



# Photo-Driven Reduction of Carbon Dioxide: A Sustainable Approach Towards Achieving Carbon Neutrality Goal

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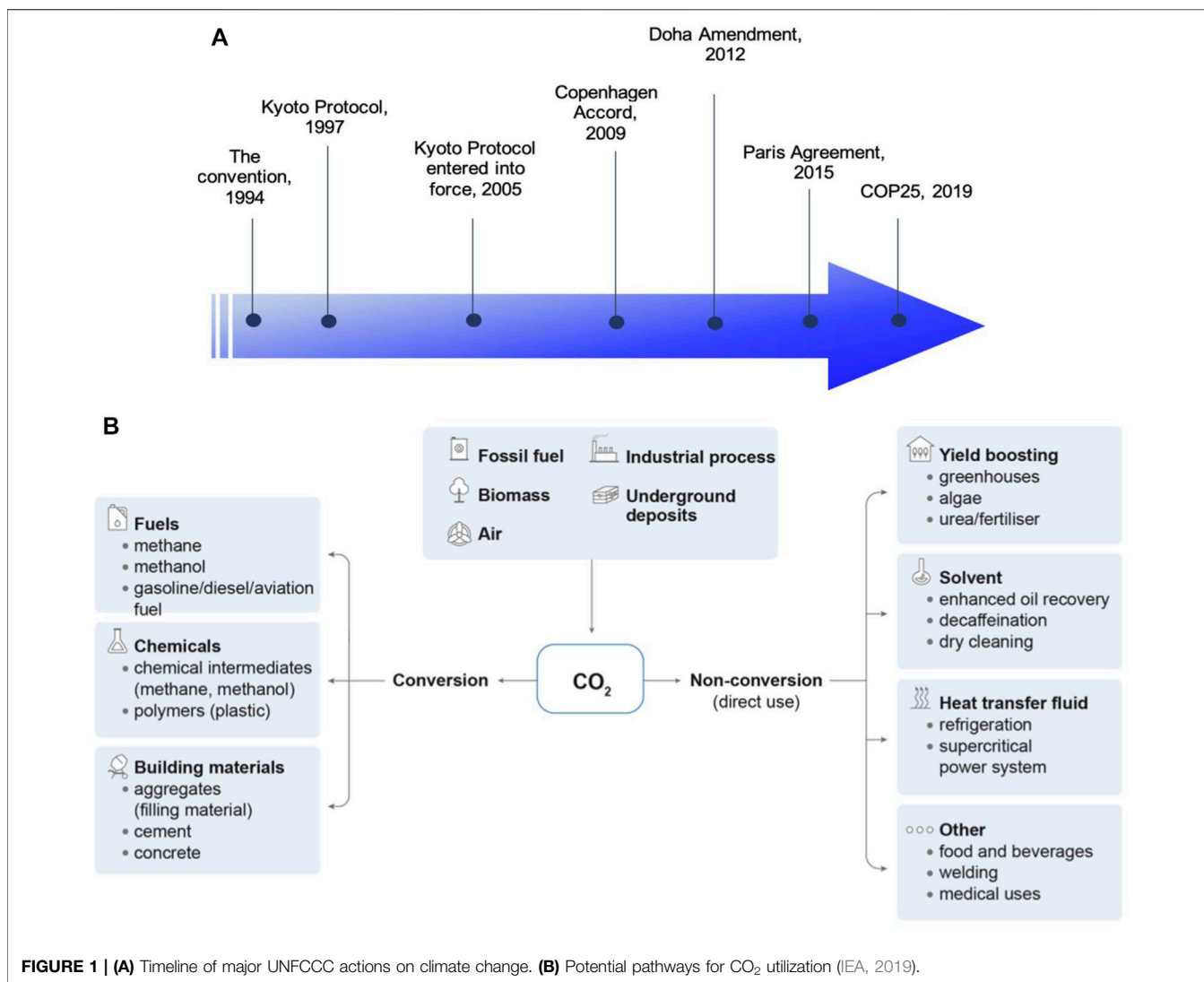
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The photo-driven reduction of carbon dioxide (CO<sub>2</sub>) into green and valuable solar fuels could be a promising solution to simultaneously address energy- and environmental-related problems. This approach could play an integral role in achieving a sustainable energy economy by closing the carbon cycle and allowing the storage and transportation of intermittent solar energy within the chemical bonds of hydrocarbon molecules. This Perspective discusses the latest technological advancements in photo-driven CO<sub>2</sub> conversion *via* various pathways, namely photocatalysis, photoelectrocatalysis and photovoltaic-integrated systems. In addition to providing an outlook on unresolved issues concerning the said technologies, this Perspective also spotlights new trends and strategies in the structural engineering of materials to meet the demands for prominent CO<sub>2</sub> photoreduction activity as well as spearhead the ground-breaking advances in the field that lead to the translation of CO<sub>2</sub> photo-driven technologies from the laboratory to industrial-scale applications.

**Keywords:** carbon dioxide, photocatalysis, photoelectrochemical, photovoltaic, environmental remediation

## INTRODUCTION

Greenhouse gases (GHG) emission has been identified as the major cause that leads to climate change and has remained as the primary challenge in the effort to control the pace of global warming. The United Nations Framework Convention on Climate Change (UNFCCC) entered into force in 1994 with a pivotal role to oversee and control the emission of GHG as a global effort. In the same year, the impact of climate change and the need for mitigations to tackle this issue were recognized and highlighted for the first time in the Convention. **Figure 1A** depicts key development of climate change actions by UNFCCC since its establishment. The Kyoto Protocol was initiated in 1997 with all party members came into agreement to limit and reduce the emission of GHG with individual targets tailored for their respective countries. The initiative only entered into force 8 years later and was amended in 2012 (known as the Doha Amendment) with emission targets renewed for the second commitment period from 1 January 2013 to 31 December 2020 (United Nations Climate Change). On the other hand, Copenhagen Accord was established in December 2009 where a target of global



**FIGURE 1 | (A)** Timeline of major UNFCCC actions on climate change. **(B)** Potential pathways for CO<sub>2</sub> utilization (IEA, 2019).

temperature rises of not more than 2 C above pre-industrial level was introduced (United Nations Climate Change, 2009). The global target was highlighted again in Paris Agreement, 2015, to drive social and economic transformation to control global warming at preferably 1.5 C above pre-industrial levels (United Nations Climate Change). In response to the Paris Agreement, both EU and United Kingdom have set their targets to achieve at least 55 and 68% reduction in GHG emission, respectively by 2030 with the final goal of zero-carbon emission by 2050 (Gov, 2020; European Commission). This ambitious zero-carbon emission target requires effort from several stakeholders across the nations, up from technology advancement and transformation of current industrial activities down to the reforming of policies and regulations to facilitate socioeconomic development sectors associated with GHG emissions.

To control the emission of carbon dioxide (CO<sub>2</sub>), the main culprit of global GHG emission, new technology pathways related to CO<sub>2</sub> capture, utilization and sequestration (CCUS) have been

widely studied over the past decades. **Figure 1B** illustrates the potential pathways for the utilization of CO<sub>2</sub> in the industry. In general, CO<sub>2</sub> collected on-site can be either: 1) directly used as a heat transfer fluid or feedstock/solvent in manufacturing processes or; 2) converted into other derivatives, such as fuels, hydrocarbons, and building materials following respective chemical synthesis routes. Currently, the established technologies are carbon capture with amine process, direct capture from air with underground deposit, and carbon capture integrated with bioenergy plant (IEA, 2021a). According to the recent report by IEA, the overall cost of carbon capture can be in a broad range of USD15-25/t CO<sub>2</sub> (for CCUS from natural gas processing) to USD130-340/t CO<sub>2</sub> (for direct capture), subject to the quality of CO<sub>2</sub> streams and the technology applied (IEA, 2021). These technologies are costly but necessary to achieve a zero-carbon emission goal. Therefore, the continuous advancement in CCUS technologies is an on-going process to improve their availability and cost effectiveness for large-scale, practical deployment.

## RECENT TECHNOLOGICAL ADVANCEMENTS IN THE PHOTOREDUCTION OF CO<sub>2</sub>

The transformation of CO<sub>2</sub> into energy bearing hydrocarbon compounds has gained incessant research interest in recent years (Creutzig, et al., 2017; Alsayegh et al., 2020). Converting CO<sub>2</sub> generated from a combustion process into hydrocarbon fuels offers attractive solution to close the carbon-fuel cycle (Ulmer et al., 2019). Ideally, for the derivation of completely renewable hydrocarbon fuels from CO<sub>2</sub>, the entire synthesis route should have minimal carbon emission and be free from fossil fuel usage. With this aim, much attention has been placed on using solar energy as the future energy source. Nevertheless, the practical implementation of CO<sub>2</sub> photoreduction technologies necessitates the development of highly efficient, robust, photo-driven materials and systems; these have been hot research areas in recent years. Thus, in this perspective, the latest technological advances in the photo-driven reduction of CO<sub>2</sub> are summarized and discussed with comments of their respective advantages and existing limitations. The three primary CO<sub>2</sub> reduction systems covered are: 1) the photocatalytic CO<sub>2</sub> reduction; 2) the photoelectrochemical (PEC) pathway as well as 3) the photovoltaic-integrated systems.

### Photocatalytic CO<sub>2</sub> Reduction

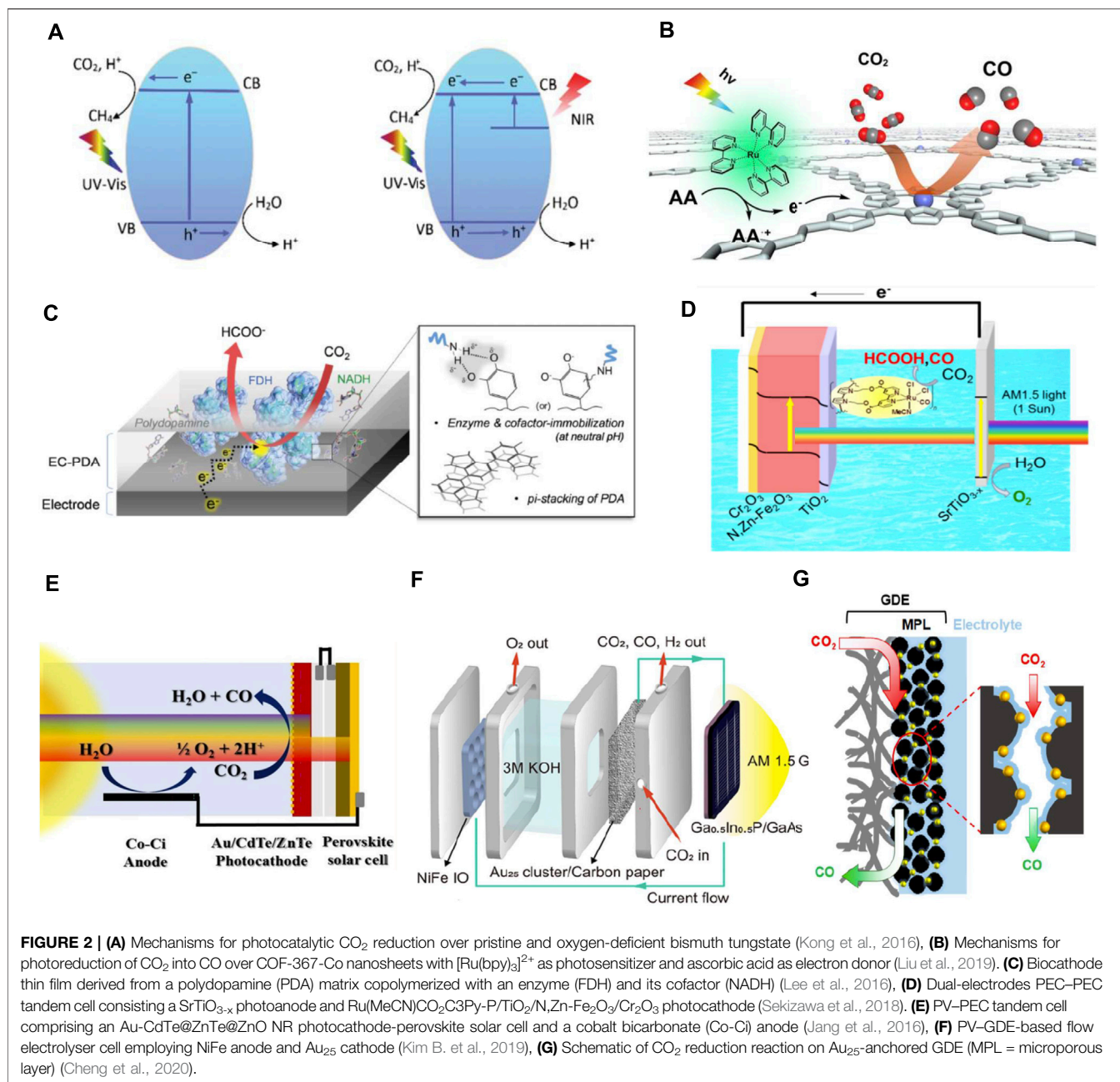
The efficiency of photocatalytic CO<sub>2</sub> reduction lies in the design of the photocatalyst. Therefore, it is of paramount importance to tailor the electronic structures of photocatalysts, with efforts to modulate the reaction paths and activation energy barriers, thereby enhancing the photoactivity and product selectivity (Wang H.-N. et al., 2021). In recent years, significant research progress has been witnessed in photocatalysis. A myriad of novel photocatalysts with 0D (zero-dimensional, *i.e.*, quantum dots), 1D (one-dimensional, *i.e.*, nanotubes, nanorods), 2D (two-dimensional, *i.e.*, atomic layers, nanosheets, nanoplates) and 3D (three-dimensional, *i.e.*, hollow nanostructures, nanospheres, microspheres) structures have been developed (Kong et al., 2017; Kong et al., 2019; He et al., 2019; Kong et al., 2020; Sun et al., 2020; Wang J. et al., 2020; Ke et al., 2021; Li L. et al., 2021). The photoactivities are highly dependent on the architectures of the photocatalysts as the light trapping capability, surface active sites, electron-hole pairs separation and transportation pathways of photocatalysts are greatly affected by the structures. To improve the photoresponse of catalysts, a number of strategies have been adopted to reduce the bandgap of photocatalysts by integration of dopants, sensitizers, co-catalysts, heteroatoms or inducing surface defects (Chen et al., 2021; Chen et al., 2020; Lee et al., 2020; Miao et al., 2021; Pan et al., 2021). For instance, introducing oxygen vacancy (OV) onto bismuth tungstate could expand the light absorption spectrum of the photocatalyst from UV to near infrared (Kong et al., 2016). The OV-induced defect states play indispensable role to trap photoinduced electrons, hence improving the electron-hole pair separation and inhibiting the direct recombination of photogenerated charge carriers. In this photocatalyst structure, near infrared light was harvested through sub-bands excitation

from OV-induced defect states to the conduction band of bismuth tungstate (Figure 2A). To promote the separation and transportation of photoinduced charge carriers, Type-II, Z-scheme or p-n heterojunctions are often introduced to the composite photocatalysts to prolong the lifetime of charge carriers so that more electrons can take part in the CO<sub>2</sub> reduction reaction (Jiang et al., 2020; Li S. et al., 2021; Wu et al., 2021). Since CO<sub>2</sub> reduction can only take place on the surface active sites, many recent researches focus on crystal facet engineering in order to maximize the exposure of most active facets (Kong et al., 2018; Jatav et al., 2021). Besides, decreasing the thickness of 2D materials to ultrathin-nanoscale or even atomic layers can endow ultrafast transportation of charge carriers from interior to the surface of photocatalysts along with large exposure of active sites to boost CO<sub>2</sub> adsorption and activation (Han et al., 2021; Teh et al., 2021).

Other than the conventional semiconductor photocatalysts, molecular photocatalysts have received much attention in the past few years. In another reported work, CO production rate of up to 10,162  $\mu\text{mol g}^{-1} \text{h}^{-1}$  was obtained using COF-367-Co nanosheets under visible light illumination with [Ru(bpy)<sub>3</sub>]Cl<sub>2</sub> (bpy = 2,2'-bipyridine) as the photosensitizer (Figure 2B) (Liu et al., 2019). The Photosensitizer plays a pivotal role where it absorbs incident light and converts it to photon energy that activates the nearby photocatalyst. Very recently, a molecularly engineered, scalable photocatalyst sheet with solar-to-formate conversion efficiency of 0.08  $\pm$  0.01% and product selectivity of 97  $\pm$  3% for formate were reported. Notably, in this setup, the photoreduction of CO<sub>2</sub> was realized without using any sacrificial reagents; however, the photocatalytic system is complex, which comprises of Rh, La, SrTiO<sub>3</sub>, BiVO<sub>4</sub>, RuO<sub>2</sub>, Au, and phosphonated cobalt(II) bis(terpyridine) (Wang Q. et al., 2020).

### PEC CO<sub>2</sub> Reduction

PEC CO<sub>2</sub> reduction, which uses electricity as an aide, enables higher conversion efficiencies and offers more design room owing to the wider selection of exploitable materials and configurations. For a commercially competitive device, PEC materials and cell design should approach high product selectivity with photocurrent of 7 mAcm<sup>-2</sup> (corresponding to a 10% solar-to-fuel efficiency) and minimum bias requirement in the simplest possible system (Kumaravel et al., 2020). Although single-semiconductor configurations in photoanode- or photocathode-driven CO<sub>2</sub> reduction reactions provide the greatest simplicity, they face elevated requirements, needing larger biases to achieve high system efficiencies (Kim J. H. et al., 2019). It is worth recognizing that higher photocurrent and lower onset potential values have generally been linked to catalytically-active materials which can overcome the high kinetic overpotential of the CO<sub>2</sub> reduction half-reaction, and by a lesser degree, the optical and charge transport properties of the photoelectrode. Single-junction expensive PV-grade materials such as Si (Fung et al., 2020; Hu et al., 2018; Rao et al., 2018), GaN (DuChene et al., 2018; Sekimoto et al., 2016), InP (Kaneco et al., 2006a; Kaneco et al., 2006b; Qiu et al., 2015; Zeng et al., 2015) and ZnTe (Jang et al., 2014; Jang et al., 2015) with excellent optical and charge transport properties have been extensively



studied. Nevertheless, these materials possess poor catalytic sites for CO<sub>2</sub> reduction, which usually offer little to no redeeming improvements in the PEC activity as compared to those of more catalytically-active copper-based (Ghadimkhani et al., 2013; Won et al., 2014; de Brito et al., 2015; Kang et al., 2015; Kang and Park, 2017; Lee et al., 2018) or molecular metal-complex (Arai et al., 2011; Jeon et al., 2014; Huang et al., 2016; Kumagai et al., 2017) systems, whose activities have hitherto remained unparalleled.

Recently, integrating microbes and enzymes into biocathodes (Figure 2C) have shown massive success in lowering bias requirements due to their innate capacity to catalyze a range of CO<sub>2</sub> metabolic processes (Fu et al., 2018; Kuk et al., 2019; Sokol et al., 2018; Xu et al., 2021). Despite the high bias requirement in

single-junction configurations, bias-free photoanode-driven CO<sub>2</sub> reduction were realized by the microbial TiO/CdS-*Methanobacterium* (Xiao et al., 2020) and enzymatic CoPi/BiVO<sub>4</sub>-PDA/NADH/FDH (Lee et al., 2016) hybrid systems, reaching unprecedented single-junction solar conversion efficiencies of 1.28 and 0.042%, respectively. Moreover, higher conversion efficiencies and lower biases have also been achieved in the more complex, dual photoanode-photocathode tandem configurations, owing to their improved spectral absorption, cumulative photo-potential and Z-schematic band arrangement which better meets the voltage and band-edge requirements for CO<sub>2</sub> reduction. A seminal work by Arai et al. reported among the highest solar-to-chemical conversion



efficiency of 0.14% to formate, which approaches that of the biological plant switchgrass (0.2%), using a SrTiO<sub>3</sub> photoanode and InP[RuCP] photocathode (Arai et al., 2013). While more recently, the combination of SrTiO<sub>3-x</sub> photoanode and Ru(MeCN)CO<sub>2</sub>C3Py-P/TiO<sub>2</sub>/N,Zn-Fe<sub>2</sub>O<sub>3</sub>/Cr<sub>2</sub>O<sub>3</sub> photocathode (Figure 2D) can yield a comparable efficiency of 0.15%, by alternately employing more abundant and cheaper multilayer metal oxides with efficient interfaces (Sekizawa et al., 2018).

## Photovoltaic-Assisted CO<sub>2</sub> Reduction

Photovoltaic-photoelectrochemical (PV-PEC) schemes are another attractive option to achieve spontaneous CO<sub>2</sub> reduction at the expense of higher system complexity. This is since voltage requirement for CO<sub>2</sub> reduction can be offset by the additional bias generated from the PV cell. The WO<sub>3</sub>/dye-sensitized solar cells tandem photoanode with Cu<sub>x</sub>O wire-array cathode, is by far, one of the most efficient PV-PEC systems with a solar-to-CO efficiency of 2.5% (Nath et al., 2016). On the other hand, an Au-decorated triple layered ZnO@ZnTe@CdTe core-shell nanoarray photocathode in tandem with CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite solar cell and a Co-Ci anode, which produced CO with a 0.35% conversion efficiency, was reported (Figure 2E) (Jang et al., 2016). Balancing light absorbance in the PV and PEC cell however remains a challenging aspect which has prohibitively restricted conversion efficiencies to mostly below <0.1% (Kuk et al., 2019; Zhou et al., 2019; Andrei et al., 2020). Zhou et al., on the other hand, reported an exceptionally high efficiency of ~10% by PV-PEC using a buried III-V tandem photoanode GaAs/InGaP/TiO<sub>2</sub>/Ni and a Pd/C cathode by means of a bipolar membrane (Zhou et al., 2016). The use of a bipolar membrane enabled the seamless coupling between two electrodes and electrolytes at different pH values, each optimized respectively for oxygen-evolution (pH 13.7) and CO<sub>2</sub> reduction reaction (pH 8.0), which overall lowered the combined cell overvoltage.

A higher efficiency range of 5–20% has usually been achieved in PV-EC configurations, where semiconductor-liquid-junctions are eliminated (Schreier et al., 2015; Bullock et al., 2017; Arai et al., 2019). Some notable works were by Schreier et al. which attained a solar-to-CO efficiency of 13.4% by pairing a three-junction GaInP/GaInAs/Ge PV cell to a CO<sub>2</sub>-to-CO electrolyser equipped with a bipolar membrane (Schreier et al., 2017). More recently, the applications of gas diffusion electrodes (GDE) in electrolyser flow cell designs have afforded record-breaking efficiencies. The use of GDEs permit a semi-gas-phase operation at the cathode compartment, allowing higher current densities by circumventing the mass-transfer limitation of CO<sub>2</sub> in aqueous system. Through directly connecting a high-efficiency PV cell and a GDE-based flow electrolyser (Figures 2F,G), Kim B. et al. (2019) and Cheng et al. (2020) have achieved a CO<sub>2</sub>-to-CO efficiency of as high as 18.0 and 19.1%, respectively. Though it should be noted that while higher efficiencies have been achieved, this comes at the price of greater device complexity and cost, and a techno-economic assessment is required to ensure that these enhanced performances offer worthwhile advantages over the simpler single-junction devices. Ultimately, the ideal choice for PEC materials and cell design should strike the perfect balance

between maximizing conversion efficiency and minimizing system cost.

## CHALLENGES AND FUTURE PROSPECTS

Despite its great significance, the photoreduction technology is still far from commercialization and the complex reaction system curtails its practical applications. Technology readiness level (TRL), a measurement system that assesses the maturity level of a particular technology, can be used as an indicator to gauge the readiness of the photoreduction technology for full commercial deployment. Currently, the TRL of the photoreduction of CO<sub>2</sub> remains low at TRL 3 to 4 (Jarvis and Samsatli, 2018). Most of the research findings reported are limited to the laboratory-scale and revolve around the development of photocatalytic materials. There are only a handful of work on pilot-scale operations at low capacity. To make this technology feasible for commercial scale production, the technology must achieve at least TRL 6 or 7 for demonstrating in the relevant environment. It is anticipated that at least five to ten years of further research is needed before carbon photoreduction technology can be practically deployed (The Global CO<sub>2</sub> Initiative, 2016).

Another key aspect is the fabrication of catalyst from inexpensive, non-toxic and abundantly available elements. To date, a diverse range of photo(electrochemical)catalysts has been reported to be efficient for CO<sub>2</sub> photoreduction in various reactor set-ups. The vast majority of these materials are carefully tailored using multiple dopants to achieve high efficiency and selectivity. The introduction of rare earth elements as dopants or co-catalysts, such as Ce/TiO<sub>2</sub> (Xiong et al., 2015), monometallic cerium layered double hydroxides (Ye et al., 2017), La/g-CNT (Muhammad et al., 2020), La<sub>0.225</sub>Bi<sub>0.775</sub>O<sub>1.5</sub> (Wang Y. et al., 2021) and yttrium-doped H-Titanate (Lu et al., 2019), Re(CO)<sub>3</sub>(bpy)Cl (Adams et al., 2018) etc., will increase the overall cost of the photoreduction and pose environmental issues.

Another challenge of CO<sub>2</sub> photoreduction lies within the product selectivity and yield. Taking methane (CH<sub>4</sub>) as an example, CO<sub>2</sub> photoreduction under visible light irradiation could yield 3–12 times more CO than CH<sub>4</sub>. (Cheng et al., 2017; Raziq et al., 2017; Thompson et al., 2020). The stark difference in yield is attributed to the preferred formation of CO over CH<sub>4</sub>, as the former only requires two electrons while the latter requires eight. Interestingly, other researchers have also reported contrasting results where a higher yield of CH<sub>4</sub> was observed over CO (Wang et al., 2021; Wu et al., 2021). The difference in the results is due to the unique characteristics of the photocatalysts used. This prompts the need for selectivity studies to tailor the photocatalyst to maximise its yield for the desired products. Additionally, water plays an essential role in CO<sub>2</sub> conversion as it serves as both the electron and proton donor. Therefore, in catalytic CO<sub>2</sub> reduction reactions, be it photocatalysis, PEC or PV-EC, the competing reaction of water reduction is fundamentally unavoidable. The aforementioned reaction effectively reduces the yield of the

intended product by reducing the electrons available for CO<sub>2</sub> photoreduction.

The CO<sub>2</sub> photoreduction pathway can be regarded as a green process since the reaction is merely powered by light energy. This, however, poses an important challenge to the commercialization of the technology. The incident light intensity is known to be one of the most important factors that controls the efficiency of a CO<sub>2</sub> photoreduction process (Tan et al., 2017). As such, the potential for industrial-scale operation of CO<sub>2</sub> photoreduction is largely constrained by regional solar intensity which is dependent on geographical factors. Commercialization of CO<sub>2</sub> photoreduction could be relatively more challenging in countries or continents with lower solar intensity. For instance, the United Kingdom is reported to have an average solar irradiance of 101.2 W m<sup>-2</sup>, ranging from 71.8 W m<sup>-2</sup>–128.4 W m<sup>-2</sup> depending on the geographical location in the country (Burnett et al., 2014). This value is much lower as compared to tropical countries which has an average annual solar irradiance of approximately 10-fold higher, ranging between 600 and 900 W m<sup>-2</sup> (Mohammad et al., 2020). A higher level of solar irradiance is important to ensure that sufficient photon energy is available for activation of the photocatalyst to drive the CO<sub>2</sub> reduction reaction (Sichel et al., 2017). As such, a higher performance photo(electrochemical)catalyst is necessary to overcome this barrier for the countries with lower solar irradiance.

With the increase in the awareness of sustainability processing, circular economy of the photoreduction system could be another focus of study. The efficiency of the photo(electrochemical) catalyst no doubt plays key role to these reaction pathways. However, one should not overlook the potential environmental impact of the use of the materials, if we are to upscale these technologies for large-scale reduction of CO<sub>2</sub>. In principal, a suitable photocatalyst material should not have significant impact to the environment, is green (or less toxic), and easy to handle. Extensive studies on the lifespan and reusability of photocatalyst materials could be the next focus to make this technology more economical and environmentally viable.

## CONCLUSION

Photo-driven technologies are undoubtedly the most sustainable and green solution for the conversion of CO<sub>2</sub> into energy-rich hydrocarbon derivatives. These processes

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employ the power of the sun as the only resource to attain the Gibbs free energy of the CO<sub>2</sub> reduction reaction; thus rendering it fossil fuel-free with markedly lower carbon footprint as compared to the conventional hydrothermal reactions or electrolysis. With the global pledge to achieve net zero-carbon emission by 2050, the development of emerging technologies in CO<sub>2</sub> utilization must be fast-tracked. In addition, with the recent interest in the exploration of Mars, a planet consisting of 95% CO<sub>2</sub> and a solar irradiation of 586 W m<sup>-2</sup>, it would be extremely beneficial to utilize its atmosphere to produce sustainable fuels for interplanetary travels. Despite great strides made in the field, the implementation and up-scaling of these photo-driven technologies for commercial applications remain a great hurdle even up till today. As highlighted in the previous sections, this is mainly ascribed to the constraints of solar energy potential as well as low product selectivity and yield. Intensified research is needed in the areas of materials discovery and innovative photoreactor designs. The resolution of these obstacles could bring about the successful industrialization of CO<sub>2</sub> photoreduction technologies in the near future, which could ultimately pave the way for a greener and more sustainable tomorrow.

## DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

## AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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