Energy Technology
Generation, Conversion, Storage, Distribution
www.entechnol.de

Recent Advances in Electrochemical Water Splitting and Reduction of CO_2 into Green Fuels on 2D Phosphorene-Based Catalyst

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Recently, monolayer or few-layered phosphorene nanosheets and other phosphorene-based materials have gained huge attention due to their unique semiconducting and optical properties. But there is a huge unexplored area of research where phosphorene-based catalysts could find new applications. Among these are the urgency of making new functionalized phosphorene materials and studying their catalytic (including photo and electrocatalytic) activities in various types of reactions. Phosphorene has a wide range of emerging applications in the field of energy conversion and storage, photovoltaic devices, and biomedical applications. However, the application of 2D phosphorene-based catalyst in the hydrogen evolution reaction (HER), oxygen evolution reaction (OER), reduction of CO_2 into green fuels, and the nitrogen electroreduction reaction (N_2RR) have not been explored further. Herein, state-of-the-art synthetic strategies of 2D phosphorene produced from allotropes of black phosphorus with their significant role as nanocatalysts in the HER, OER, hydrogen storage, electrochemical reduction of CO_2 into valuable green fuels, and the N_2RR are focused on. This review provides a wide range of knowledge on the current scenario to the researcher, scientist, industrialist, and professional working in the field of material research for energy applications.

1. Introduction

With the elevation in the utilization of energy and enhanced energy demands, major problems developed around the globe, termed as "energy crises." The major source of energy in the modern civilized world is fossil fuels. Fossil fuel sources are limited and are being exploited by humans for centuries. Renewable and sustainable energy resources are required to sustain life. Scientists and researchers around the world have shown keen interest in developing and designing alternate paths for energy resources. Hydrogen energy, biofuels, and other renewable energy sources tend are promising candidates for future energy technologies with research and development. Fossil fuel combustion produces various harmful and toxic end products such as carbon dioxide and other harmful substances that are responsible for natural calamities such as global warming, ocean acidification, and greenhouse gases. The alteration of fossil fuel source is the necessity for our generation. Herein, we provide an overview discussion of an alternate pathway for the production of energy, i.e., the electrochemical pathway. There are

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/ente.202000741.

DOI: 10.1002/ente.202000741

countries with developed status, such as the United States, Japan, and those in Europe, where the industrial revolution began in the 18th century and they were the major contributors to CO_2 emission until the end of the 20th century. These developed countries are termed as the OECD (Organization for Economic Cooperation and Development), and have an estimated population of 1.2 billion. The non-OECD countries' estimated population is 6 billion.[1–8] High energy demand and rising pollution have led to the scientific community and researchers around the world designing and developing various other pathways for energy production. The current development in the field of electrocatalysts for energy production has evolved in recent years.[9] To develop a highly efficient electrocatalyst, the fundamental knowledge and proper understanding of some important characteristics, such as the physical and chemical properties of the materials, are required. One of the most recent and efficient materials is the nanomaterial-based electrocatalyst. Different classes of nanomaterials are synthesized and various potential applications have been discovered.[10] One important class of nanomaterials synthesized using 2D materials is graphene and its derivatives. Graphene-based materials have attracted the scientist community due to their exceptional properties such as mechanical, electrical, and thermal stability. Various other materials have been reported to overcome the problems of the graphene-related materials such as transition metal dichalcogenides (TMDCs; MX_2, M = Mo, W, etc. and X = S, Se, etc.), germanenes, silicones, and phosphorene. Phosphorene, the monolayer form of black phosphorus (BP), is the most recent addition to graphene-like Van der Waals 2D systems. Due to its immense interesting properties, namely, its tunable direct bandgap, high carrier mobility, and unique in-plane anisotropy, it has emerged as a promising candidate for electronic and optoelectronic devices. The main drawback of few-layered phosphorene nanosheets (and BP itself) is their sensitivity to oxygen and humidity in the presence of light, which makes their use in ambient conditions impossible. Several approaches have been made toward protecting phosphorene sheets from oxidation. However, the synthesis of phosphorene-based materials with enhanced stability to ambient conditions is still a very challenging research area. This is why the functionalization of phosphorene sheets should not only bear some new properties or active sites but it should also able to increase the stability of new nanomaterial. The other drawback is the preparation of monolayered phosphorene. It is known that with a decreasing number of phosphorene layers some of the physical properties of materials underwent changes. The most vivid example is the increase of bandgap, which reaches 2 eV for monolayer and ≈0.33 eV for bulk BP. To achieve the most active materials with the best catalytic and chemical activities, it is favorable to prepare phosphorene nanosheets with the number of layers as low as possible, ideally to obtain suspensions considering only monolayer BP. Quite a lot of work has been done in this field, yet obtaining monolayer phosphorene is still challenging.[9,10] Currently, the method of liquid exfoliation of BP is widely used to obtain few-layered phosphorene nanosheets. This method consists of successive sonication and centrifugation processes (or rather, cycles of sonication and centrifugation where the centrifugation speed is gradually increased). Liquid exfoliation of BP makes it possible to obtain bi- or even monolayers of phosphorene, but the usage of quite a harsh sonication process (which lasts dozens of hours) breaks not only the Van der Waals interactions between two layers but also the P—P bonds within one layer, so the surface area of phosphorene also decreases. Other methods, such as electrochemical exfoliation, could help to obtain high-quality phosphorene sheets with acceptable lateral size.

In recent years, the phosphorene-based catalysts have played a very important role in various energy applications.[11–16] Black phosphorus was first synthesized by Bridgman in 1914.[17] The single layer of BP is most commonly known as phosphorene. The phosphorene name resembles graphene as they have similar properties and applications.[18–20] Phosphorene-based nanomaterials have drawn immense development due to their various potential applications in the field of energy production, for example, anode materials in lithium ion batteries and other energy applications.[21–25] The other class of compounds having similar properties to graphene and phosphorene is pnictogens. These consist of BP, arsenic (As), antimony (Sb), and bismuth (Bi) elements. These pnictogens are also analogues to graphene. The electrocatalytic properties of phosphorene-based catalysts have not been explored in light of water splitting and reduction of CO_2 applications so far. Thus, in this direction this review article focuses on the utility of phosphorene as an electrocatalyst and its wide range of potential applications in the hydrogen evolution reaction (HER), oxygen evolution reaction (OER), hydrogen storage, and reduction of CO_2 to green fuels. The future potential applications and advances in the various branches of sciences are explored.[26–35] The number of publications based on phosphorene-based catalysts in the field of batteries, water splitting, and reduction of CO_2 is shown in **Figure 1**.

2. Structure of Phosphorene

One of the most explored allotropic forms of phosphorus is BP. BP generally has a layered structure (**Figure 2**). It is a type of narrow-bandgap semiconductor having a bulk bandgap of ≈0.3 eV. Phosphorene is generally known as the single-layered phosphorus. Phosphorene can be obtained by exfoliation of BP with the help of scotch tape to form single-layered phosphorene. The band width of phosphorene was predicted and it got confirmed to be ≈2 eV.[36–46] The structure of BP is similar to that the graphite 2D crystal structure having Van der Waals

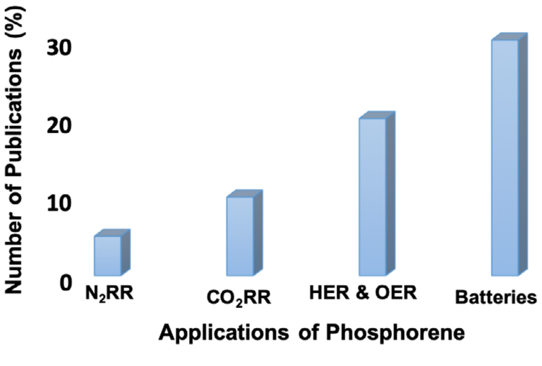

Figure 1. Studies published on 2D phosphorene from 2015 to 2020.

forces. In this structure, one phosphorus atom is bonded with three adjacent phosphorus atoms covalently and forms a puckered honeycomb-like structure in a single layer. By using quantum mechanical calculation, Bachhuber et al. showed that BP is the most stable allotrope of phosphorus, which is mostly due to the contribution of Van der Waals forces. BP and phosphorene possess various novel properties such as optical and thermal stability, conductivity, and electro/photocatalysis.[47–52]

2.1. Phosphorene and Graphene Analogous Behavior

Graphene and black phosphorene have various properties that are similar, including 2D structure. Both graphene and phosphorene have monolayer sheet structure. Graphene is a carbon monolayer; however, phosphorene is a BP monolayer structure.[40] Graphene has high carrier mobility and phosphorene has high hole mobility. The graphene and phosphorene-based materials have various applications in the energy sector but very few studies have been reported on water splitting and CO_2 reduction applications.[53–55] Both graphene and phosphorene have high-performance optical properties. One property that differs between graphene and phosphorene is their bandgap. Graphene holds zero bandgap, whereas phosphorene has a direct bandgap value from 0.3 (bulk) to 1.5 eV (monolayer). The Young's modulus (GPa) value of graphene is 1000 (GPa), while phosphorene has a value of 1000 (GPa). The fracture strain (%) values of graphene and phosphorene are also similar, 27–38 and 24–32, respectively.[53–66] **Table 1** shows the fundamental properties of graphene and phosphorene.

The basic properties of phosphorene have been explained by its crystalline and electronic band structure with the characterization of the surface via atomic force microscopy (AFM), transmission electron microscopy (TEM), and selected area diffraction pattern (SAED) (Figure 2). In addition, the Raman and photoluminance spectra of phosphorene are also shown.

3. Synthesis of Phosphorene from BP

The synthesis of 2D phosphorene (single layer) and 3D BP (bulk crystals) is discussed here. The other synthetic approaches to synthesize 2D material have been discussed as well to better understand and give direction to the future possibility of a new synthetic method for phosphorene membrane fabrication. BP consists of an orthorhombic crystalline structure according to the Cmca space group.[67–72] BP (3D) is the main source of

Table 1. The fundamental properties of phosphorene and graphene.[66]

S. no. properties	Phosphorene (semiconductor)	Graphene (semimetal)
1. Bandgap [eV]	0.3-2 eV	0
2. Young's modulus [GPa]	35–146	1000
3. Fracture strain [%]	24–32	27–38
4. Carrier mobility [$cm^2 V^{-1} S^{-1}$]	≈1000	$\approx 2 \times 10^5$
5. Thermal conductance [$W m^{-1} K^{-1}$]	**10–36**	**3×10^5**

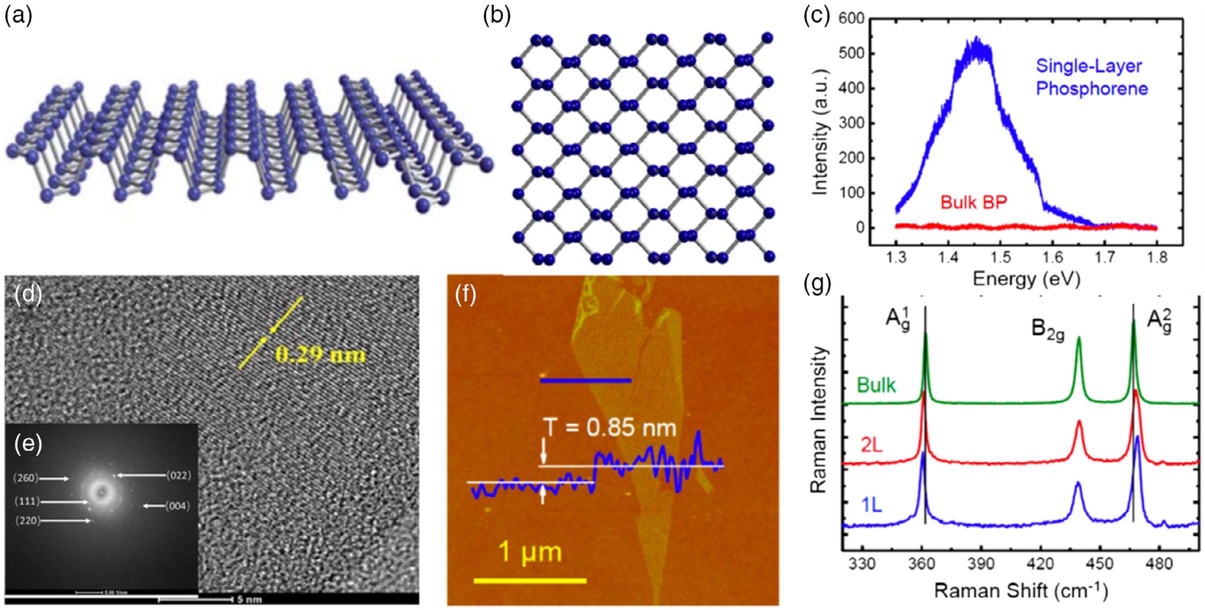

Figure 2. Basics of phosphorene. a) Phosphorene nanosheets. Reproduced with permission.[67] Copyright 2020, Royal Society of Chemistry. b) Crystal structure. Reproduced with permission.[260] Copyright 2020, Elsevier. c) Photoluminescence spectra for single-layer phosphorene showing a pronounced PL signal around 1.45 eV. Reproduced with permission.[134] Copyright 2020, American Chemical Society. d,e) High-resolution TEM image of monolayer phosphorene and inset, SAED pattern showing the orthorhombic crystal structure of the phosphorene. Reproduced under the terms and conditions of the Creative Commons CC BY 4.0 license.[261] Copyright 2018, The Authors, published by MDPI. f) Atomic force microscopy image of a single-layer phosphorene crystal. Reproduced with permission.[134] Copyright 2020, American Chemical Society. g) Raman spectra of single-layer and bilayer phosphorene and bulk BP films. Reproduced with permission.[134] Copyright 2020, American Chemical Society.

the single-layer 2D phosphorene membrane. This is a very high demand procedure, having a simple cost-effective synthesis protocol. Another way to synthesize BP is through a mineralizer-assisted gas vapor transformation. In this procedure, BP is synthesized with the heating of an allotrope of phosphorus known as red phosphorus along with tin iodide (SnI_4)/tin (Sn). The reactants are sealed packed in a glass ampoule, which leads to the synthesis of BP crystals with a quite safe and easy procedure.[73–75] The preparation of the bulk BP crystal from the red phosphorus is through a single-step methodology known as the chemical vapor transport method. Basically, in this procedure, the bulk BP crystal size is reported to be increased as investigated by different research groups.[73–78] The resultant BP materials are synthesized via mineralizer-assisted chemical vapor transport methods in a sealed ampoule, as shown in **Figure 3**.

3.1. Phosphorene Monolayer Nanosheets

Thin-film-based phosphorene materials are prepared through two common strategies. These two strategies are known as the top-down approach and the bottom-up approach, respectively. The bottom-up approach preparation of BP crystals is through thermal deposition or by combining the small phosphorus reactant. Another strategy for the preparation of BP crystal monolayer phosphorene is the top-down approach, which involves exfoliation from its 3D BP (bulk crystal) source. The strategies involved in the exfoliation technique are based upon the weak interlayer interactions;[79] amongst the sheets of the 2D phosphorene these forces can be overcome by providing an external force. On the basis of the external force provided, the exfoliation methodologies can be categorized as microwave exfoliation,[80–83] mechanical exfoliation,[84,85] and sonication exfoliation.[58] Further, the sonication exfoliation method can be distinguished as water exfoliation, organic liquid exfoliation,[86–90] or aqueous surfactant exfoliation (**Figure 4**).[91]

3.1.1. Water Exfoliation

Phosphorene is obtained from the water exfoliation method. The medium is water for the sonication to obtain various types of exfoliation. This synthetic method is a very easy and a green route to obtain phosphorene. BP powders were obtained from BP bulk

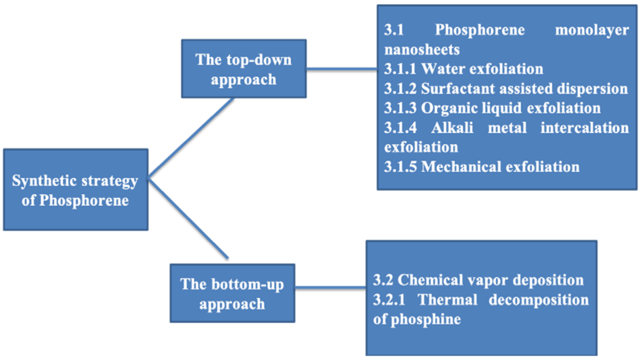

Figure 4. The synthetic methods of phosphorene.

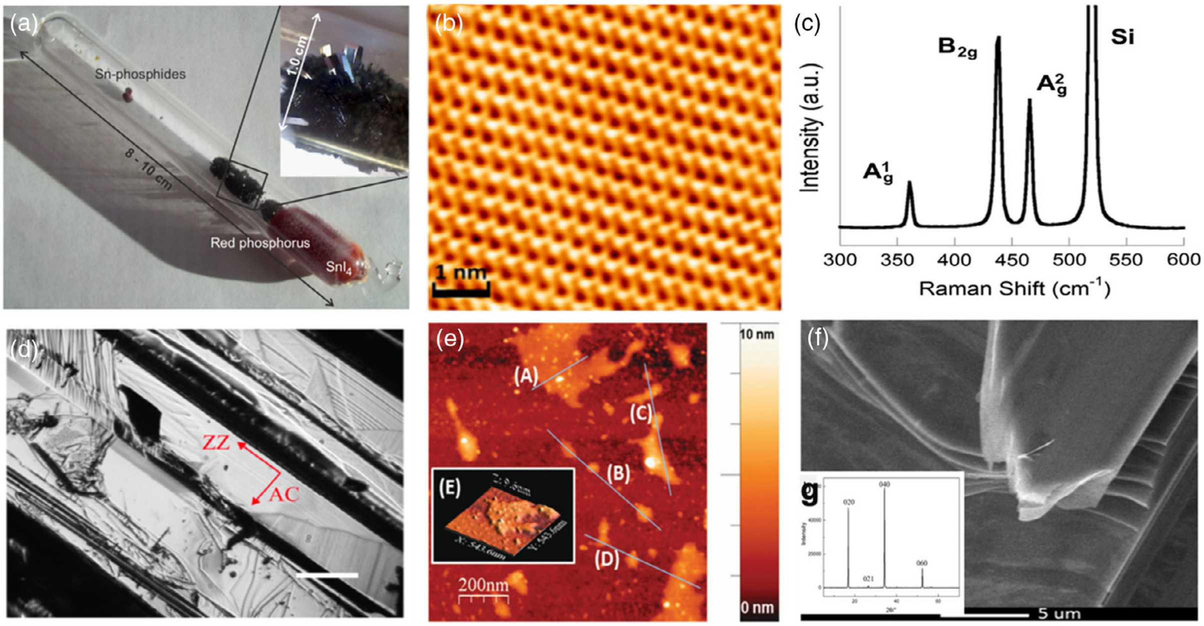

Figure 3. The synthesis of BP in a silica glass ampoule. The orange and red color are SnI_4 and red phosphorus obtained from the gas phase condensed in ampoule. Reproduced with permission.[260] Copyright 2020, Elsevier. b) Scanning tunneling microscopy image showing the atomic surface of BP. Reproduced with permission.[45] Copyright 2020, American Chemical Society. c) Raman spectra of multilayer phosphorene from liquid exfoliation of BP showing A_g^1, B_{2g}, and A_g^2 modes. Reproduced under the terms and conditions of the Creative Commons CC BY 3.0 license.[262] Copyright 2014, The Authors, published by Royal Society of Chemistry. d) Optical microscope image of BP. Reproduced with permission.[263] Copyright 2020, American Chemical Society. e) AFM image of multilayer phosphorene.[262] f,g) Scanning electron microscopy image and powder X-ray diffraction spectra of bulk BP. Reproduced under the terms and conditions of the Creative Commons CC BY 4.0 license[261] Copyright 2018, The Authors, published by MDPI.

crystals, and then BP powder (1–10 mg) was dispersed in deionized water (20 mL) by tip sonication for 30–300 min after the solution was formed, and solid particles that were dispersed got settled, and the supernatant was decanted and further centrifuged, as reported by Ren and coworkers.[92] The phosphorene may act as the anodes in secondary electrochemical batteries.

3.1.2. Surfactant-Assisted Dispersion

To obtain phosphorene as an end product from the surfactant-assisted dispersion method, chemical surfactants such as soaps and sodium dodecyl sulphate were used by Hersam and coworkers. These surfactants play a major role in breaking the interlayer interaction among the phosphorene multiple layers and help to separate the single-layered phosphorene sheets during the sonication procedure.[91] Phosphorene suspensions/dispersions are broadly used in the field of physical chemistry research, for example, in photoluminescence,[93–96] pump probe spectroscopy,[97,98] transient bleaching,[99] optical adsorption,[100–102] and exciton dynamics,[103,104] such as trion lifetime,[96,105] charge transfer,[106,107] and exciton quenching.[95]

3.1.3. Organic Liquid Exfoliation

The different types of organic solvent media, such as *N*-methyl-2-pyrrolidone, dimethylformamide,[108,109] dimethyl sulfoxide, *N*-cyclohexyl-2-pyrrolidone, and isopropanol are utilized for exfoliation of few-layer phosphorene flakes. Further, it has been discovered by Hersam and coworkers and Salehi-Khojin and coworkers that the size of the nanoflakes of phosphorene can be controlled by the strength of sonication and the processing time.[86] Various parameters were utilized to optimize the method. Bulk crystals of BP were first ground before utilization,[110,111] leading to trouble-free sonication in the following step due to the formation of the nanoparticles of the BP crystals. In comparison to bath sonication, which required 24 h, probe sonication is far more efficient and finishes the exfoliation procedure in 5 h.[86,90]

3.1.4. Alkali Metal Intercalation Exfoliation

The graphene[112] and MoS_2[113] can be intercalated with lithium to give the reduced phase with expanded lattice, and the following when immersed in the aqueous solution it produces hydrogen bubbles, which results in separation of monolayer end product, Morrison et al. reported. The lithiation of the phosphorene material results in 300% volume extension.[114,115] The interaction between the interlayer of the phosphorene starts to loosen and the layers start to split and in water lithium reaction occurs. In graphene, big sodium ions are utilized for the exfoliation and intercalation of graphene,[116,117], MoS_2,[118,119] and antimony (Sb).[120]

3.1.5. Mechanical Exfoliation

Graphene,[121–125] MoS_2,[126,127] WS_2,[128,129] h-BN,[130,131] and MXene[132,133] were successfully obtained by the microcleavage method. This is due to the simple separation of the monolayer material by scotch tape method, although the mechanical exfoliation is a very slow procedure and the end product obtained is very low in quantity.[70,134–138] The phosphorene exfoliated by this method is utilized in the field of electronic devices and fundamental physics as the obtained phosphorene generally contains a few layers of argon[139] or ozone.[140,141] This drawback can be overcome by using plasma force leaning of the surface and layer thinning and finally obtaining single-layer phosphorene. The organic residues present on the adhesive tapes must be removed; for this a better technique is adapted, which is known as the all- dry transfer technique,[142] which consists of a thermal release tape[143–145] or a polydimethylsiloxane (PDMS) stamp, as reported by Castellanos-Gomez et al.[142,146–149]

3.2. Chemical Vapor Deposition

The chemical vapor deposition method has been effectively utilized for the formation of large-area layered materials. In this method, red phosphorus thin films were prepared by heating a single piece of either BP or red phosphorus crystals in a tube furnace up to 600 °C under vacuum conditions with a silicon substrate for 30 min. The appearance of green color films on the silicon substrate surface from red is clear indication of a thin film of phosphoous.[150,151]

3.2.1. Thermal Decomposition of Phosphine

Phosphorus has been very deeply studied for n-type doping of silicon-phosphorus, for silicon microelectronics.[152–155] The silicon (100) surface terraces have been combined with phosphorus atoms through the thermal decomposition of PH_3.[156–159] Further, the addition of phosphorous films on single crystals such as Cu (111) and Au (111)[160,161] has also been studied (**Table 2**).

4. Allotropes of Phosphorus

Phosphorus is a member of the periodic table from group 15. This group is also known as the pnictogen group. Phosphorus is an earth-abundant element and generally present in the form of phosphate rocks, e.g., $Ca_3(PO4)_{2,}$ in an oxidized state. There are four main allotropes of phosphorus, which are is known as

Table 2. Synthesis of phosphorene/BP.

S. no.	Methods for synthesis of phosphorene	References
1.	Organic liquid exfoliation	[86]
2.	Surfactant-assisted dispersion	[91]
3.	Water exfoliation	[92]
4.	Alkali metal intercalation exfoliation	[114,115]
5.	Mechanical exfoliation	[139]
6.	Chemical vapor deposition	[151]
7.	Thermal decomposition of phosphine	[156–159]

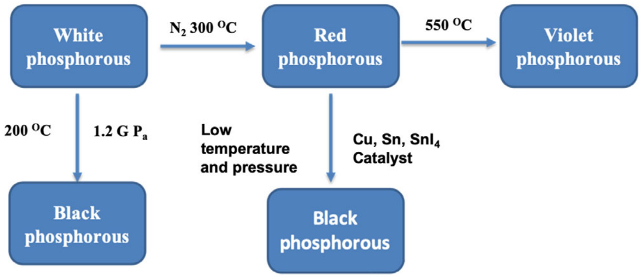

Figure 5. The allotropes of phosphorus and their conversions.

white phosphorus, red phosphorus, violet phosphorus, and black phosphorus. These are named on the basis of their outer appearance.[151] In **Figure 5** the synthesis procedure and allotropes of phosphorus have been explained.

4.1. White Phosphorus

The two major allotropes of phosphorus are white phosphorus and red phosphorus. White phosphorus is made up of the tetrahedral-shaped P_4 type of the molecule. White phosphorus has been very easily obtained by the mineral sintering of phosphate rocks with coke and silica. The main property of white phosphorus is that it is volatile and highly reactive. White phosphorus ignites in the presence of air at the temperature of 34 °C, because of which it is kept in water sealing for the purpose of storage.[151]

4.2. Red Phosphorus

The derivative of tetrahedral phosphorus P_4, in which one of the phosphorus—phosphorus (P—P) bonds breaks and it forms a fresh bond with the adjacent tetrahedron-shaped P_4 phosphorus, results in a chain type of structure that seems to resemble a polymer. White phosphorus in a N_2 atmosphere at 300 °C is utilized to synthesize amorphous red phosphorus or white phosphorus is exposed to sunlight to yield red phosphorus, and if it is heated further it will convert to crystalline red phosphorus.[151]

4.3. Violet Phosphorus

Violet phosphorus has been prepared from red phosphorus by annealing red phosphorus for a very long period of time at the temperature 550 °C in the presence of molten lead.[151]

4.4. BP

BP is generally a good conductor of electricity. It is black and shiny in physical appearance. BP is the most stable allotrope of phosphorus. It is thermodynamically very stable compared to other allotropes of phosphorus. BP is analogous to graphite in appearance. BP has been synthesized from white phosphorus under extremely high pressure (1.2 GPa) and temperature (200 °C), as reported by Bridgman.[162] The bulk crystal of BP is made up of multiple layer structures which are known to be phosphorene.[163] BP is made up of multiple layers and these layers have interlayers among them. The interaction of these layers can be easily compared to the Van der Waals force interaction.[108,164,165] These interlayer interactions have been easily broken by various methods to obtain single-layer sheets of phosphorene. Phosphorene is nowadays considered a 2D material and has become a part of the 2D membrane family.[70,86,87,146]

5. Electrocatalytic Properties of 2D Materials

The 2D materials (nanomaterials), such as graphene, have emerged as potent materials in the field of energy applications. The nanomaterials of graphene possess astonishing electronic and anisotropic physical properties. Graphene is derived from the delamination of graphite. Following graphene is a family of elemental materials such as phosphorene monolayer of BP and others, for example, bismuthine, antimonate, and arsenene.[53,166–168] The multiple-layered 2D materials are generally utilized as lubricants due to their multiple-layered structures. In modern-day science, 2D materials have been explored for a wide range of applications due to their distinguished intrinsic physical properties and electronic structure. In recent years, the scientific community and researchers around the world have been very keen to find alternative sources of energy to fossil fuels due to various disadvantages of fossil fuels as an energy resource. The 2D nanomaterials have been evaluated to be utilized in this field. Clean and green energy production is one of the major focuses in research nowadays by using electrocatalysis via an electrochemical pathway such as the OER, hydrogen oxidation reaction (HOR), oxygen reduction reaction (ORR), HER, and carbon dioxide reduction reaction (CO_2RR). The electrocatalysis properties of 2D materials are studied primarily as they have properties such as high specific surface area, mechanical properties, and conductivity (thermal and electronic) properties. The 2D nanomaterials can be the future of electrocatalysis for clean and green energy production.[53,169–173] In **Figure** 2 and **6** materials' electrocatalyst properties are shown.

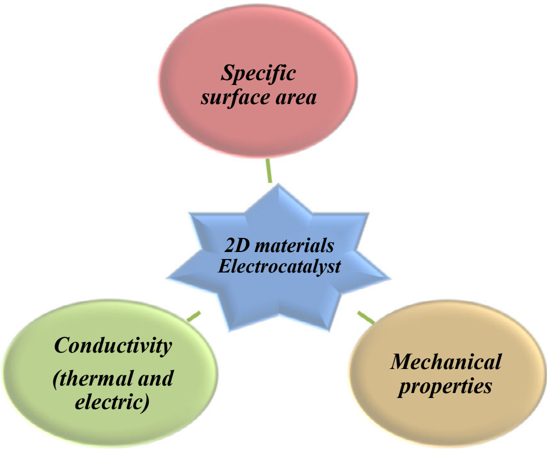

Figure 6. The electrocatalytic properties of 2D materials.

5.1. Pnictogens

The enormous interest in various types of layered materials in the form of single-layer and bulk materials for various applications occurred in 2D layered material due to a special reintroduced material known as graphene. The unique physical and chemical properties of graphene and graphene-based materials led the scientific community to look for various other 2D materials.[53,174,175] The other class of compounds that is recognized as a promising layered material is known as transition-metal dichalcogenides as they are made up of transition metals and utilized in various potential applications in the catalysis field.[168–176] The major drawbacks of the utilization of transition metal–based materials are their high cost and poor stability. However, the 2D materials have benefits as they consist of better surface area, outstanding mobility, better morphological characteristics, and tunability. That perhaps modifies the surface property of the various potential applications.[177,178] An additional purpose that led to the major interest of the scientific community in 2D layered materials is that these materials show remarkable electrochemical performance. The evolution of the electrocatalytic activity of the 2D layered materials that could provide various potential future applications such as in the field of alternate sources of energy is in urgency.[179,180] BP is a layered material that has been reintroduced[135] as a multilayered structure material that also consists of a single elemental layer, which can be easily converted by various procedures to monolayered and multilayered materials.[181] This has led group V elements (pnictides or pnictogens) to be recognized as interesting and potential 2D materials having similar semiconducting and electronic properties to those of phosphorene and graphene.[182] In **Figure 7**, the pnictogens arsenic (As), bismuth (Bi), and antimony (Sb) are shown.

5.2. Defects in 2D Materials

The defects in 2D materials and their vast importance in electrocatalytic activity are one of the most promising and interesting researches ongoing in the scientific community. There are various kinds of defects involved in 2D materials, such as topological defects, edge defects, vacancy defects, dopant-derived defects, oxygen defects, and heteroatom-dopant defects.[174,183–185] Various 2D materials have been categorized as nonmetals and metals. The nonmetals consist of graphene, black phosphorous or phosphorene, and carbon nitrides. The metals consist of TMDCs, layered double hydroxides (LDHs), metal oxides, phosphides, and MXenes [186]. Various kinds of defects and their effects on electrocatalytic activity are an astonishing approach for various electrochemical reactions and their outcomes ORR, OER, HER, and CO_2RR.[187–193] In **Figure 8**, the various types of defects in 2D materials are shown.

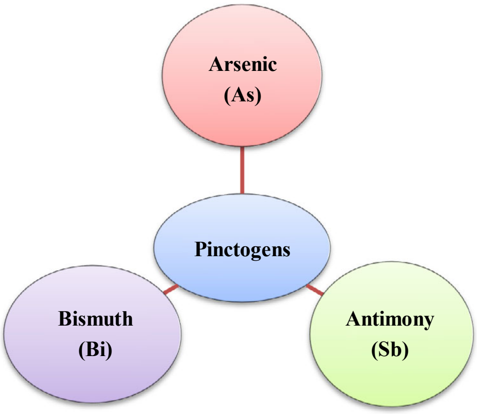

Figure 7. The pnictogens arsenic (As), bismuth (Bi), and antimony (Sb).

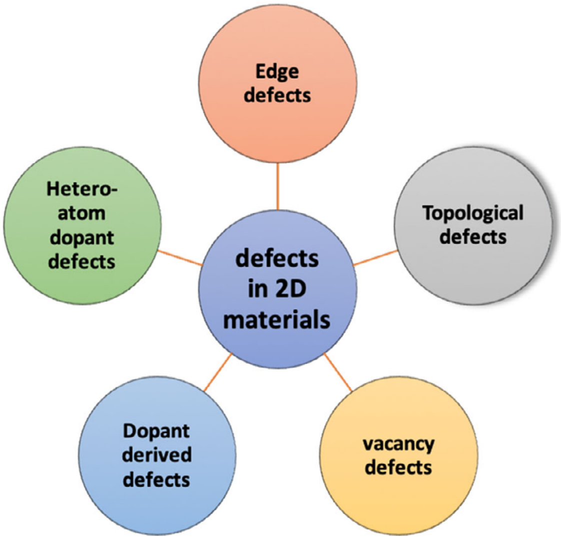

Figure 8. The various types of defects in 2D materials.

6. Phosphorene at a Glance for Various Energy Applications

Phosphorene-based materials, in recent years, have been utilized in various applications such as lithium ion batteries and secondary sodium ion batteries, energy generation, conversion, and storage, supercapacitors, photovoltaic devices, water splitting, photocatalytic hydrogenation, and the HER, OER, and ORR.[151] Single-layered phosphorene obtained from bulk BP has been utilized as an electrocatalyst in various electrochemical reactions. The design and development of various electrochemical conversion reactions and energy-storage devices have been a tremendous way to utilize renewable sources of energy. Electrocatalysis is a branch of chemistry that deals with enhancing the rate of production of electrochemical reactions (HER, ORR, and OER). The phosphorene-based materials have excellent electrocatalytic activity as they are metal-free semiconductors, and therefore BP-exfoliated phosphorene-based materials have been the center of attraction in the field of electrocatalysis.[194,195]

6.1. HER

The HER has been studied quite widely and some electrocatalytic and photocatalytic systems have been made. It is worth mentioning that pristine phosphorene sheets are not very active in HER activation, but their decoration with metal (or metal salts) nanoparticles increases catalytic activities toward water splitting (**Figure 9**). A successful example is the immobilization of nickel phosphide nanoparticles on phosphorene nanosheets and modification of an electrode by the obtained nanocomposite. Other electrocatalytic systems based on immobilized transition metal nanoparticles, such as Cu, have also been reported.

Numerous reports of the development of water-splitting photocatalysts have been made.[151,196] One of them is schematically shown in **Figure 10**. It is shown that some functionalized phosphorene sheets are active HER photocatalysts.

The HER is a cathodic type of reaction in electrochemical water splitting reaction. The phosphorene-based materials are utilized as HER electrocatalysts. Exfoliation is one of the greatest methodologies to improve and enhance the catalytically active sites of BP-based materials for hydrogen production.[196] The catalytic activity of BP was studied through density functional theory (DFT) by Sofer et al.[60] Comparative studies were performed on various types of BP materials, such as edge-plane BP, basal-plane BP, BP macroparticles, BP nanoparticles, BP–Ni_2P, BP–MoS_2, BP–NH_2, BP–Co_2P, and CoP–EEBP (electrochemically exfoliated black phosphorus). The BP edge planes have higher

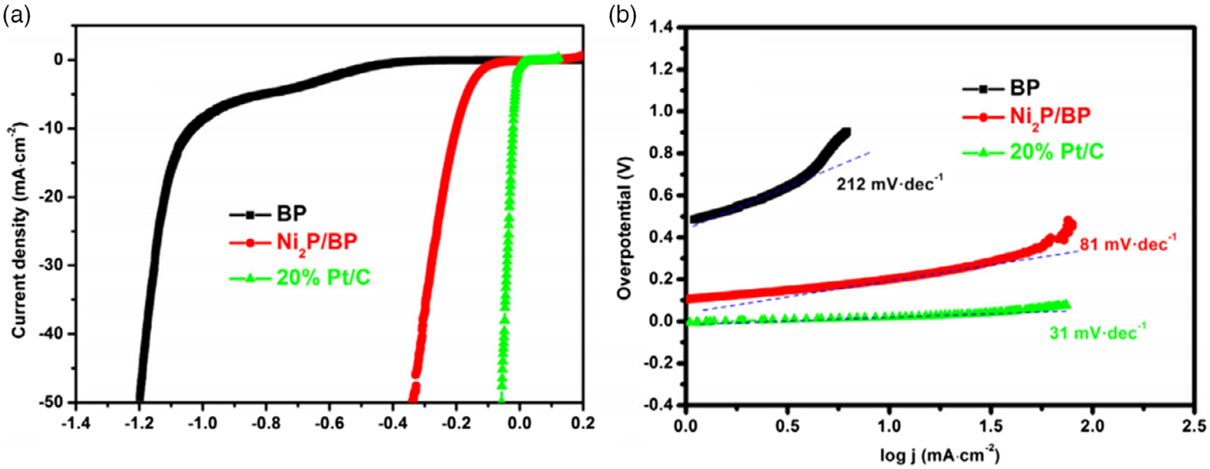

Figure 9. a) Linear scanning voltammetry curves and b) Tafel graphs for BP, Ni_2P/2D BP, and 20% Pt/C catalyst. Reproduced with permission.[195b] Copyright 2020, Elsevier.

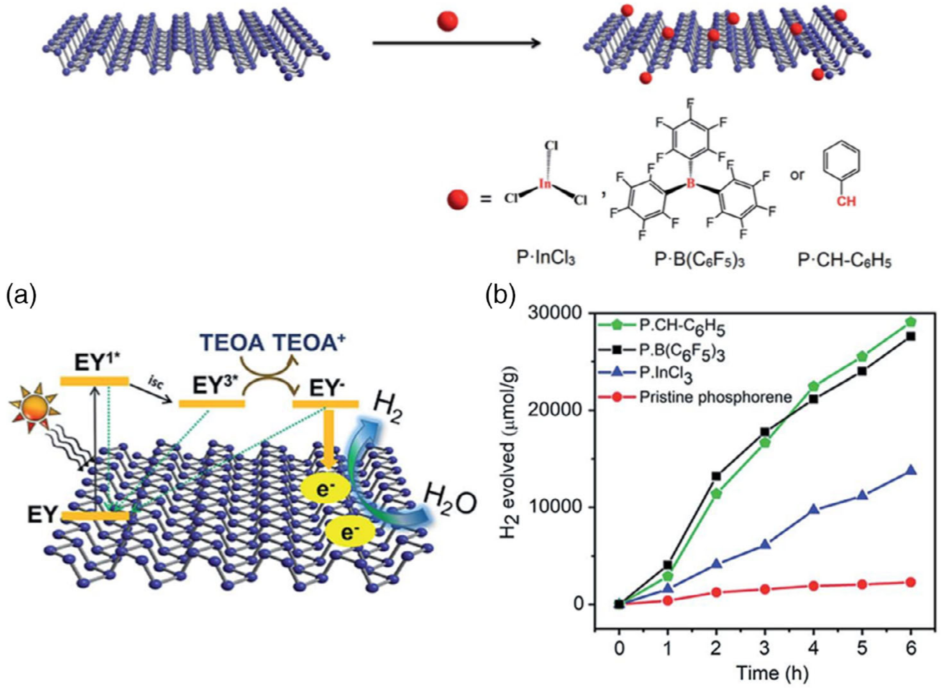

Figure 10. Photocatalytic H_2 evolution from water by phosphorene-based photocatalysts: a) schematic of a plausible mechanism of the H_2 evolution reaction and b) comparison of H_2 evolution activities of different photocatalysts for 6 h. Reproduced with permission.[196b] Copyright 2020, Royal Society of Chemistry.

electrochemical activity than the basal plane surface. The overpotential of the basal plane is -1.13 V and that of the edge plane is -0.55).[197]

In acidic condition, the HER process consists of two steps
1) Volmer step: $H_3O^+ + e^- + * \rightarrow *H_{ads} + H_2O$
2) Heyrovsky step: $*H_{ads} + H_3O^+ + e^- \rightarrow H_2 + H_2O + *$
Or Tafel step: $2 * H_{ads} \rightarrow H_2 + 2*$

The representation here is * as the catalyst and also the active site and $*H_{ads}$ is the adsorbed hydrogen ions on the active site. The two main factors are the concentration of hydrogen absorption and the coverage of the HER can be preceded from the Volmer–Tafel mechanism or Volmer–Heyrovsky mechanism.[198] **Figure 11** shows the characterization of electrocatalyst materials and electrochemical measurement data of electrocatalyst materials, a low-magnification TEM image and high-resolution TEM (HRTEM) image of a typical MoS_2–BP nanosheet, Tafel plots of Pt/C, BP, MoS_2/C, and MoS_2–BP electrocatalysts in 0.5 M H_2SO_4, Nyquist plots, exchange current densities (j_0, geometrical), and Cu stripping curves of the aforementioned electrocatalysts (**Table 3**).

Recently Dai and coworkers reported utilization of CoP nanodots on EEBP nanosheets for overall water splitting. The HER for electrocatalyst material CoP/EEBP heterostructure shows remarkable electrocatalytic activity, having an ultralow overpotential value of only 118 mV at 10 mA cm^{-2} (η_{10}).[199]

6.2. OER

The OER is basically an anodic reaction of the water splitting reaction and in the electrolysis reaction this is the most energy-intensive step as the anode reaction involves four-electron transfer. A huge number of researchers have proposed possible mechanisms for oxygen release reactions in acidic or alkaline

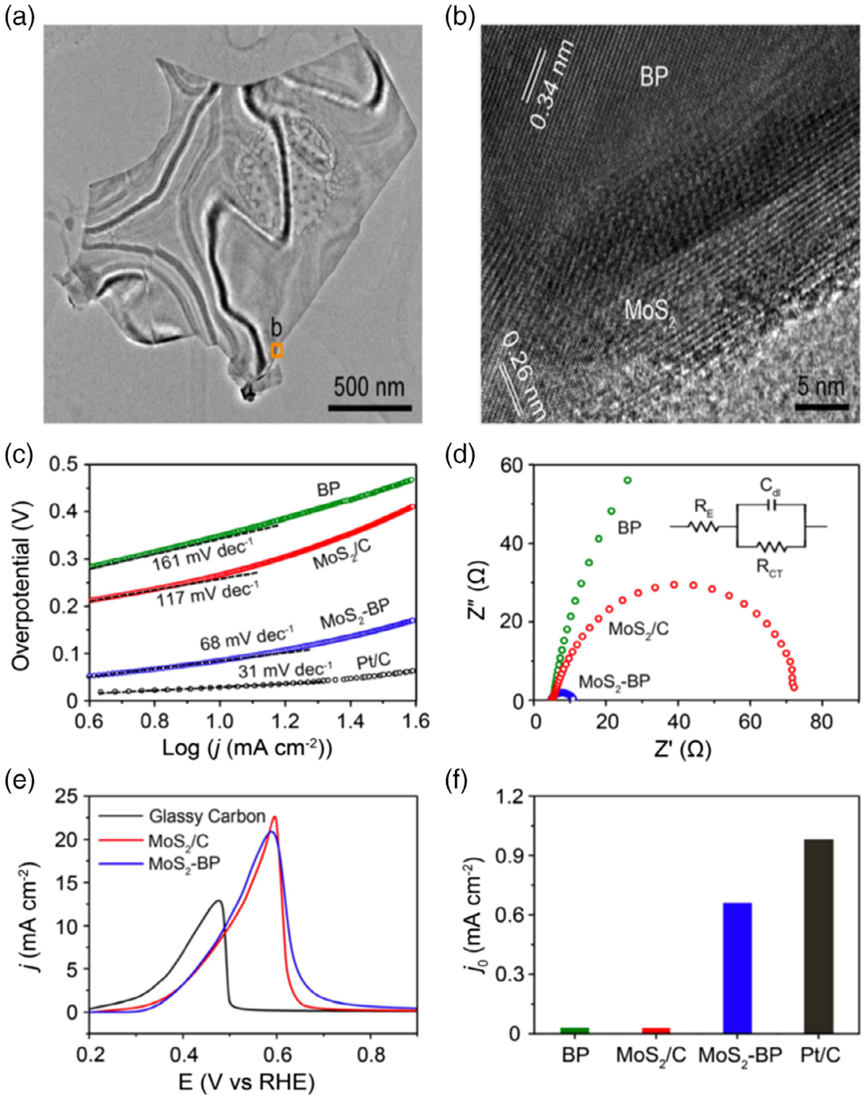

Figure 11. a) Low-magnification TEM image and b) HRTEM image of a typical MoS_2–BP nanosheet. c) Tafel plots of Pt/C, BP, MoS_2/C, and MoS_2–BP electrocatalysts in 0.5 M H_2SO_4. d) Nyquist plots and e) exchange current densities (j_0, geometrical), and f) Cu stripping curves of the aforementioned electrocatalysts. Reproduced with permission.[264] Copyright 2020, American Chemical Society.

Table 3. Summary of BP materials in the HER.

S. no.	Materials	Electrochemical activity Tafel slope/overpotential	Reference
1.	Edge-plane BP	0.55 V	[196]
2.	Basal-plane BP	1.13 V	[196]
3.	BP macroparticles	1.24 V	[197]
4.	BP nanoparticles	85 mV @ 10 mA cm^{-2} Tafel slope: 68 mV dec^{-1}	[264]
5.	BP–Ni2P	0.81 V	[266]
6.	BP–MoS2	107 mV @10 mA cm^{-2} Tafel slope: 81 mV dec^{-1}	[267]
7.	BP–NH2	290 mV @10 mA cm^{-2} Tafel slope: 63 mV dec^{-1}	[268]
8.	BP–Co2P	173 mV @10 mA cm^{-2} Tafel slope: 78 mV dec^{-1}	[269]
9.	CoP–EEBP (CBP-3 composite)	118 mV @10 mA cm^{-2} Tafel slope: 79 mV dec^{-1}	[199]

conditions at anode electrodes. A brief overview of the OER process on the surface of transition metal oxides can help to understand the activity trends observed.[200–202]

BP was first used as an electrocatalyst in the OER by Jiang et al. The electrocatalyst materials were synthesized by using titanium and BP and a carbon nanotube (CNT) network, i.e., black phosphorous and carbon nanotubes (BP–CNTs). The reported materials showed great electrochemical activity due to excellent conductivity and the larger surface area. Facile liquid exfoliation methods were used by Ren et al. to synthesize few-layered BP and study the OER. They also studied the effects of the thickness of BP on OER activity.[203,204]

In alkaline conditions, the OER procedure can be explained as follows.[205]

1) $OH^- + * \rightarrow *OH_{ads} + e^-$
2) $OH^- + *OH_{ads} \rightarrow H_2O + *O_{ads} + e^-$
3) $OH^- + *O_{ads} \rightarrow *OOH_{ads} + e^-$
4) $OH^- + *OOH_{ads} \rightarrow O_2 + H_2O + e^- + *$

* represents active sites of the catalyst and $*OOH_{ads}$, $*OH_{ads}$, and $*O_{ads}$ are adsorbed OOH, hydroxide, and atomic oxygen on the active site, respectively.[205]

The application of few-layer exfoliated black phosphorus (Ex-BP) as an electrocatalyst because of its excellent and promising applications is attractive for the OER. Zhang et al. [206] reported a robust carbon-stabilization strategy to prepare a carbon-coated Ex-BP/N-doped graphene nanosheet (Ex-BP/NGS@C) as a highly efficient electrocatalyst, which showed remarkable OER activity under alkaline conditions at room temperature.

Figure 13 shows the morphological structure determined by TEM and HRTEM images of the 2D Ex-BP/NGS@C hybrids. The interlayer spacing of 0.21 nm corresponding to the 2D structure of Ex-BP/NGS@C is shown in **Figure 12**a. However, the interlayer spacing of 0.21 nm corresponds to the (002) lattice plane of Ex-BP nanosheets. The HRTEM image as shown in Figure 12b corresponds to the (002) lattice plane of Ex-BP nanosheets. The HRTEM image (**Figure 13**b) reveals that the NGS nanosheets contained about five layers with an interlayer spacing of 0.37 nm and Figure 12b is also a signature of the amorphous carbon that was coated on the Ex-BP nanosheets. The elemental mappings for C, N, and P of the Ex-BP/NGS@C hybrid structure are shown in Figure 13c–e. The uniform distribution of all elements clearly indicates good contact of Ex-BP with NGS and amorphous carbon.

Figure 14 shows electrochemical measurement data of electrocatalyst materials and the OER of phosphorene nanosheets, polarization curves of phosphorene nanosheets in KOH electrolyte with various concentrations (ranging from 0.05 to 1 M), with the inset showing the Tafel plots, the calculated Tafel slope as a function of the OH^- concentration, at a current density 10 mV cm^{-2} and potential of 1.6 V, an electrochemical cell based on a three-electrode system with a glassy carbon electrode for contacting the working materials, a photograph of the working electrode with O_2 bubble generation in the electrochemical cell, and rotating ring-disk electrode voltammograms for determining the reaction pathway of electron transfer (**Table 4**).

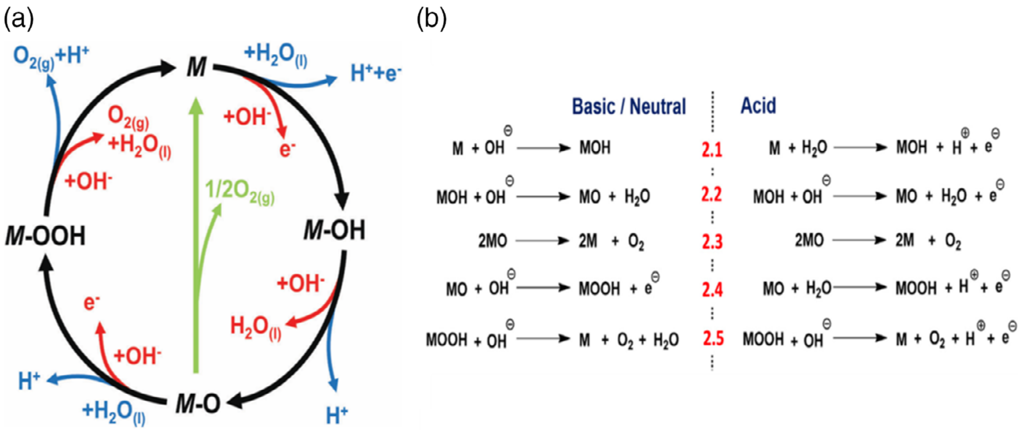

Figure 12. a) The OER mechanism for acid (blue line) and alkaline (red line) conditions. The black line indicates that the oxygen evolution involves the formation of a peroxide (M—OOH) intermediate (black line). b) Scheme of the proposed mechanism for the OER. a,b) Reproduced with permission.[200] Copyright 2020, Elsevier.

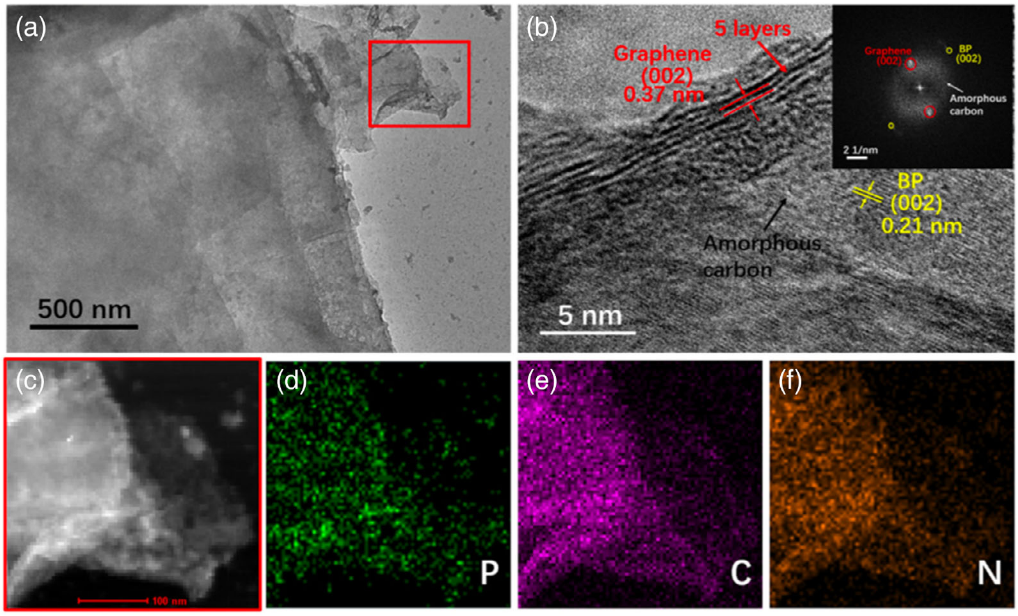

Figure 13. a) TEM image of Ex-BP/NGS@C, b) HRTEM image of Ex-BP/NGS@C to the edge location in (a). c–f) STEM-EDS elemental mapping images of Ex-BP/NGS@C. Reproduced under the terms and conditions of the Creative Commons CC BY 4.0 license.[206] Copyright 2020, The Authors, published by MDPI.

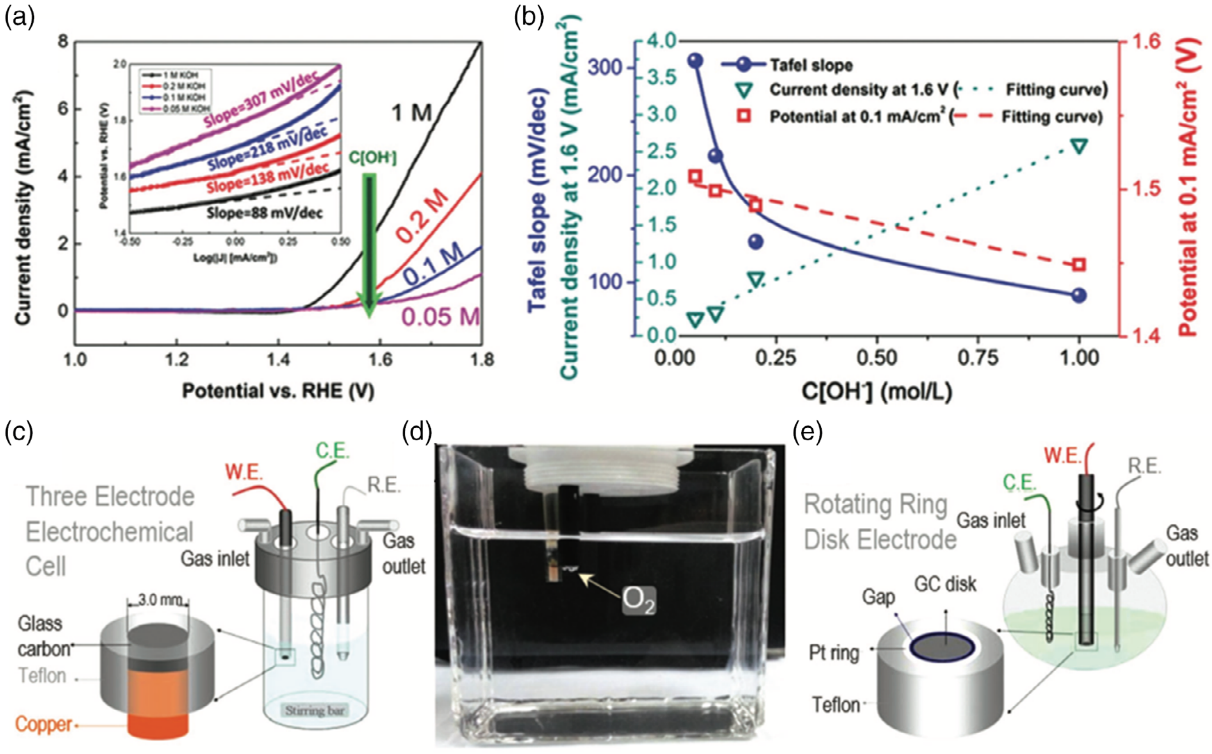

Figure 14. The OER of phosphorene nanosheets. a) Polarization curves of phosphorene nanosheets in KOH electrolyte with various concentrations (ranging from 0.05 to 1 M); inset shows the Tafel plots. b) Calculated Tafel slope as a function of the OH^- concentration, at a current density 10 mV cm^{-2} and potential of 1.6 V. c) Electrochemical cell based on a three-electrode system with a glassy carbon electrode for contacting the working materials. d) Photograph of the working electrode with O_2 bubble generation in the electrochemical cell. E) Rotating ring-disk electrode voltammograms for determining the reaction pathway of electron transfer. Reproduced with permission.[265] Copyright 2020, Wiley-VCH.

Table 4. Summary of BP materials in OER.

S. no.	Materials	Electrochemical activity	Reference
		Tafel slope/onset potential	
1.	BP nanosheets	1.92 V @10 mA cm^{-2} Tafel slope: 78 mV dec^{-1}	[269]
2.	CoP–EEBP (CBP-3 composite)	310 mV mA cm^{-2} Tafel slope: 61 mV dec^{-1}	[199]
3	BP–CNT	1.49 V @10 mA cm^{-2} Tafel slope: 72.88 mV dec^{-1}	[203]
4	BP–Ti	1.48 V @10 mA cm^{-2} Tafel slope: 91.52 mV dec^{-1}	[203]
5	BP–Co_2P	1.45 V @10 mA cm^{-2} Tafel slope: 88 mV dec^{-1}	[204]
6.	MXene BP–QDs	360 mV $mAcm^{-2}$ Tafel slope: 64.3 mV dec^{-1}	[270]
7.	Phosphorene–N–graphene	310 mV mA cm^{-2} Tafel slope: 89 mV dec^{-1}	[271]
8.	N–BP–QDs	430 mV mA cm^{-2} Tafel slope: 48 mV dec^{-1}	[272]
9.	S-phosphorene	310 mV mA cm^{-2} Tafel slope: 75 mV dec^{-1}	[273]
10.	Te–phosphorene	260 mV mA cm^{-2}	[274]
11.	$Co(OH)_2$–BP	276 mV mA cm^{-2} Tafel slope: 57 mV dec^{-1}	[275]
12.	Co–BP	315 mV mA cm^{-2} Tafel slope: 75 mV dec^{-1}	[276]

6.3. Hydrogen Storage

Lithium (Li)-decorated phosphorene has been utilized for hydrogen storage applications.[207] DFT is a theoretical study performed by the scientific community to understand the interactions among the hydrogen molecules and phosphorene-doped materials. The partial density of state analysis, electron density difference, Hirshfeld population, and adsorption energy are orderly studied to clarify the distinguished effects of various dopants such as nonmetallic and metallic atoms on phosphorene with a hydrogen molecule. The studies proved that metallic dopants such as cobalt, nickel, and platinum have increased hydrogen capture when doped with phosphorene and nonmetallic dopants show no such effects.[208–210]

6.4. Carbon Dioxide Reduction

CO_2 emissions have been steadily increasing over the past century, and atmospheric concentrations have recently surpassed the safety limit of 350 ppm (https://www.CO2.earth), with potentially disastrous consequences to both humans and the environment. An effective route for alleviating this problem is to catalytically convert this abundant CO_2 to value-added chemicals, which will provide an effective pathway for CO_2 utilization and monetization. However, CO_2 is chemically inert in nature and can only be broken by high-energy protons and electrons. Therefore, its reduction is quite difficult and will require a complex electrocatalyst.[211] Numerous strategies for CO_2 reduction have been discussed in the literature, with various degrees of success and applicability.[212]

There is not much to say about CO_2 conversion on phosphorene-based catalysts because to date there is literally no study where the activity of phosphorene compounds toward carbon dioxide reduction has been investigated. Sometimes this means that this particular compound is not active (supposing that the research already had been done but all the results were negative, and therefore not publication-worthy). But not in this case. The thing is that phosphorene is only five years old and it gained almost all of the researchers' attention in regard to its application in electronic devices but not as a potential catalyst. This is why the field of CO_2 reduction might be unexplored and attempts have to be made. In recent years, the WSe_2 electrocatalyst has been utilized for the reduction of carbon dioxide to carbon monoxide fuel. However, due to the rapid electron transfer and less overpotential value, the TMDCs immerged as promising candidates for carbon dioxide reduction. Phosphorene is considered to be analogous to the WSe_2 electrocatalyst, due to the high electron transport and electrocatalytic activity of phosphorene they can be utilized for carbon dioxide reduction.[213] The carbon dioxide is converted into methane by using g-C_3N_4 and the red phosphorous hybrid material that shows a suitable energy band diagram under solar irradiation.[214]

In the past years, there has been a tremendous amount of work done for exploring selective catalysts for the reduction of carbon dioxide (CO_2) to carbon monoxide (CO). The transition metals and noble metals such as Au,[215] Ag,[216] Pd,[217] and Zn[218] can produce CO with high selectivity and current efficiency. The other metals that have been explored and studied for carbon dioxide reduction are Cu and Bi by adjusting certain compositions and electrolytes.[219] In recent years, the nitrogen-doped carbon nanomaterials and the transition metal-based single-atom catalysts have been some of the most promising candidates for carbon dioxide reduction.[220,221] Recently, electrochemical reduction of carbon dioxide has been done by utilizing the catalyst prepared by using electrospinning and thermal annealing method as a transition metal single atom is dispersed on the surface of a graphene shell. [222] Because graphene is analogous to phosphorene, in the future we can certainly utilize phosphorene as an alternate of graphene for carbon dioxide reduction.

6.4.1. Mechanism of Electrochemical Reduction of CO_2

The electrochemical reduction of carbon dioxide is a multistep reaction procedure. This process generally involves two, four, six, or eight-electron reaction pathways.[223] The reaction generally takes place at the electrode–electrolyte interface, as a heterogeneous electrocatalyst is involved in carbon dioxide reduction. Generally, the heterogeneous electrocatalyst procedure involves three major steps: 1) the adsorption of the carbon dioxide on the electrocatalyst, 2) transfer of an electron or migration of a proton to cleave the C—O bonds and also to form a C—H bond, and 3) the rearrangement of the configuration of products to desorbs them from the surface of the catalyst and further diffuse it into the electrolyte.[224] The electrode potential that is applied and the electrocatalyst attached have a very significant effect on the end reduction products. The products are mixtures of carbon compounds having different oxidation states such as methane (CH_4), ethylene (C_2H_4), ethanol (C_2H_5OH), methanol (CH_3OH), carbon monoxide (CO), and formate ion ($HCOO^-$) or formic acid (HCOOH).[225,226]

The thermodynamic point of view of the reduction of carbon dioxide suggests that the equilibrium potential of the carbon dioxide reduction is comparable to the HER reaction, as shown in reactions. The reaction conditions are pH-7 in aqueous solution versus standard hydrogen electrode (SHE) at 25^0C, 1 atm, and 1 M concentration. This coincides very well with the fact that the hydrogen (H_2) is the main side product. However, the potential difference value is very small for the carbon dioxide electroreduction end products; this might be the reason why it is difficult to reduce carbon dioxide to the required product having great selectivity. The real electrode potential that is needed to drive the reaction is more negative than the equilibrium ones.[227]

$$CO_2 + 2H^+ + 2e^- \rightarrow CO + H_2O \quad E^o = -0.52\,V \tag{1}$$

$$CO_2 + 2H^+ + 2e^- \rightarrow HCOOH \quad E^o = -0.61\,V \tag{2}$$

$$CO_2 + 4H^+ + 4e^- \rightarrow HCHO + H_2O \quad E^o = -0.51\,V \tag{3}$$

$$CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \quad E^o = -0.24\,V \tag{4}$$

$$2CO_2 + 12H^+ + 12e^- \rightarrow C_2H_4 + 4H_2O \quad E^o = -0.34\,V \tag{5}$$

$$CO_2 + 6H^+ + 6e^- \rightarrow CH_3OH + H_2O \quad E^o = -0.38\,V \tag{6}$$

$$2H^+ + 2e^- \rightarrow H_2 \quad E^o = -0.42\,V \tag{7}$$

$$CO_2 + e^- \rightarrow CO \quad E^o = -1.9\,V \tag{8}$$

6.5. Nitrogen Electroreduction Reaction (N_2RR)

Ammonia, including its derivative urea, is a widely produced chemical that represents the basis of most fertilizers. The nitrogen-to-ammonia conversion is one of the most important and challenging processes in chemistry.[228] Currently, ammonia is derived from fossil fuels, obtaining the hydrogen precursor from natural gas, which creates concerns on environmental safety. Thus, an urgent need is there to develop sustainable approaches to its production. The electrochemical nitrogen reduction reaction (NRR) is currently being intensely investigated as the basis for future mass production of ammonia from renewables, a synthesis from molecular nitrogen and hydrogen itself. As stated by Nørskov and his team,[229] the development of an efficient, realistic machinery for electrochemical N_2 conversion could provide an alternative route to NH_3 on a distributed basis, avoiding the key factors that require the Haber–Bosch process, which involves intense conditions of temperature and pressure, combined with a considerable, complex plant infrastructure. Efficient NRR has proven extremely challenging to achieve in practice, underlining the fact that nitrogen is a highly stable and nonpolarizable molecule. This typically shifts the potentials needed to reduce nitrogen to the region where proton reduction to hydrogen becomes facile as a competing reaction.[230] Multiple strategies have been explored to minimize the occurrence of the HER, both from a catalyst and an electrolyte perspective.[231] Nonetheless, selectivity remains quite low in aqueous solution–based NRR, typically not more than 15%. In this section, we will discuss the catalyst design based on black phosphorene that may help guide the search for an electrocatalyst that can achieve selective nitrogen.

The search for an electrocatalytic NRR center in recent years has mainly focused on transition metal–based materials, which is strongly influenced by the understanding of nitrogenase enzymes, focusing on the metal complex centers of these enzymes, namely, Fe–Mo, Fe–Fe, and Fe–V. Around 30% of the electrocatalysts reported for the NRR under ambient conditions are based on Fe and Mo. It is worthwhile to note that for an excellent NRR catalyst, it requires an unoccupied nonbonding orbital and electron donor site with abundant electron cloud density; however, the d-orbital electrons in transition metals also benefit the formation of metal–hydrogen bonds, which will intensify the competitive HER, thus restricting the nitrogen reduction selectivity and catalytic efficiency. Compared to transition metals, the weak hydrogen adsorption of nonmetallic elements and their abundant valence electrons should provide a more ideal nitrogen activation center to obtain higher activity toward ammonia production, as reported by Wang and coworkers,[232] who utilized multiple nanosheet orthorhombic BP as the electrocatalyst for nitrogen reduction under ambient conditions. The BP metal-free single element emerges as an interesting electrocatalyst in this reaction due to its valence electron structure, which is very similar to that of nitrogen ($3s^23p^3$and $2s^22p^3$, respectively) with the in-plane anisotropic lattice structure that allows this material to be exfoliated into monolayer phosphorene or few-layer BP via Van der Waals interaction, thus offering the maximum number of intrinsic active sites possible for nitrogen activation via the chemisorption process. A layered Van der Waals material such as BP is characterized by inequivalent high-symmetry direction structure, notably the zig zag and armchairs, which is attributed to the enhancement of the electrical conductivities,[233] which is crucial to providing enough electrons for activation of the inert nitrogen molecule triple bond. As reported in the same work by Wang and coworkers,[232] few-layer BP nanosheets have a faradaic efficiency and ammonia production rate for the NRR in an acidic aqueous solution of 5.07% and 31.37 $mg\,h^{-1}\,mg^{-1}$, respectively.

7. Future Perspectives

Phosphorene and phosphorene-based materials have various potential applications in the fields of energy and biology. BP bulk, BP nanosheets, and BP quantum dots were utilized in biomedicine for drug delivery, cancer therapy, electrochemical sensors, fluorescent sensors, colorimetric sensors, transistors, and cancer imaging.[215] Also, other potential applications have been discovered for phosphorene-based materials in the fields of thermoelectric power, photovoltaic cells, nanoenergy generators, rechargeable batteries, solar fuel production, solar water steam, supercapacitors, and the oxygen reduction reaction.[234–259] In **Figure 15** and 16, the potential applications of BP and phosphorene in biomedical and energy applications are shown.

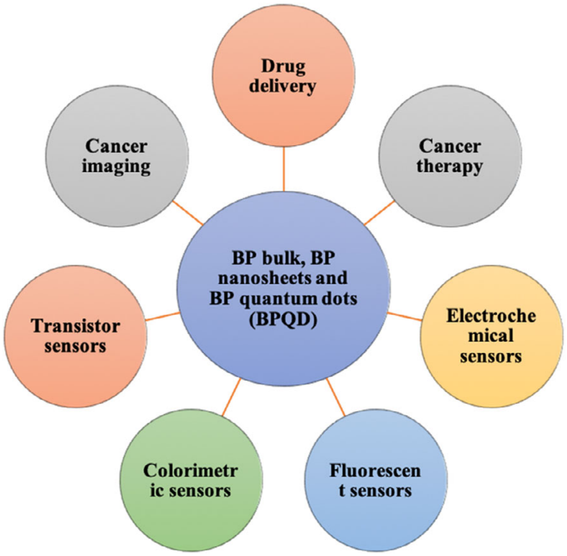

Figure 15. The potential applications of BP 0and phosphorene in the biomedical field.

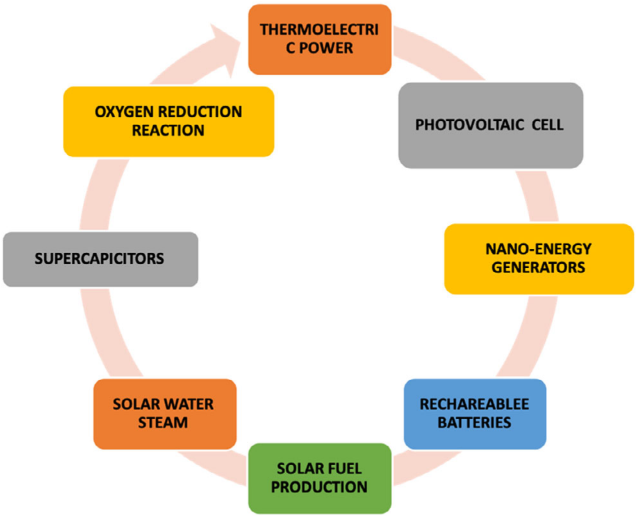

Figure 16. The potential applications of BP and phosphorene in energy production.

8. Conclusion

The rapidly growing studies on phosphorene have ignited a great interest to investigate the characteristics and potential applications of 2D materials. The free dangling bonds and van der Waals interlayer forces make 2D BP suitable for forming heterostructures with other 2D materials. Moreover, the good electronic properties, in-plane anisotropy feature, and tunable bandgaps in a wide range enable 2D phosphorene, which can work as a highly efficient electro/photocatalyst. To the best of our knowledge, this is the first review on electrochemical reduction of CO_2 and water splitting on 2D phosphorene–based catalysts. In this review, we have briefly summarized several key aspects dealing with the chemistry and application of bulk and exfoliated BP, including an overview of synthesis, to obtain 2D phosphorene-based catalysts for water splitting and CO_2 reduction. This review focuses on the progress in the synthetic strategies and electrocatalytic applications of phosphorene-based materials for the HER, OER, hydrogen storage, CO_2RR and N_2RR applications. This article will provide vast knowledge to researchers on the phosphorene materials for CO_2RR and water splitting applications.

Acknowledgements

R.S. would like to thank PDPU, India, for providing the required infrastructure and start-up research grant. This research work was funded by SERB-DST, New Delhi, under the scheme "ASEAN-India Research & Development" (Grant No.: IMRC/AISTDF/CRD/2018/000048).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D materials, black phosphorus, electrocatalysts, energy production, hydrogen evolution reaction, oxygen evolution reaction, phosphorene

Received: August 20, 2020
Revised: October 6, 2020

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