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Opinion

Reflections on Photocatalysis Progress Since the Inspiration of Prof. David Ollis in 1992

Lianfeng Zhang [1], Mehrab Mehrvar [2], Zisheng Zhang [3] and William A. Anderson [4,*]

[1] Laboratory of Ecology and Environmental Protection, Research Institute of Tsinghua University in Shenzhen, Shenzhen 518057, China; tyou6@hotmail.com (L.Z.)
[2] Department of Chemical Engineering, Toronto Metropolitan University, Toronto, ON M5B 2K3, Canada; mmehrvar@torontomu.ca (M.M.)
[3] Department of Chemical and Biological Engineering, University of Ottawa, Ottawa, ON K1N 6N5, Canada; zzhang@uOttawa.ca (Z.Z.)
[4] Department of Chemical Engineering, University of Waterloo, Waterloo, ON N2L 3G1, Canada

* Corresponding author. E-mail: wanderson@uwaterloo.ca (W.A.A.)

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ABSTRACT: Why has photocatalysis not gained the wide-ranging commercial applications in environmental purification of air and water that seemed promising 30+ years ago since the first international conference on TiO_2 photocatalytic purification and treatment of water in 1992? The primary reason lies in its low intrinsic efficiency. The progress of R&D to enhance this efficiency has been slow, possibly due to an incomplete understanding of the underlying mechanisms of photocatalysis. There is also the possibility that certain factors, with effects comparable to those of the band gap, significantly influence photocatalytic performance but remain underexplored. Additionally, challenges such as mass transfer limitations and surface contamination hinder the industrial application of photocatalysts. It may be time for scientists to reconsider and address the limitations and practical application scenarios of photocatalysis.

Keywords: Photocatalyst; TiO_2; Mechanisms; Water treatment; Air purification; Electron-hole recombination

1. Introduction

In 1992, the 1st International Conference on TiO_2 Photocatalytic Purification and Treatment of Water and Air was held in London, ON, Canada, with the attendance and participation of many pioneers in the field. A book of conference proceedings was edited and published by Ollis and Al-Ekabi in 1993 [1]. In the preface to that book, the editors encouraged researchers to C.I.T.E photocatalysis, meaning Compare (with other technologies), Integrate (with complementary technologies), Transfer (results into developments and applications), and Evaluate (the economics and optimization of photocatalysis). In memory of Prof. Ollis, it is perhaps useful to reflect on those encouragements and where the field now stands in relation to where it was in 1992. Of particular interest is a reflection on why photocatalysis has not gained the wide-ranging environmental and commercial applications that seemed very promising 30+ years ago. Although photocatalysis topics have been reviewed many times over the years, it is perhaps useful to provide a higher level reflection on broad issues such as mechanistic understandings, efficiencies, wavelengths, competing technologies, and other theoretical and practical barriers. Sometimes, these reflections are lost in the more detailed scientific reviews on materials and chemistry.

In the preface to their book, Ollis and Al-Ekabi [1] mention that publication activity in the photocatalysis field has included more than 200 papers per year since about 1982. Figure 1 shows an updated annual publication count using a Scopus database search on the keyword “photocataly*” in the title, abstract, and keywords of publications from 1993 to 2024.

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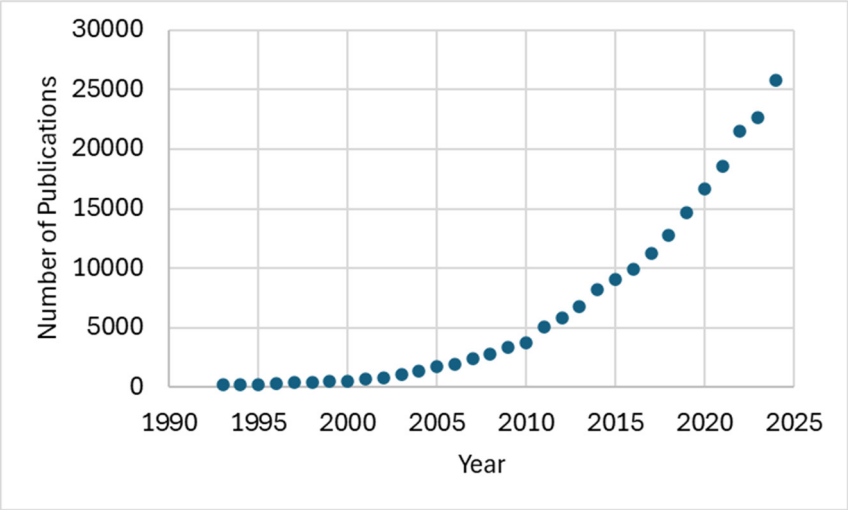

Figure 1. Number of publications each year with "photocataly*" (*i.e.*, photocatalysis, photocatalytic, *etc.*) in the title, abstract, or keywords, as counted using the Scopus database from 1993 to 2024 (as of February 2025).

Clearly, in Figure 1, the research and publication activity dealing with photocatalysis has grown substantially, with over 20,000 papers per year in the last three years. The cumulative total in this dataset is over 210,000 and it has followed almost exponential growth until recent years. If there has been a lack of commercial success for photocatalysis in environmental applications, it does not appear to be due to a lack of effort. A glance through the literature suggests that there has been plenty of effort to Compare (in the C.I.T.E. scheme) and to Integrate. There have also been efforts to Transfer and Evaluate (including optimizing). Then, what are the continuing barriers to success?

In the 1992 conference proceedings, photocatalysis was heralded as a promising "clean and green purification technology" [1]. However, over the following 30+ years: (a) the application of photocatalytic pollutant removal still remains primarily confined to laboratory or occasional pilot-scale tests (for example [2–15]), and is still frequently described with the same adjective "promising" as it was 30+ years ago [1,15]; and (b) the progress in developing a theoretical framework for photocatalysis has been limited, remaining insufficient to support R&D efforts for commercialized industrial applications.

TiO_2 is still in the leading position as the photocatalyst of choice in most studies, and in the proposed industrial market, choices other than TiO_2 are scarce. Although some TiO_2-based pre-commercial or actual products using photocatalysts are available on the market [1,15,16], doubts about their actual effectiveness persist. For example, for indoor air purification or disinfection devices, the applications of photocatalysts seem to always be in combination with other methods, such as filtration, activated carbon, ionization or others. Such applications of photocatalysts serve two purposes: (1) delivering the potential functional effects of photocatalysts, and (2) showcasing the device's association with sophistication and advanced technology. Evaluating the effectiveness of such reactors poses challenges in distinguishing the contributions of other methods from those of photocatalytic decomposition, as well as separating the effects of adsorption from those of photocatalytic decomposition. In water treatment, the situation is similar, where photocatalysts are often combined with ozone, H_2O_2, or others [17]. Therefore, a question occurs: Why has photocatalysis not yet gained the wide-ranging commercial applications in environmental purification (air and water treatment) that seemed promising 30+ years ago? Some opinions, which might be a shift from conventional perspectives, are offered here to stimulate thought and discussion in the research and commercialization community that might help shift efforts towards productive outcomes.

2. Intrinsic Efficiency of Photocatalyst

In Fujishima and Honda's seminal paper on photocatalysis [18], it was stated that "shorter than 415 nm, that is 3.0 eV, which corresponds to the band gap of TiO_2", implicitly establishing the mechanism for photocatalysis. This explanation corresponded well with observed photocatalytic phenomena. This band gap theory remains valid to this day and is routinely cited or illustrated in Figure 2 [15,16]. However, it should not be forgotten that the paper [18] also suggested that wavelengths below 1000 nm can drive materials with a band gap larger than 1.23 eV to decompose water. After half a century of scientific efforts, it is clear that this is not the reality. Therefore, it is worth noting that while the band gap theory provides some superficial explanations, it fails to effectively guide or predict outcomes in the research of photocatalysts, as illustrated below.

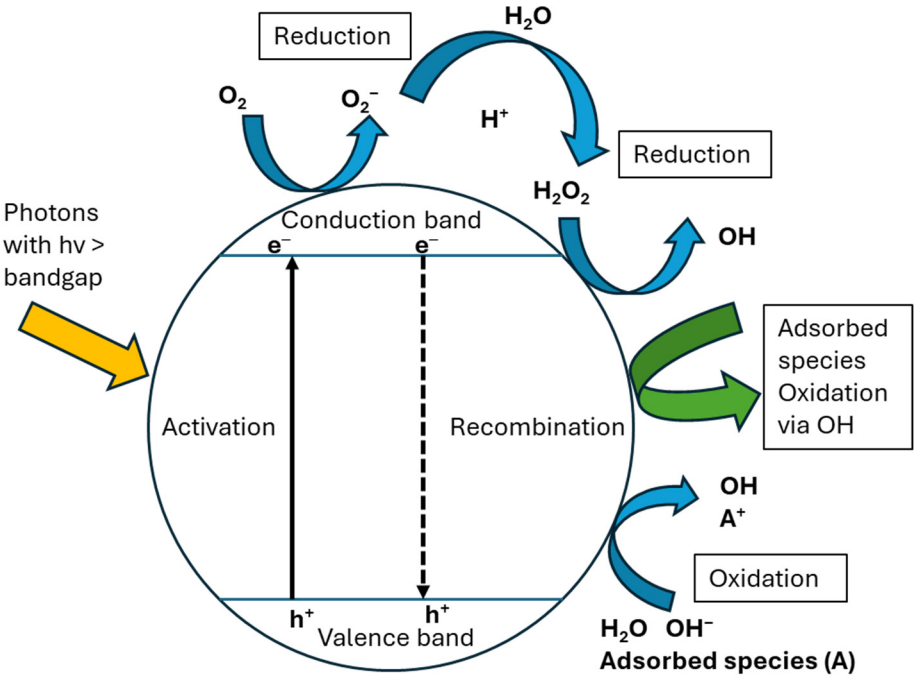

Figure 2. The band gap theory of photocatalysis, illustration inspired by [16] and others.

2.1. Low Efficiency and Insufficient Knowledge of the Mechanism

According to the band gap theory, the efficiency of photocatalysts should be significantly higher than what is commonly observed. For example, in a recent study, it was reported that using a 250 W lamp, 50 mL of a 20 mg/L phenol solution, and 800 mg/L of photocatalyst, the phenol concentration could be reduced to nearly zero within one hour [19], corresponding to a degradation rate of approximately 1 mg/h (0.0106 mmol phenol/h). This degradation rate appears remarkably low, far from the needs of industrial applications if considering the consumption of 250 W of electrical power. Theoretically, 250 W of electrical power could decompose up to 602 mmol of phenol/h (assuming that the wavelength is 400 nm, UV production efficiency is 20%, and all UV photons arrive at the photocatalyst). The prevailing explanation for the observed low efficiency is the recombination of excited electrons and positive holes, as illustrated in Figure 2. Following this reasoning, photocatalysts linked to an electrical bias might be considered the most viable approach to enhancing efficiency [20]. However, in the field of photocatalytic environmental purification, the reports on photocatalysts employing a bias remain somewhat scarce. In organic synthesis, the term "electrophotocatalysis" is mentioned, which describes the concept of merging electrosynthesis with photochemistry, but notably, it does not involve solid semiconductor catalysts [21]. Are there other factors that predominantly control the recombination of excited electrons and positive holes?

Researchers have never been confined to the band gap theory alone. To enhance the efficiency of photocatalysts, they have developed numerous new or modified photocatalytic materials. Their focus has never been limited to the band gap; instead, they have consistently conducted analyses using advanced characterization techniques such as X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), high-resolution TEM (HRTEM), Fourier-transform infrared spectroscopy (FTIR), and others [7,10,19,22–26]. This suggests an implicit acknowledgment that band gap theory alone is insufficient to explain, predict or enhance the photocatalytic performance of materials fully.

2.2. Efforts to Elevate Efficiency

Researchers have been focusing their efforts on enhancing the efficiency of photocatalysts in three main directions, as described in the following subsections.

2.2.1. Adding Additional Elements or Factors to TiO_2

It is well-reported that doping metals or attaching metal oxides onto photocatalysts is one approach to enhancing their efficiency [4,24,26,27]. However, this method lacks a solid theoretical foundation and relies heavily on empirical observations. Figures similar to Figure 3 frequently appear in the literature on photocatalysis, presenting the prevailing

mechanistic explanation for the effects of "doping". Unfortunately, these representations do not adhere to a consistent conceptual framework. The energy band describes the possible distribution range of electron energy, and the band structure diagrams derived from band theory are theoretical representations that do not accurately reflect the actual geometry of photocatalysts. In contrast, the doped metal scenario depicted in Figure 3 suggests a geometric structure. Consequently, Figure 3 conflates the energy band diagram with a geometric representation, leading to an incorrect visualization or paradigm. Thus, it suggests that no reasonable theory currently exists to explain the effectiveness of doping adequately, and in this regard, scientists have been operating in the dark.

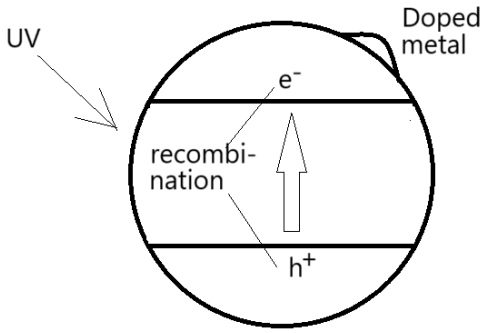

Figure 3. Illustration of the band gap theory of photocatalysis with doped metal.

2.2.2. Synthesizing New Materials with Photocatalytic Functionality

To date, TiO_2 remains in the leading position among various materials for photocatalysts, particularly those directed towards industrial applications, due to its low cost relative to more exotic formulations. Progress towards finding or making new photocatalysts with significant performance improvements has been slow, in part because the mechanism of photocatalysis is not yet fully understood. For example, if the band gap theory were sufficient to guide photocatalyst research, it would be relatively straightforward to formulate or identify new photocatalysts from materials with known band gaps and suitable band positions for the desired redox chemistry. Some insights into heterojunction photocatalysts have been generated in recent years [28], but major advances continue to be elusive. The primary issue lies in the fact that while band theory provides valuable insights, it is not sufficient on its own. This points to an incomplete understanding of the photocatalytic mechanism.

2.2.3. Longer Wavelengths

Figure 4 illustrates the band gaps of the potential materials excited by light with various wavelengths. Utilizing longer-wavelength light, particularly visible light, presents two significant advantages: it is safe for human exposure and allows for more efficient utilization of solar energy compared to relying solely on the UV spectrum. However, the current mechanism contradicts this approach, as will be discussed in detail in the following section. Specifically, visible light might not be considered a viable pathway for significantly enhancing the intrinsic efficiency of photocatalysts, even if it has advantages in terms of broader spectrum use.

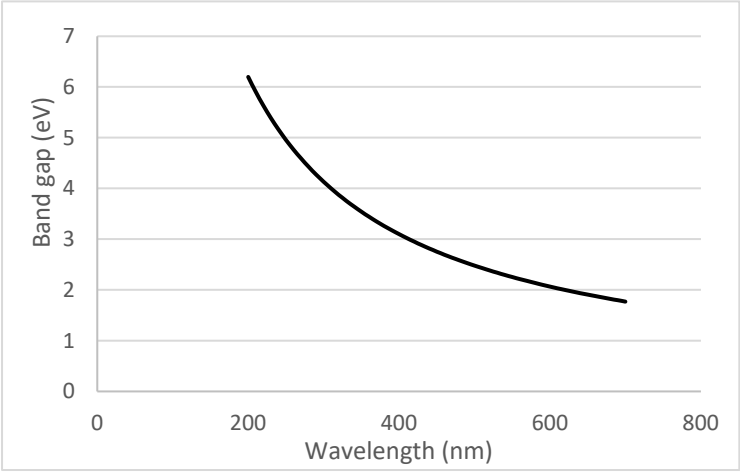

Figure 4. Band gap and corresponding wavelength.

2.3. Two Key Points of the Band Gap Mechanism

The band gap is a key property that distinguishes conductors, semiconductors, and insulators. In photocatalysis, if the energy of a photon is equal to or greater than the band gap, the material can theoretically be excited to act as a photocatalyst. From this perspective, there should be no inherent difference between semiconductors and insulators—all could potentially function as photocatalysts if excited by photons with suitable energy. Notably, no scientific reports seem to have explicitly linked the electrical conductivity of materials to their photocatalytic properties.

The band gap mechanism of photocatalysis is related to two critical factors: the size of the band gap and its position (potential). Current understanding suggests that the excited electron and positive hole generated by photon absorption drive the reaction. However, this theory does not sufficiently address the roles of the size and position of the band gap, nor does it clarify whether the band position has an influence. For example, Figure 5 illustrates the mechanism of a type of hybrid photocatalyst. While this appears to provide a reasonable explanation, it also raises questions: Why do the electrons jump as depicted in Figure 5? How do these jumps achieve "efficient charge separation" [17]? What is the relationship between these jumps and the size and position of the band gaps? These questions highlight flaws in the theory, which seems to be far from fully developed.

It has been hypothesized that higher-energy excited electrons lead to more reactive photocatalytic processes [4]. Until this hypothesis is proven incorrect, TiO_2 and materials with even larger band gaps theoretically represent the upper limit of photocatalytic efficiency. Therefore, before acquiring a deeper understanding of the photocatalytic mechanism, scientists may need to reconsider whether pursuing longer wavelengths, such as visible light, is a viable approach to improving photocatalyst efficiency.

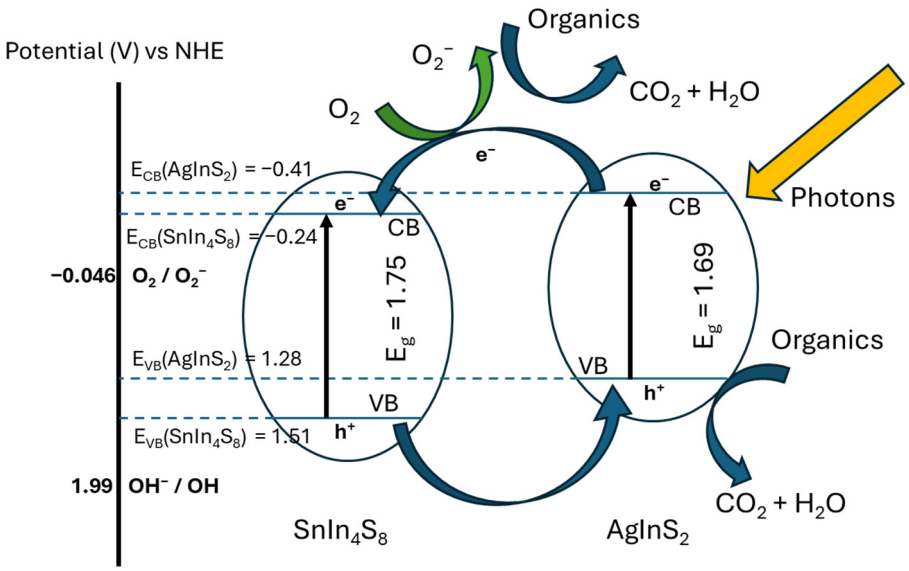

Figure 5. The position of the band gaps in a hybrid photocatalyst, adapted from [17].

In short, are there one or more factors, in addition to the band gap, that significantly influence the mechanism of photocatalysis? It is well known that anatase TiO_2 exhibits a higher photocatalytic efficiency than rutile TiO_2, but what accounts for this difference? If a mere 0.2 eV difference in band gaps is accepted as the primary reason, then logically, all efforts to pursue visible light-driven photocatalysts would be futile. However, numerous scientific reports on visible light-driven photocatalysts indicate that researchers recognize the band gap size as one factor, but they do not accept it as the only dominant reason, as discussed in Section 2.1. In any case, the current theoretical understanding of the photocatalytic mechanism is insufficient to guide R&D in this field effectively, and greater mechanistic insights are needed.

3. Photoreactor Designs

3.1. Mass Transfer and Fluid Mechanics

Photocatalytic reactions can only occur on the surface of the photocatalyst. For air or water purification, pollutants must undergo three steps to be decomposed: adsorption onto the photocatalyst surface, decomposition when photons simultaneously reach the surface, and desorption of the reaction products. Therefore, designing a reactor with an effective flow pattern that allows pollutants in air or water to consistently and rapidly contact the photocatalyst is crucial.

The main challenge is ensuring that all pollutants have the opportunity to interact with the photocatalyst surface while minimizing diffusion barriers (*i.e.*, boundary layer thickness at the surface) [29–31]. The solution to mass transfer limitations for immobilized photocatalysts is to enable air or water outside the reactors to recirculate through them at reasonably high velocities with low reaction conversion per pass. From a mass transfer perspective, single-pass flow reactors appear to be often unsuitable for photocatalyst applications, especially for water phases where the diffusional resistance at the surface is significant. Therefore, efforts to improve reactor design have generally been focused and limited to recirculating flow systems rather than single-pass flow reactors. Low surface reaction rates due to electron-hole recombination and photon transfer limitations further limit the overall system and likewise lead to the need for recirculating systems.

Furthermore, compared to photocatalysis, ultraviolet (UV) photolysis, especially vacuum ultraviolet (VUV) photolysis [32,33], seems to offer significant advantages because the reactions occur homogeneously and not on the surface of a solid such as a photocatalyst. For example, in the production of ultrapure water in the semiconductor industry, VUV photolysis technology has already been widely used industrially for many years [34], whereas photocatalysis has never been employed in such processes. Attempts at microbial disinfection using photocatalysis have similarly suffered from mass transfer problems [35], whereas the use of homogeneous germicidal UV in water treatment has been highly successful.

3.2. Light Source and Photon Transfer

The applications of visible and ultraviolet (UV) light sources differ significantly in design, as the latter requires considerations to prevent potential harm to humans. Currently, excitation light sources for photocatalysts are still primarily ultraviolet light. The harmful properties of UV light necessitate that the reactors be sealed to prevent UV leakage, and for water purification, UV transparency must also be considered and may be a limiting factor. The exciting news is that new types of light sources (UV-LEDs) can provide more geometric flexibility in design than the older mercury UV lamps. The cost of UV-LED in the UVA band has reached a level which now may be good enough for large scale industrial applications. As discussed in Section 2.3, based on the current theory of photocatalysis, achieving success with visible light should logically occur after the success of UV light, which may permit the use of other lower cost LED sources.

3.3. Contamination of the Surface of Photocatalyst

Photocatalytic reactions occur on the surface of photocatalysts, which raises the issue of surface contamination. In air purification, inorganic contaminants such as dust are prevalent and may need to be filtered out. In contrast, water purification faces a significant challenge with chemical contaminants: metal ions can transfer from the water and reduce onto the surface of the photocatalyst through a reaction with electrons. This scenario represents a specific photocatalytic application aimed at recovering metals from metal ions through processes such as doping [4]. However, this situation will often be detrimental to the use of photocatalysts for the decomposition of organic compounds. The only solution is to avoid using photocatalysts in water containing significant amounts of metal ions, which limits their effectiveness in water purification.

3.4. Oxygen

In the photocatalytic decomposition of pollutants, oxygen is essential [36] for its role as an electron acceptor, adding another influencing factor to water purification. The requirement for oxygen complicates the design of reactors for this process. When UV light is the driving force, photocatalysts struggle to compete with photolysis, as oxygen is not necessary for photolytic reactions for compounds susceptible to photolysis. Furthermore, low or no oxygen is a favorable condition for metal ions in the water (if present) to contaminate the surface of the photocatalysts via reduction reactions.

4. Other Challenges

4.1. Biosafety of TiO_2

TiO_2 is still the leader among various photocatalyst materials. But on 8 February 2020, the EU issued a Commission Regulation (EU) amending classification, labeling, and packaging rules, categorizing titanium dioxide as a Category 2 carcinogen [37]. It has significantly impacted related industries globally, especially as the EU mandates warning labels

for titanium dioxide products. Although some scientists have voiced opposition to this regulation [38], researchers and engineers pursuing commercialization must contend with the established classification of TiO_2 as a carcinogen. This raises the dilemma of either abandoning TiO_2 as a photocatalyst or dealing with the biosafety concerns in applications.

4.2. Use of Solar Irradiation

Early research on photocatalysis aimed to use solar radiation as a "free" energy source for environmental treatment [39]. The work targeting this goal has been continuing since then [40] and has been a driving force for the development of photocatalysts with longer wavelength activity. Commercially, the use of solar energy would appear to be a limiting feature for several reasons. In many locations, the availability of sunlight is limited due to clouds and seasonal changes. Potentially, even more importantly, even in optimally sunny locations at the equator, the availability is limited to 12 h, meaning that the effluent to be treated must either be stored for batch processing or inherently limited to daytime operations. From a cost perspective, the efficiency of only using the invested capital cost of the process equipment for half the time or less needs to be considered. Potentially, the energy used by artificial light in a smaller continuously operating system may not be offset by a larger intermittently operating system using "free" energy. If potential commercial applications of solar-driven photocatalysis exist, they appear to be likely limited to specialized or niche scenarios.

4.3. Competing Technologies

The low energy efficiency (due to recombination), reactor design limitations, and scalability challenges appear to have generally held back photocatalytic water treatment, especially in comparison to alternatives. Membrane filtration, activated carbon adsorption, and biological treatments are frequently found to be cost effective and easier to implement solutions to a wide variety of wastewater problems. If photocatalytic solutions are to be implemented, they may continue to be only appropriate for very specific and lower volume problems. To minimize the effects of interfering substances, photocatalysis will often be appropriate only as a final step to treat compounds that are poorly removed by preceding other methods.

5. Conclusions and Prospects

Scientists have made significant efforts in both academic research and potential industrial applications for photocatalytic air and water treatment over the past 30+ years. While some recent topical reviews continue to promote photocatalysis as a new and promising environmental technology [41], others have shown more critical insights into the limitations that need to be addressed [35,42]. The focus of academic efforts has often been on empirical factors and influences such as those related to the mechanism of surface reactions [16,43], but the pressing need is to achieve a more effective understanding of the underlying mechanisms of photocatalysis to aid in materials design. In industrial applications, the emphasis should be placed primarily on demonstrating and maximizing the actual effectiveness of photocatalysts within a treatment device or process rather than using photocatalysis as a marketing point. Other practical or limiting factors that need to be addressed include:

- The intrinsic efficiency of photocatalysts is too low to meet the requirements for industrial applications. Scientists should prioritize gaining a thorough understanding of the photocatalytic mechanism and elevate its efficiency before engineers invest efforts in industrial applications. Currently, the energy band theory serves as the dominant framework for understanding TiO_2 photocatalysis. However, it remains more empirical or speculative than a complete theoretical system. For example, energy band diagrams are often and mistakenly equated with the actual atomic geometry of TiO_2, and there is no clear mechanistic explanation for electron transitions between energy bands.
- The question of where photocatalysts could be effectively applied should continue to be a focus for everyone involved in photocatalysis research and development.
- Theoretically, photolysis seems to be capable of performing nearly all the functions that photocatalysts can for air and water purification. However, it requires UV light, which is harmful to humans. In this context, visible-light-driven photocatalysts appear promising. Nevertheless, based on our current understanding of the photocatalytic mechanism, it is the least likely that visible-light-driven photocatalysts will achieve success any sooner than their UV-based counterparts. This negative cycle hinders the advancement of photocatalysis.
- In practical applications, metal contaminants and other substances can significantly limit the use of photocatalysts in water purification, although such concerns are sometimes not pursued in laboratory research. A focus on

photocatalytic final purification after pre-treatment with other technologies to remove interfering substances will continue to be needed.

- Single-pass flow reactors do not appear to be promising due to limitations in mass transfer, even if the intrinsic efficiency of the photocatalyst is significantly improved.

As originally inspired by Prof. David Ollis in the early 1990s, researchers should continue to deeply consider the current and existing challenges, limitations, and realistic opportunities for the development and applications of photocatalysts. Researchers are encouraged to continue to reflect on the broader issues, opportunities and challenges for their own ideas on photocatalysis development, as well as a deeper understanding of the mechanisms at work.

Author Contributions

Conceptualization, W.A.A.; Investigation, W.A.A., L.Z., M.M., Z.Z.; Writing—Original Draft Preparation, L.Z.; Writing—Review & Editing, W.A.A., M.M., Z.Z.; Visualization, L.Z.; Supervision, W.A.A.

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Informed Consent Statement

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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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