence spectra of multiphase 2D HOIPs by comparing the optical characteristics of single-phase and multiphase 2D HOIPs.^[405] The transition of 2D HOIPs, including many phases from 2D to 3D behavior, results in a more significant electron–phonon interaction, making them an excellent material for the active photovoltaic layer.^[405] Additionally, the generation of excitons in a self-trapped state within the bandgap results in a superior photodetection performance.^[406,407] By substituting organic spacer cations, the optical and electrical characteristics of 2D HOIPs can also be adjusted.^[380] By using conjugated anilinium cation, Rodríguez-Romero et al. adjusted the dielectric contrast between the inorganic and organic layers of $(\text{C}_6\text{H}_5\text{NH}_3)_2(\text{CH}_3\text{NH}_3)_{n-1}\text{PbI}_{3n+1}$ with *n* being either 3 or 5 to lower the binding energy of excitons and improve the performance of the resulting solar cell.^[380] Because the inorganic and organic layers have a distinct HOMO–LUMO energy gap, the 2D HOIPs revealed a well-barrier structure that can be easily changed by varying the chain length of organic cations and the number of inorganic layers sandwiched between two organic layers.^[394,407] Blancon et al. took advantage of the quantum well structure's potential and developed a scaling law for the excitons' binding energy at various layer thicknesses.^[408] It was discovered that when the number of layers grows from one to five, both the reduced mass and binding energy of excitons decrease, resulting in the excitons' and free carriers' optical resonances.^[408]

Metal-organic frameworks (MOFs), another intriguing inorganic–organic hybrid material category whose 2D version

has recently attracted enormous attention, are well-defined 3D porous structured materials composed of organic ligands linking inorganic clusters.^[409,410] MOFs with a porosity of 90% of the free volume displayed an unprecedented internal specific surface area (over 10 000 m² g^{−1}).^[411–416] Due to their structure, MOFs are excellent candidates for gas storage and separation, sensing, catalysis, and drug delivery applications.^[409,417–426] The diameter of the micropores (general diameter < 2 nm) can be modified from several angstroms to nanometers by selecting suitable organic linkers for MOFs.^[409,427–429] 2D MOFs with a large specific surface area are expected to have more accessible surface active sites and perform better in catalytic and sensing applications than their 3D counterparts.^[430] However, the fabrication of freestanding 2D MOFs from bulk crystals via exfoliation proved difficult due to the inability of the resultant nanosheet to preserve its pristine structure during the process.^[430–432] Peng et al. and Rodenas et al. reported the 2D form of MOFs with a thickness of several nm in 2015, using bottom-up techniques to preserve their structural integrity (Figure 2i).^[431,432] Numerous other 2D MOFs have been synthesized, including zinc 1,4-benzenedicarboxylate (ZnBDC), zinc tetrakis(4-carboxyphenyl)porphyrin (Zn-TCPP), copper 1,4-benzenedicarboxylate (CuBDC), and nickel 2,3,6,7,10,11-hexaiminotriphenylene (Ni3(HITP)₂).^[430,432–434]

Numerous studies have been conducted on the mechanical characteristics of 2D MOFs. Hermosa et al. reported Young's modulus and breaking strength of 2D $[\text{Cu}(\mu\text{-pym}_2\text{S}_2)(\mu\text{-Cl})]_n \cdot n \text{ MeOH}$.^[435] The reported work identified 2D MOFs with Young's modulus of 5 GPa and a breaking strength of 10.4 GPa, respectively, 200 and 150 times lower than graphene.^[435] It is worth noting that 2D $[\text{Cu}(\mu\text{-pym}_2\text{S}_2)(\mu\text{-Cl})]_n \cdot n \text{ MeOH}$ is nevertheless sufficiently robust to withstand capillary forces and retain a sheet-like morphology in solution.^[435] Zeng et al. later reported Young's modulus and yield strength of 2D CuBDC with a nanoporous structure by nanoindentation.^[436] Along the out-of-plane direction, Young's modulus and yield strength of 2D CuBDC were determined to be 23 GPa and 450 MPa, respectively.^[436] The hardness of the 2D CuBDC was also discovered to be dependent on the interlayer sliding.^[436] Additionally, the same work postulated and investigated three failure modes for 2D MOFs: sliding/slippage, fracturing, and delamination.^[436]

2D MOFs are electrical insulators in general due to the interaction between metal ions and hydrogen or oxygen atoms from redox-inactive organic ligands, which results in a slight overlap between the metal ions' *d*-orbitals and the organic ligands' orbitals.^[437–439] Due to their low electrical conductivity, 2D MOFs have limited performance and potential in fuel cell and supercapacitor applications.^[437] Recently, it was discovered that certain 2D MOFs with a specific selection of organic ligands and dopants are electrically conductive.^[438,440] Sengupta et al. demonstrated that doping 7,7,8,8-tetracyanoquinodimethane (TCNQ) into 2D MOF increased the electrical conductivity by order of magnitude by generating a charge-transfer complex.^[438] Su et al. enhanced the electrical conductivity of a 2D MOF by using tetrathiafulvalene (TTF)-based linkers.^[440] Electrical conductivity was previously attributed to in-plane charge delocalization and prolonged π -conjugation in these 2D MOFs.^[441–443] Day et al. investigated the electrical transport properties of single-crystal 2D MOFs in detail.^[444] The metallic

behavior of the single-crystalline 2D MOFs was markedly different from that of the polycrystalline 2D MOFs.^[444] However, additional research is necessary to determine the effect of structural anisotropy and grain boundaries on electrical transport in polycrystalline 2D MOFs.^[444]

Additionally, the electrical conductivity of 2D MOFs enables their application in optoelectronics. Due to their well-defined porosity structure and abundance of luminescent centers, 2D MOFs containing rare-earth elements make good candidates for white-light emission with a long lifetime and sharp emission.^[445–453] Intriguingly, it was discovered that the emission color of 2D MOFs is dependent on the interlayer exciton emission pathway.^[454] Liao et al. described a dual-emission pathway for 2D Ca-MOFs that can tune the materials' luminescence color.^[454] The scientists asserted that by repeating the exfoliation-recombination process on 2D Ca-MOF, such properties could be utilized to create and manipulate optical memory states, as the exfoliation-recombination process resembles optical memory's writing-reading-erasing process.^[454] Cheng et al. investigated the nonlinear optical characteristics of 2D MOFs containing Cu and Zn metal centers.^[455] Cu atoms' unoccupied *d*-orbital enables interaction between *d*- π orbitals, increasing the nonlinear coefficient of 2D Cu-MOF.^[455] On the other hand, Zn atoms with fully occupied 3d orbitals cannot form such conjugation, and the resulting nonlinear optical effect is barely visible.^[455]

2.6. Biocompatibility and Biosafety of 2D Materials

Biocompatibility of 2D materials has attracted increased attention since they are increasingly used in biomedical detection and therapy, including wearable sensors,^[456–458] tumor imaging,^[459] photodynamic therapy (PDT),^[460–462] and photothermal therapy (PTT).^[463–466] Biocompatibility can be defined as a material's capacity to interact with living organisms without causing harm to them.^[467] Numerous characteristics, including cytotoxicity and long-term cell viability, can be utilized to assess the biocompatibility and biosafety of 2D materials, both in vitro and in vivo.^[464,467–469] Although graphene and its family of nanomaterials have been widely studied, inconsistent results regarding their toxicity to human bodies have been recorded under various testing situations.^[469–471] Current research indicates that the toxicity of graphene family nanomaterials is highly dependent on the materials' synthesis methods, size, thickness, purity, dose, and surface chemistry.^[471–476] While some graphene-based nanomaterials, due to their sharp edges and small size, are capable of penetrating the cell membrane and causing toxicity once within the cell,^[477] on the other hand, other researchers discovered no substantial cytotoxicity associated with CVD-grown graphene used as a cell growth substrate.^[478–482] 2D BP is less toxic and more compatible with biological cells than graphene and has been widely used in in vivo PDT for antitumor purposes.^[483–486] The unique degradation of BP nanosheets into biocompatible phosphorus oxides during light irradiation enhances their biosafety.^[485] However, in vivo tests on the biocompatibility of BP nanoplatelets revealed a more significant loss of cell viability than 2D TMDCs, implying that BP is more nanotoxic than layered TMDCs.^[475,483] As with graphene family nanomaterials, whose biocompatibility and biosafety are

highly dependent on manufacturing methods and material properties, the cytotoxicity of TMDCs and MXenes varies according to their composition.^[467,468,487–489] Although TMDCs have low toxicity against biological cells in general, chalcogen atoms have a significant influence in defining TMDC toxicity.^[459,468,490–495] In general, TMDCs containing sulfur atoms are less poisonous than those containing tellurium atoms, whereas NbTe₂ and TaTe₂ were shown to be less cytotoxic than VTe₂.^[495,496] On the other hand, MXene, especially 2D Ti₃C₂, has good biocompatibility.^[463,465,497–499] The biocompatibility and biosafety of pristine, surface-modified MXene and MXene-based composite, both in vitro and in vivo, showed negligible cytotoxicity or no abnormal behavior.^[465,498,500–502] The use of hydrofluoric acid in the synthesis, on the other hand, may constitute possible harm to the human body.^[498]

Numerous approaches, including chemical modification^[503] and incorporating with other materials,^[465,476,504] have been proposed to tailor the electrical and chemical properties of 2D materials and improve their aqueous solubility as well as effective interactions of 2D materials with biocomponents in order to reduce the potential toxicity of these materials and increase their biocompatibility.^[496,505,506] By distributing graphene in a Pluronic block copolymer, Duch et al. increased its biocompatibility.^[476] When graphene was inhaled into the lungs, the nonionic amphiphilic polymer decreased the local fibrotic reaction and minimized lung damage.^[476] By improving the exfoliation process with aminopropyl-lysozyme (AP-LYS), Siepi et al. improved the biocompatibility of MoS₂ nanoflakes.^[507] Because the positively charged surface is more favorable for associating with negatively charged molecules such as DNA and RNA, the AP-LYS-coated MoS₂ nanosheets dispersed better in water.^[507] The cytotoxicity of MoS₂ could also be reduced by surface modification of polyethylene glycol,^[508–510] chitosan (CS),^[511] or integrated into poly (lactic-co-glycolic acid) (PLGA).^[512] Liu et al. confirmed the lower toxicity of PEGylated MoS₂ than its pristine form with less dihydroethidine (DHE) positive cells when treated with MoS₂-PEG and MoS₂, suggesting lower induced oxidative stress of MoS₂-PEG to HeLa cells.^[508] Superior biocompatibility of MoS₂-CS was also observed by Yin et al. when KB cell was treated with the surface-modified MoS₂.^[511] Qu et al. revealed that when BP nanosheets were treated with a titanium sulfonate ligand (TiL₄), they possessed increased biocompatibility.^[503] The TiL₄-modified BP (TiL₄@BP) showed lower inflammation potential than bare BP, resulting from reversing the negative zeta potential of BP into the positive zeta potential and thus reducing the cell uptake by macrophages.^[503]

Numerous studies have been conducted to identify the crucial process–structure–property link underlying 2D materials that contribute to their biocompatibility. Li et al. explored the method by which few-layer graphene (FLG) enters the membrane of a cell.^[513] Both theoretical simulation and experimental observation indicate that FLG penetrates the cell membrane edge-first or corner-first and that the activation barrier for penetration is inherently unrelated to the lateral size of FLG.^[513] Moore et al. also investigated the role of morphology in the cytotoxicity of MoS₂.^[514] When the cell was treated with smaller and thinner MoS₂ flakes, a greater endotoxin level was found, which might be explained by the larger effective surface area of smaller and thinner MoS₂ flakes, which results in a stronger

inflammatory response.^[514] Chng et al. reported a similar result when they examined the cytotoxicity of MoS₂ nanosheets exfoliated using various lithium intercalating agents.^[488] While using *n*-butyllithium and *tert*-butyllithium increases the efficiency of chemical exfoliation of MoS₂, the larger surface area and active edge sites of the resulting MoS₂ nanosheet have a more significant cytotoxic effect.^[488,515] Zhang et al. also reported on the cytotoxicity of reduced graphene oxide (rGO) in a pathway-dependent manner.^[516] Compared to rGO reduced by light, rGO reduced by Fe²⁺ ion demonstrated decreased cytotoxicity due to the generation of less highly reactive graphene-based radicals.^[516] The size-dependent biocompatibility of 2D BP, on the other hand, followed a different trend than that of 2D TMDCs.^[509,517,518] Zhang et al. observed that larger BP nanosheets were more cytotoxic than smaller ones.^[518] As the size of the BP nanosheet increases, the integrity of the cell membrane deteriorates, dramatically reducing the biocompatibility of bigger BP nanosheets.^[518]

Additionally, efforts have been made to distinguish critical properties and aspects of the cytotoxicity of 2D materials. Marchwiany et al. recently conducted a theoretical investigation on the toxicological features of 2D MXene, utilizing machine learning models to ascertain the critical elements affecting 2D MXene's cytotoxicity in the human body.^[519] The investigation revealed that the surface oxide, MXene's surface modification, and external chemicals contribute to toxicity. The study's predictions paved the way for subsequent experimental optimization of surface functional groups and intrinsic oxides.^[519] Mei et al. examined the interactions of 2D materials with tissues.^[520] The authors demonstrated the complexity of 2D materials bio-nano interactions by introducing several variables into the analysis, including cell culture media and surface protein coating. They also suggested that the downstream cellular effects are highly dependent on the choice of 2D materials and cell types, necessitating the development of distinct strategies and designs to mitigate and reduce the cytotoxicity of 2D materials.^[520] Although the biocompatibility of the 2D materials family is still largely unknown, both *in vivo* and *in vitro*, the studies have provided critical information for assessing, mitigating, and preventing contamination and hazards on the job when conducting 2D materials-related research, as well as reducing injury and harm to the human body when used in biotherapeutic and biomedical applications.^[475,476,503]

3. Preparation of 2D Materials

Since the first exfoliation of graphene by adhesive tapes in 2004, many ways for fabricating high-quality, pristine 2D materials with precise control over dimension, composition, and characteristics have been developed.^[76,521] We shall explore state-of-the-art techniques for preparing 2D materials in this part, categorizing them as top-down or bottom-up approaches.

3.1. Top-Down Approaches

Exfoliation of bulk, layered materials has been a widely used technique for obtaining few-layered 2D materials.^[522–525] To

isolate the layered structures in a typical exfoliation procedure, the bulk materials are either immediately applied with shear force (mechanical exfoliation) or submerged in the appropriate solution first (liquid-phase exfoliation, LPE).^[526,527] Nonoselov demonstrated mechanical exfoliation of 2D materials on graphene isolation using sticky tapes,^[76] which was later applied in synthesis and isolation of MXene,^[527] h-BN,^[528] pnictogen nanosheets^[529] and 2D TMDCs.^[530] Although it is a straightforward and cost-effective process for isolating high-quality, pristine 2D materials, the random distribution of sample thickness and low monolayer yield significantly limit its mass production capability.^[521,531]

LPE is a versatile and low-cost technique that results in minimal 2D material aggregation and high yield (exceeding 1 g L⁻¹).^[532] Additionally, spray coating, inkjet printing, and drop-casting can apply dispersed 2D materials to various applications.^[91,533–535] LPE is primarily categorized into the following categories: exfoliation aided by sonication,^[536] ion-intercalated exfoliation,^[537] and ion-exchange exfoliation,^[538,539] based on the working mechanisms of the process.^[524,531] It should be emphasized that no single LPE technique applies to the exfoliation of all 2D materials and that the appropriate LPE process and solvents vary amongst materials.^[523,524,531]

3.1.1. Sonication-Assisted Exfoliation

Ultrasonication, whether by sonication tip or bath sonication, can be effective for LPE.^[540–545] Ultrasonic waves impart sufficient mechanical energies to the process, allowing the bulk materials to be broken down into a few-layered film (Figure 3a).^[525] Mechanical strength is generated by shock waves and turbulence caused by the collapse of microbubbles created during ultrasonic wave compression and rarefaction cycles.^[525,546] The resulting shear force, compressive stress, and tensile stress are sufficient to overcome the van der Waals interaction between the layers and exfoliate the 2D materials in the solvent.^[525] Mechanical energy sources such as rotor-stator mixers or blenders with rotor blades have also been proved to be feasible for low-cost LPE of 2D materials.^[547–549]

Hernandez et al. reported on the LPE preparation of graphene using the organic solvents *N*-methyl-pyrrolidone (NMP) and dimethylformamide (DMF) to obtain maximum concentrations of 0.01 mg mL⁻¹ and a monolayer yield of 1% wt.^[91,536] The authors stated that a monolayer graphene yield of 7–12 wt% can be attained with additional recycling and processing of the silt.^[91] Bourlinos et al. investigated alternative solvents in the LPE of graphite into graphene.^[550] It was discovered that, depending on the nature of the solvent molecules, charge transfer between graphene layers and aromatic molecules via π - π stacking determines the maximum concentration of graphene dispersion in the solvent and that graphene layers can act as either an electron donor or acceptor.^[550] A similar technique has been applied to TMDCs by Coleman et al.^[551] Several TMDCs, including MoS₂ and WS₂, had been exfoliated from bulk crystals using NMP and isopropanol (IPA).^[551] Zhou et al. have proposed LPE of MoS₂ and WS₂ using a water-ethanol mixture to obtain a more cost-effective synthesis with less toxicity than utilizing organic solvents.^[552] Shen et al. conducted a

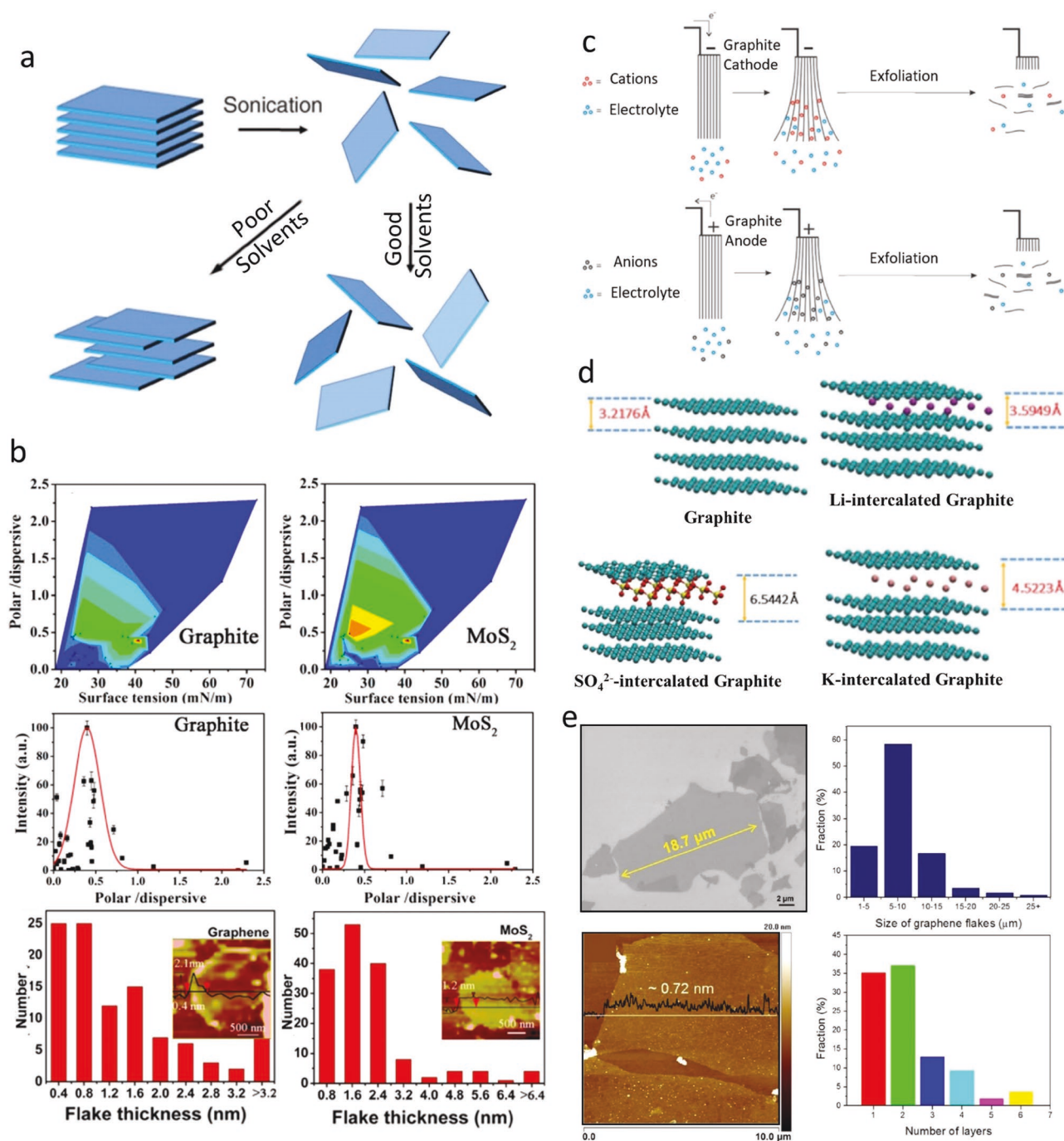


Figure 3. Top-down approaches in 2D materials preparation. a) Schematically illustration of ultrasonic-assisted exfoliation. Reproduced with permission.^[524] Copyright 2013, American Association for the Advancement of Science. b) Top and middle rows: dispersibility of graphene and MoS₂ as a function of total surface tension and polar to dispersive ratio. Bottom row showing the thickness histogram of graphene and MoS₂ exfoliated using solvents with the optimal surface tension and polar to dispersive component ratio. Reproduced with permission.^[523] Copyright 2015, American Chemical Society. c) Detailed schematic of cathodic exfoliation (top) and anodic exfoliation (bottom) of graphene from graphite. Reproduced with permission.^[1184] Copyright 2015, Elsevier. d) DFT calculation results of ion-intercalation exfoliation of graphite into graphene using SO₄²⁻ ion (bottom left), Li ion (top right) and K ion (bottom right). Reproduced under terms of the CC-BY license.^[565] Copyright 2017, The Authors, published by the Royal Society of Chemistry. e) Left: SEM image (top) and AFM image (bottom) of a typical graphene obtained by ion-intercalated exfoliation. Right: Lateral size (top) and thickness (bottom) distribution of graphene through ion-intercalation exfoliation. Reproduced with permission.^[543] Copyright 2014, American Chemical Society.

theoretical investigation into the effect of surface tension component ratios between the polar and dispersion components of applied solvents and 2D materials (Figure 3b).^[523] The results

indicated that a solvent suited for certain 2D materials LPE should have surface tension component ratios similar to the 2D materials.^[523] However, direct exfoliation using sonication and

shear force in solvents was difficult to control and resulted in a decreased yield of monolayer-thick 2D materials, limiting its use as a versatile and scalable 2D materials preparation.^[531,553]

3.1.2. Ion-Intercalated Exfoliation

A more controlled synthesis approach called ion-intercalated exfoliation was developed to compensate for the dispersion of the sample thickness during direct liquid-phase exfoliation.^[531,553–555] Ion-intercalated exfoliation was achieved by inserting ions into the interlayer spaces of bulk materials, including cations (e.g., Li^+ , Na^+ , and K^+), anions (SO_4^{2-} , for example), and radicals (OH and O radicals, for example), followed by sonication or thermal shock to exfoliate the layered materials (Figure 3c).^[524,556–558] By combining the well-developed intercalation chemistry of the bulk materials with the exfoliated materials, ion insertion may be carefully controlled to obtain high monolayer yield while retaining the materials' structure and chemical composition.^[547,555] Ion-intercalated materials with higher layer separation have a weaker van der Waals interlayer interaction, resulting in a lower exfoliation energy barrier.^[524,559] Leveraging the lower exfoliation energy barrier, exfoliation by ion intercalation is frequently used to synthesize few-layer graphene, graphene oxide, TMDCs, and MXenes.^[91,556–558,560–564] Zeng et al. optimized the cathodic Li-intercalated exfoliation of TMDCs by examining the cut-off voltages and discharge currents for various materials.^[537,555] Later work by the same group demonstrated that the same approach might provide a high yield (up to 93%) of monolayer TaS_2 .^[554] Wang et al. compared and examined the exfoliation of graphene using various ionic salts.^[565] While cations play a significant role in increasing the yield of graphene exfoliation, larger anions enable more cations to be inserted between graphite layers (Figure 3d).^[565] The calculation validated the fabrication of few-layer graphene with SO_4^{2-} as an intercalant in anodic intercalation–exfoliation.^[558,564] In the presence of inorganic ions, Parvez et al. demonstrated the exfoliation of graphite into graphene.^[543] The resulting graphene exhibited lateral dimensions of up to 44 μm and a low degree of oxidation (17.2 C/O ratio), proving the viability of ion-intercalated exfoliation for large-area (lateral size >5 μm) and few-layered (3 layers) graphene synthesis on a large scale (Figure 3e).^[543] Shih et al. also reported using other intercalants such as iodine chloride (ICl) and iodine bromide (IBr) to control the thickness of exfoliated graphene, as these ionic chemicals would insert every second or third layer.^[556] While ion-intercalated exfoliation was shown to be a versatile and scalable method for synthesizing high-quality 2D materials, it was discovered that introducing additional phases during ion intercalation could significantly alter the product's electrical characteristics.^[557,566,567] Such a process can be observed in the intercalation of MoS_2 with alkali ions (Li^+ , Na^+ , and K^+), as the production of A_xMoS_2 , where A denotes the alkali ions, in the product would convert the semiconducting 2H MoS_2 to the metallic 1T phase.^[553,567–570] Numerous studies have been published to solve this issue, including postsynthesis annealing and the use of anion in the process rather than Li-ions.^[557,571] Eda et al. demonstrated a reversible transition from the metallic 1T to the 2H phase in exfoliated MoS_2 through

Li-ion intercalation followed by mild annealing at 300 °C.^[557] Lin et al. substituted the Li-ion with ammonium molecules as intercalants by controlling the number of molecules inserted into the MoS_2 interlayer spacing.^[571] This anionic intercalation–exfoliation technique may create pure 2H- MoS_2 nanosheets with a narrow thickness distribution while preserving a high on/off ratio and charged carrier mobility.^[571]

3.1.3. Ion-Exchange Exfoliation

Exfoliation by ion exchange is suited for materials with exchangeable ions intercalated into the interlayer gap. Most fall under the transition metal oxide, clay, and layered double hydroxide categories (LDH).^[524,538,539,572–576] As with ion-intercalated exfoliation, the interlayer space in bulk host materials was enhanced by adding bigger molecules, such as amino acid and iodine ions, via ion exchange, resulting in a weaker van der Waals interaction between layers.^[572,573] Ion-exchange exfoliation is an effective method for synthesizing one-unit-cell thick 2D materials whose bulk counterparts exhibit stronger interlayer interaction due to intrinsic ions in the bulk structure and are sensitive to oxidizing acids and alkali metals, making both mechanical exfoliation and ion-intercalation ineffective.^[577]

3.2. Bottom-Up Approaches

3.2.1. Chemical Vapor Deposition (CVD)

CVD has been widely used in synthesizing 2D materials, particularly for wafer-scale growth.^[578–581] CVD processes are often characterized by chemical reactions between precursors that are carried in the vapor phase.^[578] The precursors, the reaction chamber temperature and pressure, and the substrate temperature are all crucial growth parameters that must be properly regulated to produce products with the correct morphology, thickness, and orientation (Figure 4a).^[579,582,583]

Since the discovery of graphene in 2004 by Scotch tape exfoliation, CVD development of graphene has been intensively explored, both with and without catalyst usage.^[584–590] Several metals, including Pt, Cu, Ni, and Ru, have been employed as catalysts in the CVD growth of graphene, leveraging the understanding of CNT CVD growth.^[584,591–595] Somani et al. reported the first CVD development of multilayer polycrystalline graphene over a Ni substrate in 2006.^[584] Ismach et al. and Wei et al. have since demonstrated CVD development of graphene using copper as the catalyst.^[596,80] Shang et al. reported the catalyzer-free CVD synthesis of multilayer graphene on a Si substrate.^[587] As confirmed by transmission electron microscopy, the final products were discrete graphene nanoflakes with a thickness of 23 nm.^[587] Chen et al. later demonstrated the creation of few-layer graphene with a maximum lateral size of 30 μm using a catalyst-free, near-equilibrium CVD method.^[589] Although several works have demonstrated the ability to grow few-layer graphene with high reproducibility and performance comparable to exfoliated graphene, the polycrystalline graphene grown via the CVD process has significant application limitations due to the large number of grain boundaries that promote charge carrier

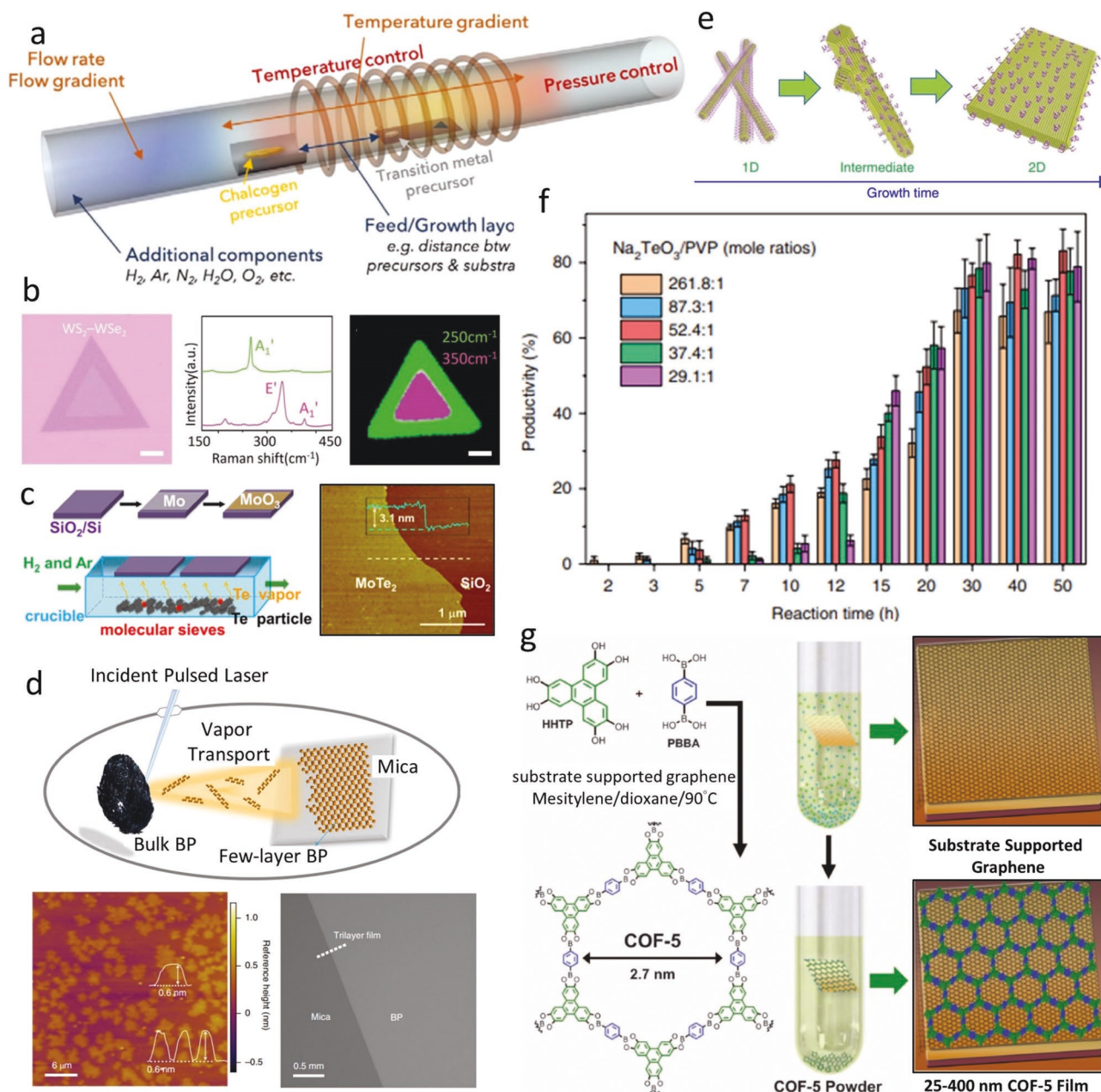


Figure 4. Bottom-up synthesis of 2D materials. a) Schematically illustration of a typical CVD process setup. Reproduced with permission.^[580] Copyright 2020, Elsevier. b) Optical microscopy images (top left), Raman spectra (top middle), spatially resolved Raman mapping images (top right), photoluminescence spectra (bottom left) and spatially resolved photoluminescence mapping images (bottom middle and right) of 2D WS_2/WSe_2 lateral heterostructures grown using CVD process. Reproduced with permission.^[636] Copyright 2017, The Authors, published by American Association for the Advancement of Science. c) Left: schematically illustration of PVD process of 2-H $MoTe_2$. Right: AFM image with height profile (inset) of PVD 2-H $MoTe_2$. A scratch was intentionally introduced to show the SiO_2 substrate. Reproduced with permission.^[634] Copyright 2015, American Chemical Society. d) Top: schematically showing the PLD process of BP fabrication. Bottom: AFM images of pulsed laser deposited monolayer BP with the height profile of monolayer BP. Reproduced with permission.^[683] Copyright 2021, The Authors, published by Springer Nature. e) Morphology evolution of 2D Te nanoflakes from 1D Te nanostructure. f) Productivity of hydrothermally synthesized 2D Te nanoflakes with different reaction time and Na_2TeO_3/PVP ratio. Error bars represent standard deviation. Reproduced with permission.^[281] Copyright 2018, Springer Nature. g) Schematically showing the synthesis of COF-5 through a graphene-templated liquid/solid interface. Reproduced with permission.^[796] Copyright 2011, American Association for the Advancement of Science.

scattering and impair carrier mobility.^[583,585,597] To accomplish the large-area synthesis of single-crystal graphene for industrial manufacture, scientists investigated and optimized graphene's

CVD growth mechanism.^[581,583,586,594,598,599] Li et al. investigated graphene's nucleation and growth mechanisms on a Cu sheet using chemical vapor deposition.^[594] The flow rate of

methane, the carbon precursor, was found to contribute differently to nucleation and growth: a low methane partial pressure is preferred for nucleation to minimize the number of crystals, whereas surface coverage demands a larger methane flow rate.^[594] Yu et al. further proved that the polycrystalline structure of the Cu films does not preclude the formation of single-crystal graphene and that nucleation may be induced during the CVD process using predetermined seeds to achieve larger single crystals.^[585] Wood et al. investigated the effect of the Cu facet on the graphene quality produced by CVD.^[583] The surface energy of the substrate facets was shown to be dependent on the number of defects in the deposited graphene, as high-index Cu facets result in more defects due to reduced diffusion and adsorption of carbon atoms.^[583] Murdock et al. established the critical role of aligned Cu facets in the quality of CVD-grown graphene.^[598] The Cu film facets directly affect the deposited graphene's orientations, shapes, and edges.^[598] Wang et al. have investigated the requirements for grain boundary-free coalescence of separate graphene domains into a single crystal on various surfaces.^[581] Besides the substrate's contribution to single-crystalline graphene formation, hydrogen was predicted to affect the quality of graphene deposited.^[586] Vlassioun et al. discovered that hydrogen worked as both an activator and an etching reagent for controlling the size and thickness of monolayer graphene.^[586] Further investigation found that when the hydrogen to methane partial pressure ratio exceeds 400, it is possible to obtain regular hexagonal graphene.^[586] Based on their understanding of the nucleation and growth of deposited graphene, the same group presented a potentially scalable CVD growth of single-crystalline graphene by pulling the Cu substrate forward.^[599] These advances in the nucleation and growth of graphene using a CVD technique toward large-area synthesis significantly advance the creation of single-crystalline 2D materials on a wafer-scale via CVD processes.

CVD growth of 2D TMDCs, such as MoS₂,^[600–603] MoSe₂,^[604–607] MoTe₂,^[608–610] and WS₂,^[611–615] has also been extensively reported previously. The typical CVD procedure for growing 2D TMDCs involves the direct reaction of chalcogen vapor with transition metal films or precursors.^[600,616–620] Unlike graphene growth in CVD, which often requires the application of a metallic layer over the growth substrate as a catalyst, CVD of 2D TMDCs can be performed directly on the substrate, such as sapphire, h-BN, graphene, or SiO₂, forming heterostructures without further transfer.^[621–623] Numerous studies have been conducted to optimize the growth conditions for 2D TMDCs to achieve large-area, high-quality single crystals. Lee et al. presented a CVD development of large-area MoS₂ via reduced graphene oxide (rGO) seeding employing MoO₃ and sulfur powder as Mo and S sources, respectively.^[600] Under high temperatures (650 °C), sulfur vapor can convert MoO₃ to the more volatile MoO_(3-x), reacting with sulfur to form MoS₂.^[600] The resulting MoS₂ film may have a lateral size of up to 2 mm due to the coalescence of the star-shaped MoS₂ film over time, while the thickness may vary between monolayer and several layers.^[600] Ling et al. studied the role of seeding promoters in the manufacture of large-area, monolayer MoS₂, including rGO, PTAS (perylene-3,4,9,10-tetracarboxylic acid tetrapotassium salt), and PTCDA (3,4,9,10-perylene-tetracarboxylic acid dianhydride).^[624] The inclusion of seeding promoters during the CVD of TMDCs

improves the nucleation sites and surface adhesive force of MoS₂, resulting in a more layered growth behavior that is more conducive to monolayer fabrication.^[624] Chen et al. suggested a novel technique for fabricating a large-area (lateral size of 350 μm) MoS₂ monolayer with a reduced number of MoS₂ nucleation sites via oxygen-assisted CVD.^[603] As the authors assert, oxygen as a carrier gas can shield MoO₃ from poisoning while also acting as an etchant for MoS₂.^[603] Wang et al. investigated the shape of 2D MoS₂ synthesized by CVD by changing the atomic ratio of the Mo and S precursors during growth.^[625] It was discovered that when the atomic ratio is stoichiometric, the film is hexagonal due to the similar growth rates of the Mo- and S-terminations, however when the stoichiometry is broken, the film forms truncated triangles or triangles due to the lower growth rates of either termination.^[625] CVD's nonstoichiometry affects not only the morphology but also the optical and electrical performance of 2D TMDCs.^[626] Kim et al. synthesized and evaluated MoS₂ with varying sulfur evaporation temperatures and exposure times and discovered that the sample with the least sulfur content displayed the greatest field effect mobility due to the presence of the most S vacancies that act as electron donors.^[626] By substituting WO₃ for MoO₃, the same procedure used to synthesize MoS₂ may also be used to synthesize 2D WS₂ with controlled shape and thickness.^[612,613,627,628] Zhang et al. reported the synthesis of large-area (lateral size of 50 μm), single-crystalline WS₂ on a sapphire substrate.^[628] The grain boundary propagation analysis of CVD-grown WS₂ revealed the critical role of early nucleation in fabricating large-area, single-crystalline WS₂.^[628] Rong et al. later demonstrated that adding sulfur vapor to the vaporized tungsten precursor over the growth substrate had an effect on the final morphology of the sample.^[627] Introducing sulfur vapor 20 minutes before the substrate reaches growth temperature results in thicker WS₂ films with a possible WS_(2+x) composition in the film's center, whereas delaying sulfur vapor introduction (after the substrate reaches growth temperature) results in an incomplete reaction and a greater number of defects.^[627] Tan et al. recently demonstrated CVD development of WS₂ on a wafer scale (2 in. in diameter) using precise control of precursor evaporation and growth temperatures in a multizone furnace.^[614] As with MoS₂, other chemicals may be added as promoters of WS₂ development.^[611] Li et al. presented an alkali halide-assisted synthesis of both WS₂ and WSe₂ by employing alkali halide compounds as growth promoters to enhance transition metal precursor delivery to the substrate.^[611] The resulting products can form monolayers with no detectable promoter doping.^[611]

Similar to the fabrication of MoS₂, selenization and tellurization of MoO₃ can be used to synthesize MoSe₂ and MoTe₂.^[604,609,629,630] Numerous publications have described the fabrication of large-area MoSe₂ and MoTe₂ films and discussed the critical growth parameters.^[605,610,619,631] Shaw et al. reported the first CVD synthesis of MoSe₂ using a 300 nm SiO₂/Si substrate and selenization of MoO₃.^[605] Hydrogen as a carrier gas aids in reducing MoO₃ to suboxide and the deposition of MoSe₂.^[605] Later work by Lu et al., Chang et al., and Wang et al. confirmed the reproducibility of this process and characterized the deposited MoSe₂ film's electrical and optical properties.^[604,631,632] By tuning the evaporation temperature of tellurium powder, Li et al. investigated the phase transition of

MoTe₂ from 1-T, 2-H to 1-T' phases.^[619] Increasing tellurization velocity can obtain phase-pure 2-H and 1-T' phases.^[619] Empante et al. demonstrated that different phases of MoTe₂ can be achieved by altering the post-deposition cool-down process.^[610] While a slow cooling rate produces semiconducting 2-H phases, metallic 1-T and 1-T' phases can be synthesized by quenching the sample at 350 and 450 degrees Celsius, respectively.^[610]

The ability to deposit 2D TMDCs and graphene directly onto various substrates enables the fabrication of heterostructures using an all-CVD process.^[623,633–636] While it is possible to create 2D van der Waals heterostructures by transferring and stacking different 2D materials together, precise control of stacking orientation and interface contamination are two significant challenges associated with this process.^[633,637–640] CVD growth of van der Waals heterostructures, on the other hand, enables a scalable manufacturing process with precise control over size and orientation and a clean interface.^[634,635,641,56] To avoid thermal degradation and uncontrolled nucleation, modifications to the CVD process have been made, such as the reverse-flow CVD reactor proposed by Zhang et al.^[636] Several heterostructures have been fabricated, including MoS₂/WSe₂,^[639] graphene/MoS₂,^[635] MoS₂/WS₂,^[640,642,643] graphene/h-BN^[634,639] and WS₂/h-BN (Figure 4b).^[623] Yang et al. realized a graphene/h-BN heterostructure and observed a trigonal moiré pattern superlattice.^[638] Gong et al. demonstrated the direct growth of WS₂ over a MoS₂ monolayer to produce a high-quality vertical and lateral WS₂/MoS₂ heterostructure.^[640] The low growth temperature was found to promote epitaxial growth of WS₂ along the edge of MoS₂, rather than in the out-of-plane direction, resulting in lateral heterojunctions along the zigzag and armchair directions.^[640] Zhai et al. fabricated a NiTe₂/MoS₂ heterostructure and demonstrated that the heterostructure's optical and electrical properties were enhanced when compared to the pristine materials.^[644] This enhancement is due to the absence of interfacial defects caused by epitaxial growth of NiTe₂ and the similar symmetry of the two materials, which reduces electron scattering.^[644] Zhang et al. also investigated the MoS₂/WSe₂ bilayer heterostructure and the resulting 2D superlattices.^[645] On MoS₂, the Moiré pattern with a lattice constant of 8.7 ± 0.2 nm was observed, as well as periodic variations in the atomic registry due to the lattice mismatch between the two materials.^[645] Such lateral atomic registration would affect interlayer coupling and thus the performance of 2D electronic and optoelectronic systems.^[643,645] Yang et al. created a vertical WSe₂/SnS₂ p–n junction with a bilayer thickness and lateral size scale of millimeters.^[641] The device demonstrated a low off-current (10^{-14} A) and a high on-off ratio (up to 10^7), as well as a rapid photo-response time.^[641]

Instead of sublimating volatile solid precursors, metal-organic chemical vapor deposition (MOCVD) uses organometallic vapors as precursors.^[646,647] Compared to conventional CVD, MOCVD enables low-temperature, large-area synthesis while allowing for greater control over growth conditions and sample morphology.^[646–649] Hoffman reported the first fabrication of MoS₂ and WS₂ via MOCVD, using Mo(CO)₆, W(CO)₆, and H₂S as the Mo, W, and S precursors, respectively.^[650] In subsequent work, Etzkorn et al. and Zink et al. synthesized MoS₂, WS₂, and MoSe₂ with various nanostructures.^[651,652] Kang et al. recently reported the growth of homogeneous MoS₂

and WS₂ films via MOCVD.^[33] The technique was demonstrated to apply to industrial-scale synthesis, and the resulting devices fabricated from the prepared films demonstrated a 99% device yield.^[33] Similarly, Eichfeld et al. synthesized WSe₂ with a domain length greater than 8 μ m.^[653] Marx et al. used MOCVD further to demonstrate the growth of MoS₂ on various substrates.^[648] It was demonstrated that MoS₂ grows 3D with a high nucleation density.^[648] Cun et al. proposed growing MoS₂ on sapphire and SiO₂ substrates on a wafer-scale by pre-depositing Na₂MoO₄ and reacting it with (C₂H₅)₂S in the furnace.^[654] Kim et al. conducted additional research on suppressing nucleation in MOCVD.^[649] Alkali halide salts may be used to enhance the domain of the MoS₂ by acting as seeding promoters and stabilizing the 2D nuclei for further growth.^[649]

3.2.2. Physical Vapor Deposition (PVD)

Physical vapor deposition (PVD) deposits atoms and molecules vaporized from solid or liquid sources in a vacuum or low gaseous plasma environment.^[655] PVD is widely used in the growth of a variety of materials with a variety of nanostructures due to its low growth temperature (which can reach as low as 150 °C), high throughput, and ease of control, such as ZnO nanowires,^[656,657] transition metal nanowhiskers,^[658,659] and carbon nanotubes.^[659,660] PVD techniques commonly used to fabricate 2D materials include evaporation deposition, sputter deposition, and pulsed laser deposition (PLD).^[661–669]

Evaporation deposition of 2D materials is typically carried out in a quartz tube with the substrate placed downstream of the sources and the carrier gas being an argon (Ar)/hydrogen (H₂) mixture.^[661–663] Atoms and molecules are directly vaporized at high temperatures (300 °C for MoS₂ and up to 850 °C for In₂Se₃, depending on the materials) and deposited onto substrates that are typically kept between 650 and 700 °C for 2D material growth.^[662,663,670] Sputter deposition employs a different mechanism for vaporizing atoms and molecules, bombarding the sources with high-energy ions or low-pressure plasma to vaporize the atoms in a vacuum environment (less than 30 mTorr).^[655,665] The materials' "sputter yield," defined as the number of atoms ejected from the source per incident ion hit, can be precisely controlled to achieve one atom per incident ion hit by adjusting the chamber pressure and the incident ion's and substrate's applied electrical potentials.^[665,671] Deng et al. demonstrated an uncatalyzed RF sputtering deposition of few-layer graphene on a carbon nanotube (CNT) and silicon wafer.^[672] However, it should be noted that high-energy incident ions may cause damage to the deposited graphene structure.^[665,672,673] The Raman spectrum of sputter-deposited graphene revealed a stronger defects peak (D peak) than the 2D peak, indicating a higher point defect density introduced by incident ions.^[665,672,673] 2D TMDCs, including MoS₂,^[664,674,675] MoTe₂,^[676,677] WS₂,^[678] and WTe₂,^[679] have been widely synthesized using sputter deposition using their bulk counterparts as sources.^[665] Huang et al. demonstrated that sputtering could deposit MoS₂ over a large area up to 20 cm² while maintaining good control over the number of layers deposited.^[675] Zhou et al. synthesized phase-pure 1T' and 2H MoTe₂ using Mo(1T'-MoTe₂), MoO_x (1T'-MoTe₂), and MoO₃ (2H-MoTe₂)

as precursors while maintaining the chalcogen vapor pressure (Figure 4c).^[634] However, because chalcogen atoms have a lower vapor pressure, bulk TMDCs as a precursor in the sputtering deposition and postsynthesis annealing processes may result in sub-stoichiometry of the TMDCs sources and chalcogen defect.^[665,680] While the former can be mitigated by utilizing sources with a transition metal-rich surface, the latter has proven difficult to resolve, particularly for MoTe₂, which is unstable and readily decomposes at a high temperature, Te-deficient environment.^[676,634] Huang et al. proposed and deposited a SiO₂ capping layer on the sputter-deposited MoTe₂ film to prevent chalcogen atoms from sublimating.^[676] Using the same strategy, the authors have also deposited additional TMDCs, including MoS₂ and WTe₂.^[676] The results demonstrated that the proposed SiO₂ capping layer provides adequate protection while maintaining the TMDCs' stoichiometry.^[675]

Rather than impinging the source with magnetrons, pulsed laser deposition (PLD) uses a focused high-energy laser (such as an ultraviolet laser) to vaporize atoms and molecules for vapor-solid deposition.^[665,681,682] Due to the laser-bombarded, high-kinetic-energy atoms and molecules that can migrate and diffuse on the substrate surface, the substrate temperature for the PLD process is lower than for sputtering deposition.^[682–686] PLD has been demonstrated to be a versatile bottom-up synthesis technique due to the pulsed laser's ability to vaporize most materials with localized, instantaneous high temperatures.^[682,687] Several 2D materials, including TMDCs,^[684,685,688–694] BP,^[668,683] and h-BN^[695,696] have been fabricated using PLD. Sajjad et al. demonstrated the deposition of h-BN using a short-pulsed laser with an average lateral size of over 10 nm and a thickness of 1.7 nm.^[695] Wafer-scale BP was also synthesized using PLD by Wu et al. with good hole mobility up to 617 cm² V⁻¹ s⁻¹ (Figure 4d).^[683] Serna et al. previously reported on the large-area, controlled-thickness synthesis of MoS₂ over various substrates.^[690] The morphology of the deposited MoS₂ film was tuned by adjusting the source materials' particle size, composition, and hot-pressing temperature.^[690] Additionally, control of the pulse frequency, laser power, substrate materials, substrate temperature, and cooling rate is critical for the quality, thickness, and surface roughness of the deposited 2D materials.^[665,682,688,691,697,698] While both the laser pulse frequency and power play a significant role in determining the atoms' departure rate, parameters related to the substrate can directly affect the quality of deposited films.^[685,697,699–701]

3.2.3. Molecular Beam Epitaxy (MBE)

Molecular beam epitaxy (MBE) is a widely used technique for growing 2D materials.^[702–708] A typical MBE process involved impinging atomic beams onto a heated substrate in an ultrahigh vacuum environment and monitoring the growth in situ using reflective high energy electron diffraction (RHEED).^[709–713] In comparison to CVD and PVD, MBE growth in an ultrahigh vacuum chamber has the potential to minimize surface contamination.^[709] Through instantaneous shut down of the effusion cells, molecular beams decouple sources sublimation from growth. It enables more precise surface morphology, doping level, and composition control.^[703,709,714] Garcia

et al. demonstrated the MBE growth of monolayer graphene on h-BN flakes.^[703] It was discovered that while the influx of source atoms in MBE has a negligible effect on sample quality, the substrate surface morphology plays a critical role in graphene epitaxy.^[703] Vishwanath et al. demonstrated the MBE of MoSe₂ over graphite, graphene, and CaF₂.^[715] It was discovered that the mobility of Mo adatoms is critical for the domain of MoSe₂.^[715] Poh et al. demonstrated that while lattice mismatch between the MoSe₂ epilayer and the h-BN substrate is tolerable, the ultra-flat, dangling-bond-free surface of h-BN is more critical for MoSe₂ epitaxy.^[714] Additionally, Roy et al. synthesized MoSe₂ and MoTe₂ via MBE.^[716] To compensate for the increased Se atom desorption from the substrates, the precursor beam equivalent pressure flux ratio between chalcogens and Mo was set to 15.^[714,716] Other pristine 2D materials, such as WSe₂,^[717–719] blue phosphorus^[720–722] and NbSe₂,^[704,723] can also be grown using MBE. Similar to CVD growth, MBE can be used to fabricate 2D heterostructures.^[713,724] Liu et al. reported on a ternary group III–VI composite GaTe_xSe_(1-x) grown via MBE and the corresponding GaTe_xSe_(1-x)/Si heterostructure.^[713] Diaz et al. demonstrated the MBE of a MoTe₂/MoS₂ heterostructure with a high number of mirror-twin boundaries (MTB).^[724] According to the authors, the high MTB density may be caused by chalcogen defects caused by a faster rate of desorption of chalcogen atoms during growth, which can be avoided by increasing the ratio of chalcogen to transition metal beam equivalent pressure flux ratio during growth.^[724]

3.2.4. Hydro/Solvothermal Synthesis

Hydro/solvothermal synthesis has been extensively studied and applied to the preparation of nanomaterials.^[725–732] Chemical reactions were carried out in the solution phase inside sealed reaction vessels at a reaction temperature greater than the solvent's boiling point and a vessel pressure greater than atmospheric pressure in typical solvothermal synthesis.^[733–735] Hydrothermal synthesis refers to using water as a solvent in a solvothermal reaction at a temperature greater than 100 °C.^[733] Hydro/solvothermal synthesis is controlled by reaction temperature and pressure, reaction vessel filling percentage (the ratio of solvent volume to reactor volume), solvent properties, other dissolved salts, and heating methods.^[733,734,736,737] Numerous modifications have been made to the hydro/solvothermal process to improve reaction kinetics, including using microwave heating, applying magnetic or electric fields, and mechanical mixing.^[727,738–740] In general, the temperature and pressure at which chemical reactions occur are determined by the reaction vessel's limits, the properties of the reactants, and the heating modes used.^[737]

Choucair et al. reported a solvothermal synthesis of gram-scale graphene (0.1 g graphene per 1 mL solution) using an equal molar ratio of ethanol and sodium.^[741] The precursor graphene was synthesized via solvothermal synthesis and then pyrolyzed and sonicated to form a graphene dispersion in water.^[741] In a sealed autoclave, an equimolar sodium/ethanol mixture formed clathrate-like structures. The ethanol was encapsulated within the metal alkoxide, and these ethanol-rich regions also served as ignition points for the nucleation of

graphene nanosheets.^[741] Xu et al. conducted additional research on self-assembled graphene hydrogels (SGH), utilizing graphene oxide (GO) dispersion as graphene precursors and establishing relationships between the as-prepared SGH properties and experimental parameters.^[742] By reducing the dispersion of aqueous graphene oxide at a high temperature (180 °C), the resulting graphene nanosheets can be further self-assembled into 3D hydrogels.^[742] With a sufficient concentration of GO and enough reaction time, 3D networks with cross-linking between partially overlapped graphene nanosheets can be formed.^[742] Deng et al. also presented a large-scale solvothermal synthesis of N-doped graphene using the reagents cyanuric chloride, lithium nitride, and tetrachloromethane.^[743] It was believed that the N doping occurred during the transformation of sp³ hybridized carbon in tetrachloromethane to sp² hybridized carbon in graphene, during which intermediates containing C=N and C=C groups were formed and coupled to form small domains during the subsequent dechlorination process.^[743] Chen et al. later reported on the hydrothermal synthesis of an N-doped graphene hydrogel using GO and an organic amine as precursors.^[744] Additionally, the nitrogen precursor acted as a structural modifier, increasing the pore size of the resulting graphene hydrogel and thus improving the performance of the graphene-hydrogel supercapacitor.^[744] Xing et al. used a similar structure tailoring strategy to synthesize N-doped graphene for lithium-ion battery anodes.^[745] Hydro/solvothermal synthesis could also be used to synthesize graphene-based composites.^[730,746,747] Zhang et al. demonstrated a one-pot hydrothermal synthesis of P25-graphene composites.^[748] Chemical bonding of TiO₂ nanoparticles to a reduced graphene nanosheet was used.^[748] Additionally, Perera et al. synthesized graphene-TiO₂ nanotube composites using GO and P90 as precursors for graphene and TiO₂, respectively.^[749] Apart from TiO₂/graphene composites, various graphene-based composites may be made using one-pot hydrothermal synthesis, including 2D hexagonal α -Fe₂O₃ nanoplates/graphene composites and VS₂/graphene composites.^[747,750]

Hydro/solvothermal synthesis was used to synthesize 2D TMDCs from a range of precursors.^[751–754] Zhan et al. described a solvothermal synthesis of MoSe₂ via hydrazine monohydrate reduction of MoO₃.^[755] Peng et al. later established the hydrothermal synthesis of MoSe₂ and MoS₂.^[756] Ammonium molybdate was used as the molybdenum precursor, combining elemental chalcogens and hydrazine monohydrate in the synthesis.^[756] Additionally, Huang et al. and Chaudhary et al. demonstrated the hydrothermal synthesis of MoS₂ utilizing sodium molybdate as the molybdenum source.^[757,758] Feng et al. studied the morphologies of MoS₂ nanosheets via solvothermal synthesis with various solvent combinations.^[759] Substituting N-methyl pyrrolidone for ethylene glycol in the water/ethanol/ethylene glycol mixture results in a change in the morphology of the as-prepared MoS₂ from fullerene-like (ethylene glycol) to nanosheets (N-methyl pyrrolidone), owing to the adsorbed organic molecules' different structure-directing ability on the MoS₂ surface.^[759] Miao et al. investigated the effect of gravity on the formation of nanostructured MoS₂ using a Ti-foil-assisted hydrothermal synthesis.^[760] The data indicate that the upper side of the Ti foil grew quicker than the lower side, resulting in the development of nanosheets.^[760] Takahashi et al. recently

demonstrated that by altering the reducing agent concentration and reaction duration in the presence of ascorbic acid, the thickness and phase of the resultant MoS₂ might be adjusted.^[761] The stacking of MoS₂ nanosheets can be inhibited using an ascorbic acid derivative adsorbed onto the basal plane.^[761] Besides MoS₂ and MoSe₂, hydro/solvothermal synthesis may be used to manufacture other 2D TMDCs and their composites.^[762–765] Arul and Han described a hydrothermal synthesis of 2D NiSe₂,^[764] whereas Wang et al. manufactured the 2D MoS₂/Bi₂S₃ composites utilizing a one-pot hydrothermal synthesis.^[765]

Hydro/solvothermal processes can also be used to produce 2D materials other than graphene and TMDCs.^[281,766,767] Wang et al. synthesized hexagonal Sb₂Te₃ nanoplates with lateral dimensions of several microns by using hexadecyltrimethylammonium bromide (CTAB) as a structural directing agent during the solvothermal synthesis.^[768] Shi et al. later reported on the hydrothermal synthesis of single-crystalline, impurity-free Sb₂Te₃ nanoplates without a structure-directing agent.^[769] It is worth noting here that structure-directing agents such as polyvinylpyrrolidone (PVP) and CTAB can be utilized to influence the development kinetics of specific facets by being adsorbed onto the crystal surface.^[281,770] Using PVP as a structure-directing agent, Xu et al. synthesized Bi₂Te₃ nanoplates by hydrothermal synthesis.^[771] The nonionic surfactant can adsorb onto the Bi₂Te₃ nuclei, preventing them from aggregating during the initial stages of production.^[771] A similar technique was used in Wu et al.'s recent study on the synthesis of tellurene nanoflakes.^[281] Adjusting the Na₂TeO₃/PVP ratio during synthesis makes it possible to create 2D Te flakes with an edge length of up to 100 μ m and a monolayer thickness (Figure 4e,f).^[281] Zhu et al. recently presented a solvothermal method for fabricating 2D black phosphorus utilizing red phosphorus as a precursor.^[766] The phase transition percentage of red phosphorus to phosphorene can reach 100%, and a batch yield of 2.1 g can be attained, indicating a highly scalable technique.^[766]

3.2.5. Interface-Assisted Synthesis

Interface-assisted synthesis is a commonly used process for fabricating 2D materials, particularly 2DP, 2D COF, and 2D MOF.^[772] Numerous techniques have been devised, including the air/liquid interface (Langmuir-Blodgett, LB) approach, the liquid/liquid interface method, and the liquid/solid (templating) method.^[773–775] This section will discuss the recent progress made by these techniques in creating 2D materials.

The LB method is a simple method for fabricating a large area, ordered 2D polymerized nanosheets at the air-water interface by trapping water-insoluble monomers in an LB trough and polymerizing them into a 2D monolayer.^[775–777] The content and structure of the resulting 2D polymerized materials can be modified by adjusting the surface pressure of water in the LB process.^[778–782] Numerous 2DP, 2D COF, and 2D MOF syntheses have been carried out using LB techniques.^[783–786] Motoyama et al. used the LB approach to fabricate layered MOFs containing 5,10,15,20-tetrakis(4-carboxyphenyl)porphyrin (H₂TCPP) and copper ions.^[787] After annealing at 200 °C, the resultant MOF remained highly crystalline and retained its structural integrity.^[787] Makiura et al. reported the monolayer

MOF NAFS-1 by layering a CHCl_3 /ethanol solution containing monomers on top of a Cu^{2+} aqueous solution, resulting in the production of pyridine-capped 2D arrays.^[788] Dai et al. reported on the assembly of 2D imine-linked COF using the LB technique.^[789] Two aromatic organic monomers were polymerized into a polyimine monolayer by spreading a dilute chloroform solution over water.^[789] The polymer sheet coalesced and formed a homogenous monolayer at the interface when the surface pressure exceeded 3 mN m^{-1} .^[789] Murray et al. also described the creation of a synthetic 2DP monolayer at the air/water boundary.^[784] Compressing the monomer film and then photopolymerizing it results in a μm^2 -scale 2DP film.^[784]

The liquid–liquid synthetic approach involves dissolving the monomers of 2D materials in two distinct, immiscible solvents and polymerizing them at the phase boundary.^[772,790] Monomer diffusion from the bulk solution to the liquid/liquid interface is critical for synthesis.^[791,792] Takada et al. produced electrochromic MOF nanosheets containing Fe^{2+} and Co^{2+} using dichloromethane and water.^[793] At the water/dichloromethane interface, another study by the same group produced a photoactive, multilayered bis(dipyrinato)zinc(II) complex nanosheet.^[786] The scientists highlighted that monolayer and few-layer nanosheets could also be generated by switching to air/liquid interface synthesis.^[786] Rodenas et al. constructed a free-standing CuBDC MOF nanosheet using a three-layer solvent system comprised of three distinct volume ratios of DMF/ CH_3CN .^[432] The intermediate segment was included to control the growth of MOF by limiting direct contact between the monomer-containing solution and the monomer-containing solution.^[434] The resulting MOF nanosheet showed lateral size and thickness ranging from $0.5\text{--}40 \mu\text{m}$ and $5\text{--}25 \text{ nm}$, respectively.^[434] Dey et al. reported a liquid/liquid interfacial production of a 2D COF thin-film employing dichloromethane and water as solvents.^[794] The described approach may be used to synthesize a variety of 2D nanoporous COFs, including Tp-Tta, Tp-Bpy, and Tp-Azo.^[794] At the pentane/water interface, Zhong et al. demonstrated a wafer-scale synthesis of 2D porphyrin polymer with monolayer thickness.^[795] The innovative laminar assembly polymerization enables the scalable synthesis of covalent and coordination 2D polymers with uniform thickness across the film by regulating the monomers and polymerization kinetics.^[795]

The liquid/solid interface approach fabricates 2D materials by utilizing solid templates. Chemically or physically adsorbed precursor molecules adhere to the template surface and are ordered following the substrate's orientation and pattern.^[772] Graphene, ionic salts, and metals are frequently reported substrates for liquid/solid interface synthesis.^[772]

Colson et al. demonstrated the formation of 2D COF-5 on monolayer graphene supported on a substrate using a liquid/solid interfacial fabrication method (Figure 4g).^[796] With a sample thickness and edge size of $\approx 7 \text{ nm}$ and $\approx 50 \text{ nm}$, respectively, the resultant COF-5 exhibited good crystallinity and long-range order.^[796] It was later suggested that oxidized graphene with oxygen-containing functional groups might facilitate the nucleation and development of 2D noble metals more effectively than chemically inert pristine graphene.^[797–801] Huang et al. used graphene oxide as a template to produce few-layer Au square nanosheets with an edge dimension of $200\text{--}500 \text{ nm}$.^[799]

When the resulting Au nanosheet thickness is around 2.4 nm , it exhibits hexagonal closed packing (hcp), transforming into face center cubic (fcc) packing as the nanosheet thickness grows.^[799] Later work by the same group demonstrated the creation of Au@Ag, Au@Pd, and Au@Pt core–shell nanosheets based on the hcp Au square nanosheet using graphene oxide as a template.^[801,802] Additionally, ionic salts such as NaCl and Na_2SO_4 have been employed as templates for forming 2D metal oxides and disulfides.^[803–808] Ren et al. used NaCl particles as a template to fabricate small MoS_2 nanosheets composites for lithium-ion battery anodes.^[804] Ma et al. also described the creation of MoS_2 /graphene nanosheets using a Na_2SO_4 template for hydrogen evolution.^[803] In both cases, the ionic salt particles were combined with precursors and then washed away following the completion of the synthesis.^[803,804] In liquid/solid interfacial 2D material synthesis, metal substrates functioned as both template and catalyst, analogous to the growth of graphene on Cu and Ni foil via CVD.^[809–812] Graphdiyne, a 2D carbon allotrope, and other 2D graphitic materials have previously been synthesized utilizing liquid metals and copper foil.^[812–814]

4. 2D Materials Based Wearable Energy Harvesting Technologies

In this section, we will comprehensively review the application of 2D materials in wearable energy harvesting from solar,^[46,815,816] thermal,^[817,818] mechanical,^[819,820] and other sources of energy.^[821–823] Table 1 summarizes the working mechanism and the corresponding desired materials properties of various energy harvesting techniques and suggests a list of potential candidates.

4.1. 2D Materials-Based Photovoltaic Energy Harvesting

4.1.1. Working Mechanism of Photovoltaic Energy Harvesting

A photovoltaic or solar cell turns sun irradiation, the most plentiful renewable energy source, into electricity. Photocurrent generation can be summarized in two steps: 1) Incoming light is absorbed by the specified materials, resulting in the formation of electrons and holes; and 2) the carriers are separated and collected at two terminals in the presence of an electrical field (Figure 5a).^[824–826] Numerous solar cell designs have been developed to increase the cell's conversion efficiency.^[824,888–890] Typical photovoltaic absorbing materials include silicon, metal halide perovskite, and other semiconducting materials such as $\text{Cu}(\text{In,Ga})\text{Se}_2$, all of which have absorption spectra that are reasonably close to those of the sun.^[827–832] Massive efforts have been made to increase photovoltaic cell efficiency by tuning the electrical and optical properties of the photovoltaic absorbing material, optimizing the photovoltaic cell's design parameters, and converting the phonons that could not be adsorbed into suitable wavelength through a spectral converter.^[891–897] Due to their unique electrical and optical features, 2D materials have generated considerable interest as candidates for next-generation solar cells.^[898] By leveraging their superior flexibility and stretchability over stiff, traditional solar cell materials, 2D

Table 1. Summary of various energy harvesting techniques, the corresponding desired material properties, and reported demonstrations based on 2D materials.

Energy harvesting techniques	Working mechanism	Desired materials properties	Reports based on 2D materials
Photovoltaic cell	Electron–hole pairs generated upon light illumination are separated and collected at the two terminals under an electrical field. ^[824–826]	• Absorption spectra close to the sun spectrum^[827–832] • Strong light–matter interaction^[833] • Long exciton and charge diffusion length^[834–836] • Long carrier lifetime^[836] • High charge mobility^[836] • Optical transparency^[837] • High absorption coefficient^[838,839]	• Graphene^[840,841] • TMDCs^[842,843] • Bp^[844–846] • MXene^[847,848] • 2D perovskite^[849,850] • 2D polymer^[851,852]
Thermoelectric energy harvesting	An electrical output is generated with a temperature gradient across the materials. ^[853]	• High Seebeck coefficient^[818,854] • High electrical conductivity^[818,854] • Low thermal conductivity^[818,854,855] • Wide operation temperature range^[856]	• Graphene^[818] • TMDCs^[857] • Tellurides^[858–860] • Tellurene^[297,298] • Bp^[109,861]
Pyroelectric energy harvesting	A current/voltage is generated by a temperature change with time. ^[862,863]	• Noncentrosymmetric crystal structure^[864,865] • Large pyroelectric coefficient^[866,867] • Low thermal resistance to heat transfer^[868] • Low volume specific heat^[866,879]	• Graphene^[869] • TMDCs^[870] • 2D perovskite^[871]
Piezoelectric energy harvesting	Electrostatic potential generated by mechanical deformation along the polarization direction of the piezoelectric material. ^[23,872,873]	• Noncentrosymmetric crystal structure^[874] • Nonzero bandgap^[874] • Large piezoelectric charge coefficient^[819]	• TMDCs^[875,876] • Bp^[877] • h-BN^[878] • 2D perovskite^[879,880]
Triboelectric energy harvesting	An electrical output was generated and collected by combining triboelectrification and electrostatic induction. ^[21,881]	• High charge density^[882] • Triboelectric pairs made of a triboelectric positive material and a triboelectric negative material are favored.^[50]	• Graphene^[883] • TMDCs^[884] • Bp^[885] • MXene^[886] • MOF^[887]

materials-based photovoltaic cells have opened the door to integrating wearable light energy harvesters into clothing and accessories.^[46,899]

4.1.2. Performance of 2D Materials-Based Photovoltaic Cells

Graphene has been extensively investigated as a semimetal with exceptional optical transparency and excellent flexibility for use as transparent conductive or counter electrodes in flexible solar cells.^[900–906] Due to the scarcity of tin and indium and the brittle nature of metal oxide, conventional indium tin oxide (ITO) and fluorine-doped tin oxide (FTO) electrodes are expensive to make. They are not suitable for flexible solar cell electrodes.^[840,900,907–911] Additionally, ITO is transparent only when exposed to visible light, owing to its high bandgap of 4 eV and free carrier absorption.^[912] On the other hand, graphene has high transmittance of 99.7% across the solar spectrum and a low transparency degradation of 2.3% per layer, making it an outstanding transparent electrode for flexible solar cells.^[48] De Arco et al. demonstrated CVD-grown graphene as the flexible organic photovoltaic cell electrode in place of ITO.^[900] The graphene electrode-based cell achieved a power conversion efficiency (PCE) of 1.18%, comparable to the ITO electrode-based cell.^[900] Yin et al. constructed a flexible organic photovoltaic cell employing reduced graphene oxide electrodes and a polyethylene terephthalate (PET) substrate.^[913] The device was reported to be capable of withstanding 2.9% tensile strain for 1000 bending cycles.^[913] However, due to the hydrophobic nature of graphene, the success

rate of producing graphene-based solar cells remained difficult since the homogeneous deposition of poly(3,4-ethyldioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) onto graphene electrodes was difficult to achieve.^[910] Park et al. doped graphene with AuCl₃ to overcome this issue by enhancing graphene's surface wetting and overall device performance.^[910] Lin et al. also synthesized graphene doped with boron as the back electrode of a CdTe solar cell.^[914] The material's work function increases by doping CVD-grown pure graphene with boron, resulting in increased solar efficiency and hole-collecting capabilities.^[914] Miao et al. doped graphene with bis(trifluoromethanesulfonyl) amide (TFSA) to create graphene/n-Si Schottky junction solar cells with an 8.6% PCE.^[107] Yang et al. enhanced solar cell performance by using a graphene oxide interlayer between graphene and silicon.^[915] Although the PCE of the dope-free cell is somewhat lower (6.8%), its superior air stability (95% PCE retention at 22°C and 60% relative humidity after one week) outperformed the chemically doped cells (which preserved just 50% efficiency under the same conditions).^[915] Jiao et al. demonstrated a flexible graphene–Si Schottky junction solar cell employing an ultrathin Si sheet (thickness 10.6 nm), which demonstrated superior flexibility in comparison to bulk analogs.^[899] Additionally, the polymethylmethacrylate (PMMA) film that served as the substrate for graphene CVD was preserved to act as an antireflective and protective layer for the graphene.^[899] Liu et al. used graphene as electrodes to create a flexible perovskite solar cell.^[916] The solar cell demonstrated excellent mechanical flexibility while achieving a high PCE and specific power density of 11.5% and 5 W g^{−1}, respectively, which are significantly

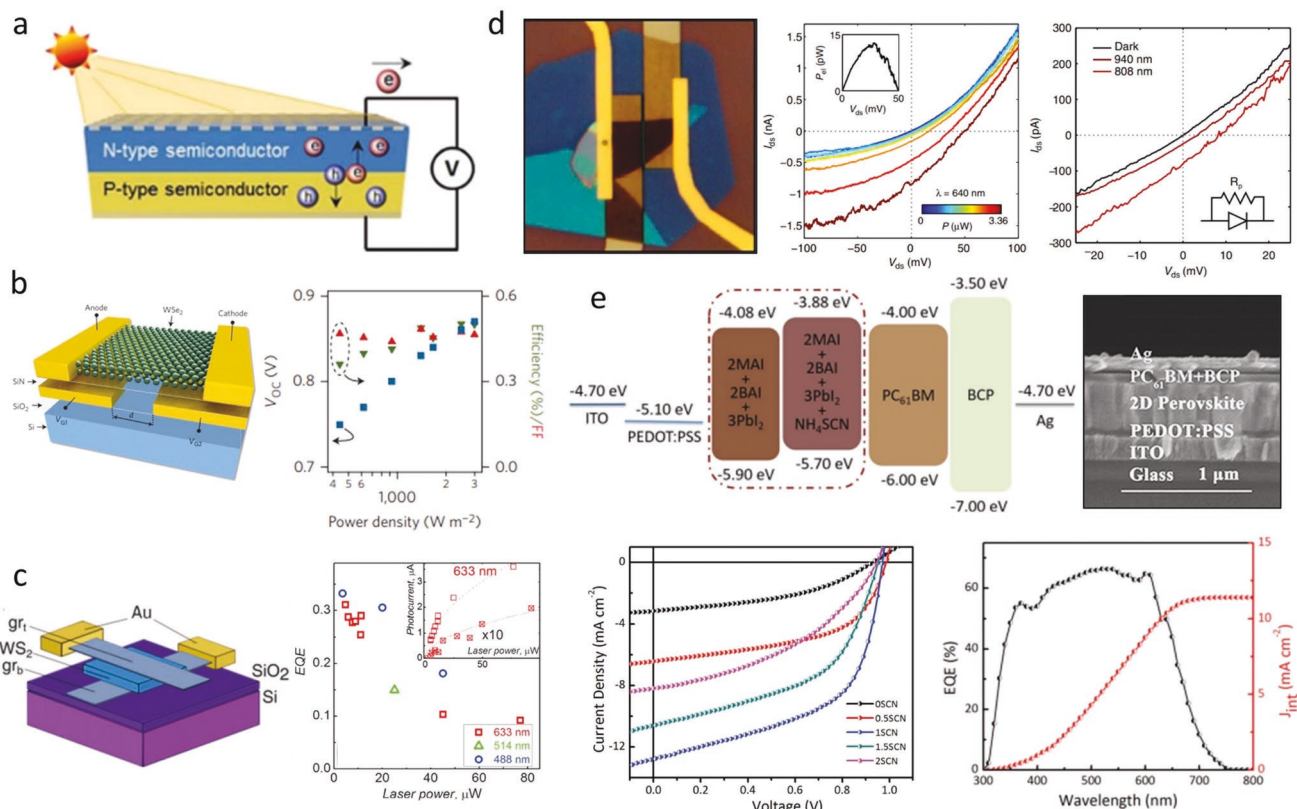


Figure 5. Working mechanism, performance and demonstration of 2D materials-based wearable photovoltaic cell. a) Schematically showing the working mechanism of a typical photovoltaic cell. Reproduced with permission.^[1164] Copyright 2016, the Royal Society of Chemistry. b) Left: schematic illustration of monolayer WSe₂-based p-n junction diode with split-gate configuration. Right: open-circuit voltage (blue), fill factor (FF, red) and PCE (green) of the WSe₂-based diode versus the incident light intensity. Reproduced with permission.^[920] Copyright 2014, Springer Nature. c) Left: schematically showing the architecture of WS₂-based photovoltaic cell. Right: the external quantum efficiency of the MoS₂-based photovoltaic cell under different laser power and wavelength. Inset: photocurrent of MoS₂-based photovoltaic cell under 633 nm laser with different power. Open symbols and cross symbols represent devices on Si/SiO₂ substrate and flexible substrate, respectively. Reproduced with permission.^[918] Copyright 2013, American Association for the Advancement of Science. d) Left: the optical image of the locally gated h-BN/BP p-n junction. Output characteristics of the locally gated h-BN/BP p-n junction as a function of incident light intensity (640 nm, middle) and in dark (black line in the right subfigure) and under different excitation wavelength at fixed light power (0.33 μ W). Inset schematically showing the equivalent circuit. Reproduced with permission.^[845] Copyright 2014, Springer Nature. e) Top left: energy band alignment of (BA)₂(MA)₂Pb₃I₁₀-based planar-structured 2D perovskite solar cell (PVSC). Top right: SEM image of the PVSC configuration based on (BA)₂(MA)₂Pb₃I₁₀ 2D perovskite film. Bottom left: J-V curves of (BA)₂(MA)₂Pb₃I₁₀-based PVSC with different amounts of NH₄SCN under 1 sun condition. Bottom right: the external quantum efficiency (EQE) spectrum and the corresponding integrated photocurrent of (BA)₂(MA)₂Pb₃I₁₀-based PVSC as a function of irradiation wavelength. Reproduced with permission.^[849] Copyright 2017, Wiley-VCH.

higher than those found in commercial solar cells.^[916] Bae et al. previously reported on the roll-to-roll fabrication of graphene electrodes and subsequent wet chemical doping over Cu substrates.^[917] A 30-inch-long (in diagonal) graphene film may achieve 97.4% transmittance while maintaining a low sheet resistance of 125 Ω sq⁻¹ for pure graphene, and further doping graphene using HNO₃ can reduce the sheet resistance to 30 Ω sq⁻¹.^[917] The technique has demonstrated significant potential for large-scale graphene electrode production for flexible solar cells.^[917]

2D TMDCs with a tunable bandgap and a strong light-matter interaction are potential photoactive layer candidates.^[815,816,918] Monolayer TMDCs have been demonstrated to absorb 5–10% percent of incident sunlight, an order of magnitude more than GaAs and Si.^[919] Bernardi et al. demonstrated a high PCE (up to 1%) when a bilayer MoS₂/WS₂ heterojunction was used as the active layer.^[919] Pospischil et al. produced a lateral p-n

junction diode with split-gate electrodes for solar cells and light-emitting diodes using monolayer WSe₂ (Figure 5b).^[920] While functioning as a photovoltaic cell, the final device had a PCE of 0.5% and a transparency of 95%.^[920] Britnell et al. examined the light-matter interaction in 2D TMDCs/graphene heterostructures and discovered enhanced light-matter interactions due to TMDCs' van Hove singularities (Figure 5c).^[918] Due to the unique electrical structure of TMDCs, photon absorption and electron-hole generation can be enhanced, increasing the external quantum efficiency by over 30%.^[918] Tsai et al. later fabricated a type-II heterojunction of MoS₂/p-Si.^[898] The monolayer MoS₂ introduced an electrical field into the proposed device, which increased carrier separation.^[898] The device has a PCE of 5.23%, which is the highest value among monolayer TMDC photovoltaic solar cells.^[898] Wi et al. proposed a photovoltaic system with an ITO/n-doped MoS₂/p-doped MoS₂/Au electrode that utilizes CHF₃ plasma to transform the

surface layer of MoS₂ to p-type semiconductors.^[921] The device had a decreased reverse dark leakage current, a high short circuit current density of 20.9 mA cm⁻², and a PCE of 2.8% due to the plasma-induced doping of MoS₂.^[921] Vikraman et al. proposed using a WSe₂/MoS₂ heterostructure in place of Pt as the counter electrode for dye-sensitized solar cells (DSSCs).^[922] While Pt electrodes are extensively employed in traditional DSSC, their instability in iodine-containing electrolytes and high cost severely limits DSSC performance.^[922–924] When used as the photoanode, the WSe₂/MoS₂ heterostructure with low charge transfer resistance at the electrolyte/electrode interface can achieve equivalent performance to traditional DSSC, indicating great potential for next-generation DSSC electrodes.^[922] Zhou et al. demonstrated an all-2D material photovoltaic cell using graphene electrodes and a MoS₂/WS₂ heterostructure as photoactive layers.^[925] When sandwiched between two graphene electrodes, the MoS₂/WS₂ bilayer enabled rapid and direct photogenerated carrier transfer to the top and bottom graphene electrodes, resulting in the photovoltaic effect.^[925] Additionally, the photovoltaic effect of this device may be tuned by adjusting the electron-hole recombination rate at the MoS₂/WS₂ interface when an external back-gate voltage is applied.^[925]

Other new 2D materials, such as BP and MXene, can be employed as photoactive materials and electrodes.^[844,846,847,926–928] Buscema et al. reported a split-gate BP FET with h-BN as the gate dielectric (Figure 5d).^[845] Electrostatic control can be used to convert the ambipolar BP channel to a p–n junction.^[845] The BP p–n junction can generate an open-circuit photovoltage of 50 mV and a short-circuit photocurrent of 1 nA with local-gating control.^[845] Tang et al. synthesized an MXene/silver nanowire (AgNW) hybrid film as the electrode in flexible organic solar cells.^[928] The obtained electrode was used to fabricate a flexible organic photovoltaic cell with an 8.3 percent PCE.^[928] Further mechanical durability testing revealed that after 1000 bending-unbending cycles, the as-fabricated cell with MXene/AgNW electrodes retained 84.6 percent of its maximum performance, confirming its high mechanical stability and flexibility.^[928] Ahmad et al. also proposed replacing the counter electrode in a DSSC with 2D Ti₃C₂.^[929] Additionally, it was demonstrated that when 2D Ti₃C₂ is used as the counter electrode, a twofold increase in PCE may be attained compared to the traditional DSSC configuration.^[929]

Although few have reported its flexibility or stretchability, 2D perovskite and 2D polymer photovoltaic cells have garnered much attention and demonstrated significant promise for wearable solar energy harvesting. Cohen et al. presented a 2D/3D mixed dimension (PEA)₂(MA)_{n-1}Pb_nBr_{3n+1} photovoltaic energy harvesting material (PA = phenyl ethyl-ammonium and MA = methyl-ammonium).^[850] Compared to pure 3D HOIPs, the 2D/3D HOIPs demonstrated a larger open-circuit voltage due to the 2D HOIPs' reduced charged carrier mobility and transport.^[850] Additionally, Zhang et al. developed a vertically oriented perovskite solar cell using 2D layered (BA)₂(MA)_{n-1}Pb_nI_{3n+1} (BA = CH₃(CH₂)₃NH₃ and *n* = 3, 4).^[849] As-fabricated solar cells demonstrated a PCE and open-circuit photovoltage of up to 8.79% and 0.98 V, respectively (Figure 5e).^[849] Additionally, the authors highlight that the proposed solar cell demonstrated excellent stability in a pure N₂ environment, as the solar cell's PCE remained unaltered compared to that under atmospheric conditions.^[849] Chen et al. also used a 2D HOIP interfacial layer between the perovskite and the hole transporting layers in a 2D perovskite solar cell.^[930] By utilizing the hydrophobicity of 2D HOIPs and the interfacial layer, Chen et al. improved the stability and output performance of the perovskite solar cell.^[930] The α-CsPbI₃ solar cell constructed by Zhang et al. employs a similar protective technique.^[931] The application of EDAPBI₄ as a protective layer efficiently prevents the production of undesirable δ-CsPbI₃ at room temperature while retaining the solar cell's output performance.^[931]

Due to their small weight and low manufacturing cost, polymer solar cells (PSCs) with exceptional flexibility have generated significant attention.^[932,933] By lowering the HOMO level in PSCs, the usage of 2D conjugated polymers can increase the open-circuit voltage.^[934,935] Huo et al. enhanced the PCE of PSCs by employing alkylthienyl groups as side chains in BDT (benzo[1,2-b:4,5-b']dithiophene) monomers.^[934] The decreased HOMO and LUMO energy levels increased the mobility of the charged carriers, hence increasing the short current density of the resultant PSC.^[934] Additionally, 2D conjugated polymers containing additional side chains such as linear alkylthio groups, alkylsilyl groups, and chlorine substitutions were synthesized to enhance the performance of PSCs by modifying the HOMO energy level.^[851,852,936] Recent progress of 2D materials-based wearable photovoltaic energy harvesting is summarized in Table 2.

Table 2. Summary of recent progress in 2D materials-based wearable photovoltaic energy harvesting.

Materials	Substrate	V _{OC} [V]	Conversion efficiency	Refs.
Graphene	PDMS	–	–	[909]
Graphene	ITO/PET	0.48	1.18	[900]
rGO	PET	0.56	0.78	[913]
CdSe QD-modified graphene	PET/ITO	0.52	0.60	[906]
Graphene	ITO	0.68	7.10	[937]
Graphene	Si film	0.44	5.09	[899]
Graphene	PET	1.04	11.50	[916]
Ti ₃ C ₂ T _x MXene	PTB7-Th:PC71BM	0.90	8.30	[928]
MoS ₂	TiO ₂ film	0.57	4.21	[938]
MoS ₂	Graphene	0.92	12.12	[939]

4.2. 2D Materials-Based Thermoelectric Energy Harvesting

4.2.1. Working Mechanism of Thermoelectric Energy Harvesting

Thermoelectric devices can harvest heat from the environment, such as geothermal energy, power plant waste heat, and automotive exhaust heat, and convert it to electricity (Figure 6a).^[817,940–942] The Peltier and Seebeck effects are two fundamental principles that govern the development and optimization of thermoelectric materials and systems.^[853,943] The Seebeck effect refers to the voltage difference caused by a temperature gradient between two materials. In contrast, the Peltier effect is the heat absorbed or rejected at a junction when an electrical current is carried through.^[853] The dimensionless figure of merit, $ZT = S^2\sigma T/\kappa$, is used to evaluate the thermoelectric materials' performance, where S denotes the Seebeck coefficient, σ denotes the electrical conductivity, κ is the thermal conductivity, and T denotes the absolute temperature.^[818,940] A good thermoelectric material, in general, should have a high Seebeck coefficient, a high electrical conductivity, and low heat conductivity.^[818,944] According to calculations, semiconductors having a charged carrier mobility of 10^{19} – 10^{21} cm^{−2} would exhibit the highest ZT values (Figure 6b).^[941,945,946] While the Seebeck coefficient, thermal conductivity, and electrical conductivity of conventional bulk thermoelectric materials are all interdependent and cannot be controlled independently due to the Wiedemann-Franz law, shrinking the dimensions of thermoelectric materials results in the quantum confinement effect and phonon scattering at the interface, which aid in enhancing and tuning the Seebeck coefficient and electrical conductivity, as well as reducing thermal conduction.^[947–949] Due to their reduced thermal conductivity and relatively higher Seebeck coefficient and electrical conductivity compared to their bulk counterparts, 2D materials are thought to be excellent candidates for wearable thermoelectric nanogenerators.^[857,858,942,950–952]

4.2.2. Performance of 2D Materials-Based Thermoelectric Nanogenerator

Pristine graphene is not an appropriate thermoelectric material due to its high electrical and thermal conductivity.^[818] Numerous attempts have been made to tune the band structure of graphene to increase the Seebeck coefficient and decrease thermal conductivity, including the formation of nanostructures, the introduction of dopants or defects, and the formation of nanocomposite materials with other materials such as PEDOT:PSS.^[818,953] Park et al. demonstrated a flexible thermoelectric nanogenerator based on graphene/PEDOT:PSS (RCG/P).^[953] By coating graphene with PEDOT:PSS, a conductive polymer with high electrical conductivity, the four-fold decrease in thermal conductivity compensates for the slightly lower Seebeck coefficient of RCG/P films compared to pure graphene and is projected to demonstrate a high ZT value.^[953] Novak et al. demonstrated nonoxidative exfoliation of n-type graphene (NOGF) with a high power factor ($P = S^2\sigma$) of 673 $\mu\text{W m}^{-1} \text{K}^{-1}$ and excellent mechanical stability (Figure 6d).^[954] PVP adsorption onto graphene during exfoliation was crucial for improving thermoelectric performance, as PVP-adsorbed

graphene exhibited a significantly more significant n-type Seebeck coefficient of $-45.3 \mu\text{V K}^{-1}$ than pristine graphene ($S = 1.2 \mu\text{V K}^{-1}$).^[954] Additionally, when a 14 °C temperature differential is introduced to the wearable NOGF-based thermoelectric nanogenerator, an output voltage of 2.2 mV is obtained.^[954] Li et al. also used Cu₂Se as a host material to insert graphene to improve thermoelectric performance at high temperatures (870 K).^[955] A ZT value of 2.44 ± 0.25 was attributable to the increased phonon scattering caused by the addition of graphene to Cu₂Se.^[955] Phosphorene has gained considerable interest in thermoelectric energy harvesting due to its high Seebeck coefficient (335 $\mu\text{V K}^{-1}$ at ambient temperature), strong carrier mobility, and variable bandgap.^[109] The in-plane anisotropy of phosphorene results in a prominent electrical conducting path that is orthogonal to the direction of heat conduction, resulting in a large σ/κ ratio that favors a high ZT value.^[956,957] Due to the distinct orthogonal significant electrical and thermal conduction directions, it was projected that the ZT value of monolayer phosphorene might surpass 1 at room temperature in the armchair direction.^[956] Further calculations by Lv et al. indicated that by applying strains in the zigzag (5%) and armchair (8%) directions, the ZT value can reach 1.65 and 2.12, respectively.^[958] Due to the band convergence caused by the applied strain, both the Seebeck coefficient and electrical conductivity increase, which both contribute to the increase in ZT.^[958] Saito et al. recently reported on a gate-tuned phosphorene-based thermoelectric harvester employing a double-layer electric transistor architecture (Figure 6c).^[959] The Seebeck coefficient can be increased to 510 $\mu\text{V K}^{-1}$ by adjusting the gate voltage, compared to 340 $\mu\text{V K}^{-1}$ for the bulk analog.^[959] However, no investigation of the device's mechanical properties was conducted.^[959]

Monolayer TMDCs with a thermal conductivity lower than graphene have garnered considerable attention for thermoelectric applications.^[960–964] The X–M–X sandwiched structure's weaker M–X bonds and the TMDC-based heterostructures contribute significantly to the lower thermal conductivity.^[965,966] Additionally, Buscema et al. demonstrated the photothermoelectric effect on monolayer MoS₂ when a laser was applied to the MoS₂ FET.^[952] The photocurrent generated in this work was attributed to the difference in Seebeck coefficients between the MoS₂ and metal electrodes caused by photothermal irradiation.^[952] When an external electric field was added, a tunable Seebeck coefficient ranging from -4×10^2 to $-1 \times 10^5 \mu\text{V K}^{-1}$ was determined.^[952] Huang et al. also conducted thermoelectric calculations on MoS₂, MoSe₂, WS₂, and WSe₂ using a variety of dopants and crystal orientations.^[857] In contrast to phosphorene, which exhibits in-plane electrical and thermal conductivity anisotropy, the crystal orientation of TMDCs had a minor effect on both electrical and thermal conductivity in monolayers of TMDCs.^[857] Further calculations indicated that the ZT values of p-type WS₂, n-type WSe₂, and MoSe₂ do not increase linearly with temperature due to the formation of valley states in band structures with energies close to the conduction band minimum, which may promote electron transport at elevated temperatures.^[857] Xie et al. proposed a flexible thermoelectric nanogenerator with an active material composition of MoS₂/graphene.^[967] The resulting MoS₂/graphene composite demonstrated superior electrical conductivity and comparable thermal conductivity to pure MoS₂, resulting in a roughly

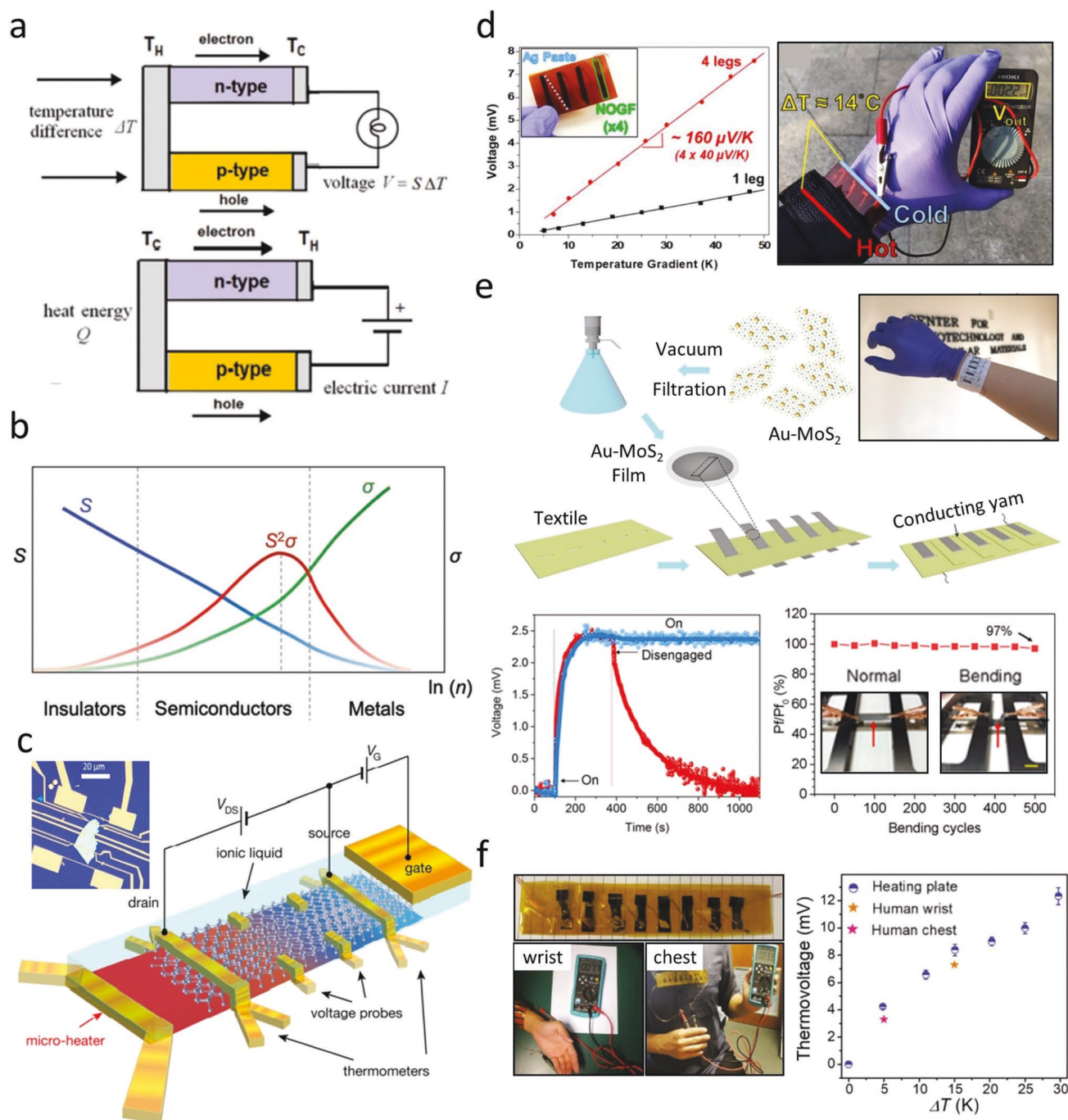


Figure 6. Working mechanism, performance, and demonstration of 2D materials-based wearable thermoelectric energy harvesters. a) Schematic illustration of working mechanism of thermoelectric generator (top) and thermoelectric cooler (bottom). Reproduced with permission.^[950] Copyright 2017, the Royal Society of Chemistry. b) Schematically showing the Seebeck coefficient (S), electrical conductivity (σ), and power factor ($S^2\sigma$) as a function of carrier density (n). Reproduced with permission.^[817] Copyright 2019, Wiley-VCH. c) Schematic illustration and optical microscopy image (upper left corner) of BP-based device for thermoelectric measurements. Reproduced with permission.^[959] Copyright 2016, American Chemical Society. d) Left: thermovoltage output of a 4-leg nonoxidized graphene flakes (NOGF)-based thermoelectric nanogenerator as a function of temperature gradient. Right: demonstration of 4-leg NOGF thermoelectric nanogenerator under a 14°C Celsius temperature gradient. Reproduced with permission.^[954] Copyright 2019, IOP Publishing. e) Top: schematically showing fabrication of Au-decorated 2D MoS_2 -based wearable thermoelectric device (upper right corner). Bottom left: thermovoltage output of the Au-decorated 2D MoS_2 -based thermoelectric wrist band. Blue: The wrist band stayed on the wrist. Red: The wrist band was taken off from the wrist once the output voltage reached a maximum. Bottom right: power factor change of the Au-decorated 2D MoS_2 -based thermoelectric wrist band after 500 bending cycles. Reproduced with permission.^[968] Copyright 2018, American Chemical Society. f) Left: photos of the FTE device and demonstration of applying the FTE device to wrist and chest. Right: thermovoltage of the FTE applied to heating plate, wrist, and chest. Reproduced with permission.^[976] Copyright 2020, Elsevier.

twofold increase in the Seebeck coefficient.^[967] Although the thermoelectric device proved flexible when mounted to a laboratory glass vial, the wearable demonstration was not carried out.^[967] Guo et al. later developed a highly flexible Au-decorated MoS₂-based thermoelectric energy harvester (Figure 6e).^[968] As a p-type dopant, Au nanoparticles can boost both the Seebeck coefficient and the electrical conductivity of MoS₂, resulting in a high power factor of 166.3 $\mu\text{W m}^{-1} \text{K}^{-1}$.^[968] Additional demonstrations demonstrated a 2.4 mV output voltage when the proposed thermoelectric device was attached to a human wrist with a temperature differential of 5 K across the device.^[968] Wang et al. demonstrated fullerene/TiS₂ nanosheet composites as highly flexible n-type thermoelectric materials.^[969] The resulting composite material had a ZT of 0.3 at 400 K and can be further processed via ink-jet printing to manufacture large-area, flexible thermoelectric devices.^[969]

Beyond 2D TMDCs, 2D semiconductors such as Te, Bi₂Te₃, and Sb₂Te₃ have long been considered to be excellent thermoelectric materials.^[298,858–860,970–972] However, these bulk materials' poor mechanical properties preclude their use in wearable thermoelectric energy harvesting.^[973,974] The issue could be resolved by thinning these chalcogen-based semiconductors to a few layers for use in flexible thermoelectric modules.^[942,973,974] Naghavi et al. calculated the thermoelectric characteristics of monolayer Pd₂Se₃, revealing ultralow lattice thermal conductivities of 1.5 and 2.9 $\text{W m}^{-1} \text{K}^{-1}$, respectively, along the *a*- and *b*-axes.^[951] The presence of Pd²⁺ cations in a square-planar crystal field results in the “pudding-mold” band structure, which contributes to Pd₂Se₃'s high power factor.^[951] Jin et al. later used sputter deposition to manufacture a carbon nanotube-supported Bi₂Te₃ thermoelectric device with a power factor of 1600 $\text{W m}^{-1} \text{K}^{-1}$ at ambient temperature.^[972] The resulting composite had a thermal conductivity of $0.53 \pm 0.05 \text{ W m}^{-1} \text{K}^{-1}$ and a ZT value of 0.9 over the ambient temperature to 373 K temperature range.^[972] Fu et al. synthesized the ternary bismuth antimony telluride (BST) alloy over sapphire.^[973] The epilayer was then transferred to curved glass and flexible polyethylene

terephthalate (PETE) substrates, demonstrating the deposited BST alloy's excellent elasticity and stretchability.^[973] The BST alloy's average ZT value was found to be 1.2, owing to its low thermal conductivity of 0.29–0.38 $\text{W m}^{-1} \text{K}^{-1}$.^[973] Shang et al. also developed Bi_{0.5}Sb_{1.5}Te₃ films as thermoelectric energy harvesters with high mechanical durability and flexibility.^[975] At a temperature differential of 40 K, a power density of 897.8 $\mu\text{W cm}^{-2}$ can be achieved.^[975] Besides pure 2D semiconductors, thermoelectric devices have been made using composites containing various materials. Wang et al. described the fabrication of Bi_{0.5}Sb_{1.5}Te₃/PEDOT:PSS composites containing BST enclosed within the CuTe layer (Figure 6f).^[976] At room temperature, a peak power factor of 312 $\text{W m}^{-1} \text{K}^{-1}$ and a thermovoltage of 7.7 mV may be achieved when the Bi_{0.5}Sb_{1.5}Te₃/PEDOT:PSS composites-based wearable thermoelectric nanogenerator is placed on the human wrist.^[976] Shang et al. reported on flexible Ag-modified BST films.^[977] The flexible thermoelectric module based on Ag_{0.005}Bi_{0.5}Sb_{1.5}Te₃ can generate an output voltage of 31.2 mV with a temperature differential of 60 K, owing to the increased Seebeck coefficient and electrical conductivity caused by structural defects.^[977] Table 3 summarizes the use of 2D materials in wearable thermoelectric energy harvesting.

4.3. 2D Materials-Based Pyroelectric Energy Harvesting

4.3.1. Working Mechanism of Pyroelectric Energy Harvesting

Pyroelectric conversion is another frequently used technique for converting heat energy to electrical energy. Unlike thermoelectric energy harvesting, which requires a temperature gradient (dT/dx) between the thermoelectric materials, pyroelectric energy harvesting requires a temperature change (dT/dt).^[862,863] This makes pyroelectric more favorable than thermoelectric for small-scale thermal energy harvesting, as establishing a spatial temperature gradient at the micro/nanoscale may be challenging.^[862,982] A pyroelectric material's structure must

Table 3. Summary of recent progress in 2D materials-based wearable thermoelectric energy harvesting.

Materials	Output voltage [V]	Temperature difference [K]	PF [$\mu\text{W m}^{-1} \text{K}^{-2}$]	Refs.
TiS ₂	–	–	450	[963]
Graphene/PEDOT:PSS	–	–	57.9	[953]
TiS ₂	–	–	904	[955]
Au-MoS ₂	2.4	5	166.3	[968]
MoS ₂ -graphene	0.24	16	–	[967]
Bi ₂ Te ₃	–	–	1600	[972]
Graphene	2.2	14	673	[954]
SWCNTs/Sb ₂ Te ₃ (p-type), rGO/Bi ₂ Te ₃ (n-type)	135	70	55 (p-type), 108 (n-type)	[978]
rGO/Bi ₂ Te ₃	14	20	460	[979]
Bi ₂ Te ₃	–	–	350	[980]
Ag ₂ Se	4.3	6.7	1882	[974]
Bi _{0.5} Sb _{1.5} Te ₃	–	–	23.2	[975]
Ag-Bi _{0.5} Sb _{1.5} Te ₃	31.2	60	12.4	[977]
Bi _{0.5} Sb _{1.5} Te ₃ /PEDOT:PSS	7.7	15	312	[976]
SWCNTs/Bi ₂ Te ₃	–	–	1.38	[981]

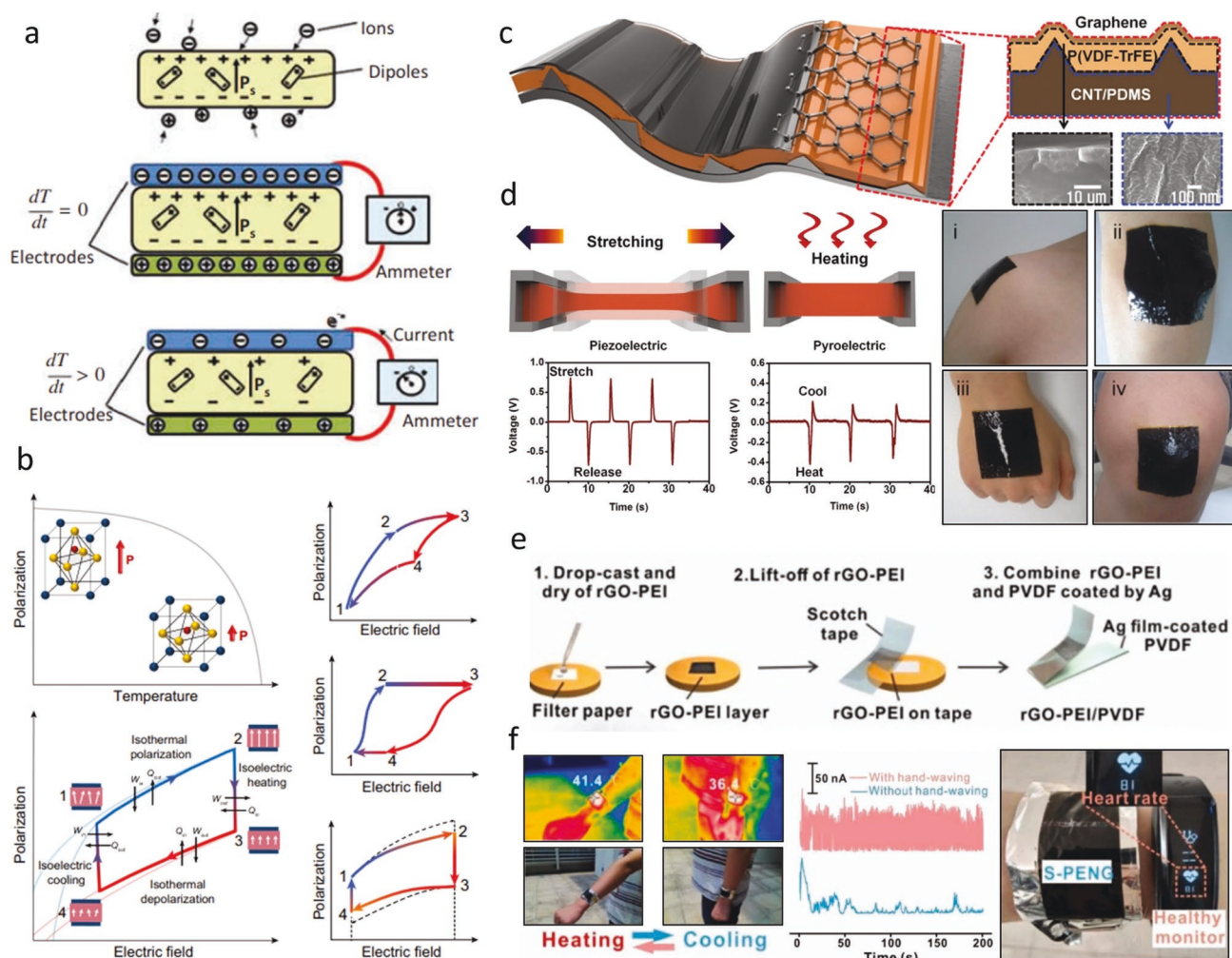


Figure 7. Working mechanism, performance, and demonstration of 2D materials-based wearable pyroelectric energy harvesters. a) Schematic illustration of working mechanism of pyroelectric nanogenerator. Reproduced under terms of the CC-BY license.^[982] Copyright 2013, The Authors, published by Taylor & Francis. b) Schematically showing pyroelectric effect as a change of polarization with temperature (top left). Pathway of pyroelectric effect-induced polarization as a function of the electric field in Ericsson (Olsen, bottom left), Carnot (top right), Stirling (middle right) and Brayton (bottom right) cycle. Reproduced under terms of the CC-BY license.^[987] Copyright 2019, The Authors, published by Springer Nature. c) Schematic illustration and SEM images of the graphene-based hybrid piezoelectric-pyroelectric nanogenerator. d) Piezoelectric output (left), pyroelectric output (middle) of the graphene-based hybrid piezoelectric-pyroelectric nanogenerator and photographs of applying the device to various locations on the human body (right). Reproduced with permission.^[993] Copyright 2013, Wiley-VCH. e) Schematically showing the fabrication process of rGO-PEI pyroelectric nanogenerator. f) Left: infrared images of the rGO-PEI pyroelectric nanogenerator under outdoor conditions when directly exposed to the sun. Middle: short-circuit current of the rGO-PEI pyroelectric nanogenerator with (top) and without (bottom) hand-waving motion. Right: photograph showing the human fitness tracker powered by the rGO-PEI pyroelectric nanogenerator. Reproduced with permission.^[994] Copyright 2020, Elsevier.

be noncentrosymmetric to generate spontaneous polarization (P_s) without an external electric field.^[983,984] When pyroelectric materials are subjected to a temperature change, the change in spontaneous polarization results in the generation of an electrical current (Figure 7a).^[983,25] The following formula denotes the electrical current created by the pyroelectric effect:

$$i_p = pA \frac{dT}{dt} \quad (1)$$

where i_p , p , and A represents the generated current, pyroelectric coefficient, and surface area of the materials.^[983,25,985,986] The pyroelectric coefficient is used to determine a material's

pyroelectric performance under constant stress and electric field, and is defined as follows (Figure 7b):^[985,986]

$$p = \left(\frac{dP_s}{dT} \right)_{\sigma, E} \quad (2)$$

Generally, pyroelectric energy harvesters can be seen as thermodynamic heat engines using an external electric field as pressure and polarization as the volume of the working fluid.^[987] When different thermodynamic cycles are applied to pyroelectric nanogenerators, Figure 7b depicts the relevant polarization-electric field pathway.^[987] Numerous 2D materials were anticipated to be pyroelectric, as are all ferroelectric and

piezoelectric materials.^[870] Additionally, the atomically thin thickness of 2D materials is considered to improve pyroelectric performance by allowing for rapid temperature changes.^[988]

4.3.2. Performance of 2D Materials-Based Pyroelectric Nanogenerator

Several highly adaptable and flexible pyroelectric energy harvesters have been reported for application in wearable waste heat energy harvesting.^[864,989–993] For instance, Lee et al. demonstrated a highly stretchable nanogenerator based on both piezoelectric and pyroelectric effects by combining a polydimethylsiloxane-carbon nanotube (PDMS-CNT) composite with graphene nanosheets (Figure 7c,d).^[993] The graphene nanosheet was chosen as the top electrode due to its high thermal conductivity, enabling quick temperature changes throughout the device.^[993] By combining the pyroelectric and piezoelectric effects in the reported hybrid nanogenerator, a synergetic combined output voltage of 1.4 V may be generated, with voltages from compress-release and temperature change measuring 1.0 and 0.4 V, respectively.^[993] Li et al. reported on a wearable pyroelectric nanogenerator based on rGO and PVDF film (Figure 7e,f).^[994] By combining the solar-thermal harvesting layer made of polyethyleneimine modified rGO (PEI-rGO) and the PVDF pyroelectric layer, the pyroelectric nanogenerator's output power can be increased from less than 5 to 21.3 mW m⁻².^[994]

Not only can a pyroelectric nanogenerator harvest heat from solar radiation but it may also be utilized to measure the level of infrared radiation.^[869] To detect mid-infrared irradiation, Sassi et al. combined single-layer graphene with pyroelectric LiNbO₃.^[869] The graphene-based photodetector, which has a greater temperature coefficient of resistance (TCR) of 4–11% K⁻¹, can also be utilized as a bolometer with a TCR of up to 900% K⁻¹ and a temperature resolution of 15 μK at room temperature.^[869] Chang et al. recently presented pyroelectric photodetectors based on SnS nanoflakes/CdS nanorods heterojunctions.^[995] When lit at 650 nm at 130 K, the pyroelectric current was three times that of the photocurrent, showing the significant potential of pyroelectric materials in cryogenic photodetector applications.^[995]

4.4. 2D Materials-Based Piezoelectric Energy Harvesting

4.4.1. Working Mechanism of Piezoelectric Energy Harvesting

Harvesting kinetic energy from human motions is a critical method for powering wearable devices, as it is less reliant on the environment and is more user-friendly.^[1,50,996] Piezoelectric nanogenerators (PENG) are a frequently reported approach for collecting mechanical energy from body motions. They utilize insulating materials with a piezoelectric effect to convert mechanical energy to electricity.^[872,997] Piezoelectricity is generated in two steps: 1) mechanical deformation along the polarization direction of the piezoelectric material breaks electrical neutral by separating positive and negative charge centers; and 2) an electrostatic potential is built up in the material due to the polarized charges, resulting in the balancing electron flow

through the external circuit (Figure 8a).^[23,872,873] The second rank strain tensor, $S_{ij} = d_{ijk}E_k$, can be used to determine a material's piezoelectric performance, where d_{ijk} and E_k denote the piezoelectric charge coefficient and electric field, respectively.^[819,872] Wang's group developed the first piezoelectric nanogenerator in 2006, utilizing zinc oxide (ZnO) nanowire arrays.^[997] Duerloo et al. calculated theoretically that 2D TMDCs and h-BN displayed a high piezoelectric effect compared to their bulk counterparts.^[876] While the antiparallel stacking of TMDCs and h-BN monolayers results in the bulk materials' centrosymmetry, monolayer TMDCs and h-BN lack an inversion center, resulting in a non-zero piezoelectric tensor.^[876] By further connecting the piezoelectric effect with the electrochemical processes of the materials, it was shown that the piezo-electrochemical effect is an ideal method for harvesting low-frequency mechanical energy (5 Hz).^[822,998,999] A change in the electrical potential of an ion-intercalating material due to the piezoelectric effect results in the insertion/desorption of intercalants and the resulting balancing electrical current.^[1000–1002] Thus, energy harvesting can be accomplished by stabilizing the electrical potential of ion-intercalated materials when mechanical force is applied.^[1001,1003]

4.4.2. Performance of 2D Materials-Based Piezoelectric Nanogenerator

In 2014, Wu et al. reported the first measurement of the piezoelectricity of monolayer MoS₂.^[1004] Under a 0.53 percent strain along the armchair direction, the exfoliated monolayer MoS₂ flakes could deliver a peak output of 15 mV and 20 pA, corresponding to a power density of 2 mW m⁻² and a conversion efficiency of 5.08%.^[1004] More notably, a smaller output signal was generated when the strain was applied in a zigzag direction. As expected by the simulation, no signal was detected in bilayer MoS₂ flakes, demonstrating the anisotropic piezoelectric effect and development of centrosymmetry in even-number layered MoS₂.^[876,1004,1005] Qi et al. later demonstrated the piezoelectric effect of CVD produced triangle monolayer MoS₂ and showed the in-plane charge polarization of MoS₂ caused by local strain.^[1006] Zhu et al. also confirmed the piezoelectric effect of free-standing MoS₂.^[1007] Kim et al. used PET substrates to create a flexible MoS₂-based PENG (Figure 8b).^[1008] Due to the anisotropic piezoelectric action of MoS₂, it is possible to produce a twofold increase in the peak voltage and current output of the flexible PENG when strain is applied in the armchair direction rather than the zigzag direction.^[1008] Han et al. developed a MoS₂-based 3D structure by combining MoS₂ monolayers with vertically grown hollow MoS₂ nanoflakes (ν-MoS₂ NF) in order to increase the output of flexible PENG (Figure 8c).^[1009] Apart from the junction strain and the strain created in the MoS₂ basal plane, the contact strain between the ν-MoS₂ NFs increases the output voltage of the PENG during the bending process.^[1009] Additionally, when the proposed PENG was attached to commercially viable shoes, it demonstrated a maximum output voltage of 18 V and a matching power density of 1.62 mW m⁻² when walking.^[1009] Another study by the same group used a polystyrene (PS) nanosphere as a template to build a 2D MoS₂ hollow sphere with multiple

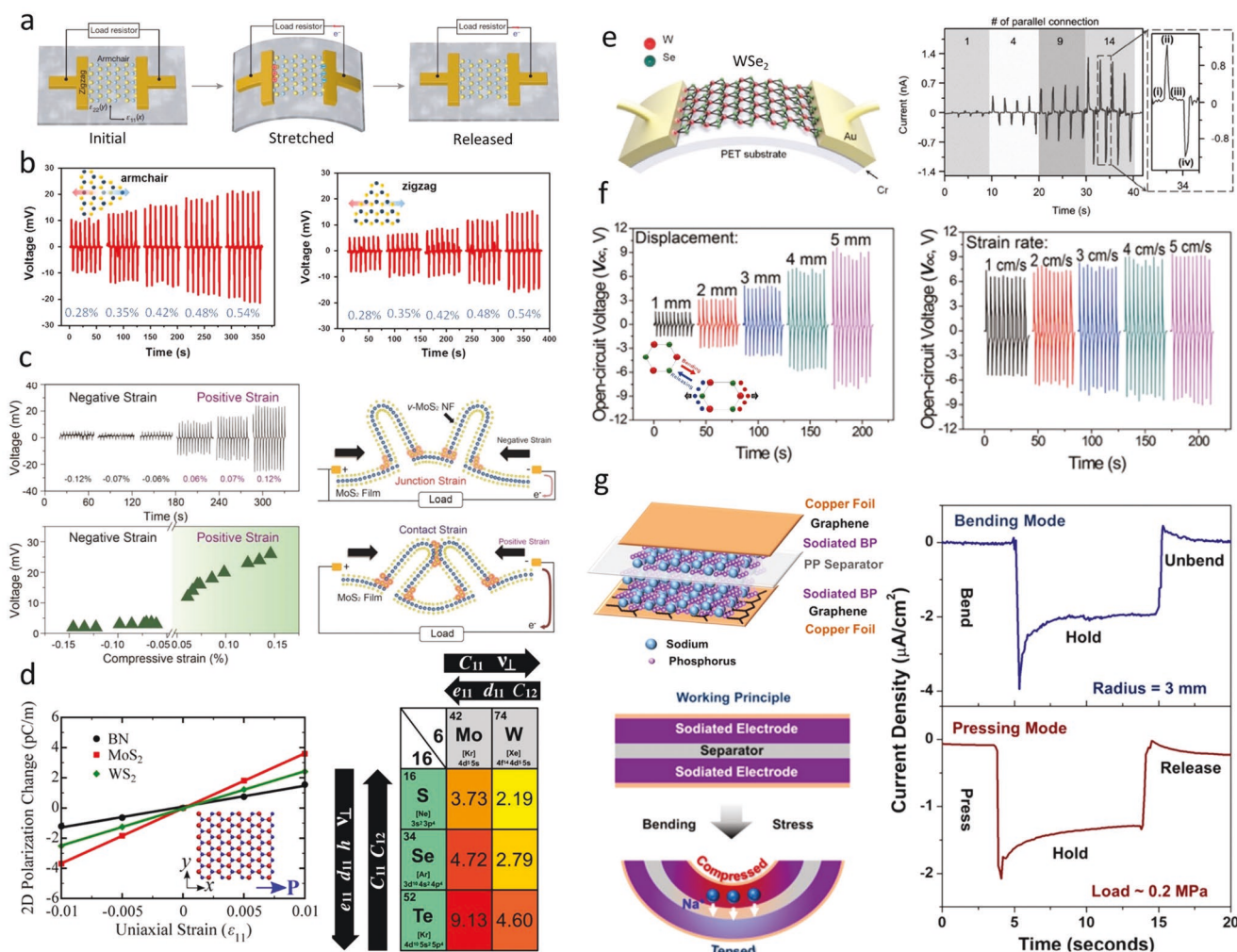


Figure 8. Working mechanism, performance, and demonstration of 2D materials-based wearable piezoelectric energy harvesters. a) Working mechanism of flexible monolayer MoS₂-based piezoelectric device. Reproduced with permission.^[1004] Copyright 2014, Springer Nature. b) Output voltage of monolayer MoS₂ PENG polarized along the armchair (left) and zigzag (right) direction. Reproduced with permission.^[1008] Copyright 2016, Elsevier. c) Left: output voltage of vertically grown hollow MoS₂ nanoflakes (ν -MoS₂ NFs)-based under different positive and negative strain. Right: schematic illustration of voltage generation mechanism when a negative (top) or positive (bottom) strain is applied. Reproduced with permission.^[1009] Copyright 2019, IOP Publishing. d) Left: polarization of monolayer BN, MoS₂, and WS₂ under applied uniaxial strain along the x-direction. Right: trend in structural, elastic and piezoelectric properties of various 2H-MX₂. The number listed are relaxed-ion d_{11} coefficient. Reproduced with permission.^[876] Copyright 2012, American Chemical Society. e) Left: schematic illustration of monolayer WSe₂-based PENG. Right: Output current of monolayer WSe₂-based PENG as a function of the number of devices connected in parallel. Reproduced with permission.^[1011] Copyright 2017, Wiley-VCH. f) Open-circuit voltage of flexible 2D h-BN-based PENG as a function of displacement (left) and strain rate (right). Inset schematically showing the in-plane polarization mechanism of 2D h-BN. Reproduced with permission.^[878] Copyright 2019, American Chemical Society. g) Left: schematically showing the configuration of BP-based piezo-electrochemical nanogenerator (top) and the working principle of the piezo-electrochemical nanogenerator (bottom). Right: short-circuit current response of BP-based piezo-electrochemical nanogenerator during bending mode (top) and pressing mode (bottom). Reproduced with permission.^[822] Copyright 2017, American Chemical Society.

layers of wall thickness.^[1010] The arrays of MoS₂ spheres were used as the piezoelectric layer of the flexible PENG, with the size of the MoS₂ sphere having a direct effect on the PENG's piezoelectric output.^[1010] As predicted by simulation, the increased piezoelectric output could result from the larger structural changes in the larger MoS₂ sphere when a force is applied.^[1010] The resulting PENG can produce a peak output voltage of 1.2 V at 4.2 kPa applied pressure.^[1010]

Additionally, other 2D TMDCs other than MoS₂ have been studied for flexible PENG (Figure 8d).^[873] Lee et al.

demonstrated the piezoelectric effect of Bilayer WSe₂ utilizing turbostratic stacking (Figure 8e).^[1011] In contrast to conventional exfoliated bilayer TMDCs, which retain an inversion center, the turbostratic-stacked bilayer WSe₂ was not centrosymmetric and displayed piezoelectricity.^[1011] The WSe₂ bilayer can withstand strains of up to 0.95% while maintaining an output voltage of 85 mV.^[1011] By introducing surface corrugation, Kang et al. examined the out-of-plane piezoelectricity of MoTe₂.^[1012] The degree of surface corrugation may be controlled by altering the substrate and MoTe₂ film thickness, and the few-layer MoTe₂ film

Table 4. Summary of recent progress in 2D materials-based wearable piezoelectric energy harvesting.

Materials	External force	V_{OC} [V]	I_{SC} [$\mu A\ cm^{-2}$]	Power output	Load resistance	Refs.
PVDF-BTO/n-Gr/PVDF-BTO	2 N	10	0.31	5.8 μW	1 M Ω	[1018]
SnO ₂ /PVDF	0.3 MPa	42	6.25	–	30 M Ω	[1017]
WS ₂	13 kg	8.7	–	–	–	[1019]
TOCN/MoS ₂	–	4.1	0.044	–	–	[1020]
MoS ₂	0.36% strain	0.03	–	62.72 pW	8 M Ω	[1021]
MoSe ₂	0.60% strain	0.06	–	7.5 pW	5 M Ω	[1014]
WSe ₂	0.39% strain	0.05	–	–	1 G Ω	[1011]
v-MoS ₂ NFs	0.12% strain	18	–	0.06 mW m ⁻²	400 M Ω	[1009]
SnS	0.70% strain	0.15	0.002	–	1 G Ω	[1016]
g-C ₃ N ₄	–	1.67	0.141	–	–	[1015]

can generate out-of-plane piezoelectricity.^[1012] Another way to induce high out-of-plane piezoelectricity using TMDCs is to add defects into the 2D TMDCs film, further disrupting the crystal's symmetry.^[1013] Kang et al. demonstrated that out-of-plane piezoelectricity may be achieved without changing the degree of surface corrugation by thermally annealing the 2H-MoTe₂ film and infusing it with Te vacancies.^[1013] Li and Zhang used flexible monolayer MoSe₂-based PENG to power a chemical sensor and a photodetector.^[1014] The MoSe₂-based PENG was shown to capture muscle vibrations from speech and finger movement, demonstrating excellent mechanical-to-electrical power conversion.^[1014] Lee et al. demonstrated the use of h-BN as the piezoelectric layer in a flexible PENG (Figure 8f).^[878] Exfoliated h-BN flakes with a lateral size of 0.82 μm and a thickness of 25 nm are capable of delivering a 50 mV open-circuit voltage and a 30 pA short-circuits current.^[878] Further construction of h-BN-based PENG allows for a voltage of 9 V and a current of 200 nA.^[878] Other 2D materials such as graphene, SnS, SnO₂, and g-C₃N₄ have previously been anticipated to be promising candidates for PENG. However, only a few actual demonstrations have been made utilizing these materials.^[1015–1018] The use of 2D materials for wearable PENG was listed in **Table 4**.

Muralidharan et al. presented an electrochemical-mechanical strain energy harvester using sodiated BP as the active layer (Figure 8g).^[822] During the bending process, the sodium ions diffuse from the compressed to the tensed side of the device, owing to the device's differing mechanochemical response to compressive or tensile stress. This results in the formation of a potential across the separator.^[822] Because this electrochemical-mechanical strain energy harvester has a response time many orders of magnitude faster than traditional PENG or TENG, it is regarded to be a promising option for ultralow frequency mechanical energy harvesting.^[822]

4.5. 2D Materials-Based Triboelectric Energy Harvesting

4.5.1. Working Mechanism Triboelectric Energy Harvesting

Another prominent approach for converting mechanical energy from human motions to electrical current is the triboelectric nanogenerator (TENG), which collects energy using

a combination of triboelectrification and electrostatic induction.^[21,881] The operating principles of generally reported TENG could be categorized into four categories: contact-separation (CS), relative sliding (RS), single electrode (SE), and free-standing (FS). Each operating mode has distinct advantages while also having distinct disadvantages.^[1022] The CS mode employs two different insulating materials as a triboelectric pair for electrification and induction.^[1023,1024] When the two insulating materials come into contact, charges transfer and collect on the surface of the triboelectric pair, creating a potential across the device and causing an external circuit to balance the flow of electrons (**Figure 9a**).^[21] While the CS mode TENG has a simple design and produces a high voltage output, the output is not continuous due to the contact separation process.^[1022,1025,1026] Triboelectrification in RS mode TENG was accomplished by horizontally moving the triboelectric pair.^[1027,1028] Charge separation was triggered during the process by a change in the contact area between two insulating materials, resulting in a continuous output.^[1027,1028] However, repetitive sliding of the triboelectric pair in RS mode TENG offers a problem for the materials' surface nanostructures since the sliding friction may damage the materials' surface nanostructure.^[1022] FS mode TENG incorporated the same concept as RS mode TENG except that the electrodes of the TENG are stationary, and the triboelectric pairs do not come into direct contact.^[1029,1030] The potential effectiveness of FS mode TENG can reach 100% while avoiding damage to the nanostructure on the surface.^[1022] However, the geometry of FS mode TENG is somewhat intricate, making integration for further applications challenging.^[1022] SE mode TENG can be considered a simplified version of CS mode TENG by substituting human skin or other items for triboelectric layers.^[1031–1033] While the SE mode TENG structure is reasonably straightforward, the output is not comparable to other TENG types.^[1022]

Fan et al. showed TENG for the first time in 2012 by bringing two different polymers into contact (Figure 9a).^[21] Kapton-PET was found to be the optimum combination, delivering a peak voltage of 3.3 V and a volumetric power density of 10.4 mW cm⁻³.^[21] The strength of the charges accumulated on triboelectric materials during contact separation can be quantified using a triboelectric series, which quantitatively ranks the material's inclination to gain or lose electrons.^[897,1034,1035] It

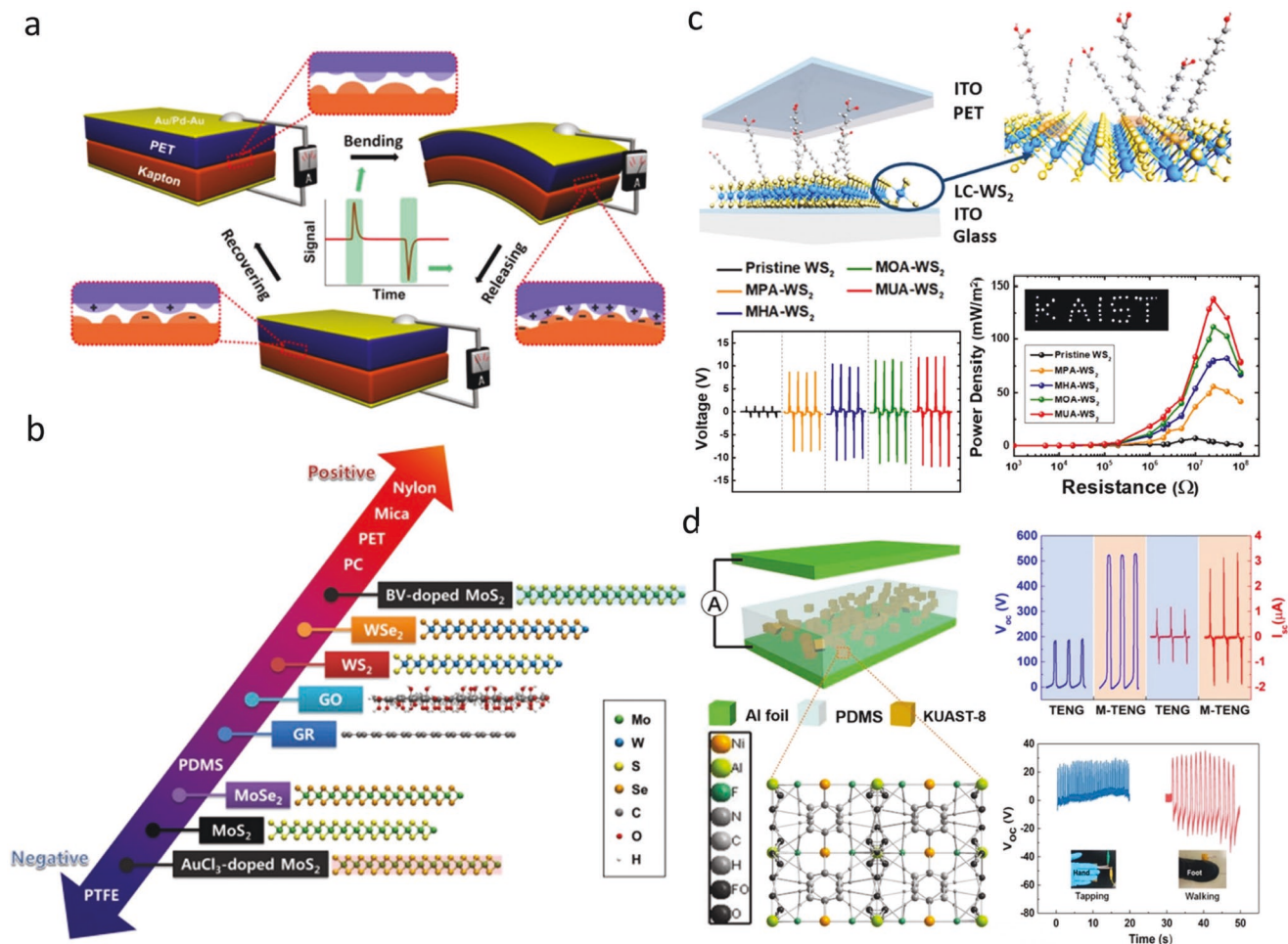


Figure 9. Working mechanism, performance, and demonstration of 2D materials-based wearable triboelectric energy harvesters. a) Schematic illustration of the structure of the first TENG. Reproduced with permission.^[21] Copyright 2012, Elsevier. b) Triboelectric series of commonly reported 2D materials, with the corresponding molecular structure illustrated to the next. Reproduced with permission.^[897] Copyright 2018, Wiley-VCH. c) Top: schematic illustration of the configuration of TENG based on thiolated ligand-conjugated WS₂. Bottom: Open circuit voltage (left) and power density (right) of WS₂-based TENG conjugated with different ligands. Reproduced with permission.^[1058] Copyright 2021, American Chemical Society. d) Left: schematic illustration of the structure of PDMS@F-MOF-based TENG (M-TENG) and the crystal structure of F-MOF. Upper right: open circuit voltage and short circuit current of PDMS-based TENG (TENG) M-TENG. Bottom right: open circuit voltage output of M-TENG using hand tapping and during walking. Reproduced with permission.^[1071] Copyright 2020, Elsevier.

should be emphasized that all materials exhibited a triboelectric effect regardless of their performance.^[897] Numerous studies have been conducted to establish triboelectric series for various materials. Diaz et al. presented a semi-quantitative triboelectric series for various regularly reported polymers, whereas Zou et al. quantified the triboelectric series using liquid metal as a reference.^[1034,1035] Seol et al. investigated 2D materials, such as graphene and 2D TMDCs, and compared their triboelectric properties to those of other commonly used polymers, such as polytetrafluoroethylene (PTFE), polydimethylsiloxane (PDMS), polycarbonate (PC), PET, mica, and nylon (Figure 9b).^[897] The 2D materials' effective work function was critical for their triboelectric order: a higher work function results in a more triboelectric negative material.^[897] Additionally, the charge transfer characteristic is not dependent on the production method and can be changed chemically.^[897]

4.5.2. Performance of 2D Materials-Based Triboelectric Nanogenerator

Due to graphene's capacity to hold surface charges for a lengthy period, it has been projected theoretically to be a good candidate for TENG.^[883,1036–1039] Due to the ripples and wrinkles on the surface of graphene nanosheets created during the wet transfer process, graphene is a common triboelectric material for flexible TENG.^[883,1036,1039–1044] Kim et al. demonstrated a TENG based on graphene and PET as triboelectric materials with outstanding transparency and flexibility.^[883] The resulting TENG had an output voltage and current density of 5 V and 500 nA cm⁻² using monolayer graphene, which could be increased to 9 V and 1.2 A cm⁻² by stacking monolayer graphene nanosheets regularly.^[883] Chandrashekar et al. presented a roll-to-roll graphene transfer method for fabricating flexible and transparent TENGs.^[1045] After transferring CVD-produced

graphene on an ethylene-vinyl acetate (EVA)/PET substrate, transparency of 97.5% may be attained.^[1045] The TENG can deliver a maximum output of 22 V and 0.9 A under a 4.3 Hz vibration using micropatterned PDMS as the counter and dielectric layers.^[1045] Xia et al. created a flexible and stretchable triboelectric material using aligned graphene sheets embedded in a PDMS matrix (AGS@PDMS).^[1046] Individual parallel-plate capacitors were inserted in the PDMS matrix, allowing the TENG to tolerate a greater breakdown voltage while minimizing dielectric loss.^[1046] Thus, the output power of the AGS@PDMS-based TENG was increased by threefold when compared to pure PDMS.^[1046] Shin et al. developed a water-based assembly procedure and an isopropanol-assisted direct transfer (IDT) method for graphene to the substrate.^[1047] Compared to conventional transfer processes, the authors claimed that the suggested IDT approach does not require additives for graphene deposition onto substrates, which is advantageous for substrates that cannot tolerate high-temperature annealing for polymer removal.^[1047] The TENG as-fabricated can generate an open-circuit voltage of 1.08 V and a short-circuit current of 0.25 A.^[1047] Chen et al. examined the surface nanostructure and roughness of crumpled graphene to fabricate a stretchable TENG.^[1048] By raising the degree of roughness in crumpled graphene by 300%, an output voltage of 83.0 V, a current of 25.78 A, and a power density of 0.25 mW cm⁻² can be reached, which is 20 times that of planar graphene-based TENG.^[1048] Chen et al. later demonstrated that the performance of TENG may be enhanced further by doping AuCl₃ into crumpled graphene.^[1049] A doping concentration of 1.2 mg mL⁻¹ AuCl₃ results in a maximal TENG output of 80.6 V and 11.9 A cm⁻².^[1049] AuCl₃ nanoparticles improved graphene's surface roughness and conductivity, both of which are favorable for charge transfer.^[1049] Pace et al. recently doped nitrogen into graphene and found comparable results.^[1050] The improved performance of TENG based on nitrogen-doped graphene has been attributed to the difference in electron affinity between the N-graphitic and N-pyridinic sites.^[1050] Additionally, graphene electrodes with high conductivity and elasticity have replaced metal electrodes with low stretchability in flexible TENG.^[1051,1052]

2D TMDCs have demonstrated significant potential as triboelectric active materials.^[1053,1054] Wu et al. produced a proof-of-concept MoS₂-based contact-separation TENG by suspending MoS₂ in polyamic acid (MoS₂-PAA).^[1054] MoS₂ was utilized in this work as electron acceptors because it effectively traps electrons and prevents them from recombining with surface-adsorbed positive molecules, hence increasing the performance of TENG by 120 times when compared to TENG without MoS₂.^[1054,1055] Kim et al. built the flexible MoS₂-based TENG by directly employing MoS₂ as the depletion layer.^[1056] Triboelectric-active materials that produced Schottky contacts and p-n junctions with MoS₂ performed better than those that formed Ohmic contacts with the material.^[1056] This boost was attributed to the diffusion of charged carriers over the Schottky contact, forming a depletion layer, and the rectifying behavior caused by the Schottky contact and p-n junction creation.^[1056] Wang et al. confirmed the WS₂'s triboelectric property using Kelvin force microscopy (KFM).^[1057] WS₂-based TENG was recently fabricated by Kim et al. with surface modification of thiolated ligands to enhance the TENG performance (Figure 9c).^[1058]

Thiolated ligands acted as a p-type dopant for WS₂, increasing the potential difference between the two triboelectric electrodes, resulting in a 10-fold increase in TENG performance compared to TENG using pure WS₂.^[1058]

Additionally, other 2D materials such as BP, MXene, and g-C₃N₄ can be employed as triboelectric active layers.^[1059–1065] Dong et al. demonstrated in 2018 that Ti₃C₂T_x is more triboelectrically negative than PTFE.^[886] Unlike PTFE, which has a low electrical conductivity, metallic Ti₃C₂T_x has high electrical conductivity, allowing for the usage of a two-electrode configuration while maintaining a high open-circuit voltage (V_{OC}).^[886] The resulting TENG demonstrated excellent flexibility when linked to human fingers and harvested energy from basic muscle motions.^[886] Jiang et al. produced a TENG based on MXene via the electrospinning technique.^[1066] MXene nanosheets were disseminated in polyvinyl alcohol (PVA) solution and electrospun onto an Al foil for flexible TENG. The constructed TENG has a peak output of 10876 mW m⁻² and has been proven to power several tens of LED lamps simultaneously.^[1066] Bhatta et al. also produced flexible TENG with a power density of 11213 mW m⁻² using MXene-contained PVDF nanofibers using an electrospinning process.^[1067] He et al. demonstrated that a single-electrode TENG with an active layer of MXene/PDMS might function as both an energy harvester and a biomechanical motion sensor by analyzing the TENG's output peaks to determine the position or motion of a human.^[1068] Cao et al. developed a stretchable single-electrode TENG using a cellulose nanofiber (CNF)/MXene liquid electrode in a silicone rubber mold.^[1069] MXene's superior dispersion in water and its surface functional groups greatly contribute to the enhanced TENG performance by forming hydrogen bonds with water molecules, which boosts the charge transfer process.^[1069] TENG is a stretchable and shape-adaptive energy harvester that can power up to 230 LED bulbs by palm tapping.^[1069] Xiong et al. developed a textile-based TENG including BP nanosheets that exhibits good durability when in contact with the skin.^[1070] To prevent BP degradation, the electron acceptor was coated with BP nanosheets and hydrophobic cellulose oleoyl ester nanoparticles (HCOENPs). When the BP-contained single-electrode TENG is applied to human skin and clothing, it harvests energy from voluntary and involuntary muscle activity to provide a maximum instantaneous output of 880 V and 1.1 A cm⁻².^[1070] He et al. investigated solution-grown Te nanoflakes as a triboelectric active layer for flexible TENG and shown that Te-based TENG can harvest energy successfully and at a rate equivalent to that of other 2D materials.^[1060] Additionally, Bayan et al. described the utilization of g-C₃N₄ in a flexible TENG. It should be mentioned that the device presented by Bayan et al. can also harvest energy from UV radiation due to the photoabsorption capability of g-C₃N₄.^[1059] Guo et al. improved the output performance of TENG by including a fluorinated MOF (F-MOF) into a PDMS matrix (Figure 9d).^[1071] By adding 0.5 weight percent F-MOF to the PDMS matrix as charge trapping sites, the number of induced charges can be increased, resulting in an 11-fold increase in the power density output of the PDMS-based TENG. With increasing research interest in 2D materials other than graphene and 2D MoS₂, the use of 2D materials as triboelectric active layers is projected to surge. Recent progress of wearable 2D materials-based TENG was summarized in **Table 5**.

Table 5. Summary of recent progress in 2D materials-based wearable triboelectric energy harvesting.

Triboelectric pairs	Applied force	V_{OC} [V]	I_{SC}	Power density	Load resistance	Refs.
Graphene-PET	1 kgf	5	$0.5 \mu A cm^{-2}$	$2.5 \mu W cm^{-2}$	10 M Ω	[883]
Graphene/PDMS	–	1.08	$0.25 \mu A$	–	–	[1047]
Crumpled graphene/PDMS	–	83.0	$25.78 \mu A$	$0.25 mW cm^{-2}$	2 M Ω	[1048]
Graphene/EVA/PET-PDMS	–	22	$0.9 \mu A$	–	–	[1045]
h-BN/ Al_2O_3 -graphene	1 kgf	1.2	$150 nA cm^{-2}$	–	–	[1042]
LIG/PI-Al	2 N	3500	$60 \mu A$	$2.4 W m^{-2}$	70 M Ω	[1072]
AuCl ₃ doped CG-PTFE	30 N	21.3	$2.1 \mu A$	$32 \mu W cm^{-2}$	10 M Ω	[1049]
HCOENPs/BP/PET-PET	5 N	880	$1.1 \mu A cm^{-2}$	$520 \mu W cm^{-2}$	100 M Ω	[1070]
MXene/ITO-PET	15 N	500	–	$0.5 mW$	–	[886]
PVA/MXene-silk	10 N	118.4	–	$1.09 W m^{-2}$	5 M Ω	[1066]
CNFs/MXene-skin	–	300	$5.5 \mu A$	$0.50 W m^{-2}$	50 M Ω	[1069]
MXene/PDMS-skin	12 N	225	$30 \mu A cm^{-2}$	$10 mW cm^{-2}$	100 M Ω	[1068]
PVDF/MXene-Nylon 6/6	–	724	$163.6 \mu A$	$11.2 W m^{-2}$	2 M Ω	[1067]
MXene/PVA hydrogel-Kapton	–	230	$270 nA$	$0.33 W m^{-2}$	1 G Ω	[1064]
PI/MoS ₂ :PI/PI-PET	–	120	$180 mA m^{-2}$	$25.7 W m^{-2}$	5 M Ω	[1054]
MoS ₂ /PVDF-Ppy/PET	5 kPa	36	$2.8 \mu A cm^{-2}$	$445 \mu W cm^{-2}$	10 M Ω	[1056]
LC-WS ₂ -PET	6 N	12.2	–	$138 mW m^{-2}$	10–25 M Ω	[1058]
ZnSO NFs/PDMS-PET	8 N	156	$2.98 mW cm^{-2}$	$2.98 mW cm^{-2}$	40 M Ω	[1073]

4.6. 2D Materials-Based Wearable Energy Harvesting from Other Sources

Apart from the generally reported energy harvesting from solar, waste heat, and mechanical energy, additional forms of energy harvesting, such as osmotic energy harvesting and biofuel cells, have been investigated, employing 2D materials as electrodes or active layers.^[1074–1078] In the reversible electro-dialysis (RED) process, osmotic energy harvesting harvests energy from the concentration difference between the two sides of the membrane.^[1079–1083] Due to the small spaces between stacked nanosheets, using 2D materials as the membrane could increase ion selectivity.^[1084] Recent work done by Ji et al. proposed a GO-based membrane pair for the RED system.^[1085] The surface charge polarity of the GO-based membrane can be changed from negative to positive for cation and anion selectivity by altering the GO surface using 1-aminopropyl-3-methylimidazolium bromide (APMIB). The GO-based membrane demonstrated excellent flexibility while providing a 54% increase in output density over a commercial ion-exchange membrane.^[1085] Ding et al. also harvested osmotic energy using a $Ti_3C_2T_x$ membrane with nanofluidic channels.^[1086] The nanochannels, which can be positively or negatively charged, transport ions with opposing charges through the membrane. The device effectively collects electrochemical energy via selective charge separation by integrating a pair of positively and negatively charged MXene membranes.^[1086] These flexible 2D material-based membrane demonstrations show tremendous potential for using a wearable osmotic energy harvester to harvest energy from physiological fluids such as sweat.

Recent years have seen a surge in interest in biofuel cells (BFCs) in biological energy harvesting. BFCs are frequently reported to harvest energy by catalytically converting glucose

and carbohydrates from the human body via cathodic or anodic reactions in the presence of biological enzymatic components.^[1074,1075,1,1087] Due to the superior mechanical qualities and unique electric properties of 2D materials, it is possible to construct a flexible biofuel cell that can be directly applied to the human body while maintaining a high output performance.^[1088] For example, Shen et al. demonstrated using a 2D graphene paper (2D-GP) supported flexible BFC to capture energy from glucose oxidation.^[1088] While retaining catalytic activity, the adsorbed enzyme may be successfully immobilized on the flexible 2D-GP bioelectrodes.^[1088] The resulting BFC may have a comparable power output to previous stiff carbon-based BFCs while demonstrating exceptional flexibility.^[1088] Additionally, Rewatkar et al. reported the development of a flexible graphene-based BFC capable of harvesting energy from glucose oxidation using a laser-induced graphene bioelectrode.^[1076,1089] With an adequate surface modification of glucose oxidase and laccase, the flexible graphene electrode may be further incorporated into a microfluidic device to deliver a $13 W cm^{-2}$ output power density. With their high degree of adaptability and high output performance, 2D materials-based BFCs are expected to be a growing field for wearable biochemical energy harvesting.^[1076]

4.7. Integration of Energy Harvester for Wearable and Human-Related Applications

Given the tremendous advancements and improvements made to the 2D materials-based energy harvester in output power, voltage, and conversion efficiency, 2D materials-based energy harvesters in wearable and human-related applications have also grown in popularity. Numerous studies have proved the viability of 2D materials-based energy harvesters

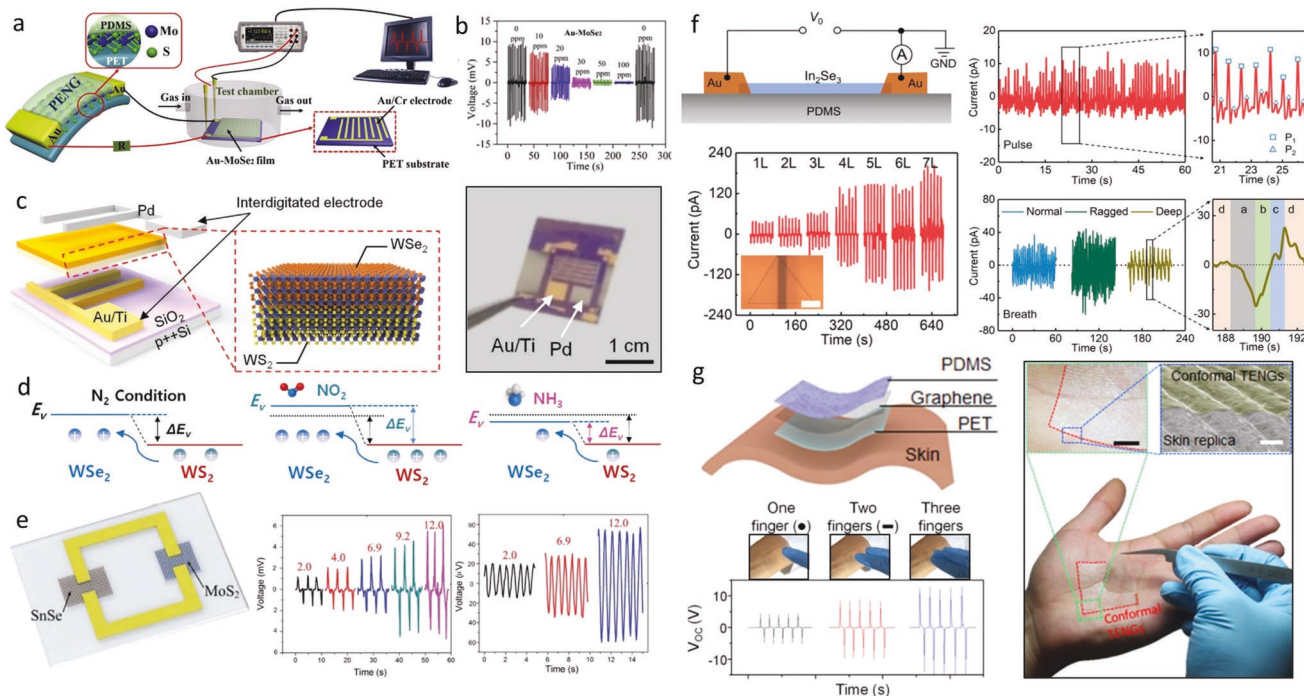


Figure 10. Integration of 2D materials-based wearable energy harvester for human-related applications. a) Schematic illustration of self-powered Au-MoS₂-based NH₃ gas sensor powered by a MoS₂-based PENG. b) Output voltage of the self-powered NH₃ gas sensor under different NH₃ atmospheres. Reproduced with permission.^[1021] Copyright 2019, American Chemical Society. c) Schematic device architecture (left) and optical image (right) of WSe₂/WS₂ gas sensor. d) Energy level diagram of charge transfer at WSe₂/WS₂ gas sensor when exposed to N₂ (left), NO₂ (middle), and NH₃ (right). Reproduced with permission.^[1104] Copyright 2020, Wiley-VCH. e) Schematic illustration of SnSe PENG integrated with a MoS₂-based sensor for the self-powered sensing unit (SPSU, left), pH sensing performance of SPSU powered by periodic bending (middle) and heartbeats (right). Reproduced with permission.^[1122] Copyright 2021, the Royal Society of Chemistry. f) Left: schematic illustration (top) self-powered α -In₂Se₃-based piezoelectric sensor and short circuit current of α -In₂Se₃ piezoelectric device with different layers of α -In₂Se₃. Right: real-time monitoring of pulse (top) and breath signal (bottom). Top right: zoomed-in signals shown in the black rectangle. Bottom right: output current changes throughout a deep breath (a: breath in, b: breath gap, c: breath out and d: breath gap). Reproduced with permission.^[1108] Copyright 2019, American Chemical Society. g) Left: schematic illustration of graphene-based conformal TENG (top), photographs (middle), and the corresponding voltage output (bottom) when the sensor was touched by different number of fingers. Right: photographs and SEM images (magnified, upper right corner) of the conformal TENG attached to the palms. Scale bars are 5 mm (magnified photograph) and 300 μ m (SEM image). Reproduced with permission.^[1058] Copyright 2016, Elsevier.

by powering sensors and wearable electrical devices with nanogenerators.^[1090–1097]

With the continued pursuit of device miniaturization and the increasing demand for energy consumption from wearable electronic devices and sensors, developing sustainable and self-sufficient energy sources to replace batteries is critical for developing wearable devices for personal health care, motion monitoring, and human-machine interfaces.^[23,1098] Numerous energy harvesting techniques, such as piezoelectric, triboelectric, photovoltaic, thermoelectric, and pyroelectric, have been demonstrated to provide a stable energy source for wearable devices sensors.^[1014,1091,1093,1099–1101] Zhang et al. developed a MoS₂-based PENG to power an Au-decorated MoSe₂-based ammonia sensor (Figure 10a,b).^[1021] By incorporating the MoS₂-based PENG into various human body parts, the nanogenerator could power the ultra-sensitive Au-MoS₂-based NH₃ gas sensor with a quick response/recovery time.^[1021] Additionally, Li and Zhang used a PENG based on MoSe₂ to power a WSe₂/MoS₂ heterojunction pH sensor.^[1014] Wang et al. recently developed a MoSe₂-based PENG for powering a humidity sensor.^[1102] These studies established that energy harvested from the human body via mechanical nanogenerators is sufficient to power sensors used in health monitoring.

Integrating nanogenerators, sensors, and electronics into a single device to create a so-called “self-powered” device has garnered considerable attention. It has proven to be an effective strategy for device design.^[1103] Lee et al. recently published a paper describing a graphene-based heterojunction photovoltaic cell-based chemical gas sensor.^[1099] Lee et al. controlled the chemical potential of graphene using adsorbed gas molecules as the counter electrode of the solar cell.^[1099] When light is irradiated onto the photovoltaic cell, the photocurrent increases or decreases depending on the redox characteristic of the adsorbed gas molecules, while the sensor does not respond in the dark.^[1099] Kim et al. developed a p-type (WSe₂/WS₂) and n-type (MoS₂/WSe₂) 2D TMDCs heterostructure-based photovoltaic cell capable of detecting NH₃ and NO₂ (Figure 10c,d).^[1104] As previously demonstrated, charge transfer from adsorbed gas molecules to 2D TMDCs significantly affects the output photocurrent.^[1104] Niu et al. later showed that the n-MoS₂/p-GaSe heterojunction photovoltaic cell might be used as a self-powered NO₂ gas sensor with an even lower detection limit (down to 20 ppb) when exposed to 405 nm light at ambient temperature.^[1105] Additionally, a self-powered UV photodetector (392 nm) and NO₂ gas sensor based on 2D g-C₃N₄ nanosheets and GaN nanorods were reported.^[1106]

Lee et al. reported the development of a self-powered graphene-based touch sensor that exhibited exceptional flexibility, stretchability, and wearability.^[1107] Due to the auxetic mesh architecture, the TENG-powered touch sensor could distinguish between simple touch and touch-sliding and offer the proper sliding trajectory.^[1107] Dai et al. used 2D α -In₂Se₃ to manufacture a self-powered piezoelectric sensor for real-time human health monitoring (Figure 10f).^[1108] Due to the remarkable flexibility and stretchability of the piezoelectric sensor based on 2D α -In₂Se₃, the device can be mounted to the wrist and chest for pulse and breath signaling.^[1107] Wang et al. reported on self-powered strain sensors based on piezoelectric and thermoelectric energy harvesting.^[1109] By combining the mechanoelectrical and thermoelectric properties of SnSe₂ and graphene heterostructures, the device achieved a record ZT value of 2.43 and functions as a strain sensor by measuring the variation of the output thermoelectric current.^[1109] Wang et al. also established the self-powered pressure sensor capability of MXene-enhanced PENG.^[1110] Du et al. demonstrated a self-powered human motion sensor using a single-electrode mode Ti₃C₂T_x-based TENG.^[1098] The TENG had an open circuit voltage range of −65 to 45 V, a short circuit current of 29 A, and was highly flexible, stretchable, and self-healing.^[1098] Recently, the concept of merging TENGs and FETs to construct a TENG-gated/driven FET (T-FET), sometimes referred to as “tribotronics,” has garnered considerable attention in the 2D materials community.^[1111–1116] Additionally, the transistor matrix and arrays constructed from many T-FETs can be employed as sensory systems for biological stimuli and artificial synapses.^[1112,1117–1120] However, the majority of these studies use 2D materials as transistor channels rather than triboelectric layers, which is beyond the scope of this review.^[1112,1114,1120]

Roy et al. used graphene oxide nanosheets and PVDF-contained nanofibers to create a multifunctional pyroelectric energy-driven breath sensor and piezoelectric pressure sensor.^[1121] The PVDF/GO energy harvester demonstrated a pyroelectric output power density of 1.2 nW m^{−2} and the ability to sense temperature changes caused by respiration.^[1121] Li et al. integrated a SnSe-based PENG with a MoS₂-based multifunctional sensor for pH, optical, and humidity sensing to create a self-powered sensing unit (SPSU) (Figure 10e).^[1122] The SPSU, driven by the vibration energy of the heartbeat via PENG, demonstrated excellent response to pH, light, and humidity, showing the feasibility and potential of a self-powered, atomic-scale human-health monitoring system.^[1122] By altering the surface functional groups of GO paper, a TENG was converted into a self-powered humidity sensor.^[1123] The hydrophilic surface of the modified-GO paper may facilitate the adsorption of water molecules, resulting in a considerable voltage and current reduction when humidity increases from 20% to 90%.^[1123]

Chu et al. previously reported using a triboelectric nanogenerator to power wearable electronics (Figure 9g).^[1041] The device could be integrated with a communication device to send Morse code using graphene as the electrode in the single electrode-mode TENG.^[1058] Additionally, Liang et al. described a GO sheet-based nanogenerator for data storage and expression.^[1124] The nanogenerator could gather energy from moisture, and by

weaving the harvester into a facial mask, it could be converted into an electronic label. These studies cleared the way for the advancement of wearable and human-health applications powered by wearable nanogenerators.^[1124]

5. Conclusion and Outlook

This review detailed the present research on 2D materials for wearable solar, thermal, and mechanical energy harvesting (Figure 11). Due to the fascinating features of 2D materials, such as tunable electrical and optical properties, superior flexibility, and stretchability, 2D materials are ideal for wearable energy harvesters. The structure and chemistry of 2D materials, such as their thickness, phases, and surface functional groups, significantly influence their mechanical, electrical, optical, and thermal properties. By accurately controlling the preparation of 2D materials via top-down or bottom-up procedures, it is possible to synthesize high-quality 2D materials with the needed structure, chemistry, and characteristics for various forms of energy harvesting. Despite considerable effort and success in related domains, more work remains to be done to advance 2D materials-based wearable energy harvesting. Beyond graphene and MoS₂, the process–structure–properties–performance relationship of emerging 2D materials is still lacking in some situations, posing a challenge for scientists interested in creating and modifying materials properties through process control.

To this purpose, we envisage the following research challenges, which are accompanied by an abundance of research opportunities:

- 1) Scalable manufacturing of large-area, substrate-agnostic, high-quality designer 2D materials. 2D materials-based wearable energy harvesting is projected to benefit significantly from developing large-area, high-quality 2D materials that can be prepared at low cost and with good control over the shape and dimensions through cost-effective, holistic design. However, the substrate-agnostic manufacturing of 2D materials on a large scale and at a reasonable cost with precise control has remained a challenge. Current synthetic techniques are constrained by product quality, process yield, growing conditions, and/or material stability. The size, shape, and/or thickness of 2D materials generated through exfoliation and hydro/solvothermal synthesis cannot be adequately controlled, resulting in a wide variety of sample sizes and thicknesses. Meanwhile, growth by CVD, PVD, or MBE processes results in low yields and needs tight control of substrate selection, chamber temperature, and pressure.^[1125] Repeatability and reliability in the scale-up production and integration of 2D materials-based wearable energy devices must be controlled to ensure device quality, including layer-to-layer registration, surface contamination, and morphologies of as-assembled 2D materials on flexible substrates. A more comprehensive approach that takes advantage of both fundamental understandings and technological capabilities is needed to remove significant manufacturing obstacles, such as controlling defects during process scaling,^[171,236] revealing non-linear scaling relationships for potential mass productions,^[550,1126] and reducing the strict requirements for growth

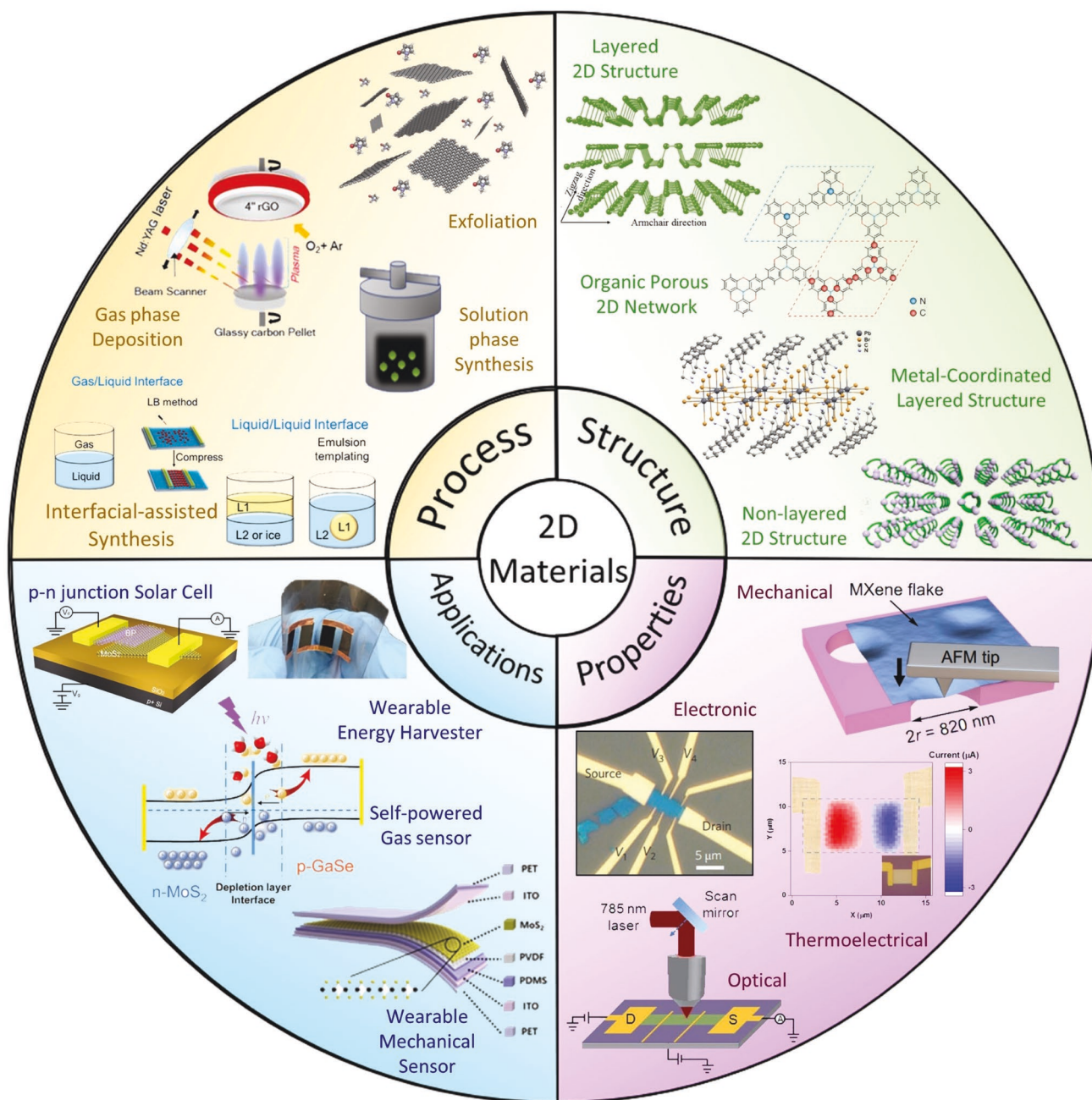


Figure 11. Overview of 2D materials with unique structure, properties and prepared by various techniques in energy harvesting and application for human-related applications. Reproduced with permission.^[111,131,142,297,301,404,542,669,757,772,927,969,1056,1185] Copyright 2014, the Royal Society of Chemistry. Copyright 2020, American Chemical Society. Copyright 2018, American Chemical Society. Copyright 2018, American Chemical Society. Copyright 2020, Springer Nature. Copyright 2016, Wiley-VCH. Copyright 2014, Springer Nature. Copyright 2019, American Chemical Society. Copyright 2019, Elsevier Ltd. Copyright 2014, American Chemical Society. Reproduced under terms of the CC-BY license.^[248,1105] Copyright 2018, The Authors. Published by the American Association for the Advancement of Science. Copyright 2021, the Authors. Published by Wiley-VCH.

substrates (for example, epitaxy).^[603,1127] Design and control of the manufacturing process must be holistic for multiple manufactured device scales to be reliably engineered.^[1128–1135] A combination of machine learning-enabled closed-loop control and inline characterization approaches could give real-time inspection and control of product quality, allowing the process to self-optimize toward manufacturing materi-

als with the desired properties.^[1136–1138] Additionally, phase engineering of 2D materials could lead to discovering new phase-dependent features of 2D materials and the expansion of 2D materials in wearable, sustainable energy harvesting.^[1139,1140] In many 2D materials, the atomic thickness also contributes to their instability in the environment.^[1141,1142] Encapsulation and device structural optimization would be

necessary to address this issue.^[781,1143,1144] Understanding the underlying relationship between the 2D functional material preparation process and its structural properties is necessary if materials with precisely controlled dimensions, compositions, phases, and properties are produced.

- 2) The biocompatibility and toxicity of 2D materials should be better understood. Wearable energy harvesters, in particular, have the potential to cause significant harm to the human body when made from 2D materials. Biosafety studies on 2D materials beyond graphene are still in their infancy, despite the rapid rise in applying these materials in wearable energy harvesting.^[467,1145,1146] There are still many unanswered questions about 2D materials' cytotoxicity, both in vivo and in vitro, even though many earlier studies have addressed this issue.^[467,470,1147] Both short-term and long-term in vivo effects on 2D materials' cytotoxicity can be influenced by their dimensions, shape, chemical content, and surface functionalization groups.^[461] One of the most critical variables in determining the biocompatibility of 2D materials is their controlled preparation and the process-structure-properties relationship.^[475] It is also essential to understand how 2D materials interact with living organs to comprehend their long-term impacts on human health better. Biocompatibility and nanotoxicity criteria for wearable energy harvesting may not be as stringent as those for drug administration or treatments, yet little is known about the biodegradation and biopersistence of 2D materials when in contact with human tissues. In order to better understand the biocompatibility and cytotoxicity of 2D materials, a library of 2D materials having both in vivo and in vitro biocompatibility and cytotoxicity should be established.^[475,496] It is important to conduct a systematic metabolic analysis of 2D materials to better understand their long-term toxicity, especially for those with surface functional groups that are chemically or physically adsorbable during the fabrication process.^[1145,1148–1150] For wearable energy harvesting devices, the safety concentration of any 2D material allowed in the human body is crucially important. It is possible to estimate the potential biocompatibility of 2D materials using machine-learning-driven simulation and calculation on biomaterials' design and hazard reduction.^[1151–1155] What matters is whether or not 2D materials can be shown to be safe for humans to use in vivo so that the ambiguity in the biocompatibility of 2D materials may be clarified.^[475]
- 3) Making 2D materials-based wearable energy harvesters more efficient and effective. Previously reported energy harvesting systems have power density per centimeter square (PCE) values in the μW to mW range and PCE values below 30%.^[1156,1157] Due to their low power density and power conversion efficiency, state-of-the-art nanogenerators cannot be used as the only power source for wearable devices. To enhance the performance of energy harvesters, two primary tactics can be used: investigating and creating novel 2D materials and improving device configuration and architecture. By bridging the process–structure–properties and structure–properties–performance relationships, it will be possible to maximize the energy conversion performance of 2D materials and device designs. While substantial work has been done on rationalizing the chemical and physical properties of 2D materials in order to improve device performance,

a complete grasp of the science underpinning the distinctive properties of 2D materials remains lacking. Although the rising utilization of heterostructures and the incorporation of porous structures into 2D materials for energy harvesting have considerable potential, fundamental understandings remain unknown.^[912] On the other hand, optimizing the device's architecture concerning various energy harvesting technologies may allow the performance limit to be pushed forward from a device perspective. Additionally, by leveraging our understanding of energy harvesting mechanisms, we may optimize output performance by merging two or more energy harvesting systems into a single device. With appropriate material selection and device design, the synergistic effect of hybrid nanogenerators can be realized. For instance, Van Sark proposed a photovoltaic-thermoelectric hybrid system with a 23% performance increase by integrating thermoelectric components within the solar module.^[1158] Additionally, the piezo-tribo hybrid generator, which combines PENG and TENG, revealed the possibility of combining piezoelectric and triboelectric effects in a single material.^[1159] Although there is still much to optimize, these studies open new avenues for increasing wearable energy harvesters' conversion efficiency and power output.

- 4) Enhancing the stability and durability of energy harvesting devices for wearable applications. The endurance and stability of 2D materials-based energy harvesters should not be overlooked, as these devices will be in direct contact with human skin for an extended period. Daily body movement places compressive, tensile, and shear stress and strain on the devices, reducing their performance. Environmental elements such as light irradiation, humidity, and temperature fluctuations should also be considered when discussing the long-term stability and durability of wearable energy harvesters. The device's stress and degradation may also fail energy harvesters, resulting in the inability to power powered wearable devices and sensors. Although cycling tests have become a figure of merit for flexible energy harvesters, few have investigated the effect of hundreds of testing cycles on the device's microstructure. Additionally, the wearable demonstrations of nanogenerators rarely consider the device's long-term performance when friction forces between the device and the human body or clothing are added during actual use. Incorporating the new "self-healing" method into 2D material design envisaged alleviating the damage caused by daily use and external forces.^[1160,1161] Additionally, the washability of wearable energy harvesters should be considered. While washing the wearable nanogenerators regularly may effectively prevent bacteria growth, the cleaning and assembly/disassembly processes result in increased mechanical deformation of the device and the requirement for water contact. This requires the device to be completely waterproof while still keeping the device's permeability for user comfort.^[1162] Recent efforts to optimize the structure/architecture of wearable energy harvesters, such as fabricating electronic tattoos and artificially adding porous structures into the membrane, demonstrate intriguing prospects for increasing the wearability of energy harvesters.^[1162,1163] Additionally, using self-cleaning materials could minimize the frequency of nanogenerator cleaning, extending the device's lifetime.

5) Integrating energy harvesting devices based on 2D materials with energy storage modules. The low power density of a single device and the requirement for many energy harvesters to run a wearable electronic device highlight the importance of combining 2D materials energy harvesting and energy storage devices.^[1164] Separate energy harvesting and storage devices require external circuitry, making them difficult to miniaturize, prohibiting their usage in wearable applications.^[1165,1166] Additionally, power management is required to ensure optimal energy storage and output due to the many energy inputs from hybrid energy harvesters. The increased power consumed by the external circuit and power management system would further degrade the nanogenerators' output efficiency. While the development of all-in-one, self-charging sensors and electronics enable the reduction of power conversion loss caused by the external circuit and power management system, the power source remains intermittent and highly dependent on its environment. To create a "all-in-one" system, numerous recent studies have examined the idea of directly merging nanogenerators and energy storage devices, such as batteries and supercapacitors.^[1167–1169] The concept of a self-charged supercapacitor power cell (SCSPC), first presented by Wang's group in 2015 for harvesting and storing mechanical energy via a PENG, may give a solution to the problem.^[1168] Several energy harvesting systems, including solar cells and TENG, were later incorporated with the SCSPC.^[1170–1172] An interesting work done by Zhang et al. further applies the self-charging idea into a zinc-ion battery to convert chemical energy from spontaneous redox reactions into electrical energy.^[1173] However, difficulties persist in increasing the output voltage of SCP-SCs from mV to V and enhancing the device's long-term stability.^[1174–1176] Integrating many energy harvesters with a single energy storage device likewise remains challenging but demonstrates tremendous promise and application. On the other hand, the necessity of developing wearable energy storage devices should not be underestimated. The issues of poor energy density and the safety of conventional electrolytes remain unresolved. Numerous efforts have been undertaken to increase the performance of wearable energy storage devices while maintaining their safety. These efforts include replacing aqueous electrolytes with high-performance solid or gel-like electrolytes and upgrading battery and supercapacitor packaging. In summary, with advancements in both energy harvesting and storage devices for wearable devices, the vision of 2D materials-based wearable devices powered entirely by energy harvested from the ambient environment can be realized.

Despite the challenges associated with large-scale, high-quality materials synthesis and the long-term durability, wearability, and biocompatibility of both 2D materials and devices, we believe that recent exciting interdisciplinary studies combining booming fields such as data science, nanomanufacturing, and machine learning with well-developed research areas such as chemistry, materials science, and solid-state electronics are paving the way for realizing the full potential of these materials and devices.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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