Transition Metal Dichalcogenides in Photocatalytic Hydrogen Evolution

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Transition metal dichalcogenides (TMDCs) is an emerging class of two-dimensional (2-D) materials that have exhibited great potential in photocatalytic applications owing to their intrinsic properties. They are normally used as cocatalysts together with other semiconductor materials and the junction created between them facilitates charge transfer of the photogenerated electrons and holes. The photocatalytic H_{2-}evolution systems can be enhanced by lengthening the parts of uncovered surface of photocatalyst and delaying the recombination of electron-hole pairs.

transition metal dichalcogenides water splitting photocatalysis co-catalyst

1. Introduction

Society at large is prospecting for carbon-free fuel alternatives to purvey an unfailing panacea to the present energy plight of the world. Research efforts into the development of alternate clean and renewable energy sources have been at a steady rise in the last few decades. Hydrogen is credited to be an unpolluted, robust, environmentally safe, and emerging fuel, efficient to curb total reliance on petroleum-based fuels. Currently, methane reforming accounts for the largest production of hydrogen fuel, where methane gas is converted to carbon monoxide and hydrogen. This process is energy inefficient and has a negative environmental impact. Consequently, it is desirable generating hydrogen through unexhausted resources such as water and solar power. Solar light-assisted water decomposition is an emerging method for sustainable hydrogen production. Producing hydrogen through solar simulated light, water and photocatalyst is a desired technology not only because water and solar energy are abundant on the earth but also because it is environmentally friendly and economically viable. Hydrogen is attributed to have the maximum energy content per weight in comparison to other combustion fuels, the by-products are environmentally friendly which are water vapour and [1][2][3][4].

To a consumer, similar to natural gas serves many purposes, hydrogen gas could be used to heat homes and as a fuel for automobiles. Environmentally, hydrogen is exceptional among other fuels, being a carbon-free fuel, it will generate zero carbon dioxide during combustion. To the scientists, hydrogen is the most simplified and most abundant element in the world. The hydrogen atom (H) is made of one electron and one proton but as a molecule (H_2), it has a favourable physical feature. In terms of energy, it is the highest carrier of energy, having 2.4 folds higher energy content than that of natural gas. This can be put to use using fuel cell to generate electricity or by combustion to produce heat for homes use. It is also a carbon neutral energy carrier. With many countries till date still dependent on energy from fossil fuel sources which is a major source of environmental pollution, there is a dire need for cleaner energy to meet CO_2 emission reduction targets. The Asian countries are the major players in realising the hydrogen economy. Commitment is a vital key to attain the regulatory CO_2 emission targets, as it is very costly to achieve. Japan has keyed into the Paris climate agreement of 2016 and has made much commitment to be a large-scale hydrogen user for energy. Most countries in Europe have signed an agreement based on the European Green Deal to ensure zero-emissions for cars and vans and to achieve zero net emissions of greenhouse gases in Europe by 2050 [1][5]. Little work has been carried out or reported on hydrogen economy in Africa. In South Africa, the telecommunication industry is already using hydrogen fuel. This is used as support system power for petrol or diesel generators in military, hospitals and mines in case of intermittent power supply.

With water and sunlight being free and abundant in nature, this method of generating hydrogen gas is beneficial, environmentally safe, and renewable avenue which could serve as valuable resource to tackle both the energy plight and environmental pollution. Photocatalytic processes have been considered as one of the most promising methods to split water into hydrogen and oxygen. For an effective photocatalytic process, there is a need to use a sturdy and very active photocatalyst to produce good yields of hydrogen gas. Since Fujishima and Honda [6] used TiO_2 electrodes for photocatalytic water splitting, a large array of other photocatalysts have been studied for solar driven water splitting; for instance, CdS, CuS, C_3N_4, ZnO, $BiVO_4$, MoS_2 [7][8][9][10][11][12][13]. So far, numerous semiconductor photocatalysts are being explored and used for photocatalytic water splitting. Photocatalysts can be subdivided into three categories namely: metal oxides, metal chalcogenides, and metal–free photocatalysts. Despite the progress made so far on these photocatalysts, they still have some deficiencies. The metal oxide makes use of ultraviolet region of the spectrum because of their large band gap energy. Some semiconductors favour either water reduction or oxidation activity, hence they are not appropriate for effective water splitting. These semiconductors undergo quick charge co-existence of the electron-hole pair resulting in inefficient charge separation and finally, the active sites must not be covered, but in bulk they are not available to partake in photocatalytic processs [7][14][15].

Transition metal dichalcogenides (TMDCs) is an emerging class of two-dimensional (2-D) materials that have exhibited great potential in photocatalytic applications owing to their intrinsic properties. They are normally used as cocatalysts together with other semiconductor materials and the junction created between them facilitates charge transfer of the photogenerated electrons and holes. TMDCs have the general formula MX_2, where M is a transition metal (Mo, W, Ti, Zr, Hf, Nb, etc.) and X is a chalcogen element (S, Se, or Te). While TMDCs of groups V and VI elements have been extensively studied for applications in photocatalysis, in the open literature, not much information is currently available on the group IV TMDCs ZrS_2 and HfS_2 especially synthesized by colloidal methods as these are very unstable and are easily oxidized in mono or few-layers.

2. Application of Transition Metal Dichalcogenides in Photocatalytic Hydrogen Evolution

2D layered transition metal dichalcogenides have received an enormous amount of attention for their magnificent catalytic activities, narrow band gap, crystallinity, excellent properties and their use in catalysis for production of hydrogen gas [7][14][16][17][18]. They are being sought for in recent time as a result of their low cost, earth abundance and excellent catalytic activities as a possible replacement for scarce and expensive noble metals [7][14][17][18]. The transformation that occurred in the material from 3D to 2D paved the way for their novel electronic and mechanical properties. There are many active sites in 2D chalcogenides which participate during catalytic reaction and the possibility of harnessing solar energy by the atoms of the semiconductor. The chalcogenides possess right and adjustable band gap depending on the number of layers they possess. To illustrate this, the band gap of ZrS_2 may be increased from 1.5 to 2.0 eV if the bulk ZrS_2 is exfoliated to single layer of ZrS_2, hence, there is changes in the band gap energy as it moved from an indirect to a direct band gap by reason of quantum confinement [18]. A lot of these 2D chalcogenides have been proven to be good photocatalysts for water splitting, including $MoSe_2$, MoS_2, WS_2, NiS, NiSe, Wse_2, SnS_2, and ReS_2 [11][16][17][19][20][21]. Some other semiconductors, the likes of HfS_2, ZrS_2, $TcSe_2$, and TaS_2 are being anticipated as photocatalysts to be considered for decomposition of water [14][22][23].

Composite photocatalysts have been used to improve photocatalytic processes by reason of their excellent configuration. To enhance the properties of these, TMDCs are integrated with other photocatalysts such as TiO_2, ZnO, CdS, CuS, g-C_3N_4 etc. There must be an intimate interface between the semiconductor and the co-catalyst which will accelerate electron transfer within the interface of the semiconductor to the co-catalyst. The design and assembly of an exclusive junction between the photocatalyst and the co-catalyst plays a vital role for enhanced photocatalytic performance [14][24]. There are reports on the utilization of MoS_2 as co-catalyst by several researchers to greatly enhance the activity of photocatalysts. **Table 1** shows a summary of composite photocatalysts that have been employed in hydrogen evolution reactions. Ma et al. [25] constructed a CdS-MoS_2 hybrid catalyst for photocatalytic production under visible light, by ultrasonication treatment. The presence of MoS_2 greatly boosted the production of hydrogen which was about two times greater than when CdS alone was used. However, due to weak interface between them, the photocatalytic activity decreased (about 35%) after four cycles which was attributed to ease of separation of MoS_2 from CdS. CdS is a well-known photocatalyst, but it has the drawbacks of photo corrosion and great toxicity of Cd to the environment. Chang et al. [26] demonstrated that the number of layers of TMDCs played a key role in a catalytic reaction. In their study for photocatalytic hydrogen evolution reaction using MoS_2 loaded on CdS in the presence of NaS-Na_2SO_3 and lactic acid as sacrificial agents, the yield of hydrogen with lactic acid was higher than with NaS-Na_2SO_3, however the catalyst was more stable in Na_2S-Na_2SO_3 compared to lactic acid. This was expected since lactic acid solution contains abundant H^+ to Na_2S-Na_2SO_3. The researchers do not report on the number of cycles conducted on the catalyst to determine its stability. They revealed that mono, or few layer(s) of MoS_2 produced maximum generation of hydrogen compared to higher numbers (7–122) of layers. The yield of monolayer was the optimum generating 0.00259 $mmolh^{-1}$ and 0.00201 $mmolh^{-1}$ H_2, respectively, and this was associated to the following 3 factors: (a) the mono layer displayed a more negative conduction band minimum to H^+/H_2 potential (b) the edge sites are very pivotal to the hydrogen production and the mono-layer has plethora exposed active sites arising from the unsaturated (S) atom to the other layers (bulk) and (c) delayed rate of recombination of photogenerated carriers was observed in mono-layer compared to bulk which was attributed to good interface contact between mono MoS_2 and CdS. The yield of MoS_2/CdS was 5.89 times higher than Pt/Cds conducted in lactic acid. However, there is possibility of carbon monoxide generation which may lead to Pt catalyst poisoning, thus a reduced production using Pt/Cds in lactic solution. Very recently, Zong et al. [27] fabricated CdS-MoS_2 by conventional impregnation method. The H_2-evolution rate was 36 times higher when 0.2 wt% MoS_2 was loaded on CdS. The enhanced photocatalytic activity was attributed to the tight junction formed between MoS_2 and CdS and the excellent H_2 activation property of MoS_2. In similar work, Zong et al. [28] also fabricated CdS-WS_2 by impregnation method. The loading of 1% weight of WS_2 increased hydrogen evolution rates by 28 times attributed to the interface formed between CdS and WS_2 and the excellent performance of WS_2 as a co-catalyst in catalysing H_2 evolution. CdS is a well-known photocatalyst, but it has the drawbacks of photo corrosion and great toxicity of Cd to the environment. In a recent work, Reddy et al. [29] accounted for the outstanding HER activity of a nanohybrid which was reported to be highest using a ternary catalyst compared to the one available in literature. The group constructed heterostructures by loading few-layered ultrathin MoS_2-WS_2 on CdS and applied the catalyst for photocatalytic water splitting using lactic acid as holes acceptor. The HER performance of 209.79 mmolg^{-1} h^{-1} was obtained which was 1.70 folds greater than Cds-MoS (123.31 mmolh^{-1}), 1.70 folds higher than Cds-WS_2 (169.82 mmolh^{-1}), 6 folds higher than CdS-Pt and 83 times higher than bare CdS (2.54 mmolh^{-1}). The 6 wt% MoS_2-WS_2 loading was the most effective which showed robust activity, durability, and stability even up to 2.5 days. The high-level activity was attributed to the following reasons: (i) more actives sites were available by MoS_2-WS_2 cocatalyst in contrast with a single cocatalyst (ii) proper band gap of the nanohybrid (iii) tight heterojunction between the catalyst and cocatalyst which promoted ultrafast electron transfer to the cocatalyst, as well tardy re-joining of the photogenerated carriers. Swain et al. [30] synthesized MoS_2 nanoflowers and used them to decorate $CaIn_2S_4$ microflowers using double step hydrothermal method. The experiment showed that hybrid catalyst generated higher H_2 production (19 times) compared when the $CaIn_2S_4$ was used in a visible light irradiation. The catalyst showed a band gap of 2.11 eV which enabled the reaction to be conducted in visible region of light compared to UV-visible light. The MoS_2 provided more edge sites having plethora of unsaturated sulphur ions which facilitate quick capture of the H^+ ions at the interface of the p-n heterojunction of the catalyst. This activity enhancement was as a result of adequate prevention of quick reoccurrence of electron-hole pair at interface of the hybrid catalyst. This work showed that MoS_2 is a possible replacement to other promoter on a semiconductor catalyst with just 0.50% loading. With increasing loading as low as 1% the output of H_2 production declined due to the black colour of MoS_2 which prevented the transfer of photons. The catalyst also generated H_2 even in the absence of sacrificial agent though with a lower yield. Stability study showed the catalyst to be stable even after 4 cycles. The rate of H_2 production generated in the research was a great improvement over previous studies on $CaIn_2S_4$ based composite for photocatalytic water splitting as the yield is far higher compared to when other catalysts that were loaded on the semiconductor photocatalyst. Nguyen et al. [31] decorated $Zn_xCd_{1-x}S$ with MoS_2 via a photo-assisted deposition. The 3% decoration of $Zn_{0.2}Cd_{0.8}S/MoS_2$ gave H_2 evolution 210 times higher than the undecorated photocatalyst. Huang et al. [32] constructed 2D/2D $ZnIn_2S_4/MoS_2$ nanohybrid using electrostatic self-assembly method. Generally, due to the possibility of shading effect as a result of excess loading of the co-catalyst on the catalyst, low loadings of MoS_2 are required. The 0.75% loading gave the maximum hydrogen yield which was almost 50 times higher than the raw $ZnInS_4$. Only traces (0.099 mmol h^{-1}) of hydrogen were generated when pristine $ZnIn_2S_2$ was used for the photocatalytic reaction. However, when MoS_2 was loaded on it, the hydrogen yield increased linearly with increase amount of the promoter until 0.75% after which the yield deceases due to the shading effect. The result was also compared to 0.75% $Pt/ZnI_2S_{4.}$ The production of H_2 was 2.2 lesser with noble metal as a promoter. The reaction was visible light-driven possibly due to its band gap that is proper for visible light, however the researchers did not report on the adsorption edges or band gap of any of the catalyst used. The structure of a catalyst determines its efficiency in a photocatalytic reaction. The high yield could be as a result of the ultrathin structure of both MoS_2 nanosheet and $ZnIn_2S_4$ nanosheet, and the robust and close contact interface in the hetero layered enhanced the photocatalytic hydrogen generation. The fabrication of ultra-thin 2D/2D structure performs better compared to 0D/2D and 1D/2D counterparts. The catalyst was reported to be stable until fourth cycle of use in which a decrease in the activity was noticed. Pudkon et al [33] synthesized $ZnIn_2S_4$ using different sulphur sources and loaded it on WS_2 using microwave method of synthesis. The L-cysteine not just being a sulphur source but played the dual purpose as a reducing agent and as capping agent and helped in growth direction for the formation of flower-like nanomaterials. The HER activity of the hierarchical (flower-like) catalyst (145.3 $\mu molh^{-1}$) was far more than the non-hierarchical (81.6 $\mu molh^{-1}$) catalyst. The former provides diverse reflections of the incident light via its shape thus a lengthy lifetime of the incident light in its structures. This can be credited to the surface area as well, surface area is an important factor in a catalytic reactions, the higher the surface the more active site will be available for reactions to take place. The nanomaterial with flower-like morphology had a surface area 1.6 folds higher than non 3D nanomaterials fabricated. The 40%wt loading of WS_2 on flower-like catalyst was most suitable for HER with activity 2 times higher (293.3 $\mu molh^{-1}$) than the pure $ZnIn_2S_4$ and 6.67 folds than raw WS_2. The close contact contributed to good separation of charge and injection at the interface of the catalysts whereas the composite formed by mere grinding of the two catalysts gave activity of (101.3 $\mu molh^{-1}$) which was even less than activity of untreated flower-like $ZnIn_2S_4$ alone. Both $ZnInS_4$ and WS_2 are photocatalysts that absorb within the visible region of solar spectrum. The researchers did not report on the band gap of the composite however, the band edge obtained from the UV-absorption showed a slight shift to higher wavelength which is an indication of a better response of the composite to visible light. However, in their report the composite catalyst activity for H_2-evolution was more favoured in UV-visible compared to visible region up to about four folds. This may be accounted for by the band gap of the semiconductor synthesized; it had a band gap of 2.81 eV, close to that of a material that absorbs in the UV region, and the promoter did not significantly improve the response of the composite to visible light. The output of hydrogen obtained was slightly lower than previously reported on $ZnInS_4$ based heterostructure demonstrated for H_2 production. Photocatalysts that absorb in the visible region are required for visible light driven photocatalysis and they have their band gap very close to 2.0 eV [34]. Zeng et al. [35] in a manner similar to constructed metal-sulphide -metal-selenide hybrid ($ZnIn_2S_4$-$MoSe_{2)}$ system for HER using a one-pot polyol method. The synthesized $ZnIn_2S_2$ had hierarchical structures which contributed to the increased HER activity of the untreated catalyst of 1023 $\mu molh^{-1}$ in contrast with previous work in which pristine $ZnIn_2S_4$ was used for photocatalytic water splitting. The 2 wt% loading on the hierarchical structure $ZnIn_2S_4$ improved the activity by a multiple of 2.2 times. The stability test on the catalyst showed good stability even after five cycles. The result obtained with $MoSe_2$ as cocatalyst on a ternary chalcogenide is very comparable to results from previous researchers with MoS_2 loaded on the same material. This is an indication the activity of $MoSe_2$ as a promoter is as well good as MoS_2 in photocatalytic water splitting.

A lot of work has also been reported on the improvement of the photocatalytic activity of TiO_2 with TMDCs. Li et al. [36] synthesized MoS_2/TiO_2 hybrid by hydrothermal treatment and found the MoS_2/TiO_2 hydrogen evolution yield was 2.19 and 3.15 times higher than TiO_2 and MoS_2, respectively. The yield was still good after 4 cycles of use, this as a result of close interaction between the co-catalyst and the photocatalyst. Recently, Zhang et al. [37]

fabricated novel mesoporous anatase titania with high photoconductivity and photocatalytic attributes compared to nanocrystal TiO_2 such as P25 using hydrothermal method. MoS_2 nanosheets was loaded on the 3D TiO_2 using same hydrothermal process and evaluated for hydrogen evolution reaction under UV light source for irradiation. The composite showed higher photocatalytic activity for hydrogen evolution compared to undecorated TiO_2 and MoS_2. The mesocrystal of the TiO_2-MoS_2 composite produced an H_2-evolution rate of 0.55 mmolh^{-1} which was 4 times greater than MoS_2-TiO_2 (P25) hybrid. The performance of MoS_2 as a promoter on the semiconductor in work is an indication of possible replacement over precious metal such as Pt and Pd. The 1% MoS_2 loading resulted in 200 times higher H_2 evolution compared to the bare mesocrystal TiO_2. However, previous reports showed that a maximum loading of about 2% MoS_2 on semiconductor resulted in enhanced photocatalytic water splitting contradict the report in this work where up to 10% of MoS_2 was the optimal loading for high hydrogen production. The main factor was identified as annealing process. Annealing the catalyst at about 160 °C greatly improved its performance towards HER since phases of TMDCs contributes immensely to the catalyst activity towards HER. The annealing caused some phase transition of the MoS_2 from metallic phase to mostly hexagonal phase. Moreover, at annealing temperatures exceeding 240 °C the activity of the composite declined possibly due to the sulphur active sites being oxidized or loss of the metallic phase. The catalyst also showed good stability after five test cycles. In another report, Zhou et al. [38] fabricated MoS_2-TiO_2 heterostructures by hydrothermal method. MoS_2 loaded on TiO_2 nanobelts and the resulting photocatalyst exhibited outstanding H_2-evolution to a value of 1600 µmolh^{-1} compared with pure TiO_2.

Dong et al. [39] also made TiO_{2}-MoS_2 heterojunction via annealing impregnation deposition method. The hybrid generated H_2 evolution with activity 7.2 times and 17.6 higher than the bare TiO_2 and MoS_2, respectively, ascribed to that the heterojunction between TiO_2 and MoS_2 quantum dots which could suppress the recombination of electron-hole pairs efficiently and that the hollow TiO_2 nanospheres and MoS_2 quantum dots with high specific surface area and pore diameter could supply plenty of active sites. Moreover, the researchers did not experiment on different loading to determine the optimal loading and the percent of co-catalyst used was not reported and more so, the recyclability test was not reported. Feng et al. [40] fabricated quaternary nanocomposite of Mn-Cds-MoS_2-TiO_2 by hydrothermal method. The quaternary nanocomposite gave outstanding H_2-evolution up to a level of 408.27 mmolh^{-1} which was 30.08 folds that of TiO_2, 5.18 folds of TiO_2-MoS_2 and 2.52 folds for CdS-Mn-TiO_2, respectively. The photocatalyst performance was greatly improved which was attributed mainly to the synergetic effects of CdS-Mn, MoS_2 and TiO_2, forming a Z-scheme system in the CdS-Mn/MoS_2/TiO_2 electrode, which not only accelerates the interfacial charge transfer efficiency but also preserves the strong redox ability of the photogenerated electrons and holes. This is one of the highest activities reported so far for a catalyst for photocatalytic water decomposition. MoS_2 has also been used to improve the photocatalytic properties of ZnO. Yuan et al. [41] loaded MoS_2 on ZnO via hydrothermal route. The loading of MoS_2 as cocatalyst on ZnO elevated its H_2-evolution rate up to 14.8 folds than the pristine ZnO. The optimum loading of 1% MoS_2/ZnO generated 768 µmolh^{-1} of H_2. The activity enhancement was due to the suppression of the recombination of electron hole pairs of ZnO. Li et al. [42] constructed the nanohybrid of MoS_2-ZnO by dispersion method. The deposition of MoS_2 enhanced the hydrogen evolution 5 times compared to pure ZnO because of the production of more electrons and holes and reducing their recombination. The hydrogen output activity was 27.69 mmolh^{-1}. The method used to prepare to prepare the nanostructure did not produce a thorough interface between the two catalysts thus a reduced activity was noticed, and the recyclability of this catalyst was very difficult. Very recently similar research was conducted by Chang et al. [43] in which they constructed p-n MoS_2-ZnO heterostructures with large surface area using hydrothermal method. The nanohybrid with 0.03 g of MoS_2 yielded the highest activity (145.6 μmolh^{-1}) after which the performance declined with further loading. However, the activity reported was 5.3 times less than what Yuan et al. obtained [41]. Chang's group [44] also investigated the photocatalytic water splitting activity of MoS_2-SnO_2 core-shell sub microsphere using Na_2S as hole collector, the yield was slightly lower when compared to MoS_2-ZnO (117.2 μmolh^{-1}). Oxides of bismuth are good semiconductors which find application in photocatalytic processes. Khalid et al. [45] constructed nano composite of Bi_2O_3-MoS_2 by hydrothermal method with varying composition from 0–15 wt% of MoS_2. The highest HER was obtained with 11 wt% with activity (10 μmolh^{-1}) which was ten folds higher than raw Bi_2O_3 and pure MoS_2. The ability of the catalyst to absorb much visible light and delayed recombination of electrons and holes led to higher production of H_2.

The use of CeO_2 has not been explored in photocatalytic water splitting until Swain et al. [46] successfully constructed MoS_2-CeO_2 using hydrothermal approach. The best output was when 2 wt% of the promoter was deposited on CeO_2 to form p-n junction with large intimate and close junction with performance of 508.44 μmolh^{-1} which is far higher (57 times) than unloaded CeO_2. The catalyst still showed good stability even after 3 cycles.

Hollow graphitic carbon nitride (g-C_3N_4) has been a photocatalyst widely used in hydrogen evolution. Both g-C_3N_4 and MoS_2 have comparable layered structures which should help to reduce the lattice mismatch and ease the planar growth of MoS_2 layers on the g-C_3N_4 surface. Hou et al. [47] loaded MoS_2 on mesoporous g-C_3N_4 by impregnation method followed by sulfurization. The nanohybrid played a vital role in boosting the photocatalytic activity to 310 μmolh^{-1} with 0.5 wt% compared to when pristine g-C_3N_4 (108 μmolh^{-1}) and untreated MoS_2 (0.25 μmolh^{-1}) were used where the activities were much lower. The group [47] in a similar reaction also formed heterostructure of WS_2-C_3N_4 using impregnation system followed by sulfurization. The 3 wt% loading gave the highest activity (20.60. μmolh^{-1}) while with unloaded C_3N_4 the activity was very minimal. In a related research, Ge et al. [48] synthesized composite of MoS_2-g-C_3N_4 by impregnation route. The 0.5 wt% MoS_2-g-C_3N_4 gave the maximum catalytic activity with H_2 evolution of 23.10 μmolh^{-1} which was 11.3 times higher to unwrapped g-C_3N_4. Wang et al. [49] performed density functional theory calculations on hybrid g-C_3N_{4}-MoS_2 and found that photo-generated electrons migrate easily from the g-C_3N_4 monolayer to the MoS_2 sheet leading to a high hydrogen evolution activity of the hybrid over g-C_3N_4 or MoS_2 singly. Recently, Liu et al. [50] synthesized 3D/2D nanojunction of flower-like MoS_2 on graphitic carbon nitride using ultrasonication and thermal treatment method. The optimum loading of 5 wt% of MoS_2 was also used by this group to generate 867.6 μmolh^{-1} of hydrogen which exceeded that of untreated g-C_3N_4 by 2.80 times. The activity observed with 3D MoS_2 and g-C_3N_4 and was reported to be higher than other dimensions of MoS_2 composite with g-C_3N_4. The enhanced HER activity was attributed to tight interface between the catalysts which facilitated higher light harnessing, quicker bond charge separation, faster electron transport and the increased electrical conductivity of the materials. The catalysts also showed good electrocatalytic HER activity with reduced overpotential and high current densities. Zhang et al. [51] used hydrothermal method to prepare heterostructure of nitrogen doped carbon tube molybdenum disulphide (TNCT@MoS_2) for photocatalytic water splitting in the absence of electron donor. The microcomposite displayed increased performance with activity of 120 µmolh^{-1} which was several times greater than untreated MoS_2. The generation of H_2 without the use of a sacrificial agent which plays the function of acceptance of the hole suggested the micro-heterostructure catalyst to be effective in water splitting. The achieved result was attributed to higher absorption of visible light, creation of more active sites, possibly the power of the xenon lamp used (1000 W); the maximum of power of xenon lamp that was reported in literature was 400 W, as well as the nitrogen dopant which could help in fast electron transfer to the active sites of the MoS_2. The group also observed similar activity when the pH of the water used was varied from 5–11. The gas chromatography used could not detect any O_2 generated during the reaction which was later probed by fluorescence method in which the intermediate OH was picked up. The O_2 produced during reaction was reported to be absorbed by the metal of the cocatalyst to form a peroxide complex.

Gupta and Rao [52] highlighted the use of dye with TMDCs in photocatalytic water splitting. The dyes have capacity to absorb light. Common dyes that are used are noble-metal-centred and metal-free dyes. Noble metal dyes that are often used are Ru(ii) tris bipyridine ($Ru(bpy)_2^{3+}$) and hydrated iridium oxide ($IrO_2.nH_2O$). The metal free dyes such as Eosin Y or Rose Bengal are commonly used. The noble metal-centred dyes are being reported to be more effective in generation of hydrogen over the metal free dyes because they give more photocurrent upon irradiation with visible light over the metal free dye. They play a dual role as oxidizing catalyst and photosensitizer [53]. Upon excitation it undergoes some transformation to inject an electron to TMDC (MoS_2). In 2009 Zong et al. [54] conducted photocatalytic hydrogen generation in visible light using composite of dye Ru(ii) tris bipyridine ($Ru(bpy)_2^{3+}$) which play the role of an organic photosensitizer and colloidal MoS_2 in presence of ascorbic acid. Upon irradiation in a visible light source, the dye transferred electrons to MoS_2 which has many active sites, which in turn reduce the protons to H_2(g). It was observed an increase in the concentration of the dye used led to a corresponding increase in the amount of H_2(g) formed. It was also noted the amount of the H_2(g) produced increased with temperature employed in the synthesis of colloidal MoS_2 using solvothermal method. When the reaction was performed in a mixture of acetonitrile and methanol solution while ascorbic acid serves as the sacrificial agent, the hybrid catalyst gave rise to 210 mmolh^{-1} of hydrogen gas, but this got diminished after some hours due to the decomposition of the organic photosensitizer dye. The use of the dye has the drawback of photo-instability.

Other dyes that have been employed as organic photosensitizer is the metal–free dye such as Eosin Y and Rose Bengal. Min and Lu et al. [55] synthesized nano composite of MoS_2 and graphene {G} using hydrothermal method. The nano composite was sensitized by Eosin y dye during photocatalytic reaction using triethanolamine (TEOA) as sacrificial agent, showing an activity of 83.8 µmolh^{-1}. However, when EY-RGO was tested the activity was very low < 0.5 µmolh^{-1} and activity of 37.7 µmolh^{-1} with MoS_2/RGO was obtained.

Table 1. Role of TMDC as a good promoter of catalysts in hydrogen evolution reaction.

Catalyst	Synthesis Method	Sacrificial Agent	Light Source	Activity	Ref.
MoS_2-TiO_2	Hydrothermal	0.35 M Na_2SO_3 and Na_2S	Xe Lamp 300 W	1600 µmolh^{-1}	[38]

Catalyst	Synthesis Method	Sacrificial Agent	Light Source	Activity	Ref.
MoS_2-$CaIn_2S_4$	Two-step hydrothermal	0.025M Na_2SO_3 and Na_2S	Xe Lamp 150 W	602 $\mu molh^{-1}$	[30]
MoS_2-CdS	Sonication and stirring	Na_2SO_3 and Na_2S	Xe Lamp 300 W	1750 $\mu molh^{-1}$	[25]
MoS_2-CdS	Impregnation	Methanol and 10% Lactic acid	Xe Lamp 300 W	532.8 $\mu molh^{-1}$	[26]
MoS_2-CdS	Centrifugation	Lactic acid	Xe Lamp 300 W	259 $\mu molh^{-1}$	[27]
WS_2-CdS	Impregnation	10% Lactic acid	Xe Lamp 300 W	420 $\mu molh^{-1}$	[28]
CdS-MoS_2-WS_2	Hydrothermal	10% Lactic acid	-	209.790 $\mu molh^{-1}$	[29]
MoS_2-$Zn_{0.2}Cd_{0.8}S$	Photo deposition	0.25 M Na_2SO_3 and 0.25 Na_2S	Xe Lamp 300 W	2 $\mu molh^{-1}$	[31]
MoS_2-$ZnIn_2S_4$	Electrostatic self-assembly	Lactic acid	Xe lamp	4974 $\mu molh^{-1}$	[32]
WS_2-$ZnIn_2S_4$	Micro wave	Na_2SO_3 and Na_2S	Xe lamp 150 W	293.3 $\mu molh^{-1}$	[33]
$MoSe_2$-$ZnInS_4$	One polyol	0.35 M Na_2SO_3 and 0.25 Na_2S	Xe Lamp 300 W	2228 $\mu molh^{-1}$	[35]
MoS_2-TiO_2	Hydrothermal	Methanol	Xe Lamp 350 W	75 $\mu molh^{-1}$	[36]
MoS_2-TiO_2	Hydrothermal	10% Lactic acid	-	550 $\mu molh^{-1}$	[37]
MoS_2-TiO_2	Annealing and impregnation	Triethanolamine	Xe Lamp 350 W	391.1 $\mu molh^{-1}$	[39]
Mn-CdS-MoS_2-TiO_2	Hydrothermal	Methanol	Xe Lamp 300 W	408.370 $\mu molh^{-1}$	[40]
MoS_2-ZnO	Hydrothermal	0.5M Na_2SO_4	Xe Lamp 1000 W	768 $\mu molh^{-1}$	[41]
MoS_2-ZnO	Hydrothermal	0.10M Na_2S	Xe Lamp 300 W	27,690 $\mu molh^{-1}$	[42]
MoS_2-ZnO	Hydrothermal	0.10 Na_2SO_4	Xe Lamp 100 W	145.6 $\mu molh^{-1}$	[43]

Catalyst	Synthesis Method	Sacrificial Agent	Light Source	Activity	Ref.
MoS_2-SnO_2	Hydrothermal	0.10 Na_2SO_4	Xe Lamp 400 W	117.2 µmolh^{-1}	[44]
Bi_2O_3-MoS_2	Two steps hydrothermal	-	Xe Lamp 300 W	10 µmolh^{-1}	[45]
MoS_2-CeO_2	Hydrothermal	Methanol	-	508.44 µmolh^{-1}	[46]
MoS_2-g-C_3N_4	Impregnation and sulfidation	10% lactic acid	Xe Lamp 300 W	108 µmolh^{-1}	[47]
WS_2-g-C_3N_4	Impregnation and sulfidation	10% lactic acid	Xe Lamp 300 W	20.6 µmolh^{-1}	[47]
MoS_2-g-C_3N_4	Impregnation	Na_2SO_4	Xe Lamp 300 W	23.10 µmolh^{-1}	[48]
MoS_2-g-C_3N_4	Sonication and treatment	Triethanolamine	Xe Lamp 300 W	887.6 µmolh^{-1}	[50]
MoS_2-C	Hydrothermal	Na_2SO_3 and Na_2S	Xe Lamp 1000 W	120 µmolh^{-1}	[51]
MoS_2-RGO	Hydrothermal	Triethanolamine	Xe Lamp 400 W	42,000 µmolh^{-1}	[55]

excitation state because of bulky (Br) atom in the molecule. High activity was accounted for by the tight p-n junction of the composite. The graphene being heavily doped with the heteroatom nitrogen (15%) boosted electron injection capability of the graphene thus better HER activity. The intensity (power) and kind of lamp used could as well contribute the amount of H_2 produced. The amount of H_2 generated was a magnitude of four-fold higher (42 mmolh^{-1}) when 400 W xenon lamp to when 100 W Halogen lamp was used (10.5 mmolh^{-1}).

Deployment of phase-engineering has greatly help to elevate the catalytic output of MoS_2 by making both the edge sites and basal plane available for HER thus improving on the performance of the catalyst. This method helps to tune the electronic structure of the semi-conductor nanomaterial to a metallic 1T-phase. New research has proved that phase-engineered metallic 1T-MoS_2 was more effective in water splitting over 2H-MoS_2 since both the edge sites and basal plane active sites participated in the catalytic reaction. Maitra's group [56] demonstrated the use of mono 1T-MoS_2 for photocatalytic water splitting in the presence of dye (EOSIN Y) the yield was 30 mmolh^{-1} of H_2 which was several folds (600) higher compared to few-layer 2H-MoS_2 (0.05 mmolh^{-1}). With prolonged time (3 h) 1T MoS_2 generated 250 mmolh^{-1} of H_2. However, when the catalyst was dried, the activity was minimized, and observable reduction was noticed in the performance when annealed sample was applied. The H_2 yield was comparable to that of 1H-MoS_2.This suggested possible transformation to 1H-MoS_2 upon annealing the sample. The group also did further study on the metallic group VI TMDC by working with other chalcogens. When metallic 1T $MoSe_2$ was tested for HER performance was 900 times higher than 1H-$MoSe_2$. A similar observation was also recorded for 1T $MoTe_2$ which showed far better activity when compared to 1H $MoTe_2$. In related research applying phase engineering the group [52] also produced 1T $MoSe_2$ which showed high performance for hydrogen evolution using visible light. The reaction was conducted using EOSIN as a sensitizer and TEOA as holes acceptor. It was observed that the HER activity of 7500 mmolh^{-1} was far higher than for the semiconducting phase (1H-$MoSe_2$) by almost 100 times, as well more superior to both 2H and 1T- $MoSe_2$. This was attributed to ultra-fast electron transport to the active sites of the material upon excitation with photon. DFT using theoretical calculation also proved 1T-$MoSe_2$ to have a lower work function than both 1T and 2-H $MoSe_2$.

Other methods that are employed to make the basal plane of TMDC active apart from phase engineering include introducing sulphur vacancies and doping with active metal ions charge. Common methods used in introducing chalcogenide deficiencies are contact with argon (Ar) plasma, annealing in H_2 and electrochemical reduction. Doping with active metals such as Cu, Zn, Ag, Pt and Co can increase the active sites and conductivity of TMDC. Recent Investigations have shown that the basal plane of TMDC become active by activating the catalytic activity of Se atoms in the basal plane [57].

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