



Review

Photocatalysts for a sustainable future: Innovations in large-scale environmental and energy applications

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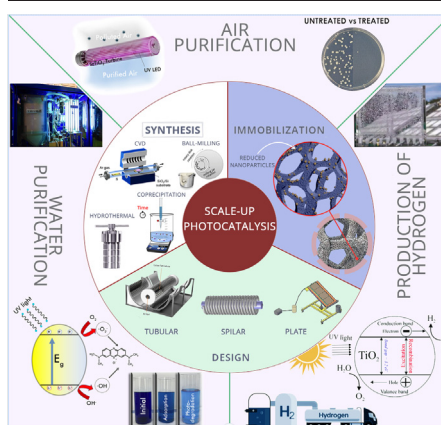
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HIGHLIGHTS

- Photocatalytic systems for solving energy and environmental problems.
- The main challenges in the development of large-scale photocatalytic systems.
- Pilot applications of photocatalytic reactors for large-scale water and air purification and solar hydrogen production.

GRAPHICAL ABSTRACT



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ABSTRACT

The growing environmental and energy crises have prompted researchers to seek new solutions, including large-scale photocatalytic environmental remediation and the production of solar hydrogen using photocatalytic materials. To achieve this goal, scientists have developed numerous photocatalysts with high efficiency and stability. However, the large-scale application of photocatalytic systems under real-world conditions is still limited. These limitations arise at every step, including the large-scale synthesis and deposition of photocatalyst particles on a solid support, and the development of an optimal design with high mass transfer and efficient photon absorption. The purpose of this article is to provide a detailed description of the primary challenges and potential solutions encountered in scaling up photocatalytic systems for use in large-scale water and air purification and solar hydrogen production. Additionally, based on a review of current pilot developments, we draw conclusions and make comparisons regarding the main operating parameters that affect performance, as well as propose strategies for future research.

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Nomenclature

PANI	polyaniline
NF	nickel foam
MIL	Matériaux Institut Lavoisier
ALD	atomic layer deposition
PE-ALD	plasma-enhanced atomic layer deposition
ICP-PECVD	inductively coupled radiofrequency plasmas - plasma enhanced chemical vapour deposition
APP	atmospheric pressure plasma
CFD	combined Computational Fluid Dynamics
DOM	discrete Ordinates Method
SFM	six-flow model
STH	solar-to-hydrogen conversion
FTO	fluorine-doped tin oxide
OWS	overall water splitting
CPC	compound parabolic collector
PMR	photocatalytic membrane reactor
PVDF	polyvinylidene fluoride
DCF	diclofenac
LSC	fluorescent solar concentrator
VOC	volatile organic compound
TOC	total organic carbon
COD	chemical oxygen demand
BEN	bentonite
VER	vermiculite
η_{Rxn}	quantum efficiency

1. Introduction

Although photocatalysis is a promising technology that could partially solve the environmental and energy problems associated with excessive use of fossil fuels, the production and application of photocatalysts capable of operating in visible light on a large scale remains an unsolved problem (J. Chen et al., 2022a; Ozin, 2022; Patowary and Kalita, 2022). At the same time, basic research on the photocatalysis process shows its marketing attractiveness for widespread application in the near future (Khan, 2023). Despite significant advancements in developing photocatalysts with high efficiency, thermal stability, purity, and environmental compatibility, low efficiency in photocatalytic reactors remains a major hurdle in their large-scale application. To enhance the efficiency of photocatalytic reactors, it

is imperative to address the fundamental challenges related to large-scale synthesis and deposition of photocatalysts on solid substrates using simple yet high-quality methods. Additionally, there is a need to develop optimal designs and geometries to overcome the existing limitations (Low et al., 2023). For example, the use of photocatalysts for water purification of different kinds of organic pollutants is a promising technology. However, due to the mass transfer of nanoparticles, it is necessary to collect nanoparticles later, which complicates the process of their repeated use. Mass transfer in turn may lead to incomplete oxidation of the pollutants on the surface of the nanoparticles due to the lack of optimum time for the process. As a result, the formation of byproducts becomes possible. When treating large volumes of water, it becomes necessary to carry out an additional time-consuming process of filtering water from nanoscale photocatalysts, which calls into question the economic viability of nanoparticles. Such technological disadvantages limit investment in the application of photocatalysis for environmental problems. One solution is the creation of special photocatalytic reactors capable of eliminating emissions of pollutants into the environment at minimum costs in comparison with similar methods. For example, the creation of photocatalysts in the form of thin films on a special surface by different methods of deposition capable of functioning in a stable mode was suggested for operation on an industrial scale (Dolai et al., 2022; L. Wang et al., 2022c). In the case of repeated use, it is important to pay attention to the mechanical stability of such films, as they are prone to deformation at high flows (Y. Liu et al., 2022d; D. R. Ramos et al., 2021b).

The development of alternative energy sources can free mankind from its dependence on oil and gas resources. Today's global environmental problems, such as the greenhouse effect, are forcing us to focus on renewable energy sources such as solar, hydrogen and wind power. In addition to being environmentally friendly, cost-effective end products are also important for large-scale production. Unfortunately, there are some disadvantages to the currently available technology that prevent a full transition to renewable energy consumption. Intensive research has been carried out on the conversion of solar energy into photovoltaic energy (solar panels) and chemical reaction energy (reduction of CO₂ into hydrocarbons and formation of H₂) (Photocatalytic Hydrogen Production: A Rift into the Future Energy Supply - Christoforidis - 2017 - ChemCatChem - Wiley Online Library, n.d.). The widespread introduction of hydrogen energy is one possible solution to decarbonization in the world economy. The search and development of innovative hydrogen technologies could lead to the creation of a quite competitive industry of large-scale hydrogen production. Currently, hydrogen is produced worldwide by the conversion of methane from fossil energy resources, which requires high temperature and pressure (Li Liu et al., 2022a; Liu et al., 2023). A developing method of hydrogen production is the photocatalytic decomposition of water. Despite the advantages of photocatalysis in their ability to work in usual environmental conditions, at the scale of hydrogen production, it is important to take

into consideration some parameters that can influence production efficiency. For example, an even distribution of radiation of suspension inside the reactor increases the amount of absorbed photons and optimum speed of agitation. Most scientific works characterize the photocatalyst efficiency using artificial light sources (different lamps) in which only a certain part of sunlight is presented. In the long term, it is necessary to use sunlight for photocatalysis considering that solar radiation is an environmentally friendly and renewable energy resource.

A number of studies on photocatalytic systems and their performances in real environments have been reported in the last few years (Jiménez-Calvo et al., 2023). However, a few factors, such as high cost, low quality, and difficulty in controlling the morphology and structure obtained in large quantities of powders, cause difficulties in producing nanosized photocatalyst particles in large volume and simultaneously with high photocatalytic activity. Consequently, the focus in the bulk production of nanoscale photocatalysts should be on cost-effectiveness, the ability to control morphology and size, high photoactivity and environmental friendliness. To realize these goals, we hope to develop the shortest, most convergent routes with reliable and sustainable methods for the synthesis of photocatalysts functioning in a wide UV-visible range using readily available and inexpensive reagents and cocatalysts without releasing harmful byproducts and waste.

Significant advances in the production and application of photocatalytic systems allow us to speak with confidence about their large-scale implementation in the promising future. This is indicated by numerous works aimed at creating effective systems under laboratory conditions. From this perspective, we believe that a comprehensive review of photocatalytic systems with the potential for large-scale implementation in areas such as water and air purification and hydrogen production is needed. This review compiles research on photocatalytic systems with respect to their practical applications. The main problems hindering the large-scale application of the photocatalytic process for environmental cleaning and solar hydrogen generation have been analyzed and discussed in detail to deeply understand and explore the relationship between their synthesis parameters and applications. Finally, the problems, solutions, and prospects for the widespread application of photocatalytic systems in areas such as water and air purification and solar hydrogen generation are discussed in conclusion.

2. Challenges for scale-up

To achieve the full efficiency of the photocatalytic process, the main problems hindering large-scale photocatalytic applications for environmental cleaning and solar hydrogen generation must be solved. The first important obstacle to realizing this goal is the production of highly active photocatalysts on a large scale through low-energy, economical and environmentally friendly synthesis methods. The next hurdle is selecting the optimal substrate structure, material and immobilization method. In addition, difficulties arise in selecting the necessary geometry and design of photocatalytic reactors depending on the conditions and mode of operation. In the following sections, the main obstacles and solutions encountered in large-scale photocatalyst particle synthesis, substrate immobilization and the selection of the optimal photocatalytic reactor design will be discussed in detail.

2.1. Large-scale synthesis of photocatalysts

Fundamentally, all methods for producing nanomaterials, including photocatalytic materials, can be divided into two types: top-down and bottom-up. In the first case, the methods are characterized by their simplicity and scalability and in many cases represent the delamination of nanolayers from more massive materials but are limited by the low yield and lack of control over the size, thickness, and morphology of nanomaterials (Burke et al., 2020; Xiudong Chen et al., 2019d; Li et al., 2016; Yusran et al., 2020). Bottom-up methods, on the other hand, have broad prospects for synthesizing pure, homogeneous, and controlled nanoparticles with a complex structure with scalability (Cui et al., 2018; Liu et al.,

2019). However, among the various bottom-up methods, economical one-pot (B. Tian et al., 2018a) and in situ growth methods (M. Liu et al., 2021b) are best suited for large-scale production. In particular, the one-pot methods are characterized by easy availability of starting reagents, a small number of reaction steps and full use of precursors (J. Chen et al., 2022a; Parthibavarman et al., 2018; L. Wang et al., 2020b), whereas the in situ synthesis technique allows size and morphology control (G. Li et al., 2019b; Xin et al., 2020).

Regarding the large-scale production of nanoscale photocatalysts, the methods should not only be simple and stable but also allow the control of size and morphology, which play a key role in improving charge carrier mobility and enhancing light collection in various photocatalytic applications. For instance, various studies have indicated that mesoporous polymorphs (Xiong et al., 2019), nanosheets, nanotubes (Zhou et al., 2018), three-dimensional hierarchical porous structures, flower-shaped structures (Huang et al., 2018) or needle-shaped structures show better photocatalytic activity than commercial samples (Mamaghani et al., 2020; H.-X. Wang et al., 2020a). On the other hand, scaling up should also be considered to increase the reaction yield and create complex heterostructures with high efficiency, as simple photocatalysts such as TiO_2 , ZnS , gC_3N_4 , and CdS are ineffective due to their wide bandgap and small conductivity (Kang et al., 2019; C. Liu et al., 2021a; Liu et al., 2019). For example, a BP/CN photocatalyst with high efficiency with respect to H_2 release (at $786 \mu\text{mol h}^{-1} \text{g}^{-1}$) and relatively low cost ($\text{€}0.235/\text{g}$) has been reported on a large scale (Min Wen et al., 2019b). The use of cocatalysts allows composite photocatalysts with higher yields and higher photocatalytic activity for H_2 release to be obtained; however, despite high results, precious metals such as Pt are often used as cocatalysts, which considerably increase the cost of photocatalysts (Ma et al., 2018).

Under laboratory conditions, it is possible to obtain photocatalysts in grams, but the subsequent increase in yields requires the introduction of larger scale reaction apparatuses with high control of heating and mass transfer (Yi et al., 2021). Transition to large-scale photocatalyst production is also possible using a combination of several methods, including the abovementioned single-step and in situ methods (Deng et al., 2023). In addition, the ecological purity of solvents should be considered when applying the synthesis methods on a large scale, as there will be byproducts and wastes from reagents on the way to making photocatalysts in large quantities (Jahanshahi et al., 2020; G. Li et al., 2019b).

To date, several studies have been conducted to produce high-performance photocatalysts, both in the laboratory and on a larger scale, with the aim of achieving full-scale commercialization of photocatalytic applications. For a seamless transition from laboratory to industrial production, modern methods of large-scale synthesis should prioritize high reaction yield, scalability, and reliability while also allowing control of the morphological and structural characteristics of nanomaterials. Furthermore, the synthesis methods should ensure the rational use of the environment and maximum environmental safety to facilitate sustainable production practices.

2.2. Immobilization of photocatalysts

Modern photocatalytic experiments for hydrogen extraction or environmental purification are carried out to a large extent using a particle suspension in bespoke laboratory reactors. These are usually quartz reactors with a total volume of 50 to 500 mL. Such systems have achieved considerable photocatalytic success on the laboratory scale by using the entire active surface of the photocatalyst, which in turn leads to high reaction rates. However, particle-suspended reactors for large-scale use have a few limitations, such as the energy cost of dispersing large amounts of liquid before and during light irradiation so that particles do not settle to the bottom of the reactor, separation, and collection of the photocatalyst from liquid for maintenance purposes, as photocatalyst efficiency decreases with time. Studies aimed at reducing the limitations (e.g., electromagnetic, mechanical agitation) could not significantly reduce the limitations of suspended photocatalysts due to the impracticality of their applications at the

production scale. Thus, particle-immobilized photocatalysts on a solid substrate are used to enable large-scale photocatalyst applications, the main advantage of which is to ensure continuous operability and easy extraction of the photocatalyst from the spent solution (Bui et al., 2020; Loeb et al., 2019). Potential difficulties in the practical operation of immobilized photocatalytic systems are related to the mass transfer of reagents and products to the catalyst, both in liquid and gaseous phases, as well as the reduced illuminated surface area and fast photon scattering rate (Phan et al., 2018).

Depending on the photocatalyst application, the substrate design can vary, e.g., for hydrogen production, solid substrates are used, whereas for water and air purification systems, emphasis is placed on porous membranes and substrates because in addition to increasing the active centers, the bandwidth for unimpeded transport of pollutants is improved (Schwarze et al., 2022b). For large-scale photocatalytic hydrogen production, thin film photocatalysts immobilized on various substrates, such as 2D glass (Cerrato et al., 2019), PANI/cotton (Yu et al., 2019), and nonwoven fiber (Kacem et al., 2017; Schwarze et al., 2022a), are mainly used. Compared to suspended photocatalysts, thin-film photocatalysts are the most efficient in producing solar hydrogen due to the following advantages: thin films absorb photons uniformly with low scattering, contributing to efficient generation of charge carriers in large amounts; good scalability compared to suspended photocatalysts due to continuous H_2 production and minimal consumption of immobilized photocatalyst powder with high activity (Gopinath and Nalajala, 2021). In particular, a thin film photocatalyst with immobilized CdS and Ni_3S_2 particles as cocatalysts on a flat foam surface showed excellent results with a maximum hydrogen extraction rate of $100.5 \text{ mmol g}^{-1} \text{ h}^{-1}$, which corresponds to obtaining 132 L H_2 using 1 m^2 of CdS- Ni_3S_2 /NF photocatalyst under irradiation with light in the visible spectrum for 12 h (Chao et al., 2018). Unlike 2D photocatalytic purification of water and air from various pollutants, 3D substrates are applied, which are the most practical. The advantages of such 3D substrates are a large active specific surface area and low surface resistance, which provides improved mass transfer and photon absorption, which in turn increases the photocatalytic properties of the immobilized photocatalyst and the possibility of multiple uses.

To date, the following three-dimensional photoreactors have been reported: Duranit (Karavasilis et al., 2022), borosilicate glass (pros: continuous flow, transparent, reuse without loss of efficiency) (Karavasilis and Tsakiroglou, 2022), and ceramics with immobilized photocatalytic TiO_2 and ZnO nanoparticles (Sraw et al., 2018). Such spherical substrates for photocatalysts are mainly made of transparent materials and are notable for their durability and reusability without losses in continuous-flow systems, but the rate of pollutant decomposition (70 % chemical oxygen demand in 180 min) (Vaiano and Iervolino, 2018) is significantly lower in comparison with their suspended particle counterparts (90–95 % chemical oxygen demand in 120 min) (Kanakaraju et al., 2019). Other membrane-like substrates have rather high prospects for both water and air purification, such as graphene oxide (GO)/polysulfone (PSF) membranes (Cheng et al., 2021), ceramic membranes/ TiO_2 -P25 (Lumbaque et al., 2021), poly(vinylidene fluoride)/ TiO_2 -GO (Tran et al., 2020), porous stainless steel/ TiO_2 (Rodríguez-Chueca et al., 2019), Al_2O_3 /N- TiO_2 (Horovitz et al., 2018), and carbon nanofibers/ TiO_2 nanowires/MIL-100 (Zhang et al., 2018).

Another important factor is the choice of photocatalyst deposition method on the substrate surface, as the thickness of the immobilized layer should be thin enough to reduce photon scattering. One of the simple, economical and often used methods for both 2D (Rana et al., 2022; Schwarze et al., 2022b) and 3D substrates (Allen et al., 2018; Levchuk et al., 2016; D. Li et al., 2019a; Omerzu et al., 2021; Su et al., 2018) is the wet impregnation method, which involves immersion of the support material in a photocatalyst suspension or application of photocatalyst suspension drops to the substrate surface followed by drying at moderate temperatures up to 150°C . Although this method is simple and cost-effective, it is not suitable for large-scale deposition, nor can it accurately control the growth of thin films on the surface, and it is not suitable for thermal treatment of inorganic substrates such as matrices and polymers (Rana et al., 2022).

Immobilization of low-size particles is also possible using other low-cost and low-temperature chemical methods, such as sol-gel and hydrothermal methods, but in the case of polymeric materials, organic residues promote excessive crystallization and increase in grain size, which in turn reduce photocatalyst activity (Allen et al., 2018). Recent work has been reported on the use of gas-phase methods that can accurately control growth and thickness. In particular, TiO_2/Al_2O_3 and separate TiO_2/ZnO layers have been deposited on the surface of a polycarbonate membrane matrix by the gas-phase atomic layer deposition method (Su et al., 2018). Similarly, TiO_2 powders have been deposited on porous aluminum foams (Levchuk et al., 2016), which indicates the possibility of this method to deliver photocatalyst particles to difficult-to-access complex surfaces; however, the ALD method is limited by a low deposition rate and deposition temperature range ($120\text{--}180^\circ\text{C}$), which does not allow for the use of heat-sensitive substrates. To reduce the deposition temperature, optimization methods have been proposed by using a low-temperature plasma process. Thus, low-temperature PE-ALD (Omerzu et al., 2021), ICP-PECVD (D. Li et al., 2019a), and APP (Banerjee et al., 2020) methods have been applied for photocatalyst deposition on polymer substrates. For instance, using low-temperature plasma-controlled deposition, we managed to deposit a thin layer of TiO_2 on a complex three-dimensional porous foam surface and obtain high photocatalytic activity in a special recirculating reactor during dye decomposition with multiple use (20 cycles) capability (Uricchio et al., 2021). In view of the abovementioned works, an ideal substrate for modern photocatalytic systems for large-scale applications should meet criteria such as sufficiently strong contact with photocatalysts, high specific surface area, high adsorption to reaction products, chemical stability and structural strength. In addition, the choice of a suitable photocatalyst immobilization method is based primarily on the material and shape of the substrate used, as the quality of photocatalyst adhesion and the stability of photocatalytic activity directly depend on this.

2.3. Design of photocatalytic reactors

Research into photocatalytic environmental cleaning and hydrogen production has increased significantly in recent years. The main elements of heterogeneous photocatalysis are the light source and the photocatalytic reactor. The main function of photoreactors is to bring the illuminated photocatalyst and reaction product (contaminated water/air or product for H_2 generation) into contact. Consequently, the photoreactor for large-scale applications must have such a geometry and configuration to ensure the largest possible three-phase contact area.

The geometry of the photoreactor must be chosen according to the size and shape of the radiation source to maximize the collection of irradiated radiation. Reactor irradiation can take place outside or inside the reactor. In internally illuminated reactors, photocatalysts are irradiated by an ultraviolet or visible lamp located inside the reactor with cylindrical (Reilly et al., 2018), spiral (Jo et al., 2015) or circular geometry (Rincón and La Motta, 2019). Photocatalysts immobilized on glass spheres (Kamaei et al., 2018; Vaiano and Iervolino, 2018), clay balls (Sraw et al., 2018) or powder suspensions (Peralta Muniz Moreira and Li Puma, 2021; Rincón and La Motta, 2019) are commonly used. Such photoreactors with internal illumination are distinguished by high mass transfer, as they possess a large photocatalytic surface area per unit volume of liquid or volatile gas (Bafaqeer et al., 2018; Khan and Tahir, 2019). However, internally illuminated photoreactors for commercial applications are limited by efficiency drop due to scattering and reflection on immobilized photocatalyst layers when using photoreactors with large diameters (Ramos et al., 2019), and in suspension reactors in the case of nanoscale photocatalysts, these particles can adhere to the tube inner surface, thus also reducing light penetration efficiency (Khan and Tahir, 2019). In addition, there are cases of optimizing such photoreactors by using intensely and uniformly distributed light optical fibers (Harrisankar et al., 2021; O'Neal Tugaoen et al., 2018; Zhong et al., 2019), economical LEDs (Khodadadian et al., 2018; O'Neal Tugaoen et al., 2018) or designing reactors that are irradiated by LEDs

from both inside and outside to minimize dead spots (Manassero et al., 2023).

In contrast to photocatalytic reactors with internal illumination, externally illuminated photoreactors are irradiated by multiple external lamps, which are situated perpendicularly (Claes et al., 2019), above (Fang et al., 2019; Kassahun et al., 2020) or on the focal axis of a parabolic reflector (Casado et al., 2019). Externally illuminated reactor designs are usually transparent tubular and can be irradiated by direct sunlight rich in UV radiation (Ochoa-Gutiérrez et al., 2018). Solar collector-based photocatalytic reactor designs, namely, compound parabolic collectors (CPCs), have broad applications for photocatalytic environmental cleaning (Gutiérrez-Alfaro et al., 2018; Luna-Sanguino et al., 2020; Moreira et al., 2018) and hydrogen generation (Cao et al., 2018b; Ma et al., 2020; Ren et al., 2022). CPC photoreactors are more suitable for large-scale applications because they maximize the use of sunlight by redirecting direct and scattered rays by reflectors to the tube reactor and perfectly cross-mix suspensions due to low-cost turbulent flow (Cabral, 2022; Talwar et al., 2020). As a rule, such reactors are hermetically sealed and made of economical materials (Fendrich et al., 2019). This paper (M. Tian et al., 2018b) details the factors to consider when selecting the most efficient CPC-based photoreactor design with 2D and 3D, asymmetric, seashell-shaped and convex geometry for large-scale operation. However, externally illuminated photoreactors are limited due to the incomplete irradiation of the photocatalyst slurry through the reactor surface's depth. In addition, reactors of this type are difficult to scale in natural direct sunlight conditions.

Reactors with flat geometry in the form of fixed plates (Ahmed et al., 2022; Devia-Orjuela et al., 2019; B. Ramos et al., 2021a) or rotating disks/drums (Zelić et al., 2022a, 2022b) are more suitable for such purposes. The main advantages of planar photocatalytic reactors are as follows: control of liquid flow rate; absence of possible dead zones and provision of continuous liquid flow mode in case of contaminated water treatment; and easy maintenance and operation (very important in production scale and with the possibility of uniform irradiation by sunlight (Bahmani et al., 2020a) and illumination by additional emitters (Zhou et al., 2023)). For example, using these advantages, a thin film sludge reactor irradiated with blue LEDs for urea degradation was designed. In addition, various operating parameters, such as light source, flux rate (Ahmed et al., 2022), catalyst dosage, emitted surface area, and pH, on photocatalytic efficiency have been investigated (Babić et al., 2021; Bahmani et al., 2020b). At present, modern mathematical modeling techniques such as combined computational fluid dynamics (CFD) (Lira et al., 2022; Tong et al., 2020; Yusuf et al., 2018, 2020), the discrete ordinates method (DOM) (Armaković et al., 2020; Moreno et al., 2019), the six-flow model (SFM), and others (Tolosana-Moranchel et al., 2019; van Walsem et al., 2018) can accurately analyze internal kinetic processes to optimize photocatalytic reactors of any given photoreactor configuration if the required parameters are correctly considered. In particular, a real photoreactor simulated using CFD and the discrete element method increased the photocatalytic reaction rate by adjusting the size and shape (Tong et al., 2020). Methods such as CFD and DOM have made great progress in estimating the radiation field and contaminant removal by photoreactors with simple geometry and with laboratory artificial light sources, but they are limited in scaling solar photoreactors because they are time-consuming and require large computing resources, whereas the SFM model is more accurate in describing radiation transport phenomena in real-life solar photoreactor operating conditions. SFM has been used to simulate different geometries of photocatalytic reactors, including solar flat films, tubular CPC, and mixed multitubular photoreactors (Acosta-Herazo et al., 2020; Nchikou et al., 2021; Ochoa-Gutiérrez et al., 2018; Otálvaro-Marín et al., 2019).

Thus, the design of the selected photocatalytic reactor plays a key role in ensuring that the photocatalysts obtain the maximum benefit from light energy and provide a platform with a large contact area between the irradiated photocatalysts and the reaction products used. From the above data, it can be concluded that the choice of optimum geometry and configuration must first be based on the photocatalytic application method and the shape and variety of the radiation source. Consequently, when designing modern

high-efficiency photocatalysts for large-scale applications, researchers should focus on stable reactors operating in the visible region of radiation, using artificial radiation sources (cost-effective LEDs and optical fibers) and natural sunlight, with high reaction rates of both pollutant degradation and water splitting and the possibility of easy photocatalytic reduction and increased mass transfer. In addition, modern computer modeling techniques have made progress in optimizing the geometry of photoreactors, which will help to save time and money in designing the required large-scale photoreactors.

3. Recent developments of photocatalytic reactors for a variety of large-scale applications

3.1. Large-scale solar hydrogen generation

The rising global energy demand and depletion of fossil fuels are accelerating the transition to renewable energy sources, including green hydrogen. The direct conversion of solar energy into hydrogen through a photocatalytic process is distinguished by its simplicity and economy compared to competitive technologies such as photoelectrochemistry and photovoltaic-electrochemistry, which require more complex designs than a photocatalytic system. The photocatalytic process also has an ultra-low energy consumption as the main source of energy is solar energy, making it more cost-effective per kg of hydrogen compared to other processes, costing around 3.12 \$/kgH₂ (Frowijn and van Sark, 2021) and 1.60–3.20 \$/kgH₂ (Chen et al., 2017). Meanwhile, in photoelectrochemical and photovoltaic-electrochemical processes, under optimal conditions, the cost is expected to decrease to \$6.05/kgH₂ and \$3.72/kgH₂, respectively (Frowijn and van Sark, 2021). Additionally, the installation of local photocatalytic systems in remote locations can further reduce logistics costs. Green H₂ is usually obtained by photocatalysis using two processes: photocatalytic water splitting and biomass photoreforming (Qian et al., 2022). While many high-performance materials have been developed in both systems (Lu et al., 2023; Xia et al., 2023), the reactor design to improve photocatalytic reaction conditions are often neglected. Most laboratory-scale photoreactors are not suitable for scaling up as they do not require complex engineering efforts and are designed for testing the main photocatalyst. Therefore, the development of an efficient and simple photoreactor for large-scale applications requires more engineering effort.

Suspension photocatalytic plants, due to their simplicity and economical manufacturing, have an advantage over panel photocatalytic and photoelectrochemical plants (Hisatomi and Domen, 2019). Based on suspension photocatalytic particles, not a few scale-up pilot programs have been proposed since the 2000s (Cao et al., 2018b, 2018a; Chen et al., 2019a, 2019b, 2019c, 2019d; Jing et al., 2010; Maldonado et al., 2018; Priya and Kanmani, 2013; Ruiz-Aguirre et al., 2022; Wei et al., 2017). Parabolic photocatalytic concentrator reactor (CPC) types are popular because they effectively capture and focus direct and scattered solar rays. There are many CPCs based on different photocatalysts and different capacities (Cao et al., 2018b, 2018a; Maldonado et al., 2018; Ruiz-Aguirre et al., 2022; Toledo-Camacho et al., 2021; Wei et al., 2018, 2017). For example, tubular CPC reactors with a total volume of 25 L based on TiO₂-CuO with the addition of glycerol as a sacrificial agent can produce hydrogen with a maximum STH efficiency of 0.9 % (Ruiz-Aguirre et al., 2022), while a system of 76 tubular CPC reactors with a total light area of 103.7 m² can produce 42.84 L of H₂ a day (Wei et al., 2017). It should be noted that when designing suspension photoreactors, such parameters as flow rate, catalyst and sacrificial agent concentration, and tube radius, which directly affect the productivity of the plant (Jing et al., 2010; Wei et al., 2018), should be considered. However, despite the relatively high achievements, suspension systems in large-scale applications have a few major limitations, in particular:

- (1) More water is needed, making photocatalytic systems bulky and uneconomic;

- (2) Constant agitation is required because particles will settle to the bottom and will not receive enough solar energy, and some systems require constant availability of sacrificial agents;
- (3) Difficulties associated with the extraction of the photocatalyst from the suspension.

Thus, for instance, to maintain constant mixing of the photocatalyst and sacrificial agent suspension in a unit with a total area of 2.1 m² containing a water volume equal to 14.25 L, a pump with a flowrate of 20 L/min is necessary (Toledo-Camacho et al., 2021). To solve these problems, photocatalytic reactors with immobilized photocatalytic particles on flat plates made of a Au layer (Chen et al., 2019a, 2019b, 2019c, 2019d, 2021), carbon electrically conductive layer (Wang et al., 2017), nickel mesh (Zhang et al., 2023), nickel foam (Liu et al., 2020), 25 cm² matte steel (Xiong et al., 2014) and FTO (Zhu et al., 2022) were suggested. However, the hydrophilicity and porosity of the photocatalyst need to be controlled for the effective use of such panels, as these parameters significantly affect the safety and efficiency of photocatalytic panels (Goto et al., 2018). Another advantage of fixed-particle installations is that the pH value and gradient do not affect the photocatalytic activity of the particles because the immobilized particles are tightly fixed on the electroconductive layer and H₂ and O₂ are released in the immediate vicinity (Hisatomi and Domen, 2019).

To date, work is underway to improve the performance and efficiency of photocatalytic installations with immobilized photocatalytic particles. In 2015, for example, the development of a large-scale panel-type reactor for general water splitting under direct sunlight based on gC₃N₄ immobilized on stainless steel plates by drip coating was reported for the first time (Schroder et al., 2015). Despite 10 vol% triethanolamine aqueous content and the use of an energy-consuming pump for solution circulation, the reactor showed an insignificant STH equal to 0.12; however, it operated for 30 days and became a starting point for further large-scale development of modern photocatalytic reactors functioning in natural conditions. As the use of sacrificial agents and circulating pumps limited the large-scale use of photocatalytic reactors in vivo in 2018, a 1 × 1 m² panel reactor filled with water without sacrificial agents with a thickness of 1 mm and capable of operating without forced circulation was designed (Goto et al., 2018). A flat reactor based on immobilized SrTiO₃@Al particles demonstrated hydrogen yield with an STH efficiency of 0.4 %. A water layer depth of 1 mm did not affect the activity, and the hydrophilic surface of the panel window improved the unobstructed escape of gases. In addition, based on this work, a 100 m² pilot scale plant was established (Nishiyama et al., 2021), which operated for several months off-line with negligible loss of efficiency. It should be noted that the plant, which consisted of a system of 1600 reactor units, operated for one year without obvious safety problems. However, although the efficiency of such plants was only STH 0.76 %, this work proves the viability and prospects of large-scale hydrogen production and the possibility of putting it into operation. To this end, photocatalytic stand-alone systems with STH efficiencies >6 % and low catalyst concentrations (<0.2 g (catalyst)/L) (Hisatomi and Domen, 2019; Schneidewind, 2022) under natural solar radiation should be developed for economically viable photocatalytic hydrogen production on a production scale. The authors (Toe et al., 2022) report that developing efficient photocatalysts and engineering systems capable of increasing STH from 1 % to 5 % will reduce costs by 75 %. Moreover, reducing the cost of photocatalysts (by using base materials), increasing their service life (to at least 1 year), and introducing solar concentrators can further decrease costs by 20–30 %.

All this led to the active development of this field, and recently, an InGa_{0.5}N/GaN composite photocatalyst with Rh/Cr₂O₃ core/shell and Co₃O₄ nanoparticles as cocatalysts and deposited on a silicon wafer was developed that achieved a high STH efficiency of 9.2 % in pure water splitting under the action of concentrated natural sunlight (Zhou et al., 2023). Such high performance is due to the synergistic effect of InGa_{0.5}N/GaN-based high-efficiency composites and cocatalysts (Chowdhury et al., 2018; Y. Wang et al., 2019c) and the suppression of the recombination of separated hydrogen and oxygen gases at a moderate reaction temperature of approximately

70 °C, which helps to increase the reaction rate by enhancing mass transfer and chemical bond breaking (Tembhurne et al., 2019; Zhang et al., 2017). In addition, this composite photocatalyst showed an STH efficiency of 7 % in seawater splitting and 6.2 % in a large-scale photocatalytic OWS test with 257 W natural sunlight power on a 4 × 4 cm² plate unit.

Thus, despite the simplicity and efficiency of particle suspended photocatalyst reactors, reactors based on immobilized photocatalyst particles are the most scalable for large-scale photocatalytic production of solar hydrogen. Pilot studies of flat panels have proven the viability of safe industrial production of solar H₂ by photocatalysis. In addition, based on the study mentioned above (Nishiyama et al., 2021), a large-scale photocatalytic process has shown that hydrogen can be produced from abundant solar energy without additional energy input and without emitting by-products such as solid waste, polluted water, or harmful volatile gases. This can significantly reduce the overall cost of hydrogen production. These developments are currently the most feasible for commercialization despite the low efficiency of STH and require further upgrading of photoreactor design, operating conditions and the use of composite high efficiency photocatalysts operating over a wide range of light emissions, thereby improving overall efficiency.

Table 1 compares the above-described solar photoreactors for large-scale solar H₂ production. The overall efficiency of photocatalytic solar H₂ production directly depends on such parameters, as photoreactor geometry, type and amount of photocatalysts, availability and concentration of sacrificial agent and solar radiation intensity.

3.2. Large-scale water purification

The demand for very high purity water has posed new challenges for scientists in the field of water purification. Traditional water treatment technologies such as adsorption, biological treatment, chemical treatment with chlorine, ozone, and hydrogen peroxide, thermal and catalytic oxidation, and high-energy UV irradiation all have their own drawbacks, such as problems with waste disposal or the emission of harmful intermediate products. In contrast, the photocatalytic water purification process allows for the complete mineralization of water pollutants, making it an effective and environmentally friendly option for water treatment. In addition, the process is able to remove a wide range of pollutants, including organic compounds, heavy metals, and microorganisms. Overall, the use of photocatalytic water purification can lead to a more sustainable and cost-effective solution for water treatment compared to conventional technologies.

Photocatalytic pilot or prospective for implementation on a large-scale installation for effective treatment of formaceous, textile and other polluted water can be divided into two main groups, i.e., photoreactors either with suspended photocatalysts (Janssens et al., 2021) or with immobilized photocatalysts (Kane et al., 2022). Photocatalytic reactors with suspended photocatalysts are characterized by simplicity and relatively high efficiency: particles are effectively irradiated by light and interact smoothly with pollutants in aqueous suspension (Manassero et al., 2017). To date, numerous photoreactors with artificial illumination methods have been developed through pilot projects (Ahmadpour et al., 2022; Baaloudj et al., 2022). Such reactors exhibit improved performance in water purification compared to other types of reactors, but the high efficiency is limited by the not insignificant energy consumption costs of lamps and mixers. Suspension photoreactors operating under the action of free natural sunlight are highly scalable and economical in this respect (Saran et al., 2019). In particular, a slurry pilot plant based on a solar photocatalytic tube reactor has been tested for wastewater treatment by reusing local materials at low cost. In addition, there are many pilot developments of parabolic solar concentrator photoreactors for wastewater treatment with total volumes of 85 L (Rapti et al., 2022), 39 L (Davididou et al., 2019; Grilla et al., 2019), 35 L (Luna-Sanguino et al., 2020), 16 L (Martínez-Costa et al., 2020), 9 L (Esteban García et al., 2021), 7.7 L (Díaz-Angulo et al., 2019) and 2 L (Martín-Sómer et al., 2021). However, it should be kept in mind that such reactors, although having small pipe diameters, occupy a

Table 1

Recent applications of different photocatalytic reactors for solar hydrogen production.

Photoreactor type	Total working volume (area)	Photocatalyst	Photocatalyst concentration	Average solar intensity ($\text{W}\cdot\text{m}^{-2}$)	Efficiency	Sacrificial agent (conc.)	Ref.
CPC	207 L (32.4 m^2)	$\text{Cd}_{1-x}\text{Zn}_x\text{S}$	$2.77 \text{ g}\cdot\text{L}^{-1}$	800	$10.321 \text{ L}\cdot\text{h}^{-1}$	Na_2S and Na_2SO_3 ($0.1 \text{ mol}\cdot\text{L}^{-1}$)	(Cao et al., 2018a)
CPC	25 L (2.10 m^2)	Cu/TiO_2	$0.2 \text{ g}\cdot\text{L}^{-1}$	N/A	STH 2.6 %	Glycerol at pH = 9 (0.05 M)	(Maldonado et al., 2018)
CPC	25 L (2.10 m^2)	$\text{TiO}_2\text{-CuO}$	$0.1 \text{ g}\cdot\text{L}^{-1}$	N/A	STH 0.9 %	Glycerol (0.075 M)	(Ruiz-Aguirre et al., 2022)
CPC	720 L (103.7 m^2)	$\text{NiS}/\text{Cd}_x\text{Zn}_{1-x}\text{S}$	$0.5 \text{ g}\cdot\text{L}^{-1}$	965.5	$7.14 \text{ L}\cdot\text{h}^{-1}$ ($\eta = 0.087 \%$)	Na_2SO_3 ($0.25 \text{ mol}\cdot\text{L}^{-1}$), Na_2S ($0.35 \text{ mol}\cdot\text{L}^{-1}$)	(Wei et al., 2017)
CPC	25 L (2.10 m^2)	Au/TiO_2	$0.2 \text{ g}\cdot\text{L}^{-1}$	67.4	$1.3 \text{ mmol h}^{-1}\cdot\text{L}^{-1}$ ($\eta = 1.8 \%$)	Formic acid (0.05 M)	(Arzate Salgado et al., 2016)
SUC	70 L	$\text{NiS-Cd}_x\text{Zn}_{1-x}\text{S}$	$0.5 \text{ g}\cdot\text{L}^{-1}$	876.7	$2.148 \text{ L}\cdot\text{h}^{-1}$ ($\eta = 0.475 \%$)	Na_2SO_3 ($0.75 \text{ mol}\cdot\text{L}^{-1}$), Na_2S ($1.05 \text{ mol}\cdot\text{L}^{-1}$)	(Wei et al., 2018)
CPC	25 L	Pd/TiO_2	$0.2 \text{ g}\cdot\text{L}^{-1}$	N/A	$0.618 \text{ L}\cdot\text{h}^{-1}\cdot\text{g}^{-1}$	Glycerol (5 vol%)	(Toledo-Camacho et al., 2021)
Flat panel	10 L (0.756 m^2)	$\text{Pt}/\text{g-C}_3\text{N}_4$	$13 \text{ g}\cdot\text{m}^{-2}$	~ 800	STH 0.12 %	Triethanolamine (10 vol%)	(Schröder et al., 2015)
Flat panel	1 m^2	$\text{RhCrO}_x/\text{SrTiO}_3/\text{Al}$	$8 \text{ g}\cdot\text{m}^{-2}$	650–750	$1.016 \text{ L}\cdot\text{h}^{-1}$ (STH 0.4 %)	None	(Goto et al., 2018)
Flat panel	100 m^2	$\text{SrTiO}_3/\text{Al}/(\text{Rh}, \text{Cr}, \text{Co})$	$8.9 \text{ g}\cdot\text{m}^{-2}$	880	$216\text{--}222 \text{ L}\cdot\text{h}^{-1}$ (STH 0.76 %)	None	(Nishiyama et al., 2021)
Flat wafer + Fresnel lens	$0.0016 \text{ m}^2 + 1.21 \text{ m}^2$ (lens)	$\text{InGaN}/\text{GaN}/\text{Rh}/\text{Cr}_2\text{O}_3/\text{Co}_3\text{O}_4$	$88.1 \text{ mmol}\cdot\text{m}^{-2}$	850	STH 6.2 %	None	(Zhou et al., 2023)

relatively large volume and require large areas for such systems to avoid shading each other, and, in addition, nighttime or cloudy days reduce the efficiency of such SRS reactors. Although authors have reported the separation of photocatalyst powder by filtration techniques, the potential separation of particles in the case of dispersed powders is hampered by its energy intensity (Lee et al., 2018), which is another serious disadvantage of such systems, as toxicity and adverse environmental and health effects are not excluded (von Gunten, 2018). Alternatives to such photoreactors are fluidized bed photoreactors modified with inexpensive and economical LEDs (Surenjan et al., 2019) that do not require photocatalyst collection and a constant stirring stage (Aimeur et al., 2021). In such systems, an immobilized photocatalyst on a retained substrate is maintained inside the reactor by circulating water flow at a certain rate, preventing it from settling to the bottom of the reactor. For example, fluidized bed reactors with immobilized photocatalysts on sand (Aimeur et al., 2021), silica (Rincón and La Motta, 2019), polymers (Surenjan et al., 2019), and graphene macrostructures (Fang et al., 2019) as substrates, without the need for separation and to increase the active area of photon-irradiated catalysts, have been proposed for large-scale continuous use. In addition, fluidized bed photoreactors are well combined with ozone aeration and membrane filtration for water purification (Wei et al., 2020). Guillermo J. Rincón and others (Rincón and La Motta, 2019) reported that as the diameter of silica particles decreases, the photooxidation efficiency increases in a fluidized bed unit, i.e., the carrier particle size directly affects the unit efficiency. Consequently, increasing the carrier size to reduce high catalyst dispersibility is not effective, limiting the scalability of such systems.

Another variety of photocatalytic reactors for water treatment are immobilized catalyst photoreactors. Such photoreactors have been proposed as the most suitable for conversion from laboratory scale to full scale water/wastewater treatment due to the absence of photocatalyst collection and provision of direct continuous flow (Alalm et al., 2021; Fouad et al., 2021). The most popular are photoreactors with immobilized particles on solid fixed membranes (Lotfi et al., 2022). Based on the photocatalytic membrane reactor (PMR) made of hollow fibers of PVDF, a pilot project was implemented to treat diclofenac (DCF) from wastewater (Plakas et al., 2016). The proposed PMR had a total volume of 25 L and was fully automated to continuously produce $1.2 \text{ m}^3/\text{day}$ of treated water. In such continuous flow devices, the flow rate plays an important role, and the photocatalytic oxidation reaction occurs by passing a slurry of pollutant through the pores irradiated by the loaded photocatalyst (Pascariu et al., 2019). Recently, Chechia Hu (Hu et al., 2021) designed a photocatalytic membrane reactor (PMR) and compared flat and hollow

substrates based on Al_2O_3 fibers, where the stable flux was $4250 \pm 100 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ versus $80\text{--}100 \text{ L}\cdot\text{m}^{-2}\cdot\text{h}^{-1}\cdot\text{bar}^{-1}$ for hollow and flat substrates, respectively, and the phenol degradation efficiency reached 90 %, whereas in a similar study (Hu et al., 2019) with the same hollow substrates but with a different photocatalyst, phenol degradation was over 92 %. However, in PMR, the difficulty is related to the penetration of light deep into the porous structure and the relatively weak mass transfer, which can affect the efficiency of the whole system. In addition, such reactors face the problem of membrane clogging by degradation products due to photocatalysis (Sundar and Kanmani, 2020). An alternative is thin-film photoreactors, which have a high degree of light absorption and mass transfer regulation (Samy et al., 2020a). In such plants, water flows in a laminar stream along an inclined plane covered with a thin layer of photocatalyst in recirculation mode (Samy et al., 2020b). Mahmoud Samy et al. (Samy et al., 2021) in a pilot scale purified real agrochemical and pharmaceutical aqueous pollutants using planar photoreactors based on immobilized CNT/MOF-808 photocatalysts with 93 % and 76 % efficiency, respectively, and reusability with no loss of efficiency after 5 cycles. A study of depreciation and operating costs showed that such a large-scale plant costs $\$2.523/\text{m}^3$. However, the use of sunlight, reuse of photocatalyst film and simplicity can significantly reduce operating costs (Pava-Gómez et al., 2021).

The main problem with immobilized photoreactors is the large difference in photon-photocatalyst interaction time ($\sim 1 \mu\text{s}$) and reagent diffusion inside the porous catalyst ($\sim 1 \text{ s}$), which limits the interaction between the illuminated catalyst and pollutant molecules (Kayahan et al., 2020). Compacted bed reactors (Manassero et al., 2022) and macroporous foam reactors (Goetz et al., n.d.) operating in continuous mode are of most interest to improve mass transfer at large scales in immobilized particle photoreactors. Such photoreactors are scaled by continuously pumping contaminated water into the photoreactor and placing several units in parallel (Pestana et al., 2020; Sacco et al., 2018). The compacted structure improves the mass transfer efficiency and light absorption by reducing the light path and effective mass transfer caused by laminar flow perturbation by properly selected photocatalyst carriers (Zhang et al., 2020), thus requiring a short time for complete decomposition and making it possible to treat large amounts of contaminated water in a continuous operation mode (Sacco et al., 2019; Vaiano et al., 2020). Among many immobilized particle photoreactors, this compacted bed photoreactor (Claes et al., 2019) proved to be much more productive, with a reactor rate constant of 0.82 min^{-1} at a flow rate of 216 mL per minute, which is 3–4 orders of magnitude higher than other immobilized bed photoreactors, such as compacted flat $\sim 10^{-5} \text{ s}^{-1}$ (Vaiano et al., 2015) or immobilized foam $\sim 10^{-5} \text{ s}^{-1}$ (Kouamé et al., 2013) continuous reactors. In addition, the authors

analyzed 29 photoreactors and sorted them according to their performance, where this photoreactor was the most efficient in terms of scaling on par with suspension and microreactor systems. To reduce energy costs, flow photochemistry is combined with fluorescent solar concentrators (LSCs) (Cambié et al., 2017, 2019). LSCs are systems where phosphors such as fluorescent dyes or quantum dots are ground into a polymer or glass, with the ability to convert solar radiation into light of a specific wavelength. Based on LSCs, an optimized photomicroreactor with densely packed photocatalyst-coated glass beads (850 μm) has been designed (Zhao et al., 2020) with high performance. Such optimized photoreactors not only increase the efficiency but also represent a new direction to simultaneously improve mass and light transfer for enhanced photocatalysis.

In light of the research work described above, it can be concluded that photocatalytic installations for large-scale photocatalytic treatment of water, both with suspension and immobilized photocatalysts, are at the stage of development. Each type of photoreactor has advantages and disadvantages, and the choice of one or another photoreactor depends on the operating conditions. For example, in regions with a warm climate, solar photocatalytic installation with the possibility of working year-round would be more effective, whereas in cold regions, the most suitable reactors are economic with effective artificial light sources. In addition, hybrid photocatalytic reactors need to be developed to improve efficiency and reduce operating costs by combining the optimal qualities of these devices.

Table 2 lists recent pilot-scale photocatalytic developments and compares their operating parameters directly affecting the overall efficiency in photocatalytic water purification.

3.3. Large-scale air purification

As air pollution is a serious concern in today's society, attempts are being made to create technologies to clean the air of various kinds of pollutants. Among air pollutant control methods such as adsorption with highly porous materials, UV irradiation and nonthermal plasma causing unwanted byproducts such as ozone and thermal oxidation requiring high costs of additional energy supply, photocatalysis technology has the following advantages: it can do without external additional energy or chemical supply as it is powered by free sunlight; it can completely degrade volatile organic compounds (VOCs); it is fully capable of treating air in a safe and environmentally sound manner; and it has the ability to clean air at all times. As in the case of water purification, large-scale photocatalytic air purification is at a developmental stage and is limited by low purification rates, the possibility of photocatalyst deactivation and the difficulty of scaling (Sharma et al., 2022). However, on the way to scale, pilot developments have been proposed to clean various air pollutants ranging from small rooms to huge production facilities (Lekshmi et al., 2020; Muscetta and Russo, 2021), which will be discussed in this section.

UV disinfection is mainly used to disinfect indoor air from bacteria, viruses and other pathogens (C. Wang et al., 2019a). However, UV irradiation is not effective enough to fully inactivate bacteria with multiple layers, such as *Bacillus subtilis* spores (Zacarias et al., 2019). In contrast to UV sterilization, photocatalysis (Ahmadi et al., 2021) can fully disinfect airborne pathogens on a large scale. For example, antibacterial experiments have been carried out using a pilot-scale flat photocatalytic reactor on a continuous scale (Abou Saoud et al., 2021). Cu-Ag/TiO₂ was used as the photocatalyst and showed good results in sterilizing bacteria and removing volatile organic compounds (VOCs). Another TiO₂/MXene composite based on a dynamic photocatalytic reactor was more effective in inactivating antibiotic-resistant bacteria compared to conventional bacteria, providing a basis for industrial application of this development (Liming Liu et al., 2022b).

In addition to sterilization, photocatalytic oxidation is one of the most effective methods of degrading VOCs in indoor air (Mata et al., 2022; Sharma et al., 2022; Shayegan et al., 2022). Highly toxic VOCs such as formaldehyde (He et al., 2022), benzene (L. Chen et al., 2022b), toluene (Zhang et al., 2022b), chlorobenzene (Zhang et al., 2022a) and others can

be safely removed by photocatalysis using both UV and UV-visible lamps and energy-efficient UV-emitting diodes (Rouhani and Taghipour, 2022; Wu et al., 2022). There are not a few large-scale technologies aimed at cleaning premises from VOCs in natural conditions (Alkaabi et al., 2022; Malayeri et al., 2021; F. Wang et al., 2022a; Xu et al., 2021). Nevertheless, industrial application is very difficult due to scaling and real conditions in contrast to laboratory applications (F. Wang et al., 2022a). The results of studies on photoreactors of three scales (full scale, pilot scale and laboratory scale) for removal showed that the key operating parameters limiting the efficiency of large-scale photocatalytic oxidation are a high flow rate, high concentration and short residence time of VOCs at the inlet (Shayegan et al., 2019).

Recently, solar photocatalysis technology has attracted much scientific attention in the photocatalytic reduction of the greenhouse gas CO₂ into renewable fuels (Dilla et al., 2019; Nguyen and Wu, 2018). Although there are not few photocatalytic materials for converting CO₂ into carbon fuel (e.g., into methane (Kumar et al., 2020; Sun et al., 2022), ethane (G. Wang et al., 2022b; Xie et al., 2022), CO (Dong et al., 2022; Liu et al., 2019), HCOOH (X. Liu et al., 2022c; Zhou et al., 2022)), research has been conducted on a laboratory scale, and there is a need for photoreactors for industrial operation (Khan and Tahir, 2019). Advances in this field have contributed to the development of a mesoscale photocatalytic reactor, which can operate in continuous mode for photoconversion of CO₂ (Nabil et al., 2021, 2022). The advantages of the proposed plant for large-scale implementation are the possibility of continuous operation; productivity in a relatively large volume (several g/day); use of energy-saving LEDs; uniform illumination; high area relative to volume for efficient mass and photon exchange; and additional integration of a chilled water bath to improve CO₂ solubility. In addition, membrane photocatalytic reactors can also be operated in continuous mode and show high efficiency in the photoconversion of CO₂ to carbon fuel (Baniamer et al., 2021; Kandy et al., 2021). For example, continuous membrane photoreactors using naphion as a photocatalyst carrier are 10 times more efficient in the production of carbon fuel compared to batch photoreactors (Pomilla et al., 2018).

Another important aspect in photocatalytic air purification is the reduction of photocatalyst activity due to deactivation during the photoreaction. At the gas-solid interface, the reduction of activity is more noticeable in comparison with the liquid-solid interface because in the latter case, water by dissolution can clean the photocatalyst surface, whereas the solvent capacity of the air medium is limited. Thus, undesirable components such as decomposition intermediates can accumulate on the photocatalyst surface, which not only limits light input but also blocks the diffusion of oxygen and other products to the reaction centers, thereby seriously limiting large-scale application under realistic conditions (Weon et al., 2019). There are several methods to regenerate deactivated photocatalysts at the expense of heteroatoms and carbonaceous deposits, including UV treatment with deionized water (Pei et al., 2021; Pylynev and Wong, 2019), thermal treatment (Chen et al., 2020), and washing with aqueous H₂O₂ solution (Serge-Correales et al., 2022). However, these regeneration methods, due to their added complexity and cost, may limit their practical application on a full scale; hence, preventing and minimizing photocatalyst deactivation by optimizing photocatalysts and photoreactors is a priority (Cha et al., 2019; Kim et al., 2022; Ran et al., 2019; H. Wang et al., 2019b).

Another problem with photocatalytic air purification technology is the low purification rate. To increase the process efficiency, photocatalytic treatment is combined with other methods of air purification, e.g., thermal catalytic oxidation (Wei et al., 2021), nonthermal plasma (Guo et al., 2022; Kvss et al., 2023), and adsorption (Meicheng Wen et al., 2019a; Zou et al., 2019). This synergy can help to level out the disadvantages and take advantage of each of the methods; for example, the photocatalysis process can be used as an end stage in VOC purification, as it is more effective in purifying pollutants with low concentrations (below ppm) (Weon et al., 2019), whereas other methods are more effective for air purification only with high concentrations (Krishnamurthy et al., 2020). For example, in the plasma-photocatalytic method, plasma splits high concentration air pollutants and their decomposition

Table 2
Recent pilot-scale photocatalytic reactors for the efficient degradation of water pollutants.

Reactor type	Reactor design	Total volume, (L)	Flow rate, (L·h ⁻¹)	Pollutant	Concentration (mg·L ⁻¹)	Photocatalyst (conc.)	Light source	Efficiency	Reusability	Ref.
Slurry reactor	CPC	85	N/A	O-desmethyl venlafaxine	6074.28×10^{-6}	TiO ₂ (200 mg·L ⁻¹)	Sunlight	90 % degradation at 240 min	From 70 % to 63 % after 3 cycles.	(Rapti et al., 2022)
Slurry reactor	CPC	35	N/A	Pesticides	0.2	TiO ₂ -rGO (200 mg·L ⁻¹)	Sunlight, 30 W·m ⁻²	100 % degradation at <25 min	N/A	(Luna-Sanguino et al., 2020)
Slurry reactor	CPC	16	1584	Sulfamethoxazole, diclofenac	0.1	BEN + VER (200 mg·L ⁻¹)	Sunlight, 40 W·m ⁻²	80 % degradation at 180 min	N/A	(Martínez-Costa et al., 2020)
Slurry membrane reactor	Cylindrical	25	1200	Diclofenac, TOC	470×10^{-6}	TiO ₂ (500 mg·L ⁻¹)	UV-C, 2.08 W·L ⁻¹ , 253.7 nm	90.5 % and 52.4 % degradation at 180 min	N/A	(Plakas et al., 2016)
Slurry reactor	Cylindrical	2	N/A	COD	0.030	TiO ₂ (990 mg·L ⁻¹)	8-W UV-C 8 lamps	77 % degradation at 90 min	N/A	(Ahmadpour et al., 2022)
Slurry reactor	Cylindrical	1	0.06	Clofibric acid	20	TiO ₂ (500 mg·L ⁻¹)	150 W Ha-Hg lamp, 350–410 nm	$\eta_{\text{rxn}} = 4.59 \%$	N/A	(Manassero et al., 2017)
Slurry reactor	Cylindrical	280	60	Cefixime, cefactor, cefuroxime	10	Bi ₂ TiO ₂₀ (1500 mg·L ⁻¹)	24 W UV, 200 W·m ⁻² , 270 nm	94 %, 81 %, and 69.71 % degradation at 210 min	N/A	(Baaloudj et al., 2022)
Slurry reactor	Tubular	25	15	COD	1420	Ag-TiO ₂ (200 mg·L ⁻¹)	Sunlight, 10 W·m ⁻²	99 % degradation at 120 min	High stability after 5 cycles	(Saran et al., 2019)
Fixed-film reactor	Staircase	2	29	Flumequine	5	TiO ₂ (film thickness ~ 1 mm)	UV + H ₂ O ₂ , 38 mW·m ⁻² , 365 nm	97 % degradation at 150 min	N/A	(Kane et al., 2022)
Fixed-film reactor	Cylindrical	1	0.06	Clofibric acid	20	TiO ₂ (film thickness - 2.7 µm)	150 W Ha-Hg lamp, 350–410 nm	$\eta_{\text{rxn}} = 2.02 \%$	N/A	(Manassero et al., 2017)
Fixed-film reactor	Inclined plate	0.2	0.02	Carbamazepine, diazinon	4.8, 34.2	CNTs/MOF-808 (film thickness - 1 mm)	400 W metal-halide lamps, 511 nm	91.6 % degradation at 90 min, 99.7 % degradation at 60 min	From 91.6 % to 83.1 % after 5 cycles. From 99.7 % to 92.1 % after 5 cycles	(Samy et al., 2021)
Fixed-film reactor	Annular	0.6	90	Clofibric acid	1.8×10^{-7} mol/cm ³	TiO ₂ (film thickness - 3.2 µm)	40 UV-LEDs, 365 nm	70 % degradation at 360 min	N/A	(Manassero et al., 2022)
Fixed-film reactor	Inclined plate	5	1.20	Methylene blue	N/A	Cu/TiO ₂ -glass, Cu: TiO ₂ -LDPE	Sunlight, 609.6 W·m ⁻²	73.7 % degradation at 360 min, 68.1 % degradation at 360 min	From 73.7 % to 51.2 % after 3 cycles. From 68.1 % to 66.3 % after 5 cycles	(Pava-Gómez et al., 2021)
Fluidized-bed reactor	Cylindrical	4	0.60	Diclofenac	0.05	C-TiO ₂ polymer (500 mg·L ⁻¹)	8000 lx 27 vis-LEDs, > 400 nm	100 % degradation at 120 min	high stability after 6 cycles (washed Di water)	(Surenjan et al., 2019)
Fluidized-bed reactor	Cylindrical	0.1	0.06	Bisphenol A	5	TiO ₂ -graphene (100 mg·L ⁻¹)	350 W Xe lamp	97 % degradation at 30 min	From 97 % to 92 % after 5 cycles	(Fang et al., 2019)
Fluidized-bed reactor	Tubular	0.8	31.8	Phenol	1.0 mmol L^{-1}	TiO ₂ -silica gel (20 g·L ⁻¹)	UV-C lamp	100 % degradation at 240 min	N/A	(Rincón and La Motta, 2019)
Fluidized-bed reactor	Cylindrical	37.8	44,961	Tebuconazole	0.02	Phenalenone-sand (30 g·L ⁻¹)	1.2 W·m ⁻² , 254 nm 5-W blue LED, 435–465 nm	73 % degradation at 480 min	From 73 % to 49 % after 3 cycles	(Aimeur et al., 2021)
Packed-bed reactor	Cylindrical	1	0.06	Clofibric acid	20	TiO ₂ (film thickness ~ 0.44 µm)	150 W Ha-Hg lamp, 350–410 nm	$\eta_{\text{rxn}} = 2.96 \%$	N/A	(Manassero et al., 2017)
Packed-bed reactor	Cylindrical	0.007	0.07	Crystal violet	10	ZnO-zelite (4 g)	UV-LEDs 12 W·m ⁻¹	93 % degradation at 4.7 min	N/A	(Sacco et al., 2018)
Packed-bed reactor	Cylindrical	70.68×10^{-6}	0.01	Methylene blue	4.6	TiO ₂ -Glass beads (film thickness ~ 200 µm)	300 W Xe-lamp, ~1070 W·m ⁻²	96 % degradation at 20 c	From 93.73 % to 83 % after 25.5 h	(Zhang et al., 2020)
Packed-bed reactor	Cylindrical	0.5	3	Methylene blue, TOC	7, 5	N-TiO ₂ /polystyrene spheres	vis-LEDs, 780 W·m ⁻² , 400–700 nm	100 % degradation at 120 min, 80 % degradation at 180 min	High stability after 5 cycles	(Sacco et al., 2019)
Packed-bed reactor	Rectangular	0.05	12.96	Methylene blue	10	TiO ₂ -Glass beads (1.9 g·L ⁻¹)	3.264 W 192 LEDs, 191 W·m ⁻² , 375 nm	$k_{\text{app}} = 0.82 \text{ min}^{-1}$	N/A	(Claes et al., 2019)

intermediates, preventing accumulation on the surface, thus regenerating the photocatalyst and producing byproducts in the form of NO_x and O_3 , which are decomposed by photogenerated radicals ($\text{H}^\cdot$, $\text{HO}^\cdot$). In particular, scientists on a pilot scale using a photocatalysis/nonthermal plasma hybrid have achieved improved efficiency in the removal of ammonia, oil aldehyde (Abou Saoud et al., 2018) and toluene (Mohammadi et al., 2020) compared to using these methods alone. The combination of adsorbent and photocatalyst promotes instant adsorption of target molecules and gradual degradation by photogenerated active centers (Zou et al., 2019). Such hybridization helps to overcome the mismatch in the timing of rapid mass influx and slow photodegradation of airborne pollutants (Li et al., 2020). On the other hand, thermal photocatalysis makes it possible to utilize the full spectrum of sunlight by utilizing both solar and thermal energy, whereas the potential of traditional high-performance photocatalytic materials is in the UV and visible region, which accounts for approximately 47 % of the entire solar spectrum (Xi Chen et al., 2019c). In addition, thermocatalysis extends the lifetime of photocatalysts by additional high-temperature oxidation of undesirable intermediates accumulating on the catalyst surface (Krishnamurthy et al., 2020). In such systems, catalysts made of noble materials (Pt/TiO_2 (Vikrant et al., 2021), Pt/MO (Yu et al., 2020) and $\text{MnO}_2\text{-Au}$ (da Silva et al., 2018)) show high catalytic activity in air purification but have high cost and low thermal stability. However, transition metal oxides have recently been used as an alternative to noble metals to improve efficiency (Xi Chen et al., 2019c). In particular, a $\text{CeO}_2/\text{LaMnO}_3$ composite with a wide light absorption band (800–1800 nm) can be used as an efficient photothermal catalyst with an efficiency of 15.3 % in toluene conversion (J.-J. Li et al., 2019c). Based on this strategy, a pilot photothermocatalytic system was proposed, with simultaneous space heating and air purification under the influence of solar energy. The authors used a $\text{MnO}_x\text{-CeO}_2/\text{TiO}_2$ composite as a catalytic material, and the air-cleaning rate of the developed system was in the range of 5–23 m^3/h with the possibility of application for indoor air cleaning (Li et al., 2023).

A detailed analysis of works on photocatalytic air purification research suggests that further research is still needed for large-scale photocatalytic air purification under realistic conditions to bridge the gap between laboratory and production testing. Further strategies should be directed (1) toward the cleaning of multicomponent mixtures, which is more feasible under natural conditions, whereas only one component is mainly cleaned; (2) to develop inexpensive and simple photocatalytic systems that will prevent or regenerate the deactivation of photocatalysts; and (3) to introduce hybrid technologies together with the photocatalytic process whose advantages have been summarized above.

4. Conclusions and strategies for the future

The photocatalytic process is recognized as a promising technology for solving environmental and energy problems as a clean and safe method that does not require additional energy-consuming systems. However, although much effort has been directed to the study of photocatalysts, the practical application of photocatalysis on an industrial scale is still limited. This article details the main limiting obstacles and possible solutions for moving from a laboratory to a large-scale process, including large-scale production of high-performance photocatalysts in a simple and environmentally friendly way, efficient immobilization of photocatalyst particles on the surface of the desired material and support design, and criteria for choosing a photocatalytic reactor with the required optimal geometry to provide an effective three-phase boundary with maximum area. In addition, a review and comparison of operational parameters from recent pilot studies for large-scale photocatalytic hydrogen production and water and air purification was made. Based on the analysis of the above studies, future strategies in developing the ideal photocatalytic system for practical full-scale use should have the following directions:

1. The development of highly efficient photocatalysts with the possibility of multiple reuses with minimal loss of photoactivity. The regeneration

of photocatalysts deactivated by contaminants or prevention of photocatalyst deactivation to help increase the service life and maximize material savings. Numerous studies show that the immobilization of photocatalyst particles on a solid base is the most suitable option for reuse with minimal loss of efficiency. Despite this, more research should be carried out aimed at measuring the service life and developing a method to combat the deactivation of photocatalysts.

2. More efforts need to be directed toward the efficient use of free sunlight for the large-scale photocatalytic process through the introduction of additional systems capable of concentrating and focusing direct and diffuse beams.
3. Modifications of optimal designs of photocatalytic reactors with increased mass transfer of reaction products and maximum use of incident photons to increase overall performance.
4. Consideration of the possibility of combining the photocatalytic process with other methods for solving environmental and energy problems, thereby serving to balance the disadvantages and optimize the advantages of each.
5. To conduct a comprehensive cost estimate of a photocatalytic system for commercialization and comparison with other technologies, it is necessary to provide data on the prices of the materials used and the costs of operation.
6. Finally, the photocatalytic process is a “green” technology, and scientists need to make more effort to spread its benefits in order to address energy and environmental problems by overcoming barriers to commercialization and improving economic efficiency.

It is expected that this comprehensive review will assist future research in obtaining up-to-date insights and overcoming possible barriers to scaling up and commercializing photocatalytic systems.

CRediT authorship contribution statement

Zhengisbek Kusanov: Conceptualization, Methodology, Investigation. **Baglan Bakbolat:** Conceptualization, Methodology, Investigation. **Alzhan Baimenov:** Resources, Formal analysis. **Aidos Issadykov:** Writing – review & editing. **Mukhtar Yeleuov:** Writing – review & editing. **Chingis Daulbayev:** Writing – original draft, Project administration.

Data availability

No data was used for the research described in the article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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