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# Views & Comments Photo-Based Advanced Oxidation Processes for Zero Pollution: Where Are We Now?

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# 1. Introduction

There is a pressing need to transition into a sustainable society, committed to achieving zero pollution. Water, humanity's most precious resource, can galvanize the environmental, social, and economic transformations called for in the European Union (EU) Green Deal. Improved water use and protection are essential components for addressing the global climate and biodiversity crises. To achieve the goal of zero pollution, investment is required and specific policies need to be implemented to address certain contaminants of emerging concern (CECs), such as endocrine-disrupting chemicals, pharmaceuticals, and microplastics; this is particularly pertinent given that conventional wastewater treatment is ineffective in dealing with these pollutants [1–4]. As such, innovative solutions that facilitate the transition into processes with zero pollution discharges are highly desirable.

Among the various solutions, photo-based advanced oxidation processes (photo-AOPs), which are able to break down trace CECs, have attracted considerable research attention, as reflected by the plethora of studies in the last decade (Fig. 1). Despite the advantages of AOPs and promising technological advances in the field, large scale applications remain limited. Moreover, even at a fundamental level, the understanding of oxidation reaction pathways, specific roles of different radical species, and surface catalytic reactivities is still nascent. Therefore, further research is required to determine the role of photo-AOPs in the elimination of CECs from water. Given these considerations, it is necessary to identify both the current status of AOP research as well as research gaps and future applications. Herein, we identify and discuss the feasibility of wider photo-AOP applications as state-of-the-art solutions to provide safe, clean, and affordable water in the context of a zeropollution strategy.

## 2. Current utility of photo-AOPs

Various AOPs used in wastewater treatment have been integrated with artificial or natural light sources and have been of research interest for decades (Fig. 2). AOPs previously utilized include the use of photo-hydrogen peroxide ( $H_2O_2$ ), -Fenton, -chlorine, -persulfate ( $S_2O_8^{2-}$ ), -ozone ( $O_3$ ), and -catalysis systems.

The **photo-H<sub>2</sub>O<sub>2</sub>** AOP, typically referred to as ultraviolet (UV)-H<sub>2</sub>O<sub>2</sub>, is a conventional AOP that relies on H<sub>2</sub>O<sub>2</sub> photolysis

to generate strong and non-selective oxidants, namely hydroxyl radicals (·OH). UV-H<sub>2</sub>O<sub>2</sub> has been widely studied since the late 1970s, given its ability to degrade trace organic chemicals [5]. However, due to its low absorption coefficient and scavenging effect for ·OH, additional H<sub>2</sub>O<sub>2</sub> is inevitably required during the process, thus decreasing the process efficiency [6]. The ·OH quenching and light-shielding effects of the water matrix also necessitate excess dosing. As such, even with excess H<sub>2</sub>O<sub>2</sub>, the UV-H<sub>2</sub>O<sub>2</sub> process generates an ·OH concentration with a range of only  $10^{-13}$ – $10^{-12}$  mol·L<sup>-1</sup> in pure water [7]. Typically, only 5%–10% of the H<sub>2</sub>O<sub>2</sub> needs to be removed prior to effluent discharge. With the goal of zero pollution discharges, there is a risk that even greater quantities of excess H<sub>2</sub>O<sub>2</sub> will be needed to completely mineralize CECs.

The **photo-Fenton** AOP expands on the typical Fenton process and addresses some of its inherent challenges, such as its highly acidic environment, large reagent consumption, and ferric sludge production [9]. The addition of photons to Fenton AOPs accelerates the production of •OH through the photolysis of Fe(III) hydroxocomplexes, which simultaneously regenerates Fe(II), thereby



Fig. 1. Publication trends in photo-based AOP research (data collected from Web of Science<sup>®</sup>).







reducing reagent consumption. To address ferric sludge production, heterogeneous photo-Fenton AOPs using zero-valent iron, iron oxides, and composite materials have been developed [10]. However, metal leaching beyond the optimal pH range (typically < 4) leads to decreased catalytic performance and an increased  $H_2O_2$  dose requirement. Hence, photo-Fenton AOPs are not currently cost-effective in treating large-volume effluents in the context of zero pollution [11].

The **photo-chlorine** AOP has been explored as an emerging AOP since the excessive consumption of chlorine in swimming pools under sunlight was observed in the 1980s [12]. Photo-chlorine, which typically takes the form of UV-chlorine, produces OH and Cl- as the primary radicals through the photolysis of HOCl and OCl<sup>-</sup> [13]. The concentration of OH is one or two orders of magnitude higher than that of Cl- because of the reactions between Cl- and H<sub>2</sub>O or OH<sup>-</sup>. Secondary radicals, including Cl<sup>2--</sup> and ClO-, can also be produced during this process. Compared to OH, reactive chlorine species (RCSs) are more selective and thus less sensitive to the scavenging effects of the water matrix [13]. It is also noteworthy that the irradiated light wavelength can alter the radical species [14]. Photons with wavelengths > 300 nm can produce organic compounds via the decomposition of OCl<sup>-</sup> [15].

HOCl and OCl<sup>-</sup> can only be activated by UV light because their absorption lies in the UV range. However, the activation wavelength can extend into the visible range by incorporating heterogeneous catalysts [16]. As an emerging AOP, UV-chlorine is typically comparable to UV-H<sub>2</sub>O<sub>2</sub> in terms of target pollutant removal, energy consumption, and by-product formation. However, contrasting research findings are common, likely as a result of different target pollutant properties and water matrices [17,18].

The **photo-persulfate** AOP is a hybrid persulfate-based AOP; here, activated persulfate  $(S_2O_8^{2-})$  produces sulfate radicals  $(SO_4^{--})$ , which are strong single-electron oxidants. Certain commonalities can be seen with Fenton reactions, as transition metals in combination with photosensitized dyes, ligands, and catalysts are used to activate persulfate [19]. Thus, persulfate-based AOPs are occasionally referred to as Fenton-like AOPs [20].  $SO_4^{--}$  has a higher redox potential, longer half-life, and wider pH range compared to the major reactive oxygen species (ROS) in Fenton processes (i.e.,  $\cdot$ OH). In addition,  $SO_4^{--}$  is more selective, minimizing the loss of oxidation power due to the scavenging effects of the water matrix [21].

Photo-persulfate AOPs perform better than their non-irradiated counterparts and have seen two decades of research [22]. Moreover, the research interest in photo-persulfate AOPs has recently intensified. Various oxidants and oxidation mechanisms give persulfate-based AOPs a unique advantage, as they can degrade persistent CECs that are inert to OH (e.g., per- and polyfluoroalkyl substances (PFAS)) [23–26]. However, homogeneous systems produce metallic sludge (e.g., Fe), while heterogeneous systems could be inactivated by the sulfate layer that gradually forms on the catalyst surface [27].

The **photo-O<sub>3</sub>** AOP is an expanded form of O<sub>3</sub>-based AOPs. Given that dissolved O<sub>3</sub> has an absorption peak at 260 nm and can be photolyzed into  $H_2O_2$ , the UV-O<sub>3</sub> AOP has been developed to decompose O<sub>3</sub> into ·OH, allowing further degradation of target pollutants. Similar to other AOPs, homo- or heterogeneous catalysts based on transition metals can be added to the system, to accelerate the overall oxidation reaction.

The  $O_3$  injection rate and pH significantly affect the performance of the photo- $O_3$  AOP by determining the  $O_3$  solubility and subsequent ·OH generation [28]. Although photo- $O_3$  AOPs were originally developed for wastewater treatment in the 1970s [29], their full-scale application remains limited, owing to high capital and operational costs. A few recent studies have attempted to improve  $O_3$  utilization efficiency by decreasing the injected  $O_3$ gas bubble diameters [30]; this is an expensive operation, particularly in the context of targeting complete CEC mineralization.

**Photocatalytic AOPs** use illuminated heterogeneous catalysts and produce charge carriers, which in turn react with the water to generate ROS (e.g., ·OH). In some cases, the charge carriers (i.e., both electrons and holes) participate directly in redox reactions with pollutants. Photocatalysis has been of environmental interest since 1985, when the photocatalytic degradation of chloroform was first investigated [31]. Given that engineered photocatalysts may degrade CECs, a strong focus has been placed on designing novel catalysts to allow higher pollutant degradation efficiency through the improvement of charge carrier separation and manipulation of energy band gaps [32–34].

Despite reports of promising performance, photocatalytic AOPs have been criticized for their low adoption rates and applicability



Fig. 2. History of photo-AOP research. ROS: reactive oxygen species; RCS: reactive chlorine species; E: the redox potential.

at scale. Furthermore, the efficient recovery of photocatalytic materials and their light-shielding effects present challenges to the overall cost and scalability of the technology [35]. Fortunately, recent pilot-scale studies have endorsed the unique advantages of this technology and support its potential transition into some niche applications; nonetheless, its utility in municipal wastewater treatment applications to zero pollution discharges remains impractical [34,36].

## 3. Strategies towards zero pollution

The fundamental objective of zero-pollution photo-AOPs is to sustainably and continuously produce clean water that is free of secondary contamination at a low cost. Theoretical and technological innovation development to meet this objective is an extremely challenging task; indeed, not only are CECs found in trace amounts, but they are also persistent and stable in the aquatic environment. Nevertheless, the challenge of advancing next-generation photo-AOPs that will enable the complete degradation of water pollutants should be undertaken.

In this regard, photo-AOPs have shown great potential and success in degrading endocrine disrupting chemicals (EDCs) [37], PFAS [35], and even microplastics [38], which are considered among the most challenging pollutants to deal with when targeting zero pollution. Photo-AOPs produce various oxidation species with different redox potentials that decompose CECs at high reaction rates, for example, second-order reaction rates with a range of 10<sup>7</sup>- $10^{10}$  mol·L<sup>-1</sup>·s<sup>-1</sup> [7]. In addition, advances in material science have generated new potential heterogeneous photo-AOPs which are more effective than conventional AOPs. Despite these recent advances, several knowledge gaps remain and the photo-AOP technology still require further development to achieve the ultimate zero-pollution goal. A recent study serves as a reminder that many reaction mechanisms, including by-product formation, reaction pathways, and interference from water matrix components, remain unexplored. For instance,  $SO_4^{-}$  was shown to react with water matrix components to produce harmful aromatic organosulfates [39].

Additional water matrix interferences, including oxidative radical scavenging and the absorbance of photons, affect the performance of photo-AOPs and reduce light utilization efficiency. However, some photolysis products of the water matrix components act as oxidizing agents, offsetting the scavenger effect [40]. Photo-AOPs also alter some water matrix components (e.g., dissolved organic matter) to produce disinfection by-products [41]. The various impacts of the water matrix on the performance, mechanisms, and by-product formation (including potentially toxic intermediate compounds) of photo-AOPs necessitate a comprehensive assessment, given the objective of achieving zero pollution.

The fundamental limitation of photo-AOPs is the short light penetration path within the water matrix. As a result of this limitation, photo-AOPs tend to typically serve as enhancements for existing technologies, for example, in achieving high-quality effluent for water reclamation. However, tertiary water treatment usually requires the integration of various technologies. Indeed, processes have been proposed that integrate two or more photo-AOPs to reduce the overall energy consumption and the risk of by-product formation [42]. However, the mechanisms involved with these integrated systems remain largely unknown. Although significant advances have been made in determining pathways, especially those involving  $\cdot$ OH and O<sub>3</sub> [43], unanticipated reactions may occur during AOP operation. For example, Nnitrosodimethylamine and aldehydes were formed when O<sub>3</sub> was injected at the Edward C. Little Water Treatment Facility (El

Segundo, CA, USA) to prevent microfiltration fouling; this illustrates the need for preliminary research to ensure rational designs of integrated systems [28]. Regarding heterogeneous systems, complexity is added by the light-shielding effect and catalyst mass transfer limitations, ultimately warranting comprehensive modeling and reactor design efforts. Moreover, catalyst production cost, durability, and ease of recovery are key issues [44] that need to be resolved to meet the zero-pollution objective.

Selecting a suitable light source is a major cost consideration for all photo-AOPs. The widespread notion that photo-AOPs are costeffective has been criticized, primarily due to the high energy input required by artificial light sources. Nonetheless, technological advances in artificial light sources, as seen in light-emitting diodes (LED), may provide new opportunities [45]. UV-LEDs offer a different light distribution compared to conventional UV lamps, possibly resulting in more compact and efficient reactors. Other advantages of UV-LEDs when compared to conventional lamps include lower energy consumption, higher mechanical strength, lower cost, longer life span, less mercury use, and accurate tuning of the photon wavelength.

On the other hand, the intensity of solar light is weaker and highly dependent on weather conditions, time, and location. Therefore, a larger reaction space is required to achieve a treatment performance equivalent to that of an artificial light source system. Indeed, the larger footprint equates to a higher capital requirement, and thus, the overall treatment costs of solar light-based AOPs can be much higher than that of their UV-based counterparts [44].

The treatment costs of photo-AOPs follow their respective energy consumptions; more specifically, UV-chlorine has the lowest cost and consumption, followed by UV-persulfate, UV-O<sub>3</sub>, UV-H<sub>2</sub>O<sub>2</sub>, UV-Fenton, and finally, with the highest cost and energy consumption, UV-photocatalysis [17]. However, the treatment costs vary significantly according to the treatment goal and influent quality. With a zero-pollution target, more energy is required for the complete mineralization of organic pollutants compared to the targeting of specific discharge limits. The impact of influent quality on AOPs is illustrated in a case where increased energy consumption (i.e.,  $6.4-41.1 \text{ kW}\cdot\text{h}\cdot\text{m}^{-3}$ ) in the UV/TiO<sub>2</sub> process resulted from an increase in influent alkalinity (i.e., CaCO<sub>3</sub> content increase from 0 to 120 mg·L<sup>-1</sup>) [46]. Nonetheless, when considering the entire life cycle of zero pollution water treatment, an urgent need to reduce the energy consumption of photo-AOPs is evident.

Overall, achieving zero pollution requires several theoretical and technical challenges to be addressed directly, for example, reducing (or eliminating) water matrix interference, toxic byproducts, high energy consumption, and chemical use. Although photo-AOPs can theoretically undertake complete mineralization of any toxic chemical, innovations related to scaling up, costs, operations, and system integration are strongly recommended.

## 4. Conclusions

Photo-AOPs represent a promising technology to be used in achieving zero-pollution goals, owing to their ability to break down trace CECs. However, intrinsic properties of AOPs, particularly the photon penetration limitations, make photo-AOPs more suitable as a refinement technology, to be used in the final processing step. Although some photo-AOPs have been implemented at scale, most notably those in water reclamation facilities, promoting their wider application to achieve the zero-pollution objective will require several key issues to be carefully considered, for example, novel photocatalyst designs, efficient photoreactor development, and advances in artificial light sources. Furthermore, integrating different photo-AOPs for CEC removal could be a feasible strategy for achieving the zero-pollution objective. Hybridization with photoelectrochemical technologies may render other AOPs more costeffective [47,48]; however, to ensure rational designs and to avoid possible failure of integrated systems, a complete preliminary assessment should be conducted on performance and by-product formation. In addition, comparing the performance, treatment costs, and reaction pathways of different AOPs should be conducted at bench- and pilot-scales. Engineering design innovations are also required to address the challenges in scaling up this technology. Ultimately, in accordance with the green transition being undertaken in the water sector, energy-efficient and chemicalfree photo-AOPs should be pursued.

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