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Review of CO_2 reduction photocatalysts with a sustainability perspective

Pınar Özdemir, Beyza Yılmaz, Ramazan Yıldırım*

Department of Chemical Engineering, Boğaziçi University, 34342, Bebek-Istanbul/Turkey

**Corresponding author. E-mail address: yildirra@bogazici.edu.tr (R. Yıldırım)*

Abstract

The most frequently used semiconductors and co-catalysts for the photocatalytic CO_2 reduction ($PCCO_2R$) are reviewed with sustainability perspectives; unlike a regular review article, the most commonly used semiconductors and co-catalyst alternatives are evaluated in terms of material availability and supply risk, sustainability of production routes, CO_2 reduction performance, operational stability, and end-of-life degradation patterns as sustainability criteria. It was found that g-C_3N_4 is the most sustainable semiconductor, followed by TiO_2, among the most frequently used alternatives, while Cu_2O/CuO, ZnO, GO/r-GO, and ZnS can be considered as moderately sustainable. $BiVO_4$ cannot be treated as a sustainable semiconductor due to the supply risks associated with both Bi and V, while $CsPbBr_3$ and CdS should be avoided due to the high toxicity of Pb and Cd, respectively. Among the semiconductor alternatives, Cu, Fe, and Ni seem to be sustainable choices, while the precious metal co-catalysts may not be sustainable due to low availability and supply risks, price fluctuations, and environmental impacts they may impose during the mining and refining stages. Cobalt should also be avoided due to its high toxicity and environmental burden through its life cycle.

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1. Introduction

Catalysts are among the critical ingredients of the modern chemical industry; Ciriminna et al, report that catalysis plays a role in at least a certain point of processes involving more than 80 percent of all manufactured products[1]. Catalysis is also vital for sustainable development; any improvement in efficiency, selectivity, and stability of any chemical process by developing better catalysts will likely contribute to sustainability by reducing material and energy consumption. Some of the renewable energy technologies (like biofuel production, green hydrogen production, solar fuel production, fuel cells, electrolyzers, and so on), which are the key enablers of sustainable development, also heavily rely on catalysis. At the same time, the catalytic/photocatalytic processes are extensively used for environmental cleaning, especially wastewater treatment.

Catalysts are complex chemicals manufactured through complex processes that should also be sustainable themselves; even though the use of renewable catalytic materials (like enzymes and carbon-based solid materials) is also increasing in recent years, most of the catalytic reactions are carried out over the solid heterogeneous catalysts that contain materials (especially metals) that may have serious impacts on human health and the environment. Meeting sustainability requirements with a reasonable performance may be much harder for photocatalysts because the alternatives for both semiconductor (the electrochemical properties should be suitable to create an electron-hole pair under visible irradiation) and co-catalyst (it should be effective in the separation of charges and selectively speed up the reactions) are limited[2]. Sustainability of photocatalytic CO_2 reduction (PCCO_2R), which is one of the most widely investigated processes aiming to produce solar fuels, by using solar energy to eliminate CO_2 (Fig. 1), is especially important; it would be contradictory to use non-sustainable materials in such a process that may be considered as an example for sustainability.

Then, what criteria and guidelines should be used to decide whether a catalyst is sustainable? Various aspect of sustainability have been covered by various resources from *lifecycle assessment systems* (LCA)[3,4] and *green chemistry*[5] to *UN Sustainable Development Goals* (SDGs)[6] suggesting numerous criteria and methods that are hard to implement simultaneously; often a limited number of criteria (and methods) are used (or needed) depending on the aims and priorities of the assessment project, the characteristics of the material and process to be assessed, and the availability and quality of the data needed. Consequently, the sustainability assessments published

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in literature, even for the same material or processes, are not standard and comparable, as it is desired for such tasks that often end up comparing and selecting among the alternatives. Unfortunately, two additional difficulties have to be also considered for $PCCO_2R$. First, the process is still in the development stage; hence, there is no final list of semiconductor or co-catalyst alternatives to compare yet. Second, some of the materials (some rare earth metals and noble metals) have become important parts of national/international policies, as often discussed in the political arena in recent years. Such policies may impose some restrictions on these materials, increasing supply risks that cannot be accounted for in the future; unfortunately, some materials (especially metals used as co-catalyst) used in photocatalysis, including CO_2 reduction, may easily fall into the strategic material lists of some nations.

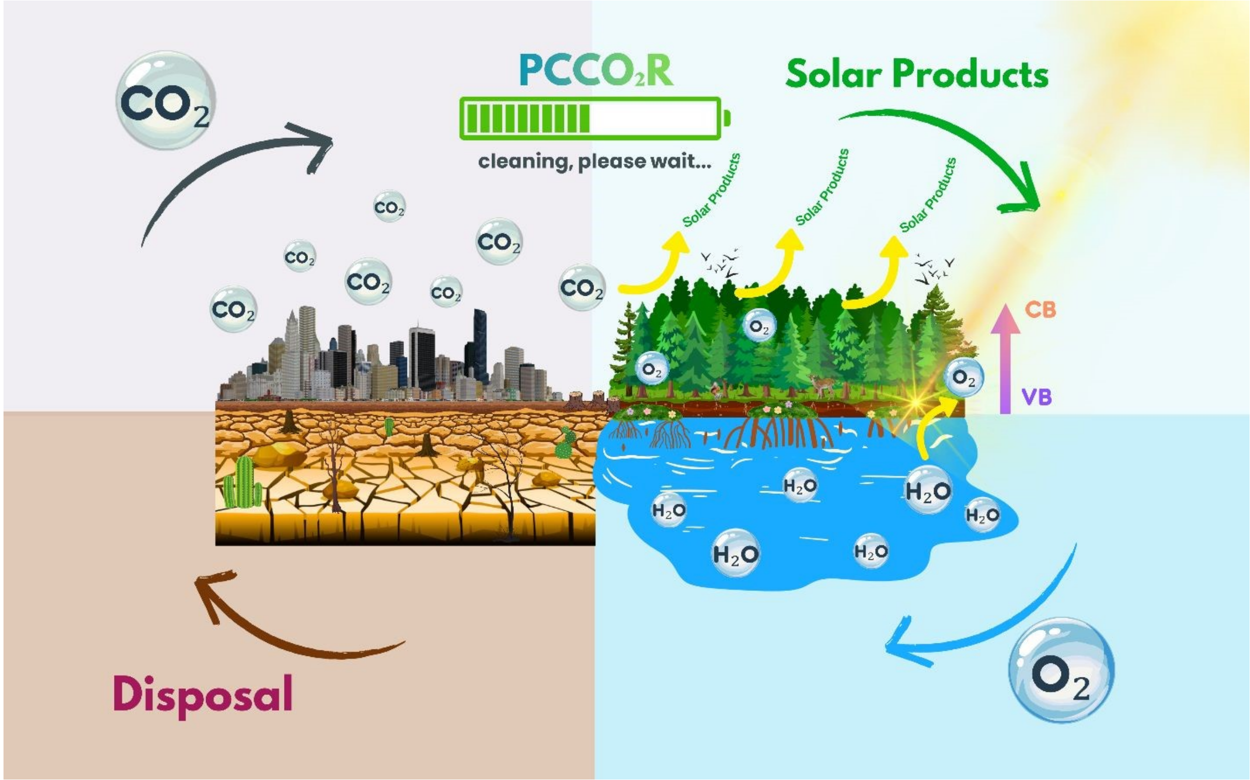

Fig.1 Photocatalytic CO_2 Reduction ($PCCO_2R$) as part of sustainable future

On the other hand, we believe that a sustainability perspective is still needed even for the technology in the development stage to direct the research and development efforts into more sustainable solutions; even though some works may be performed using the scarcest materials to improve our understanding of a subject, no technology can be commercialized if it does not rely

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on sustainably supplied materials. In one of our recent works, we extensively analyzed the material and methods used for the $PCCO_2R$[2]. Here, we assessed the most commonly used catalytic materials identified in that work from a sustainability perspective. As far as we know, no such work was published in the literature. We believe that, even though the uncertainties associated with the immaturity of the technology and lack of sufficient data do not allow a full-scale sustainability assessment, the evaluation of the most frequently used materials from a sustainability viewpoint will contribute to the material selection in future works. We presented our results for semiconductors and co-catalyst in separate sections; the co-catalyst materials create greater environmental concerns because they are mostly in metallic form; the semiconductors, on the other hand, are quite diverse in their ingredients and structures, and are less analyzed in terms of sustainability. Finally, we discussed the limitations of the current analysis and our future perspectives on the subject.

2. Selection of photocatalyst materials and criteria for evaluation

2.1 Selection of semiconductors and co-catalysts

The lists of semiconductors and co-catalysts analyzed here are based on the analysis of Web of Science (WOS) author keywords in one of our recent works on the analysis and review of current state of $PCCO_2R$[2]; the ten most commonly used semiconductors and co-catalysts were taken and analyzed in Section 4 and 5, respectively. Further details related to the frequency of these materials can be found elsewhere[2].

2.2 Criteria for sustainability evaluation

The sustainability criteria for materials or technology in general can be collected in two groups*:* *long-term availability* and *environmental impacts, including human life.* While long-term availability indicates if resources can be sustained, and assessed in terms of renewability, natural abundance, amount needed, and recyclability, the environmental impact shows if the social and physical environment can be sustained when the material or technology of interest is used, and can be assessed using various criteria describing the extent and severity of the impact. The sustainability of material technology has been treated and discussed with various perspectives in the literature. For example, the SDGs approach the subject in terms of actions to be taken and

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results to be achieved. In this respect, the sustainability of catalysis can be linked to several goals like SDG 7 (Affordable and Clean Energy) through the targets involving increase of renewable energy share and energy efficiency, SDG 9 (Industry, Innovation, and Infrastructure) promoting sustainable industries (T9.2), SDG 12 (Responsible Consumption and Production) through the sustainable use of natural resources and reduction of waste; SDG 13 (Climate Action) also indirectly pointing out the role of catalysis in addressing global environmental challenges including the photocatalytic reduction of excess CO_2[6]. On the other hand, the 12 principles of green chemistry, which are highly popular in the field of chemistry, approach the same problems with more fundamental principles such as designing safer chemicals, developing less hazardous synthesis methods, using renewable resources, and preventing/cleaning up the waste; the use of catalysts is also stated as one of the principles of green chemistry[5]. Finally, life cycle assessment (LCA) systems and tools suggest more direct criteria and tools to assess the environmental impact of material or technology in its entire life cycle.[7] Both criteria and tools of LCA are quite diverse and hard to implement for one material or process due to a lack of data; consequently, the extent of such assessment works is usually dependent on the needs, aim, or assessment, and availability of relevant information.

Here, we adopted some of the criteria, which may be relevant for our purpose, from sources in literature such as the works by Nuss and Eckelman[8] and Wand et al.[9], while we used an EU report on the criticality and supply risks for the materials[10]. We evaluated and discussed the sustainability of the semiconductors and co-catalysts in four categories as (1) *material availability and supply risk, (2) sustainability of synthesis routes, (3) CO_2 reduction performance, selectivity and operational stability* and (4) *end-of-life degradation* (Fig.2). We summarized the results in Table 1 and Table 2 for semiconductors and co-catalyst respectively using more specific criteria to describe the sustainability status of the alternatives; we used each criterion in five scales (also using color codes in Table S1) as explained in the tables. The descriptions of scales for all criteria and the source of the assessment are presented in SI (Table S2-S10 for semiconductors, Table S12-S20); Table S11 and Table S21 present the same information in Table 1 and 2 with more detailed explanations and related references.

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Fig.2 Four cycles of semiconductors and co-catalysts used in $PCCO_2R$

3. Current state of the art in photocatalytic CO_2 reduction

The $PCCO_2R$ process is initiated when the semiconductor absorbs light, promoting electrons from the valence band to the conduction band, and therefore generating electron-hole pairs. The photogenerated electrons are subsequently utilized for CO_2 reduction on the co-catalyst surface, while the holes participate in water oxidation to produce oxygen on the semiconductor (Fig. 3). In this system, the semiconductor primarily serves as the light-harvesting material and the active site for water oxidation, whereas the co-catalyst facilitates charge separation by suppressing electron-hole recombination and enhancing the reduction kinetics. Depending on the nature of the semiconductor, co-catalyst, and reaction environment, a variety of products, including H_2, CO, CH_4, CH_3OH, HCHO, and HCOOH, can be produced.

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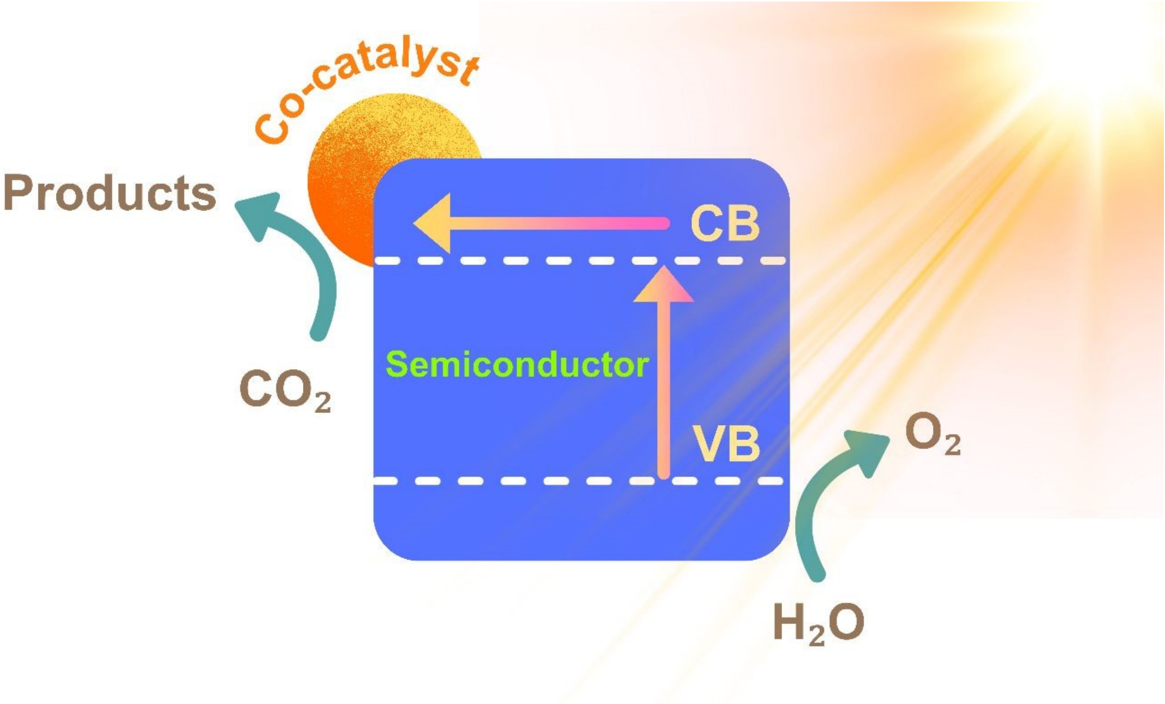

Fig. 3 Major steps of the photocatalytic CO_2 reduction occurred over a photocatalyst

Although the concept of utilizing solar energy to convert CO_2 into value-added chemicals, by mimicking natural photosynthesis, is a highly attractive and elegant route, achieving a commercially viable process remains a significant challenge due to low efficiencies and low operational stabilities of the photocatalysts. Before even starting to talk about its commercialization potential, two fundamental issues must be addressed and solved first: (1) the discovery or development of semiconductors having sufficient visible-light absorption potency, and (2) the suppression, or at least mitigation, of electron-hole recombination before their involvement in redox reactions, which is one of the primary reasons for employing co-catalysts. Among these challenges, the design of efficient semiconductor materials appears to be the more demanding task. Consequently, a growing number of papers have been published in recent years to test the suitability of semiconductor alternatives in terms of material types and formulations; meanwhile, various co-catalyst alternatives were also investigated.

In one of our recent paper[2], we investigated the annual publications trends and the author keywords to assess the current state of art in this important technology; we identified and assessed the most commonly used materials, structures, synthesis methods, modification types, sacrificial reagents,

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and product variations. Here, we took the most frequently appeared semiconductors and co-catalysts in our previous work[2] and evaluated them from a sustainability point of view.

4. Review of semiconductors with a sustainability viewpoint

Various semiconductors have been investigated for $PCCO_2R$ in the literature to convert CO_2 into higher-value solar fuels and chemicals[11]; in this section, the sustainability of g-C_3N_4 (appeared 2004 times in author keywords between 1990 and 2024) and TiO_2 (appeared 1732 times in author keywords between 1990 and 2024) were evaluated first, as they are overwhelmingly the most commonly studied semiconductors. Additionally, the following materials, which were also used quite frequently, were added to the list as the representative of their kinds: graphene oxide/reduced graphene oxides as the other examples for carbon-based materials, ZnO and copper oxides as other examples for metal oxides, ZnS and CdS as examples for sulfides, $BiVO_3$ as example for inorganic perovskites, $CsPbBr_3$ as example for halide perovskites and BiO_xBr_y as example for bismuth oxyhalides. The complete list of common semiconductors used in the field is presented in Fig. 4.

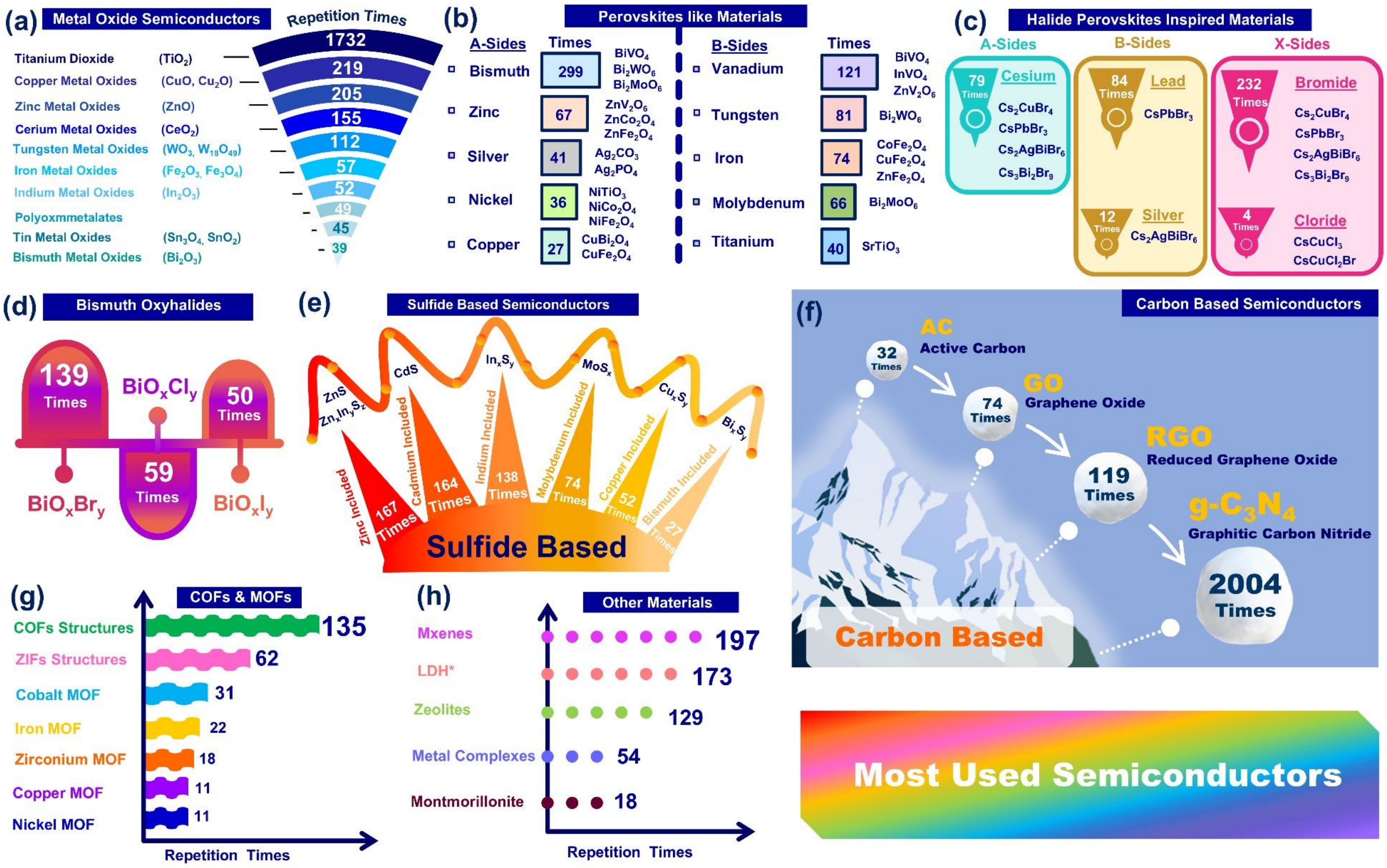

Fig.4 Frequency of use of photocatalyst materials for PCCO$_2$R. (a) Metal oxides, (b) perovskites like materials, (c) halide perovskites inspired materials, (d) bismuth oxyhalides, (e) sulfide-based materials, (f) carbon-based materials, (g) MOFS and COFs, and (h) others[2]

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4.1. Graphitic carbon nitride (g-C_3N_4)

Material availability and supply risk. g-C_3N_4 is composed of earth-abundant elements with globally established supply chains; hence, it can be considered as a widely available raw material with negligible supply risk[10]. In this respect, g-C_3N_4 can be distinguished from many widely used photocatalysts, especially those containing metals, and emerges as one of the potential sustainability semiconductors for $PCCO_2R$[12].

Sustainability of synthesis routes. As far as precursor materials and production technologies are concerned, g-C_3N_4 exhibits favorable characteristics as well. It is produced from common industrially well-established precursors such as urea, melamine, dicyandiamide, and thiourea, which also have comparatively low impact on human health and environmental especially when compared with metal salt precursors used in the synthesis route of some other semiconductors[13]. g-C_3N_4 is predominantly synthesized via thermal polycondensation, while alternative approaches, including solid-state reactions[13], solvothermal methods[14], chemical deposition[15], and supramolecular assembly strategies[16], are also employed to produce small amounts of materials for research purposes to tailor the surface area and electronic structure. However, these alternative roots may require additional processing steps and chemicals that may have serious inverse effects on human health and the environment if they were used in industrial scale. Energy demand represents the dominant sustainability consideration in industrial g-C_3N_4 production; thermal polycondensation, typically conducted at 500-600°C[17]. The electricity and heat input during calcination-especially during precursor processing and post-treatment-as dominant contributors to environmental impact[18]. On the other hand, the synthesis route is solvent-free and does not require corrosive reagents, thereby minimizing wastewater generation and direct chemical emission to the environment[17].

CO_2 reduction performance, selectivity, and operational stability. g-C_3N_4-based systems yield CO (up to 883 μmol g^{-1}cat. h^{-1})[19] and CH_4 (up to 562 μmol g^{-1}cat. h^{-1})[20], while methanol (up to 2547 μmol g^{-1}cat. h^{-1})[21] and formic acid (up to 5069 μmol g^{-1}cat. h^{-1}) [21] are reported depending on reaction conditions and catalyst modification, reflecting a tunable selectivity profile. Performance decay is typically linked to surface-related processes such as intermediate accumulation[22], modification of nitrogen functionalities[23], or inefficient charge separation[24]. g-C_3N_4 generally displays good chemical and photochemical stability under visible-light irradiation

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due to its bandgap of ~2.7eV[25]. Unlike many metal-based photocatalysts, it shows minimal photocorrosion and does not release toxic species into the reaction medium[26]. However, the performance enhancement strategies such as heterostructure formation[27], elemental doping[28], exfoliation[29,30], or co-catalyst loading[31] may increase the risk of oxidation or interfacial degradation under reaction conditions[32,33] even though it may also significantly improve CO_2 reduction rates and selectivity[12].

End-of-life degradation. End-of-life considerations also favor g-C_3N_4, which is made of carbon and nitrogen arranged in a heptazine-based polymeric framework[34]; this eliminates any concern related to heavy-metal toxicity, leaching, or bioaccumulation during use or disposal. Upon degradation or disposal, g-C_3N_4 does not generate persistent toxic by-products and ultimately decomposes into environmentally benign carbon- and nitrogen-containing species[35]. The absence of leachable metal ions reduces long-term ecological risks compared with metal-containing photocatalysts[36].

4.2. Titanium dioxide (TiO_2)

Material availability and supply risk. Titanium is an earth-abundant element supported by well-established global supply chains; according to the European Union (EU), titanium occurs far more abundantly in oxide mineral forms with very low supply risk [10]. Due to mineral abundance and industrial availability, as well as its chemical stability and low toxicity, TiO_2 is widely recognized as a benchmark photocatalyst; it is also among the most frequently used for $PCCO_2R$.

Sustainability of synthesis routes. Industrial TiO_2 production is primarily based on chloride and sulfate processes, both enabling large-scale manufacturing but differing in energy demand, chemical consumption, and waste generation[37]. Energy demand in these processes represents a major sustainability consideration for TiO_2 production. At the laboratory scale, synthetic strategies such as sol-gel, hydrothermal, solvothermal, and flame-based methods are also employed frequently to tailor crystal phase, morphology, and surface chemistry[38]. These methods enable precise control over material properties but may decrease sustainability due to the solvent use and multi-step processing[39]; the prolonged reaction times and solvent-intensive procedures also increase energy and water use in these methods[40].

CO_2 reduction activity and operational stability. Photocatalysts based on TiO_2 predominantly produce CO (up to 1223 μmol g^{-1}cat. h^{-1})[41] and CH_4 (up to 1290 μmol g^{-1}cat. h^{-1})[42]; the product

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distribution is influenced by crystal phase, defect density, co-catalyst loading, and reaction environment[43]. TiO_2 demonstrates excellent chemical and photochemical stability, exhibiting negligible photocorrosion under UV irradiation[44]. High structural robustness enables TiO_2 to retain catalytic functionality over repeated reaction cycles[43]. However, its wide band gap (~3.2 eV[45]) restricts visible-light absorption, necessitating modification strategies such as doping, sensitization, or heterostructure formation to enhance solar utilization[46]. While these modifications improve activity and selectivity, they may introduce additional sustainability trade-offs through increased material complexity; the durability of the modified systems also depends on the stability of secondary components rather than on the TiO_2 framework itself[47].

End-of-life degradation. Owing to its strong Ti-O bonding and chemical inertness, TiO_2 does not degrade into toxic secondary products upon disposal or environmental exposure. Instead, TiO_2 (particularly in nanoparticles) exhibits high environmental persistence. Recent reviews emphasize that the end-of-life risks associated with TiO_2 arise primarily from accumulation, rather than degradation or metal leaching[38]. However, the nano-sized TiO_2 particles may undergo aggregation, sedimentation, or surface transformation in soil and aquatic systems, and may have negative environmental impacts[48].

4.3. Copper Oxide (Cu_2O/CuO)

Material availability and supply risk. Copper oxides, including Cu_2O and CuO, have received significant attention as photocatalysts for solar-driven CO_2 reduction because of their favorable band structures, low material cost, and broad elemental availability. Copper is an industrially established and globally abundant transition metal, enabling scalable production routes and long-term supply security[49]. The EU classifies copper as a non-critical material with relatively low supply risk, although its strategic importance in energy-transition technologies is increasing[10].

Sustainability of synthesis routes. Copper is produced from various ores (mainly chalcopyrite-CuFeS2) as well as recycled scrap, while copper oxides are generally synthesized through industrially established processes such as thermal oxidation of metallic copper, chemical precipitation followed by calcination, electrochemical oxidation, and flame-based pyrolysis[50]. In the laboratory, it may also be synthesized from copper salts such as nitrates, acetates, chlorides, or sulfates through solution-based or thermal transformation routes such as hydrothermal[51] and solvothermal[52], precipitation[53], electrodeposition[54], and thermal oxidation[55]. Compared with noble

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metal-containing photocatalysts, copper oxides require low-cost precursors and moderate reaction conditions[56]; however, solvent use, long reaction times, repeated washing cycles, and post-synthesis treatments increase the energy and water use[57]. Strategies such as dopant incorporation, nanostructuring, or heterostructure engineering often add additional processing steps that elevate solvent and energy requirements[58] even though the catalytic activity and selectivity are enhanced[59].

CO_2 reduction performance and operational stability. Both copper oxides possess narrow band gaps, which are approximately 2.0-2.2 eV for Cu_2O[60] and 1.3-1.6 eV for CuO[52], enabling visible-light absorption and multi-electron CO_2 reduction pathways[61]. Studies in literature indicate that Cu-based photocatalysts predominantly produce CO (up to 87.7 μmol g^{-1}cat. h^{-1})[62] and CH_4 (up to 14.93 mmol g^{-1}cat. h^{-1})[63] as gaseous products, while liquid oxygenates such as CH_3OH (up to 1.2 mol g^{-1}cat. h^{-1}) [64] and HCOOH (up to 50 mmol g^{-1}cat. h^{-1}) [65] can be formed under specific conditions. Despite their high availability and good performance, however, the use of Cu_2O and CuO is strongly constrained by photo- and electrochemical instability[66]. Cu_2O undergoes partial self-photocorrosion under illumination through Cu^+ reduction or oxidation pathways, leading to gradual activity decay under illumination[61]. Composites[67] and heterostructures[65] can improve the activity and stability by stabilizing Cu^+ intermediates and enhancing charge separation, but may increase structural complexity and therefore sustainability trade-offs[61]. Hence, copper oxides should be regarded as conditionally stable photocatalysts, whose sustained activity relies on continuous suppression of redox-driven degradation.

End-of-life degradation. At the end-of-life, copper oxides exhibit environmental persistence rather than benign degradation. Cu_2O and CuO undergo oxidation and dissolution processes that can release Cu^+/Cu^{2+} ions into the surrounding media, leading to accumulation in sediments and soils[58]. Toxicological studies report elevated ecotoxicity, particularly for CuO, driven largely by ion-mediated oxidative stress mechanisms[68].

4.4. Zinc Oxide (ZnO)

Material availability and supply risk. Zinc is an earth-abundant element with globally established supply chains and high annual production volumes, ensuring long-term material availability[69]. The European Union classifies zinc as a non-critical material with low supply risk, although it is increasingly recognized as a strategically important material for energy and

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environmental applications[10]. The abundance and industrial maturity of zinc support the scalable production of ZnO-based photocatalysts.

Sustainability of synthesis routes. Metallic zinc is mainly produced from ZnS, which occurs naturally as sphalerite and is transformed into ZnO during conventional zinc production[8]. For photocatalytic purposes, ZnO is synthesized from common zinc salts such as zinc acetate or zinc nitrate using solution-based methods like sol-gel, hydrothermal, or precipitation routes[70]. These approaches are simple and allow easy control of particle size and morphology. Energy demand for zinc production from sphalerite (ZnS) is dominated by ore concentration, high-temperature treatment, and refining, while water is mainly consumed during flotation and washing operations[8]. Energy consumption for ZnO synthesis during heating, drying, and calcination constitutes the primary environmental concern, while washing and purification steps in solution-based synthesis also introduce additional water demand[71]. Green or plant-assisted synthesis has emerged as an alternative strategy to reduce the use of harsh chemicals, although challenges related to reproducibility and scale-up remain[72].

CO_2 reduction performance and operational stability. ZnO photocatalysts exhibit a band gap of approximately 3.0-3.3 eV[73], which favors UV-driven reactions. They produce a range of products, including CO (up to 3814 μmol g^{-1}cat. h^{-1})[74], CH_4 (up to 100 μmol g^{-1}cat. h^{-1})[75], CH_3OH (methanol) (up to 6700 μmol g^{-1}cat. h^{-1})[76], C_2H_5OH (up to 80 μmol g^{-1}cat. h^{-1})[76] and HCOOH (formic acid) (up to 1.233 mmol g^{-1}cat. h^{-1})[75]; the product distribution strongly influenced by morphology and catalyst modification. ZnO demonstrates moderate operational stability; long-term performance is often limited by photocorrosion. Strategies such as heterojunction formation or defect engineering have been shown to enhance durability by improving charge separation and suppressing self-oxidation processes[77] However, they may also increase structural complexity, introducing trade-offs between performance gains and long-term sustainability in terms of energy and material use.

End-of-life degradation. ZnO photocatalysts display environmentally reactive end-of-life behavior because partial dissolution in aqueous environments can release Zn^{2+} ions, contributing to ecological risks[78] such as potential adverse effects on aquatic organisms[79]. Long-term risk management, therefore, requires controlling dissolution processes and environmental release rather than relying on intrinsic material stability alone.

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4.5. Graphene Oxides (GO/r-GO)

Material availability and supply risk. Graphene oxide (GO) and reduced graphene oxide (r-GO) are carbon-based photocatalytic materials derived from graphitic carbon frameworks. Unlike metal-oxide photocatalysts, their composition relies primarily on carbon and oxygen, which are abundant with no supply risks[10]. Hence, material availability is determined by scalable graphite oxidation and reduction processes but not by elemental availability[80].

Sustainability of synthesis routes. GO is most commonly produced through chemical oxidation of graphite via Hummers or modified Hummers methods[81]. Modified routes aim to reduce hazardous gas release and improve scalability, although acidic media and oxidizing agents still contribute to environmental burden during large-scale production[82]. Reduced graphene oxide is typically obtained through chemical, thermal, or biological reduction of GO, partially removing oxygen functionalities to restore conductivity and electronic structure[83]. Conventional reductants such as hydrazine offer efficient reduction but raise toxicity concerns, motivating greener alternatives, including ascorbic acid or plant-extract-based reduction strategies[84,85]. The environmental profile of GO and r-GO production is largely determined by oxidation and reduction processes. Modified Hummers-type oxidation involves strong oxidants, acidic media, and extensive washing steps that increase water consumption and chemical waste generation[86]. Alternative strategies such as ball-milling or greener reduction methods aim to lower solvent demand, although energy consumption during processing remains a major environmental hotspot[87,88].

CO_2 reduction performance and operational stability. GO and r-GO function primarily as light-harvesting supports or electron mediators rather than standalone semiconductors in $PCCO_2R$ systems. Oxygen functional groups in GO introduce localized electronic states, while r-GO exhibits improved conductivity with an effective band structure (~2.0–2.5 eV), enabling enhanced charge transfer under visible-light[89]. Reported systems commonly produce CO (up to 760 µmol g^{-1}cat. h^{-1})[90] and CH_4 (up to 953.72 µmol g^{-1}cat. h^{-1})[91] as major gaseous products, whereas CH_3OH (up to 355.3 µmol g^{-1}cat. h^{-1})[92] and HCOOH (up to 1975 µmol g^{-1}cat. h^{-1})[75] formation is frequently observed in composite architectures[89]. Although GO and r-GO improve light utilization by facilitating electron transport and suppressing recombination, performance gains often depend on additional materials that may introduce sustainability trade-offs. Operational stability in GO/r-

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GO systems is closely related to defect chemistry and interfacial charge-transfer. GO may undergo partial photoreduction or surface reconstruction during irradiation, while r-GO acts as a conductive electron mediator that suppresses recombination and enhances durability in composites like Cu_2O/r-GO[93]. Long-term performance is strongly influenced by the reduction degree and defect density, with an optimized heterostructure improving robustness by distributing photogenerated charges across conductive carbon networks[94].

End-of-life degradation. Graphene oxide can undergo spontaneous oxidative aging in aqueous environments, where reactive oxygen species gradually fragment the carbon framework during long-term exposure[95]. Reduced graphene oxide generally shows greater environmental persistence due to lower oxygen content and a more graphitic structure[96]. Ecotoxicological studies indicate that interactions with natural organisms, such as algae, may mitigate oxidative stress and modify ecological impact[97]; long-term end-of-life risks of GO/r-GO depend on exposure pathways and ecosystem interactions rather than toxic material release.

4.6. $CsPbBr_3$

Material availability and supply risk. $CsPbBr_3$ is composed of cesium, lead, and bromine, which are supplied through established industrial raw material systems. According to the EU Critical Raw Materials assessment, neither of the elements involved was classified as a critical raw material[10]. Hence, the availability of $CsPbBr_3$ is not related to the elemental scarcity but may be influenced by lead-related regulatory considerations due to its high toxicity.

Sustainability of synthesis routes. $CsPbBr_3$ is commonly prepared via solution-based methods; in hot injection, the Cs-oleate precursor is introduced into a hot $PbBr_2$ solution containing organic solvents such as octadecene, oleic acid, and oleylamine. This approach allows superior control over nanocrystal size and phase but requires high temperatures (120–180°C), leading to increased energy demand[98]. Alternatively, antisolvent precipitation is performed at lower temperatures by rapidly crystallizing Cs and Pb salts from polar solvents like DMF or DMSO and antisolvents such as toluene or isopropanol. While hot injection offers better control over crystal properties, antisolvent methods are simpler and less energy-intensive; both involve solvent-intensive processing and multiple separation steps[99].

CO_2 reduction performance and operational stability. $CsPbBr_3$ is an all-inorganic halide perovskite that has received attention for photocatalytic CO_2 reduction due to its strong visible-

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light absorption, suitable band structure, and efficient charge transport. The reaction predominantly yields CO (up to 401.2 μmolg^{-1}cat.h^{-1}) [100] and CH_4 (up to 230.6 μmol g^{-1}cat. h^{-1})[101], with product selectivity strongly influenced by surface passivation, crystal morphology, and reaction environment[102]. $CsPbBr_3$ shows limited but controllable operational stability during $PCCO_2R$. Activity loss is mainly associated with surface degradation and halide loss under illumination, especially in humid environments. However, $CsPbBr_3$ can maintain photocatalytic activity over repeated cycles when surface passivation or composite structures are applied[103].

End-of-life and degradation. $CsPbBr_3$ can degrade upon exposure to water, oxygen, and light, which determines its end-of-life behavior. Degradation primarily starts at the surface and results in the formation of lead-containing species and soluble bromide salts. Several studies identify potential Pb^{2+} release during degradation as the main environmental concern, rather than long-term persistence of the material itself [104]. Bromine is generally converted into bromide salts with low environmental risk, as well as cesium, which mainly forms stable and highly soluble salts such as CsBr with low toxicity and no tendency for bioaccumulation[105].

4.7. ZnS

Material availability and supply risk. Zinc sulfide (ZnS) is a semiconductor made of zinc and sulfur, both of which are widely available elements. As explained above for ZnO, Zn is not a scarce or critical material[10]. Sulfur, which is mostly obtained as a by-product of metal smelting and fuel refining, is also abundant and does not require dedicated mining.

Sustainability of synthesis routes. For photocatalytic applications, ZnS is synthesized using controlled chemical routes. Common precursors include zinc salts such as $ZnSO_4$ or $ZnCl_2$ and sulfur sources like Na_2S, thiourea, or thioacetamide, which are industrially established and widely available[106]. Coprecipitation and hydrothermal methods are most frequently used methods, which use aqueous solutions and moderate reaction temperatures[106]; hence, they generally require low thermal energy but involve repeated washing and drying steps that increase water use[107]. However, the electricity and heat input during synthesis and post-treatment (like calcination) still dominate the overall environmental footprint, whereas water consumption plays a secondary role[108]. More advanced solvothermal or sol-gel routes are mainly used to enhance activity but increase processing complexity and energy demand[109].

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CO_2 reduction performance and operational stability. ZnS-based photocatalysts show strong reduction ability under UV irradiation due to their wide band gap and negative conduction band position with a wide bandgap of about (3.6–3.8 eV). Products are mainly C_1 species, with CO (up to 2075.7 μmolg^{-1}cat.h^{-1})[110] and formic acid (up to 234 μmolg^{-1}cat.h^{-1})[111] being the most common, while CH_4 (up to 53.72 μmolg^{-1}cat.h^{-1})[112] is occasionally observed depending on surface modification and co-catalyst loading[108]. A key limitation of ZnS is its weak visible-light absorption, which is widely enhanced by doping and heterostructure formation, while these strategies may introduce additional stability and sustainability trade-offs[113]. ZnS shows moderate operational stability during $PCCO_2R$; prolonged irradiation can accelerate surface degradation and promote partial release of Zn^{2+} ions into the surrounding medium[106].

End-of-life degradation. ZnS does not remain chemically inert at the end of its service life. Under environmental and aqueous conditions, ZnS can undergo oxidative degradation, leading to sulfur oxidation and changes in zinc-containing phases rather than long-term structural persistence[114]. Environmental toxicity assessments show that pristine ZnS nanoparticles generally exhibit low ecotoxicity, while degradation products may induce biological effects at higher concentrations[115].

4.8. CdS

Material availability and supply risk. Cadmium sulfide (CdS) is a visible-light-active semiconductor that has been extensively studied for $PCCO_2R$ due to its narrow bandgap (~2.4 eV) and strong reduction ability[116]. In terms of availability, cadmium is not geologically scarce; however, its practical use is limited primarily by toxicity rather than supply constraints; the EU Critical Raw Materials report highlights that Cd is subject to strict regulatory control driven by environmental and human health concerns[10]. As discussed in the previous section, there is no serious supply risk for sulfur.

Sustainability of synthesis routes. The environmental footprint of CdS production is largely governed by upstream cadmium extraction and refining, which are energy-intensive and associated with high cumulative environmental burdens[117]. CdS is typically synthesized from soluble cadmium salts such as cadmium nitrate, acetate, or chloride combined with sulfur sources via hydrothermal, solvothermal, or chemical precipitation methods[118]. These routes enable good control over crystal size, morphology, and interfacial contact in composite systems, which directly influences photocatalytic activity. However, toxic Cd-containing precursors are intrinsic to all

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reported synthesis pathways, increasing environmental and occupational risks during material preparation[119]. Furthermore, although CdS synthesis is typically performed at moderate temperatures, repeated washing, centrifugation, and solvent use lead to considerable water consumption and wastewater generation[120]. Additional synthesis steps, such as doping or co-catalyst deposition, increase the potential environmental risks and waste generation[116].

CO_2 reduction performance and operational stability. CdS exhibits strong visible-light absorption and high reduction potential, making it highly active for $PCCO_2R$[121]. Reported products mainly include CO (up to 2322.8 μmolg^{-1}cat.h^{-1})[121] and CH_4 (up to 18.53 μmolg^{-1}cat.h^{-1})[122], with selectivity strongly dependent on surface structure and reaction conditions[123]. Photocatalytic operation can induce partial photocorrosion of CdS, resulting in Cd^{2+} release into the reaction medium[119] directly linking catalytic performance to environmental risk. Operational stability is also a big challenge for CdS-based photocatalysts; under prolonged irradiation, CdS is prone to photocorrosion driven by oxidation of sulfide species, resulting in gradual activity loss and cadmium leaching[124]. Heterojunction construction and surface modification can temporarily suppress charge recombination and improve short-term stability[118]. However, these strategies do not eliminate the intrinsic chemical instability of CdS[116]; hence, the long-term operations remain problematic because structural degradation and toxic ion release are fundamentally linked to the CdS lattice itself.

End-of-life degradation. End-of-life degradation represents a major sustainability limitation for CdS. Upon degradation or improper disposal, CdS can release soluble cadmium species that persist in soils and aquatic systems, leading to bioaccumulation and chronic toxicity[115,120]. Toxicological assessments consistently show adverse effects of Cd on aquatic organisms and microbial communities even at low concentrations[119]; these risks cannot be fully mitigated through material design alone, significantly limiting the scalability of CdS-based photocatalysts.

4.9. $BiVO_4$

Material availability and supply risk. Bismuth and vanadium originate from mineral extraction and metallurgical refining routes[8]. Both are classified as a critical raw material in the 2023 EU assessment[10]. Bismuth is mainly produced as a by-product of lead and copper smelting, which limits direct control over its production volume and links availability to base-metal markets[10]; its reserves are location dependent, which introduces moderate vulnerability. In contrast, vanadium,

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which is typically obtained from titanomagnetite ore or petroleum residues, is listed as a critical element due to its economic importance and supply risk.

Sustainability of synthesis routes. The production of metallic elements generally requires significant energy during mining, purification, and refining stages[8]. $BiVO_4$ is typically synthesized from bismuth nitrate, bismuth oxide, or related Bi^{3+} salts combined with vanadium precursors such as ammonium metavanadate through like hydrothermal, solvothermal, precipitation, or microwave-assisted routes[125]. Most of the synthesis strategies require post-calcination at elevated temperatures, which increases energy input; water use is also significant for synthesis and washing steps[126]. Engineering approaches such as doping, heterojunction formation, and Z-scheme construction are frequently applied to improve activity but add material complexity and additional processing steps[127]. From a sustainability perspective, precursor selection and thermal treatment remain key determinants of the overall environmental profile.

CO_2 reduction performance and operational stability. $BiVO_4$ is widely reported as a visible-light-responsive semiconductor with a bandgap of approximately 2.4 eV, which enables absorption in the visible region[128]. It is rarely used as a stand-alone photocatalyst; it is widely utilized as an oxidation photocatalyst in Z-scheme or S-scheme systems for $PCCO_2R$ because its valence band position favors water oxidation, while its conduction band alone is not sufficiently negative for efficient CO_2 reduction[129]. In composite systems, $BiVO_4$ is coupled with reduction photocatalysts such as g-C_3N_4, Cu_2O, or sulfides to enhance charge separation and broaden visible-light absorption[127]. Reported CO_2 reduction products in $BiVO_4$-based systems include CO (up to 138.4 $\mu molg^{-1}cat.h^{-1}$)[130], CH_4 (up to 32.6 $\mu molg^{-1}cat.h^{-1}$)[131], and CH_3OH (up to 583.35 $\mu molg^{-1}cat.h^{-1}$)[132] depending on catalyst design and reaction conditions[129]. Although visible-light absorption is favorable due to its band gap, rapid electron-hole recombination limits intrinsic reduction efficiency[128]. Operational stability of $BiVO_4$ depends on electrolyte composition, pH, and illumination conditions. While $BiVO_4$ is more stable than many non-oxide semiconductors, photocorrosion and dissolution can occur during prolonged operation[133]. Structural recycling experiments in pollutant degradation systems indicate that morphology can remain stable over several cycles, although long-term durability under real operational conditions remains a challenge[134].

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End-of-life and degradation. At the end-of-life, the stability of $BiVO_4$ is mainly determined by its dissolution behavior and potential metal release. Experimental and thermodynamic analyses show that $BiVO_4$ has a very low solubility product, yet partial dissolution can occur depending on pH and illumination conditions[135]. While bismuth is generally regarded as having relatively low toxicity, dissolved vanadium species can present ecological and biological risks at higher concentrations. Therefore, the overall risk associated with $BiVO_4$ disposal is not inherently high but depends on environmental conditions, particularly long-term contact with water.

4.10. BiOX (X: Br, Cl, and I)

Material availability and supply risk. Bismuth oxyhalides (BiOX, X = Cl, Br, I) are layered semiconductors with average band gaps of roughly 3.3 eV, 2.8, and 1.9 eV for BiOCl, BiOBr, and BiOI, respectively[136]. As discussed above, Bi is classified as a critical raw material with considerable supply risk, while there is no such risk for the supply of halides.

Sustainability of synthesis routes. BiOX photocatalysts are most commonly synthesized via hydrothermal or solvothermal methods, typically at temperatures below 200°C[137] using Bi^{3+} precursors such as $Bi(NO_3)_3 \cdot 5H_2O$, $BiCl_3$, Bi_2O_3 (after acid dissolution), and bismuth acetate[138]; the inorganic salts such as KCl, KBr, and KI are used as halide sources while HCl or HBr can also serve both as halide source and acidity regulator in aqueous systems[139]. Water is the most frequently employed solvent in hydrothermal synthesis, yet mixed solvents such as ethylene glycol/water or alcohol-based systems are often introduced to control crystal growth, defect density, and morphology[140]. In addition to solvent use, repeated washing steps require significant water consumption, while the drying procedure contributes to energy demand significantly, even though reaction temperature is low[141].

CO_2 reduction performance and operational stability. BiOX photocatalysts mainly produce CO (up to 144.5 $\mu mol g^{-1} cat. h^{-1}$)[142] as the dominant product during $PCCO_2R$, while CH_4(up to 11.93 $\mu mol g^{-1} cat. h^{-1}$)[143] is typically formed in smaller amounts under similar conditions[141]. The layered structure and internal electric field are often cited as factors supporting charge separation and selective CO formation[144]. The recombination of photogenerated carriers remains a limiting factor in pristine systems, which is why recent reviews emphasize defect control and structural modulation to improve effective light use[138]. BiOX materials are generally described as structurally stable under light irradiation[138]. The internal electric field between Bi_2O_2 slabs and

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halogen layers promotes charge separation, thereby helping maintain photocatalytic activity over repeated cycles[145]. However, several studies also report that defect concentration strongly influences durability; moderate oxygen vacancies can improve electronic properties, while excessive defect formation may disturb the lattice and reduce long-term stability[146].

End-of-life and degradation. BiOX photocatalysts are generally considered chemically stable, and the formation of secondary toxic phases has not been observed under typical photocatalytic conditions. Activity loss is generally linked to carrier recombination, surface passivation, or excessive vacancy formation[144]. The halide component (Cl^-, Br^-, or I^-) may become mobile under certain aqueous conditions; however, they are naturally present in water systems and are not inherently harmful at typical concentrations. Additionally, bismuth compounds are regarded as having relatively low toxicity.

4.11. Summary of analysis

Our analysis for semiconductors is summarized in Table 1 while the details are given in SI; Table S2-S10 explains the scales used for various criteria while Table S11 provides detailed explanations for the results in Table 1. g-C_3N_4 appears to be the most sustainable semiconductor among the alternatives discussed above; it has high material availability with reasonable CO_2 reduction performance and very low environmental impact even though it is energy-sensitive due to calcination. TiO_2-based photocatalysts are also sustainable in terms of material availability and environmental impacts; however, they are more energy-dependent, and the modifications required to improve their visible-light activity may also increase their environmental burden, making TiO_2 somewhat less sustainable than g-C_3N_4. Cu_2O/CuO, ZnO, GO/g-GO, ZnS can be considered moderately sustainable as all have high availability, low supply risks; however, they may have some moderate impact on the environment, as can be seen from Table 1.

The sustainability of $BiVO_4$ can be considered as low; both Bi and V are in the critical material list of the EU with limited availability and significant supply risks, while the energy and water consumption through the life cycle of $BiVO_4$ is also high. The same is also true for BiOX due to its Bi contain even though the oxygen and halide contents pose no concern for sustainability. $CsPbBr_3$ and CdS, on the other hand, cannot be considered sustainable due to the high toxicity of Pb and Cd. Even though these elements are not considered critical in terms of availability, the EU Critical Raw Materials report highlights that both should be subject to regulatory control, which

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may impose some supply risks; more importantly, these elements possess significant risk to human health and the environment, which makes it unjustifiable to use them in large-scale operations when other more environmentally benign alternatives exist.

Table 1. Assessment of major semiconductors used in the PCCO$_2$R process

Material	**Abundance**	**Industrial maturity**	**Process complexity**	**Supply risk (EU)**[10]	**Energy intensity (relative)**	**Water consumption**	**Human/ environmental toxicity**	**PCCO$_2$R performance**	**Overall sustainability**
g-C$_3$N$_4$	🟩 **Very high**	🟨 **Moderate**	🟩 **Low**	🟩 **Very low**	🟨 **Moderate**	🟩 **Low**	🟩 **Very low**	🟩 **High - Moderate**	🟩 **Very high**
TiO$_2$	🟩 **Very high**	🟩 **Very high**	🟩 **Low**	🟩 **Very low**	🟧 **High**	🟨 **Moderate**	🟩 **Low**	🟩 **High**	🟩 **High**
Cu$_2$O/CuO	🟩 **Very high**	🟩 **High**	🟧 **High**	🟨 **Moderate**	🟧 **High**	🟨 **Moderate**	🟨 **Moderate**	🟩 **High**	🟨 **Moderate**
ZnO	🟩 **High**	🟩 **High**	🟨 **Moderate**	🟩 **Low**	🟨 **Moderate**	🟨 **Moderate**	🟨 **Moderate**	🟩 **Very high**	🟨 **Moderate - High**
r-GO/GO	🟩 **High**	🟧 **Low**	🟧 **High**	🟩 **Low**	🟧 **High**	🟥 **Very High**	🟩 **Low -Moderate**	🟨 **Moderate**	🟨 **Moderate**
CsPbBr$_3$	🟨 **Moderate**	🟨 **Moderate**	🟧 **High**	🟧 **High**	🟧 **High**	🟧 **High**	🟥 **Very high**	🟨 **Moderate**	🟥 **Very low**
ZnS	🟩 **High**	🟩 **High**	🟨 **Moderate**	🟩 **Low**	🟨 **Moderate**	🟨 **Moderate**	🟨 **Moderate**	🟨 **Moderate**	🟨 **Moderate**
CdS	🟨 **Moderate**	🟨 **Moderate**	🟧 **High**	🟧 **High**	🟧 **High**	🟧 **High**	🟥 **Very high**	🟨 **Moderate**	🟥 **Very low**
BiVO$_4$	🟨 **Moderate**	🟨 **Moderate**	🟧 **High**	🟧 **High**	🟧 **High**	🟧 **High**	🟨 **Moderate**	🟧 **Low**	🟧 **Low**
BiOX (Cl/Br/I)	🟨 **Moderate**	🟨 **Moderate**	🟨 **Moderate**	🟧 **High**	🟨 **Moderate**	🟧 **High**	🟩 **Low-Moderate**	🟥 **Very high**	🟧 **Low-Moderate**

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5. Review of co-catalysts with a sustainability viewpoint

Several co-catalysts, including noble metals such as Pt, Au, Pd, Ag, and Rh, as well as metal/metal oxide like Cu, Ni, and Cd, and alloy structures like Au/Cu and Pt/Cu, have been coupled with semiconductors to improve photocatalytic performance[147]. Even though co-catalysts are generally used in small amounts, and they have been shown to considerably enhance the efficiency of the process through improving charge separation and reaction rate, they may also have significant inverse effects on sustainability because they are mostly metals; while some of them suffers from high toxicity, some other has significant supply risks. The top 10 most commonly appearing co-catalysts in our previous work[2] in Fig.5

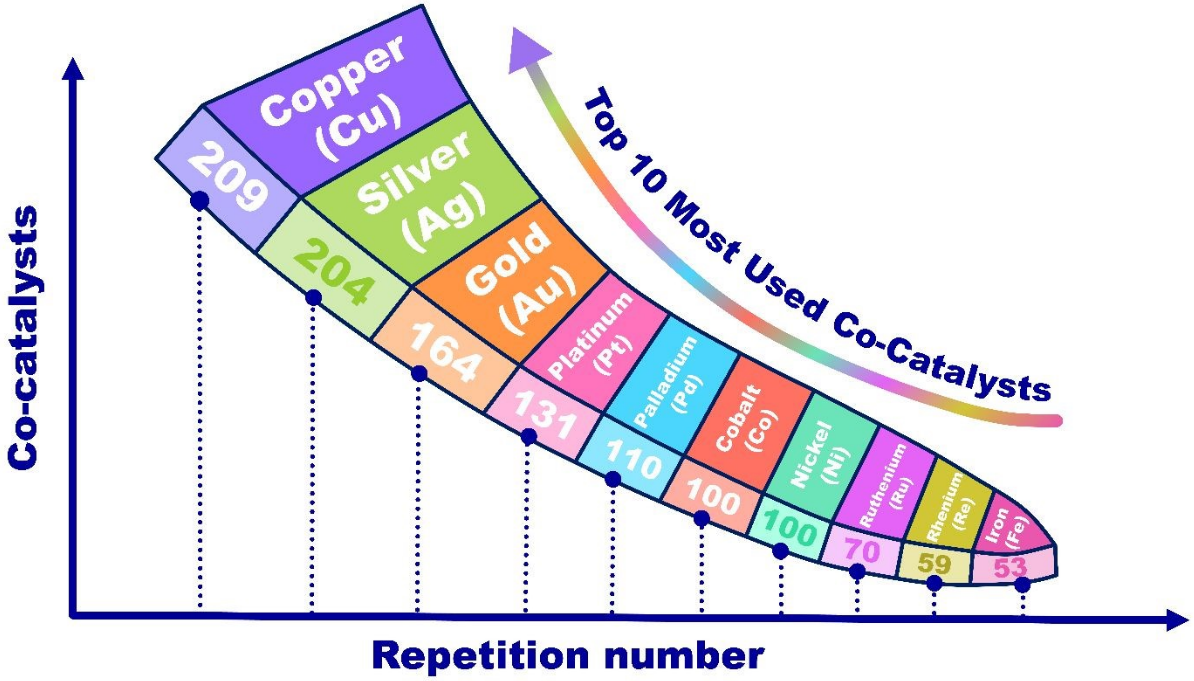

Fig.5 Top 10 most observed co-catalysts extracted from author keywords analysis of published papers on the topic of the $PCCO_2R$[2].

5.1. Copper (Cu)

Material availability and supply risk. Copper is also used as the most common co-catalyst in CO_2 reduction. As discussed above, it is a globally abundant; the EU classifies copper as non-critical with relatively low supply risk; however, it lists Cu as a strategic raw material[10].

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Sustainability of synthesis routes. As a co-catalyst, Cu is loaded on the semiconductor through photodeposition[148], impregnation[149], chemical reduction[150], or electrodeposition[151]. These routes require low metal loading and relatively mild conditions, reducing material consumption[152]; at a larger scale, wet impregnation followed by thermal or hydrogen reduction remains a widely used industrial strategy due to its compatibility with continuous catalyst production[153]. Overall sustainability depends more on the use of solvent and reducing agents, as well as post-treatment processes (including washing, drying, and calcination) that contribute significantly to water and energy consumption[154].

CO_2 reduction performance, selectivity, and operational stability. Copper co-catalysts strongly influence product distribution and light utilization by acting as electron sinks and active reduction sites. Atomically dispersed Cu centers enhance CO selectivity by stabilizing reaction intermediates[155] while alloyed systems allow tunable syngas production under visible light[156]. Reported Cu-modified photocatalysts predominantly generate CO (up to 870 μmol g^{-1}cat. h^{-1})[157] and CH_4 (up to 43.15 μmol g^{-1}cat. h^{-1})[158]. Liquid products such as CH_3OH (up to 627.66 μmol g^{-1}cat. h^{-1})[159] and occasionally HCOOH formation (up to 31 μmol g^{-1}cat. h^{-1})[160] are also reported depending on semiconductor support and reaction conditions[154]. Atomically dispersed Cu species maintain activity over repeated cycles by stabilizing intermediates and limiting aggregation[155], whereas nanoparticle-based systems may experience gradual restructuring and activity decline under prolonged illumination[161].

End-of-life degradation. At the end-of-life, Cu co-catalysts are not completely inert and may undergo oxidation, dissolution, or surface restructuring depending on environmental conditions. Cu^0/Cu^+ species can transform into more oxidized states, affecting mobility and long-term stability[154]. In composite photocatalysts, degradation behavior is often governed by interactions with the semiconductor support rather than copper itself[161].

5.2. Silver (Ag)

Material availability and supply risk. Silver is a precious metal widely used in electronic and energy technologies, yet its long-term availability is shaped more by demand pressure and material flows than geological scarcity. The EU classifies silver as a non-critical material with relatively low supply risk, although future market dynamics may alter this balance[10]; as a precious material, its price and availability are highly dependent on economic and political developments.

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Sustainability of synthesis routes. Silver is obtained as an element through mining; the life-cycle studies show that primary silver production is associated with substantial environmental burdens, reinforcing the need for circular strategies and recycling to support long-term stability[162,163]. Ag co-catalyst preparation is strongly influenced by the selected synthesis pathway; wet-chemical reduction routes remain widely used because they allow controlled particle size and dispersion, yet they require stabilizers, solvents, and multiple processing steps that contribute to environmental burdens through water and energy consumptions[164]. Emerging one-pot and bio-based approaches aim to simplify reactions and reduce hazardous chemicals, highlighting a shift toward greener synthesis concepts[164].

CO_2 reduction performance, selectivity, and operational stability. Ag co-catalysts mainly regulate reaction pathways by improving charge separation and stabilizing reaction intermediates rather than acting as the primary catalytic sites. Reported systems commonly produce CO (up to 80 μmol g^{-1}cat. h^{-1})[165], CH_4 (up to 113.84 μmol g^{-1}cat. h^{-1})[166], and liquid products such as HCOOH (up to 421 μmol g^{-1}cat. h^{-1})[165], with selectivity strongly influenced by support structure and Ag loading[167]. Ag also enhances light utilization through plasmonic effects and electron trapping, which improves reaction efficiency under irradiation[168]. Operational stability is generally governed by nanoparticle dispersion and metal–support interactions; generally, stable activity is observed over multiple reaction cycles without significant deactivation[168].

End-of-life degradation. Silver nanoparticles can undergo oxidation, aggregation, and sulfidation in aqueous environments, changing their ecological impacts[169]; nano-Ag released during disposal may transform into less soluble species such as Ag_2S or AgCl, reducing short-term toxicity but increasing long-term persistence in sediments and wastewater systems[170]. Although these transformations can stabilize the metallic phase, dissolution of Ag^+ ions remains a key factor controlling bioavailability and potential ecotoxicity[171]. Recycling strategies and controlled containment are essential to reduce long-term ecological risks associated with Ag-based co-catalysts.

5.3. Gold (Au)

Material availability and supply risk. Gold is a precious noble metal characterized by limited natural abundance and a supply chain largely dependent on global market dynamics[10]; according to the EU, Au is a non-critical element, but it may create strategic vulnerabilities under specific

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trade conditions[10]. Hence, maximizing catalytic efficiency while minimizing Au loading becomes essential to balance performance and long-term resource availability.

Sustainability of synthesis routes. Au is obtained as an element through mining that causes significant environmental concerns; the resource consumption arises mainly from mining-related activities, making metal efficiency a key sustainability parameter[8]. Au co-catalysts are typically introduced onto semiconductor supports through in situ photoreduction, deposition–precipitation, or reductive immobilization of Au^{3+} precursors. Approaches that enable direct nucleation of metallic Au on photoactive surfaces are particularly attractive because they enhance electronic contact while avoiding excessive stabilizing agents[172]. Methods relying on strong chemical reductants can offer precise size control, yet they may increase reagent demand and generate additional wastes[173]. Simplified and ligand-free deposition strategies therefore provide a more sustainable pathway for catalyst preparation.

CO_2 reduction performance, selectivity, and operational stability. In $PCCO_2R$, Au nanoparticles mainly influence reaction pathways by facilitating electron transfer and enabling plasmon-induced excitation under visible light. Enhanced formation of products such as CO (up to 8982 μmol g^{-1}cat. h^{-1})[174], CH_4 (up to 82.65 μmol g^{-1}cat. h^{-1})[174], or CH_3OH (up to 232 μmol g^{-1}cat. h^{-1})[76] has been reported depending on semiconductor composition and reaction environment[175]. Localized surface plasmon resonance promotes hot-carrier generation, extending light absorption beyond the intrinsic band gap of many supports[176]. However, improvements in activity must be balanced against increased material complexity and noble-metal usage. Particle reshaping, migration or aggregation under irradiation can alter plasmonic behavior and active surface area[177–179]. Designing strong metal–support interactions and maintaining narrow particle size distributions are therefore critical for achieving stable operation.

End-of-life degradation. Gold exhibits high chemical inertness, which generally limits dissolution during catalyst use; nevertheless, nanoscale transformations and environmental accumulation remain relevant considerations[8]. Immobilization of solid support can reduce mobility, while recycling strategies are essential to offset the high resource intensity associated with Au production.

5.4. Platinum (Pt)

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Material availability and supply risk. Platinum is a noble metal with high economic value; the EU classifies it as a critical raw materials having supply risks due to economic importance and the reserves that are located in limited regions of earth[10]. Pt is one of the most common catalysis materials; it is also deposited onto semiconductor photocatalysts to accelerate reduction kinetics and stabilize photogenerated electrons during $PCCO_2R$.

Sustainability of synthesis routes. Platinum is extracted from earth open-pit mines, refined, and produced as pure platinum[180]. Formation of Pt nanostructures on semiconductor surfaces is commonly achieved through photochemical reduction, deposition-precipitation, or reductive immobilization of precursors like chloroplatinate precursors[147]. For example, Pt-modified TiO_2 systems show that optimized dispersion can significantly enhance CO_2 reduction performance without requiring high noble-metal loading[181]. LCA studies indicate that Pt (and other Pt group elements) have a very high burden on the environment, especially through extraction and refining[8] even though they are usually needed in small fractions; the energy, solvent, and water demand in laboratory scale applications may also contribute to the environmental profile of Pt.

CO_2 reduction performance, selectivity, and operational stability. Pt primarily modifies reaction pathways by facilitating electron transfer and stabilizing reduction intermediates. Pt-decorated semiconductors typically yield CH_4 and CO as the dominant reduction products; they may be formed up to 83.6 μmol g^{-1}cat. h^{-1} [182] and up to 4.2 μmol g^{-1}cat. h^{-1} [183] respectively depending on photocatalyst support composition; CH_3OH (up to 17.85 μmol g^{-1}cat. h^{-1}) may be also formed in liquid phase[184]. Changes in activity during prolonged operation are typically associated with nanoscale restructuring or aggregation of Pt particles.

End-of-life degradation. Due to its noble character, Pt shows limited dissolution under photocatalytic conditions. However, the environmental burden associated with platinum extraction highlights the importance of catalyst recovery and recycling instead of releasing it to the environment.

5.5. Palladium (Pd)

Material availability and supply risk. Palladium is a strategic platinum-group metal (PGM) characterized by geographically concentrated reserves, creating long-term supply vulnerability[185]. The EU lists Pd, as other PGM, as critical and evaluates the supply risk through economic

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importance and sourcing concentration[10]; expanding use in environmental and catalytic technologies further increases pressure on primary resources and may influence future supply stability[186].

Sustainability of synthesis routes. Pd is also extracted from the earth through open-pit mining with significant impacts on the environment. In laboratory environments, the organometallic precursors such as $Pd(acac)_2$ are used for the controlled formation of highly dispersed metallic nanoparticles through low-temperature thermal or photochemical pathways[187]. Similar to Pt, the environmental burdens (especially energy and fresh water use) per unit mass are mostly linked to mining and refining; extraction and linking availability to sustainability considerations[188]; at the catalyst scale, deposition procedures and thermal post-treatments also contributes to environmental impacts through additional energy inputs, water consumption (due to the washing and purification steps) indicating the needs for lower Pd loadings and mild preparation conditions.

CO_2 reduction performance, selectivity, and operational stability. During $PCCO_2R$, Pd co-catalysts alter reaction pathways by favoring deeper reduction routes. Reported systems predominantly produce CH_4 (up to 39.1 μmol g^{-1}cat. h^{-1})[189], whereas CO (up to 581.9 μmol g^{-1}cat. h^{-1})[190] and occasionally CH_3OH (up to 3.17 μmol g^{-1}cat. h^{-1})[191] appear as secondary products. Operational durability is governed largely by surface chemistry; prolonged irradiation may alter the Pd oxidation state and induce partial PdO formation, gradually affecting activity[192]. Strategies that confine nanoparticles or strengthen metal-support interactions help maintain dispersion and suppress aggregation during repeated use[193].

End-of-life degradation. Pd-based photocatalysts generally maintain structural integrity during operation, yet their end-of-life impact is related to metal persistence; palladium species released from catalytic systems may accumulate in soil and aquatic environments, raising concerns regarding ecological exposure pathways[194]. Overall, end-of-life risks relate less to catalyst breakdown and more to recycling efficiency, resource recovery, and controlled containment of palladium species. Hence, as the other Pt-group metals, recycling strategies should play significant roles in maintaining resource security and minimizing environmental impacts.

5.6. Cobalt (Co)

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Material availability and supply risk. Cobalt is an important transition metal widely used in catalysts, batteries, and alloy production, but its supply is limited and unevenly distributed. The EU 2023 Critical Raw Materials Report classifies cobalt as a critical raw material because production is concentrated in a small number of countries, which increases supply risk[10].

Sustainability of synthesis routes. Cobalt is produced as a by-product of copper and nickel mining, which increases vulnerability to supply chain disruptions[195]; it is obtained from copper-cobalt ores through chemical processes such as acid or ammonia leaching, which cause considerable environmental impacts[196]. Cobalt exposure can also pose risks to nearby communities; it leads to deforestation, biodiversity loss, and landscape disturbance[197]. Cobalt-based co-catalysts, which are commonly prepared by impregnation, solvothermal processes, photodeposition, or chemical reduction, although these routes allow good control over cobalt dispersion and oxidation state[198], may also have negative impacts on environments through the use of chemicals and energy-intensive steps like calcination[199]. These factors undermine its long-term sustainability despite its catalytic usefulness.

CO_2 reduction performance, selectivity, and operational stability. Cobalt co-catalysts are widely studied because they improve charge separation and promote CO_2 activation at the catalyst surface[200]. Different cobalt structures, such as oxides, sulfides, phosphides, MOFs, and single-atom sites, can tune adsorption strength and stabilize key intermediates[201], enabling selective formation of CO (up to 2075.7 μmol g^{-1}cat. h^{-1})[110] and CH_4 (up to 20.63 μmol g^{-1}cat. h^{-1})[202]. Cobalt compounds show good resistance to photocorrosion and allow stable redox cycling, supporting long-term operation[203].

End-of-life degradation. End-of-life management of cobalt-based catalysts is important due to environmental and health concerns over toxicity and environmental burden. Recovering cobalt as hydroxide, sulfate, or carbonate can reduce environmental impacts[199].

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5.7. Nickel (Ni)

Material availability and supply risk. Nickel is also widely used in alloys, batteries, and catalytic systems, which makes it economically important. It is not classified as a critical raw material by the European Commission, but it is listed as a Strategic Raw Material due to its growing role in clean-energy technologies[10].

Sustainability of synthesis routes. Nickel is produced through mining of various ores in two general types: sulfides and oxides; processing routes may significantly influence sustainability, since some ore types require higher energy input[204]. Nickel co-catalysts can be synthesized and integrated into semiconductor surfaces mainly by photodeposition[9], wet impregnation[205], hydrothermal or solvothermal synthesis for structural control[206], and doping into the semiconductor lattice[207]. These techniques generally use moderate temperatures and accessible precursors, which support practical, scalable, and sustainable production[9]. Reducing solvent use and lowering calcination temperatures can further improve the overall sustainability of nickel-based photocatalysts[204].

CO_2 reduction performance, selectivity, and operational stability. Ni, as a co-catalyst, improves CO_2 photoreduction by enhancing charge separation and directing photogenerated electrons to surface reduction reactions[208]. In Ni-based systems, the dominant product is CO (up to 599 μmol g^{-1}cat. h^{-1})[209] and CH_4 (up to 29.65 μmol g^{-1}cat. h^{-1})[210] because Ni sites favor CO_2-to-CO pathways, and some tailored Ni catalysts can achieve very high CO selectivity while suppressing hydrogen evolution[205,211]. Nickel co-catalysts are reasonably stable under reaction conditions; their variable oxidation states and strong interactions with the support help maintain active surface sites without rapid deactivation during prolonged illumination[208]; however, they do not remain chemically unchanged after long-term operation under light irradiation and in aqueous systems. Changes in nickel oxidation state can influence stability and surface reactivity over time[9]. Small nickel particles may also aggregate, which reduces the number of accessible active sites[205].

End-of-life degradation. Environmental risks mainly depend on whether nickel is released in soluble form, since soluble nickel species are more mobile and more easily taken up by organisms, increasing potential ecological and health risks[153]. In supported photocatalysts, nickel is usually immobilized, which limits leaching and reduces environmental exposure[153]. LCA studies show

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that extraction and refining determine the overall environmental impact significantly, highlighting the importance of the recovery and recycling of used Ni[204].

5.8. Ruthenium (Ru)

Material availability and supply risk. Ruthenium (Ru) is a PGM obtained mainly as a by-product of platinum and nickel mining. Because it is geologically scarce and produced in limited regions, its supply is sensitive to geopolitical and market conditions[212]. Like other PGMs, the EU classifies Ru as critical due to its high supply risk and economic importance[10]. Despite low usage (below 1 wt% in PCCO$_2$R), supply vulnerability remains a concern for large-scale applications [213,214].

Sustainability of synthesis routes. Ru-based photocatalysts are generally prepared by impregnation, photodeposition, sol-gel processing, or encapsulation within porous supports such as MOFs. While these routes are relatively simple and scalable, their sustainability depends strongly on solvent use, reducing agents, and post-calcination temperatures. Strategies that stabilize ruthenium as highly dispersed nanoparticles or single atoms are particularly advantageous, since they maximize atomic utilization, reduce the required metal loading, and help lower the overall demand for this scarce and energy-intensive Ru metal[215].

CO$_2$ reduction performance, selectivity, and operational stability. Ruthenium is widely used as a co-catalyst in PCCO$_2$R because it facilitates interfacial electron transfer and promotes multi-electron reactions. When deposited on semiconductors such as TiO$_2$, ZnS, carbon nitride, or MOF-based supports, Ru species can act as electron sinks, suppress charge recombination, and provide active sites for CO$_2$ activation[216]. Products typically include CO (up to 124.8 μmol g^{-1}cat. h^{-1})[217] and CH$_4$ (up to 19.8 μmol g^{-1}cat. h^{-1})[217], depending on the support, oxidation state of Ru, and reaction conditions[218]. Ru-based systems generally show good resistance to photocorrosion and maintain activity over repeated cycles, although aggregation or oxidation-state changes under prolonged irradiation can gradually affect performance[219]. Ruthenium is generally stable, but long-term photocatalytic use can affect its structure and activity. During repeated redox cycles, Ru may shift between oxidation states (Ru^0, Ru^{2+}, Ru^{4+}), which can gradually change its surface chemistry and reduce active site efficiency. Another key degradation pathway is aggregation. Isolated Ru atoms or very small clusters may migrate and form larger particles over time[214].

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End-of-life degradation.

Because Ru is usually used in very low amounts, the overall environmental burden is limited; it is also quite rare that it is normally expected to be recycled. However, improper disposal may lead to metal loss and resource waste. Ru is generally considered less hazardous than some heavy metals, but soluble Ru compounds may pose health and environmental risks if released[8].

5.9. Rhenium (Re)

Material availability and supply risk. Rhenium is one of the rarest elements in the Earth's crust; it is not mined from its own ores but recovered as a by-product of molybdenum and copper processing, mainly from large copper deposits in Chile, which supply more than half of global production[220]. This strong geographical concentration increases supply risk. The European Union considers rhenium strategically important because of its low availability and limited substitution options[10]. Since rhenium production depends on copper mining activity, fluctuations in copper markets directly affect its availability. Although recycling is increasing, the primary supply remains limited. Growing demand from aerospace superalloys and catalytic applications further increases the pressure on this already constrained resource[221].

Sustainability of synthesis routes. Rhenium is most commonly applied as a molecular Re complex in photocatalytic CO_2 reduction systems[222]. These complexes are typically prepared through solution-based coordination reactions under relatively mild laboratory conditions. While such methods allow structural tunability and controlled catalytic properties, they often require organic solvents and additional reagents, which increase chemical consumption[223]. To improve stability and recyclability, Re complexes are frequently immobilized on porous supports like MOFs[224]. Although this strategy can enhance durability and facilitate catalyst recovery, additional modification steps may increase material and energy input. Therefore, the sustainability of Re-based systems depends largely on solvent choice, metal loading, and long-term catalyst stability rather than on the complexity of the coordination chemistry itself.

CO_2 reduction performance, selectivity, and operational stability. Rhenium is widely used as a molecular Re complex in $PCCO_2R$ due to its high activity and strong selectivity toward CO formation (up to 76.8 μmol g^{-1}cat. h^{-1})[225]. The preference for CO production is related to the favorable binding and activation of CO_2 at the Re center, which stabilizes key reaction intermediates[222]. In terms of operational stability, Re complexes can suffer from deactivation

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through aggregation or structural changes during long irradiation times[222]. If Re nanoparticles are weakly attached to the support, repeated redox cycles during CO_2 reduction can detach or dissolve small Re species into the reaction medium[226,227]. Immobilization on solid supports generally improves durability and recyclability while maintaining high CO selectivity[224].

End-of-life degradation. Rhenium degradation typically involves oxidation, dissolution, and structural changes. Under light irradiation and in the presence of oxygen or water, metallic Re can gradually oxidize to perrhenate, which is soluble and relatively stable in the environment[224]. Because rhenium is scarce and expensive, recycling is well developed. Optimized modern recovery processes can extract more than 98% of rhenium by applying controlled oxidation, alkaline treatment, or a combination of thermal and hydrometallurgical steps[221]. Re has low environmental toxicity compared to many heavy metals, although high concentrations may still require controlled waste management[220].

5.10. Iron (Fe)

Material availability and supply risk. Iron is one of the most abundant elements in the Earth's crust and has a well-established global supply chain [228]. Due to its wide distribution and strong industrial base, Fe is classified as a non-critical raw material in the latest EU assessment, indicating low supply risk[10]. Fe is an attractive alternative because it is abundant and cheap. Studies also identify Fe as a promising option due to its suitable electronic structure for CO_2 activation[153].

Sustainability of synthesis routes. Iron is produced through mined ore or recycled materials. Although mining has environmental impacts[229], the use of its small amounts of co-catalysts limits environmental concerns. Fe-based co-catalysts are generally prepared through simple and scalable methods such as impregnation, co-precipitation, hydrothermal treatment, and in situ growth on semiconductor supports[230]. In most cases, common and low-cost precursors such as $FeCl_3$, $Fe(NO_3)_3$, or $FeSO_4$ are used, which are widely available and industrially produced from abundant iron resources[153]; these iron salts reduce both material cost and supply risk. Many Fe-containing systems, including Fe-MOFs and Fe-doped oxides, can be synthesized under relatively mild temperatures and atmospheric pressure, lowering overall energy demand[231]. In several studies, Fe species are introduced by wet-chemical routes followed by moderate thermal treatment, avoiding complex multistep procedures[232]. Moreover, Fe doping or surface modification often requires low metal loading, which further limits resource consumption[233]. Overall, the sustainability of Fe co-

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catalyst synthesis depends mainly on solvent use and post-treatment conditions, but the abundance and low toxicity of iron make these routes more environmentally favorable.

CO_2 reduction performance, selectivity, and operational stability. Fe-based co-catalysts play an important role in improving CO_2 adsorption and activation, while also promoting interfacial charge transfer[230]. The main products reported for Fe-modified photocatalysts are CO (up to 122 μmol g^{-1}cat. h^{-1})[232] and CH_4 (up to 30 μmol g^{-1}cat. h^{-1})[234], depending on the support and reaction conditions, and CO is the dominant product due to the relatively lower kinetic barrier for the two-electron reduction pathway[230]. However, when electron density around Fe sites is optimized, higher selectivity toward CH_4 has also been observed[233]. Fe-based co-catalysts generally show good durability under light irradiation because iron is chemically stable and resistant to severe photocorrosion[153]. Deactivation, when observed, is often related to surface oxidation state changes or aggregation.

End-of-life degradation. End-of-life management of Fe co-catalysts is generally straightforward because iron is non-toxic, widely recyclable, and already integrated into large industrial waste streams[229]. Unlike noble metals, Fe does not require complex recovery systems due to it abundance. Most Fe-based photocatalysts gradually convert into iron oxides or hydroxides during use, which are stable and environmentally benign forms. LCA indicates that Fe compounds do not create long-term ecotoxic risks when properly managed[228].

5.11. Summary of analysis

Table 2 summarizes the results of sustainability analysis for co-catalysts, while the details are provided in the SI; similar to semiconductors, the scales used for various criteria were given in Table S11-S20, and the results in Table 2 are explained further in Table S21. According to our analysis, copper and iron have the highest sustainability among the most frequently employed co-catalysts. Both elements are abundant in nature with well-established supply channels without any significant risks (even though the EU declared Cu as a strategic material). Although they may have some environmental burden in the mining and refining stage, they are used in small amounts in catalysis and deposited on semiconductors using well-established precursors with relatively mild procedures. CO_2 reduction performance of Cu is high while Fe has moderate activity; their end-of-life impacts on the environment are also moderate. Ni, which is also labeled as a strategic material by the EU, can also be considered in the sustainable co-catalyst category even though its toxicity

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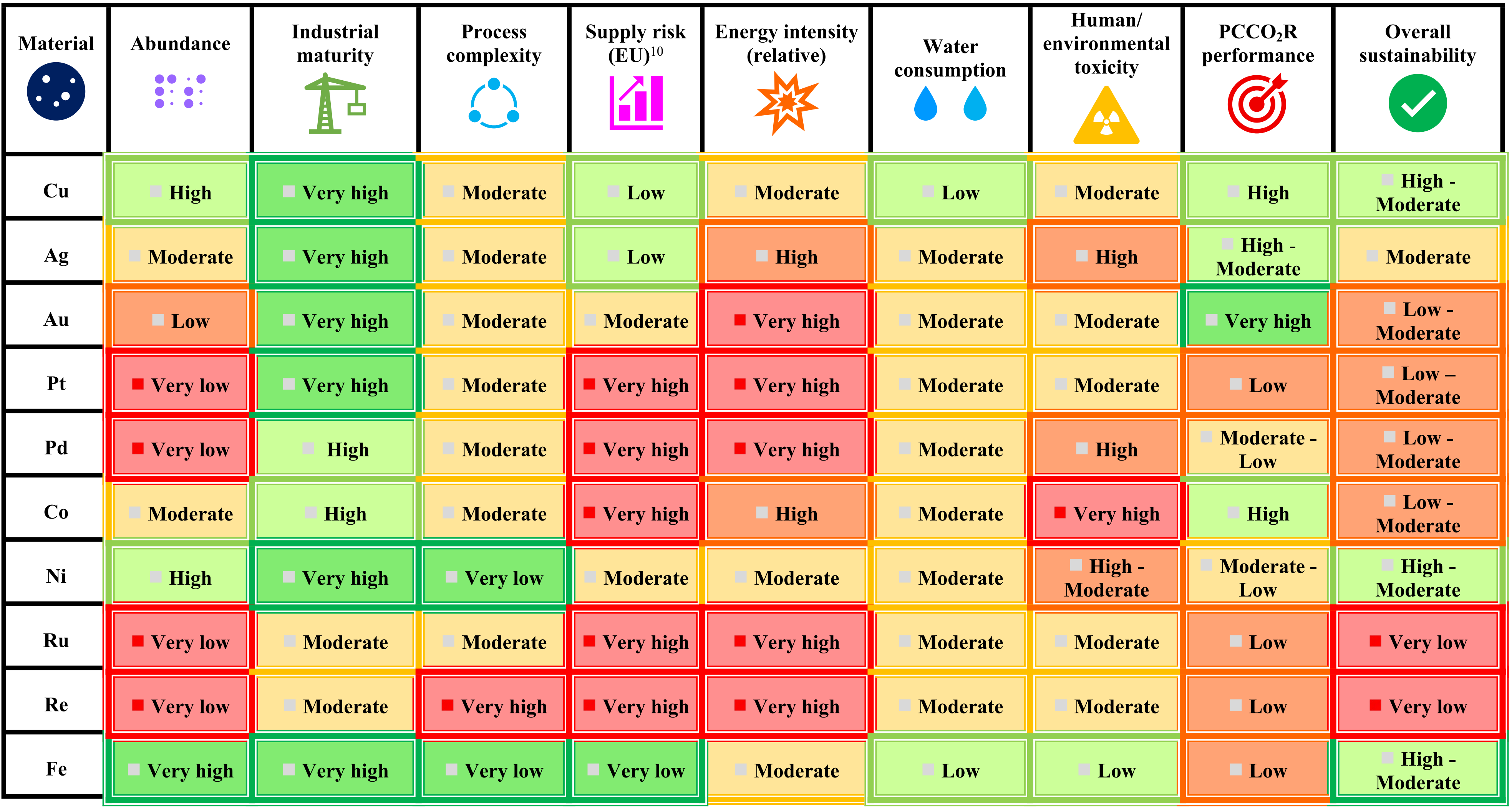
is higher than cu and Fe. Ag, as one of the most commonly utilized co-catalysts, however, can be considered only moderately sustainable due to its environmental burden through its life cycle and relatively high end-of-life toxicity of its compounds.

On the other hand, the sustainability levels of all precious metals are low due to their low availability, significant supply risks, price fluctuations, and heavy environmental burden they cause, especially during mining and refining. Especially Ru and Re cannot be treated as sustainable in commercial-level applications due to their scarcity; they should be avoided because any higher value-added industry needing these elements may create significant supply risk for a commercial CO_2 reduction, which will create low economic value by its nature. It seems that cobalt should also be avoided due to its high toxicity and serious social/environmental impacts caused throughout its life cycle.

Table 2. Assessment of major co-catalysts used in the $PCCO_2R$ process



Material	Abundance	Industrial maturity	Process complexity	Supply risk (EU)[10]	Energy intensity (relative)	Water consumption	Human/ environmental toxicity	$PCCO_2R$ performance	Overall sustainability
Cu	High	Very high	Moderate	Low	Moderate	Low	Moderate	High	High - Moderate
Ag	Moderate	Very high	Moderate	Low	High	Moderate	High	High - Moderate	Moderate
Au	Low	Very high	Moderate	Moderate	Very high	Moderate	Moderate	Very high	Low - Moderate
Pt	Very low	Very high	Moderate	Very high	Very high	Moderate	Moderate	Low	Low – Moderate
Pd	Very low	High	Moderate	Very high	Very high	Moderate	High	Moderate - Low	Low - Moderate
Co	Moderate	High	Moderate	Very high	High	Moderate	Very high	High	Low - Moderate
Ni	High	Very high	Very low	Moderate	Moderate	Moderate	High - Moderate	Moderate - Low	High - Moderate
Ru	Very low	Moderate	Moderate	Very high	Very high	Moderate	Moderate	Low	Very low
Re	Very low	Moderate	Very high	Very high	Very high	Moderate	Moderate	Low	Very low
Fe	Very high	Very high	Very low	Very low	Moderate	Low	Low	Low	High - Moderate

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6. Review of reactor structures with a sustainability viewpoint

Photoreactor structure is one of the most significant factors influencing the performance of photocatalytic CO_2 reduction by controlling illumination efficiency, mass transfer, catalyst-reactant interactions, product separation, catalyst recovery, and operational stability. These considerations are also relevant to process sustainability, as they determine energy and material utilization, catalyst handling, product separation requirements, system durability, and the feasibility of scale-up. Photoreactors employed in $PCCO_2R$ can be classified according to their mode of operation, the phases, and the catalyst beds. The way in which the catalyst is positioned within the photoreactor provides a direct basis for comparing reactor performance and sustainability. Accordingly, the most commonly encountered configurations in literature include slurry, fixed-bed, and membrane-based photoreactors[2] as presented in Fig. 6

Slurry photoreactors are largely used in liquid-phase $PCCO_2R$ owing to their simple construction, low cost, and available surface area. In these systems, CO_2 is bubbled through a reaction medium in which photocatalyst particles are dispersed under continuous mixing, which ensures an effective contact between gas, liquid, and solid phases as well as exposes a large fraction of the catalyst to irradiation[2]. Slurry reactors are convenient for laboratory-scale experiments, but large volumes of operation may compromise process sustainability due to challenges in light distribution and catalyst utilization. As the catalyst loading increases, scattering and absorption within the dense slurry reduce the light penetration, weakening the benefit of additional catalyst loading[235]. In addition, continuous stirring for catalyst dispersion noticeably increases the operational energy demand. Therefore, ensuring sustainability for slurry reactors necessitates a balance between material use and energy consumption. Another drawback of slurry reactors is the recovery of catalyst particles after reaction[236]. This energy-intensive process poses environmental toxicity risks due to the potential release of unrecovered nanomaterials, especially those containing toxic metals[237]. This limitation has promoted the development of immobilized and fixed-bed structures, which avoid catalyst recovery after reaction. On the other hand, a comparative study on the LCA of TiO_2-based photocatalytic systems indicated that slurry reactors can exhibit reasonably lower environmental footprint, as immobilization presents high electricity and material demands[238]. Gas-solid photoreactors have recently attracted interest as they can overcome some limitations of aqueous slurry systems, especially low CO_2 solubility, catalyst separation after reaction, and use of liquid solvents; however, their sustainability is still limited by relatively low conversion[239].

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Fixed-bed reactors employ catalysts fixed on solid supports such as glass plates, meshes, beads, fibers, or monoliths, retaining the catalyst within the reactor and avoiding excess water use and material loss, facilitating catalyst reuse, and enabling continuous flow operations[236]. However, simply attaching photocatalysts to substrates can result in optical and mass transfer limitations, as restricted light penetration, non-uniform catalyst coating, and reduced reactant access may cause ineffective participation of active sites during the reaction. Hence, photoreactor design has moved toward structured configurations such as honeycombs, monoliths, microchannels, and optical-fiber assemblies to maximize CO_2 conversion by improving light harvesting and catalyst accessibility[240]. On the other hand, their sustainability advantages should be carefully evaluated, as the additional material use, fabrication steps, and operational complexity required for construction and maintenance may increase the overall energy and material footprint of the reactor[241].

Membrane photoreactors integrate photocatalytic reaction and separation in a single unit, separating oxidation and reduction zones to suppress backward reactions, enhance product selectivity, and potentially lower waste generation[242]. However, their overall efficiency can be limited by mass transfer resistance, restricted charge mobility, and rapid membrane fouling[243]. In addition, stable membrane production requires considerable material and energy input, while short lifetimes and frequent cleaning may lead to environmental burdens during fabrication, operation, and maintenance[244].

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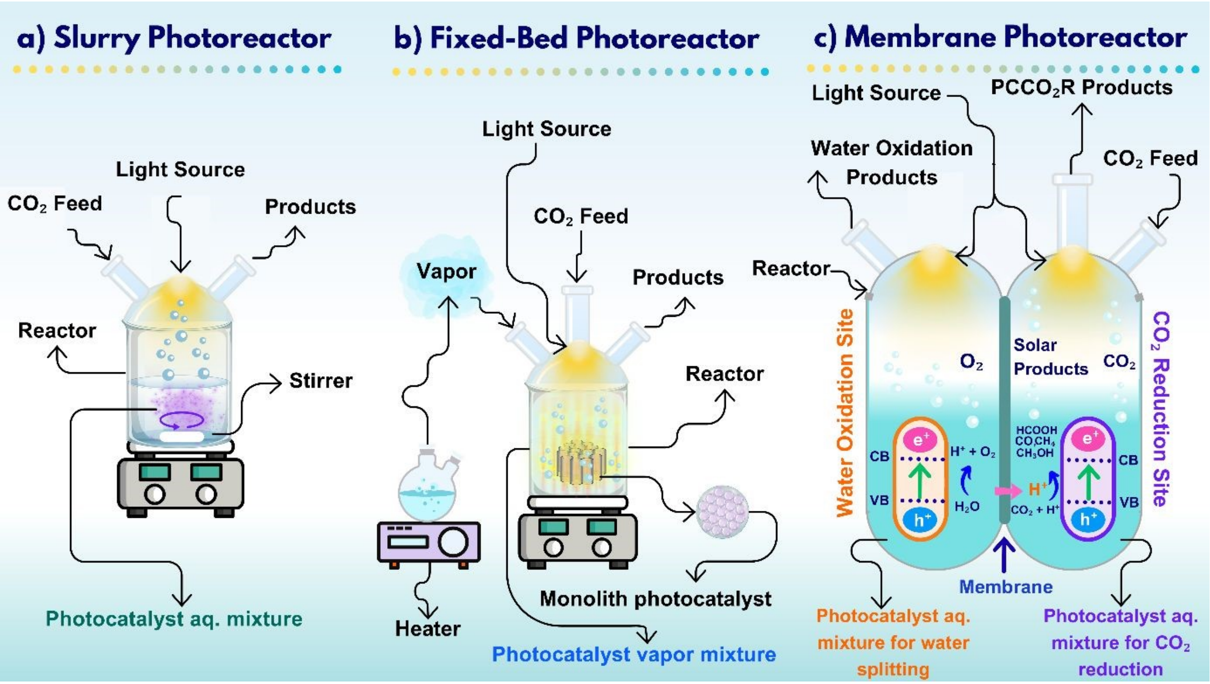

Fig.6 Major photoreactor structures used in $PCCO_2R$

7. Future perspective

7.1. Possible directions and perspectives for future

Analysis above indicated that g-C_3N_4 is the most sustainable semiconductor among the most commonly studied alternatives. TiO_2 can also be considered as sustainable; however, this material is active under UV lights, and modifications to improve its visible light activity have the risk of reducing its sustainability profile. CdS seems to be among the least sustainable semiconductors due to its persisting toxicity, while $BiVO_4$ can also be put in the same category due to supply risks associated with both Bi and V. $CsPbBr_3$, which has started to be used in the field after its remarkable success in photovoltaic applications, also suffers from the toxicity of Pb. As far as sustainability is concerned, all other common semiconductors lie between these two extremes (Table 1), suggesting that the use of semiconductors synthesized from biological sources[245] and abundant elements will increase in the future as long as the concern for sustainability continues. Even though the current works involving biotechnologies are mostly on fuels and energy production, the efforts for the new material developments are likely to increase in future because the commercial success for biotechnological processes are highly depend on the effective utilization of all parts of feedstock, and this requires the production of value added chemicals in

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addition to fuels and energy (as the basis of biorefinery concept inspired from the material efficiency of petrochemical industry). Such developments, together with the other developments in the field of photocatalysis, will increase the sustainability of the process as well.

As far as the co-catalysts are concerned, Cu can be considered sustainable with high availability and catalytic activity; however, its end-of-life degradation should be considered together with the semiconductor used. Similarly, Fe poses significant advantages as an abundant and non-toxic alternative. Sustainability of precious metals, on the other hand, is shadowed significantly due to their limited availability, supply risk, price fluctuations, and environmental burden associated with mining and refining; it seems that they can be sustainable only if they are used with effective recycling strategies. Consequently, we can expect that the use of more abundant and relatively clean metals will increase in the future, while the recycling of precious metals will also gain more attention.

It should also be kept in mind that the photocatalytic reduction is not the only route for CO_2 utilization; the progress in other alternatives will also influence the future of photocatalytic processes. Even if we limit ourselves to transformative CO_2 utilization technologies, we have to assume that similar developments will also increase the feasibility of thermal and electrochemical processes, which are already more mature that photocatalytic route. While the photocatalytic processes directly utilize solar energy, the electrochemical process relies on the premise that the electricity will be generated from renewable sources; however, both suffer from the low long-term stability of the catalysts used[246]. The thermal conversion processes (like methanol formation and syngas production), on the other hand, require more energy and renewable hydrogen production, even though they are technologically more mature than the other two categories.

Artificial intelligence/machine learning (AI/ML) has also contributed significantly to photochemical CO_2 reduction research in recent years, and this can be expected to grow in the future. With the astonishing progress in AI/ML and the ever-increasing availability of scientific data, AI/ML has been employed in material discovery with increasing frequency in recent years[247]. While the access to the external data is getting much easier with the progress in digitalization and spreading open access policies, the data generation through high-throughput computations, which are mostly based on DFT, and experimentation through automation and robotics and self-driven laboratories, as recently discussed by Orouji

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et al[248]. Consequently, the capabilities for material discovery and development, including more sustainable photocatalysts, are increasing. Additionally, AI/ML can help to understand the catalytic performance; for instance, $PCCO_2R$ over MOFs[249], perovskites[250], and halide perovskites[103] have been analyzed using ML recently to determine the best combinations of a large number of alternatives for various elements/groups in the molecules with such modular structures. Generative AI tools, which have been developed and popularized in recent years, may also contribute to the field significantly by clarifying the true potential of the materials for commercial applications and, therefore, sustainability applications. For example, the large language models (LLMs) can process a very large amount of resources by effectively utilizing all search engines, various calculation tools, and a human-like inference function[251].

In the previous section, we briefly discussed the uncertainties in photocatalysis data (including $PCCO_2R$), especially those related to varying characteristics of light sources and experimental set-ups. Unfortunately, the problem grows bigger during publications because the results are not reported in a standard manner either. For instance, there is no single measure of activity. The conversion is usually low, and cannot be accurately computed, while the other measures like turnover frequency or quantum efficiency have their own difficulties [226]. Probably because of those, the performance is often reported in terms of the production rate of individual products (sometimes even as total products). Consequently, the different product distribution and production rates may be reported for the same photocatalyst tested under similar conditions, making the comparison, which is also inevitable to determine the most sustainable alternative, impossible. To overcome this, standard protocols for testing and reporting should be developed and used in the field; this issue is also discussed in more detail in the work by Bonchio et al.[252] and some other publications[253,254].

7.2. Challenges in sustainability assessment

The biggest challenge for a reliable sustainability assessment is the availability of high-quality data, especially those involving the indirect impacts on human life and the environment. This is more critical for the materials and technologies that are not yet fully established; especially, some materials used in such processes are produced in small amounts in laboratories, and the data on these materials are far from representing those produced in industrial volumes. Similarly, the synthesis and testing conditions in well-conditioned and controlled research laboratories or pilot

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units cannot have the same impact as industrial operations. Besides, even for well-established industrial operations, the data quality is not as good as required for comparative assessment because they are often collected by different sources with different objectives, methods, and levels of precision.

Another problem in sustainability assessment is the difficulty of determining the system boundaries. Any commercial material is produced in an industrial value chain from mining and refining to its use in a final product. Each stage of the value chain uses raw materials and energy from the organizations that are also in the value chain of other industries (and possibly located in regions of the world with different regulations). Hence, it is highly difficult to identify all potential impacts and account for them in a sustainability assessment. This should be much harder for complex products and systems involving a large number of components and ingredients. Monitoring and accounting for all the potential impacts may not be practically possible.

Diversity of the sustainability assessment approaches, criteria, and tools is another challenge to be faced by anyone who tries to assess the impact of a material and technology. Various experts and companies have different sets of criteria and priorities for LCA or other sustainability-related assessments; the criteria and prioritizations also differ from country to country despite some international efforts like ISO 14044[255] and SDGs[6] for standardization. This is understandable to some point as the priorities are determined by the needs and desires of the sides, as the critical element and supply risks determined by USGS[69] and EU[10] are also different.

Finally, it is highly probable that the assessment results are significantly influenced by the beliefs and position of people or organizations, considering that the environmental issues are highly subjective and even political (often related to national interests); different groups in the same society may have different perspectives on the same issue depending on their level of interest. Even when the works are executed with great care and objectivity, unintentional bias cannot be fully avoided in the quantitative decision-making involving multiple criteria. Such approaches usually involve the evaluation and scoring of the alternative materials and technologies against certain criteria, which may result in different scores as a result of the different tools and procedures employed and the level of expertise of assessors. Worse than this, the prioritization of criteria and weighing them in terms of importance may be significantly affected by personal beliefs and priorities. This will be especially true for the new technologies, of which the benefits and pitfalls

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are not well established, especially in the fields with a variety of personal, professional, and institutional perspectives. More details regarding the bias in decision-making can be found elsewhere[256,257].

7.3. Additional limitations in present analysis

Needless to say, the current work is not free from the challenges explained above, even though it is a review with a sustainability perspective, not a full-scale sustainability assessment report. On the contrary, it has some additional limitations, mostly stemming from the fact that $PCCO_2R$ is still in the research stage. To begin with, the materials to be utilized on an industrial scale have not been decided yet; the efforts for the discovery and design of new semiconductors and co-catalysts continue with the help of new experimental characterization techniques and computational tools like DFT and machine learning. Hence, the material needs of a commercial $PCCO_2R$ process in the future may be different than those investigated today.

Another problem is that the sustainability of a process cannot be assessed without knowing the efficiency and stability of the material used. Consider a semiconductor or co-catalyst that provides superior performance for a long period, even if it is used in small amounts. That material, even if it has some environmentally undesired elements, may be more sustainable than those that are completely renewable but less effective materials; in the long run, the energy used and environmental impacts may be much higher for the second case. To prevent such problems, a quantitative and accurate assessment has to be carried out so that alternatives can be effectively compared. Unfortunately, this cannot be done at this stage, because not only is technology still immature, but also the data quality is quite poor in photocatalysis, especially data involving activity and selectivity measurements, as another limitation to be considered; the performance range for almost all photocatalysts tested seems to be quite large to make any accurate comparison. The main reason for this is the uncertainties associated with the light source; even the same type of light source with the same rated power (like 300 W xenon arc lamps) may have different irradiation patterns (and this is even true for different models of the same light sources produced by the same companies). Uncertainties increase further in practice because the energy absorbed by the photocatalyst may differ depending on the distance and position of the light, reactor material, and medium composition. In addition, the differences in mass transfer limitations arising from the use of different operational conditions cannot be fully

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accounted[258], while there are also significant concerns that some of the carbon in the products may be coming from the carbon contaminations accumulated over the photocatalyst but not from reduced CO_2, in recent years[253].

8. Conclusions

Sustainability of semiconductors, co-catalysts, and reactor structure used in $PCCO_2R$ is evaluated using criteria describing their availability and supply risk, and impacts on human life and the environment. We can summarize the conclusions we found as follows:

- g-C3N4 is the most sustainable semiconductor among the most repeatedly studied alternatives; this is also true for the pristine TiO_2; CdS seems to be among the least sustainable alternatives due to its persisting toxicity. While $BiVO_4$ has the supply risks associated with both Bi and V, $CsPbBr_3$ suffers from the toxicity of Pb.
- Cu can be considered as a sustainable co-catalyst with high availability and catalytic activity; similarly, Fe poses significant advantages as an abundant and non-toxic alternative. Sustainability of precious metals, on the other hand, is shadowed significantly due to their limited availability, supply risk, price fluctuations, and environmental burden associated with mining and refining.
- Although the strategies involving elemental doping or forming heterostructures may improve photocatalytic performance, they may significantly deteriorate the sustainability profiles of semiconductors due to the increasing complexity of the materials.
- Recycling, especially for precious metals or highly toxic materials, should be part of end-of-life strategies.
- Some of the methods used in the laboratories to have better control of morphology and structure are too complex for sustainable industrial production.
- Even though sustainability is usually associated with human health and environment, the availability and supply risks of some materials will also be more important in the future due to market fluctuations, location dependence, and political developments.

Finally, it should be kept in mind that the results reported here have some limitations; the technology is not mature enough to drive definitive conclusions for the material choices,

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while only qualitative evaluations of the materials could be done due to the uncertainties in the process and lack of a sufficient amount of high-quality data. However, an initial assessment, as presented here, can still be beneficial to direct the efforts to more sustainable alternatives.

Pınar Özdemir: Investigation, Formal analysis, Writing-original draft. **Beyza Yılmaz:** Investigation, Formal analysis, Writing-original draft. **Ramazan Yıldırım:** Conceptualization, Supervision, Funding acquisition, Writing-review & editing,

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

The authors confirm that the data used in this work was provided within the article or in Supplementary Information file.

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