Photocatalytic degradation of recalcitrant aromatic hydrocarbon compounds in oilfield-produced water: A critical review

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Review

Photocatalytic degradation of recalcitrant aromatic hydrocarbon compounds in oilfield-produced water: A critical review

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ABSTRACT

Oilfield-produced water (OPW) contains a complex mixture of toxic and hazardous organic and inorganic compounds. Refractory polycyclic aromatic hydrocarbons (PAHs) and polyaromatic hydrocarbons such as benzene, toluene, ethylbenzene, and xylene (BTEX) chemical compounds present in OPW which result from hydrocarbon sources and chemicals added during the oil recovery process, have become a source of global concern. Their continuous presence in the OPW requires suitable technology to remove them before their discharge to the environment. When the OPW contains harmful pollutants combined with oil in minute size, the treatment performance of various water technologies appears insufficient due to the generation of secondary by-products, which are also recalcitrant and toxic. To consolidate scattered knowledge of OPW in the field of water pollution control, this article critically reviews and evaluates the technical feasibility of heterogeneous photocatalysis for OPW treatment, their bottlenecks in applications, and the way forward for the removal of recalcitrant hydrocarbon compounds from OPW. This work also presents the nature, composition, and health implications of these compounds in the OPW. Pertinent factors for effective photodegradation were presented. It also critically evaluates and discusses a variety of pilot and commercial-scale applications of photocatalysis in water treatment and its limitations. It was conclusively evident from 239 published articles (1988–2022) that heterogeneous photocatalytic degradation is a powerful approach for the removal of the hydrocarbon compounds such as PAHs, BTEX, and phenol with an average of over 80% of degradation efficiency. The photocatalytic process can be scaled up to the commercial scale with the proper design of photocatalytic reactors and the synthesis of advanced photocatalysts.

1. Introduction

The United Nations (UN) Sustainable Development Goals (SDG#6) pledges to "ensure availability and sustainable management of water and sanitation for all". Due to rapid industrialization, a major effort to safeguard water bodies from pollution and maintain water quality is vital to protect the environment and public health (Mengting et al., 2022). Achieving this goal not only requires the development of sustainable and efficient technologies to make water remain clean but also entails preventive measures to ensure the sustainable use of water resources (Matta et al., 2021).

In spite of such preventive measures, contaminated water is still produced due to the extraction of oil and gas. The water, which is naturally present in the reservoir, condensed water, and floodwater previously injected during gas and oil production, makes up the complex oilfield-produced water (OPW). It is a by-product of oil and gas

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exploration and production on land or at sea (Beyer et al., 2020). Because many oil deposits are found in or near groundwater aquifers, large amounts of OPW are frequently generated during exploration (Salem and Thiemann, 2022). Their quantity can be as high as ten times the quantity of oil produced throughout an oilfield's lifetime (Al-Kindi et al., 2022).

Benzene, toluene, ethylbenzene, and xylene (BTEX) are monoaromatic hydrocarbons and common pollutants found in groundwater plumes and other water resources as a result of the disposal of contaminated industrial effluents and accidental events such as oil spills and pipeline leakages (Fayemiwo et al., 2017). In addition to their abundance in fossil fuels, they are widely employed as chemical compounds in a variety of industrial processes (Zhu et al., 2020). The high solubility of BTEX in water poses a major risk of contamination of groundwater (Zanello et al., 2021).

Because of their volatility, the chemicals exist in trace amounts in surface waters (Asejeje et al., 2021). Nevertheless, they can be found in higher amounts in groundwater, where they are regarded as priority contaminants and consequently included as substances to be tested in water analysis (Román Falcó and Moya, 2007).

Groundwater pollution due to hydrocarbons such as BTEX has become increasingly common. This necessitates the development of effective technologies to remove or mitigate the harm caused by target substances (Kurniawan et al., 2022a, 2023). In addition to their high operating costs, physical treatments can remove toxins from the environment without eliminating or changing them, resulting in harmful residue accumulation (Saravanan et al., 2021). Human exposure to BTEX compounds over time causes skin and sensory irritation, as well as respiratory problems and irritation of the central nervous system which are some of its side effects (Liang et al., 2023; Piccardo et al., 2022) Even though BTEX compounds are harmful to public health, they are neglected and untreated in drinking water. As a result, the danger of water-borne infections rises as this water is consumed by the public (Kurniawan et al., 2011).

Polycyclic aromatic hydrocarbons (PAHs) are a type of organic chemicals that are resistant to natural breakdown. Because of their genotoxic, mutagenic, and carcinogenic nature, these chemicals constitute a great risk to natural ecosystems and human health (Dutta et al., 2022). PAHs have a strong interaction with organic matter in OPW (Bolden et al., 2017; Kuppusamy et al., 2017). PAHs of all molecular weights are stable and are hardly biodegradable. Because of their mutagenic and carcinogenic effects, the United States Environmental Protection Agency (USEPA) has classified 16 PAHs as priority micropollutants (Jiménez et al., 2018). OPW is a major source of anthropogenic PAHs. PAHs are a broad and diversified group of major environmental pollutants that can arise as a result of the incomplete combustion of organic materials (Reizer et al., 2022).

PAHs may be colorless, pale yellow or white solids that are environmentally persistent having numerous structures and varying levels of toxicity, and are found in OPW. (Abdel-Shafy and Mansour, 2016). According to a recent study, chronic exposure to PAHs has been linked to cancer in aquatic animals and increased mutagenicity in sediments (Qi et al., 2017). Because PAHs are natural components of fossil fuels and are found in relatively high concentrations in petroleum products, the petroleum industry and transportation process are the main contributors to PAHs in the environment (Ding et al., 2008; Imtiaz et al., 2022). PAHs can enter the environment through various mechanisms, including bioaccumulation, volatilization, chemical oxidation, soil particle adsorption, and photooxidation (Ossai et al., 2020). As a result, BTEX and PAHs must be removed from OPW.

Some known technologies for the removal of these hydrocarbon chemical compounds and other constituents from OPW include membrane separation (Liu et al., 2021a, 2021b), cyclonic and gravity separators (Liu et al., 2020; Nunes et al., 2021), coagulation-flocculation (Li et al., 2023) biological treatment (Abujayyab et al., 2022), adsorption (Fathy et al., 2018), evaporation (Heins and Peterson, 2018), and floatation (Piccioli et al., 2020; Wang et al., 2022) are less efficient as a standalone system specifically for the removal of these hydrocarbon chemical compounds from OPW. Not only are they less efficient at separating stable oil-water emulsions, but they are also prone to fouling in the case of membrane separation (Wei et al., 2019). Due to their safety, space, cost, and efficiency, they are not specifically suited for offshore oil and gas production facilities (Liu et al., 2021c; Salem and Thiemann, 2022). In addition, existing technologies are limited in their ability to treat emulsified oil droplets with size of less than 10 μm (Zhu et al., 2014; Dickhout et al., 2017).

As a result, some finely dispersed oil and dissolved petroleum hydrocarbons remain in the OPW as saturated hydrocarbons and low molecular weight aromatic, which could be harmful to the ecosystem when released into the ocean. Petroleum hydrocarbons' solubility in seawater reduces, while their size (molecular weight) increases.

Current treatment technologies are only relevant when they are used at the right point in the treatment process. Otherwise, they have limitations in their practical applications. Various treatment technologies have been tested to remove heavy metals, oil, grease, and suspended particles to a certain level, while the elimination of aromatic hydrocarbon compounds at micro or minute concentration has not been fully addressed at an industrial scale and remains a bottleneck. Primary treatment technologies only change pollutants from one phase to another, but they are unable to eradicate compounds such as BTEX, PAHs, phenol, and other recalcitrant hydrocarbon compounds (Amakiri et al., 2022a).

As an advanced oxidation process (AOP), heterogenous photocatalysis using photocatalytic oxidation can degrade minute-sized oil contaminants and other pollutants, which remain in the OPW after the initial treatment or even as the starting treatment method (Liang et al., 2011; Samuel et al., 2022a). The photocatalytic process has demonstrated considerable potential as an environmentally friendly, sustainable, and low-cost treatment technique, that is acceptable to the water industry's "zero-waste" policy. It has been demonstrated that this oxidation treatment method is powerful in removing microorganisms and persistent organic pollutants in wastewater and does not generate additional waste since the produced radicals degrade the organic pollutants indiscriminately (Kurniawan et al., 2022b; Samuel et al., 2022a).

A preliminary literature survey by the authors reveals that several review articles on the treatment of OPW have been published earlier. A review published by Olajire (2020) focused on the current technologies for the management of oil and gas-produced water. Their work emphasized the mechanisms of the chemical reaction involved in OPW treatment and the various effects of the produced by-products from these treatment processes on the environment. Al-Ghouti et al. (2019) reviewed methods for treatment and management of OPW in addition to the various areas of reuse of the treated OPW, while Cabrera et al. (2021) focused their review on recent research advances and progress as well as discussion on the performance and evaluation of the integrated electrochemical and biological process in OPW treatment. In another review, Samuel et al. (2022b) discussed the various conventional and membrane-based technologies for OPW treatment, highlighting the potential for reuse of the treated OPW, while Coha et al. (2021) reviewed the potential of various advanced oxidation processes (AOP) to remove organic substance from OPW. In addition, Igunnu and Chen (2014) evaluated and discussed extensively the various technologies for the treatment and management of OPW, while exploring the potential of electrochemical techniques for the treatment of OPW in the future.

In spite of the novelty of the various reviews published earlier, their work mainly concentrated on the general treatment of OPW using various techniques without focusing on the treatment of recalcitrant hydrocarbon organic compounds, a source of global concern to the environment. To the best of the authors' knowledge, only very few review articles have specifically focused on photocatalytic degradation for the removal of recalcitrant hydrocarbon chemical compounds present in OPW.

To close the knowledge gap in the field of study, this article critically evaluates the composition of these hydrocarbon compounds in OPW, their characteristics, and the photocatalytic degradation process as an emerging AOP approach to remove these compounds via the photocatalytic process. Operating parameters influencing effective photocatalytic degradation of the hydrocarbon compounds in OPW, and bottlenecks in pilot and commercial scale applications of photocatalytic systems in wastewater treatment were discussed. Perspectives of this technology for the removal of recalcitrant aromatic hydrocarbon compounds in OPW are also presented.

Finally, this review identified knowledge gaps and proposed niche areas of future research to improve the removal performance of the recalcitrant hydrocarbon compounds from OPW for environmental conservation and reuse purposes. It is expected that this work benefits stakeholders' understanding of how to add value to the wastewater in the framework of water resource reclamation and recovery based on circular economy (CE) paradigm.

2. State of global focus on OPW generation and treatment

Due to rising oil demand and increased global production activity, OPW production is anticipated to grow (Osaki, 2019). The three regions with an exponential increase in OPW production volumes are Europe/Central Asia, the Middle East/Africa, and North America. OPW is an inevitable by-product of crude oil extraction that tends to rise annually as oil fields mature and new ones emerge. It poses both opportunities and challenges for hydrocarbon resource recovery. In aging oilfield production wells, the ratio of OPW to oil equivalents reaches 98%–2%. With continuous fossil fuels production, the global production of OPW is anticipated to be around 250 million barrels per day (Al-Ghouti et al., 2019; McCabe, 2020).

The Produced Water Society has projected that during the next decade, the amount of oil produced from conventional onshore and offshore fields will rise by 200% from about 300 million barrels per day to 600 million barrels per day with a daily demand reaching 104.1 million barrels per day (International Energy Agency (IEA), 2021). Oil operators are managing OPW by either reinjection for oil recovery (Petrowiki, 2018), or for reuse purposes (irrigation, livestock watering, habitat, and wildlife watering, etc) (Al-Ghouti et al., 2019; Neff et al., 2011) after a certain level of treatment or discharge (Veil et al., 2004). Discharging OPW into the environment is recommended if the first two methods of OPW management mentioned above are not feasible (Nasiri et al., 2017). Regulations in oil-producing nations have established strict limitations for OPW discharge due to environmental concerns about its toxicity (Samuel et al., 2022a).

A report by the Texas produced water consortium revealed that excess OPW from unconventional wells within the Persian Basin could offer a significant source of water if pilot treatment projects can demonstrate that the OPW can be processed economically and at a quality that protects public health and the environment. The report also implies that depending on treatment capacity and recovery rates, the potential for treating OPW could result in approximately 2 billion barrels per year (256,000 ac-ft) and as much as 4 billion barrels per year (511,000 ac-ft) of treated OPW that could be used outside of oil and gas operations. The consortium reported that the environmental impact could be substantial when being weighed against anticipated water shortages in this area in the future (Texas Produced Water Consortium (TxPWC), 2022). Over time, a reservoir's OPW quality and volume change. Fig. 1 shows the source and categories of various effluents generating OPW.

Table 1 lists the annual amounts of OPW (barrel) generated by different countries. The tremendous amount of OPW generated can be linked to the countries' expansion and growth (Alomar et al., 2022). As a result, managing OPW has become necessary.

To present an overview of the treatment of OPW, the authors examined relevant articles published in this area using relevant keywords like "advanced oxidation process", produced water treatment", 'photodegradation', "organic hydrocarbon compounds" and "heterogenous photocatalysis". About 239 articles related to photocatalytic OPW treatment were retrieved from the Web of Science (WoS) from 1988 to 2022. Relevant articles were selected based on the application of heterogenous photocatalysis for organic and chemical compound removal from OPW and according to their title and abstract to address the specific topic of this review.

Fig. 2 presents the number of published articles on OPW treatment.

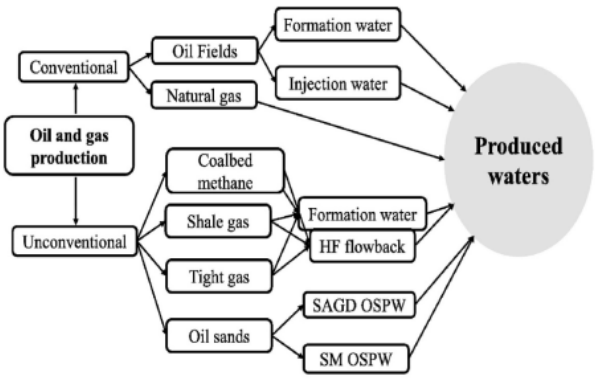

Fig. 1. Sources and categories of effluent generating OPW, Reprinted/adapted with permission from (Ganiyu et al., 2022), Copyright 2022, Elsevier.

Table 1
Amount of oilfield-produced water (OPW) generation by countries.

Country	OPW generated (bbl./year)	Year reported	Reference
USA	24,400,000,000	2015	Veil (2015)
Oman	10,331,010,000	2002	Hirayama et al. (2002)
Kuwait	730,000,000	2018	AlAnezi et al. (2018)
Australia	207,570,000	2010	Australian Government: Department of the Environment, 2014
Iraq (Rumaila Field)	198,925,000	2013	Kuraimid et al. (2013)
Qatar	50,508,816	2014	Alomar et al. (2022)

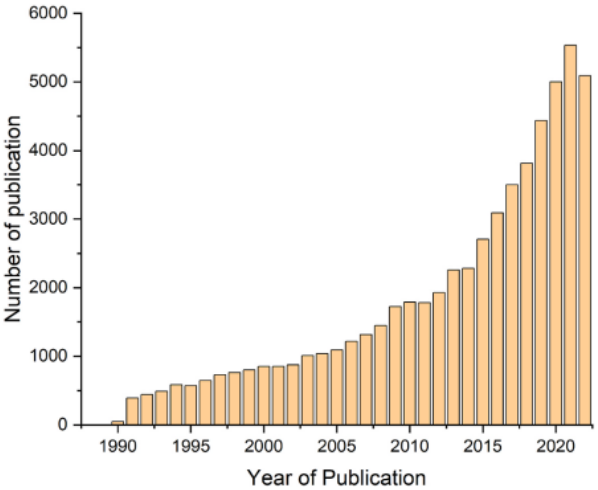

Fig. 2. OPW treatment-related publications between 1988 and 2022 (Source: Web of Science, Accessed April 2023).

They have increased drastically over the past decades. By 2022, over 60,071 OPW-related articles published have cumulatively been recorded in the Web of Science (WoS) database. Out of them, 1976 OPW-related articles focused on the photocatalytic treatment of OPW. This implies the scientific community's focus on the treatment of OPW and the growing need to address the increasing generation of OPW using emerging technologies such as heterogenous photocatalysis as a cost-effective and eco-friendly technology for OPW treatment.

3. Oilfield-produced water (OPW) composition and characteristics

The oilfield's geographic location, technique of extraction, hydrocarbon products being extracted, as well as minerals available within the geologic formation have impacts on the chemical and physical characteristics of OPW. The water typically has the chemical features of the hydrocarbons present in the OPW since they have had physico-chemical interactions for many years with hydrocarbon-bearing formations (Scanlon et al., 2020). The average OPW is saline and contains significant levels of TDS as well as magnesium, sulfate, sodium, calcium, and chloride. Grease and oils (emulsified, dispersed, or free), Semi-volatile and volatile organics like BTEX, PAHs, waxes, and organic acids are the principal contaminants in OPW (Lin et al., 2020; Samuel et al., 2022a).

Chemical additives used to enhance drilling and production, dissolved gases (such as sulfide, hydrogen, and ammonia), and naturally occurring radioactive materials that leach or precipitate from formations, resulting from water mixing, are contaminants present in smaller quantities. Additionally, OPW contains transformational byproducts and heavy metals that result from the interaction of the water and the additional chemicals (Chaudhary et al., 2019; Rodriguez et al., 2020).

In 2006, when collecting information from the literature to study hydraulic fracturing during exploration, the US Environmental Protection Agency discovered 600 distinct compounds in OPW samples (EPA, 2019). Table 2 and Table 3 summarizes the general characteristic and composition of OPW and the concentrations of specific aromatic hydrocarbon chemical components in OPW respectively. Table 3 shows that BTEX, PAHs, and phenols concentrations in OPW, range from 40 to 2148 g/L, with phenanthrene, naphthalene, pyrene, and anthracene having the highest concentrations. It is important to note that the range of values for the hydrocarbon organic compounds varies from one oilfield to another.

Table 2
Characteristics and composition of OPW.

Component	Range of values	Average values	References
pH	4.3–7.5	6.30	Janson et al. (2015); Veil et al. (2005)
Oil and grease (mg/L)	40.5–654	299.92	Benko and Drewes (2008); Tibbetts et al. (1992a)
Nitrite (mg/L)	0.05	0.05	Abou El Leil et al. (2021); Veil et al. (2005)
Potassium (mg/L)	44–2162	1194.50	Janson et al. (2015); Tibbetts et al. (1992a)
Sulfide	828.00	828.00	Janson et al. (2015)
Bicarbonate (mg/L)	144.4176–15,000	6563.18	Abou El Leil et al. (2021); Benko and Drewes (2008)
Nitrate (mg/L)	2.15–9.492	5.82	Abou El Leil et al. (2021); Veil et al. (2005)
Ammonium (mg/L)	11–14.54	12.77	Abou El Leil et al. (2021); Janson et al. (2015)
Magnesium (mg/L)	4.7943–12,341	3164.21	Tibbetts et al. (1992b); Udeagbara et al. (2020)
Sodium (mg/L)	1030–150,000	60,132.00	Benko and Drewes (2008); Çakmakce et al. (2008)
Thiosulfate (mg/L)	14.00	14.00	Janson et al. (2015)
Chloride (mg/L)	2265–250,000	92,666.97	Abou El Leil et al. (2021); Çakmakce et al. (2008)
Total phosphorous (mg/L)	0.71	0.71	Veil et al. (2005)
Ammonia (mg/L)	9.66–74	41.83	Abou El Leil et al. (2021); Veil et al. (2005)
Bromide (mg/L)	51.00	51.00	Janson et al. (2015)
Acetate (mg/L)	347.00	347.00	Janson et al. (2015)
TOC (mg/L)	491–1700	876.25	Benko and Drewes (2008); Veil et al. (2005)
BOD (mg/L)	750–957	853.50	Tibbetts et al. (1992b); Veil et al. (2005)
COD (mg/L)	1220–1910	1459.13	Çakmakce et al. (2008); Janson et al. (2015)
Salinity (mg/L)	7165–100,000	78,399.33	Abou El Leil et al. (2021); Çakmakce et al. (2008); Veil et al. (2005)
Conductivity (μS/cm)	7200–87,542	48,979.33	Abou El Leil et al. (2021); Janson et al. (2015)
Corrosion inhibitor	0.3–10	–	Igunnu and Chen (2014)
Glycol	7.7–2000	–	Igunnu and Chen (2014)
Scale inhibitor	0.2–30	–	Igunnu and Chen (2014)
Phenol	0.001–10000	–	Ekins et al. (2007); Tibbetts et al. (1992a)
Saturated hydrocarbons	17–30	–	Neff et al. (2011)
TSS (mg/L)	500.6–7820	2455.30	Abou El Leil et al. (2021); Benko and Drewes (2008)
Alkalinity	285	–	(EPA, 2019; USEPA, 2020)
Chloride (mg/L)	2265–250,000	92,666.97	Çakmakce et al. (2008); El-badawy et al., 2022
Sulfate (mg/L)	54–15,000	3969.37	Benko and Drewes (2008); Tibbetts et al. (1992b)

3.1. Structure and chemical-physical properties of BTEX

One-ring BTEX is the most prevalent hydrocarbon in OPW. BTEX can be found in OPW water from a variety of sources with concentrations ranging from 68 to 600,000 μg/L. OPW from gas wells has higher quantities of BTEX than OPW from oil wells in the North Sea (Neff and Sauer, 1996; Stephenson, 1992; Lin et al., 2010). The most abundant BTEX chemical in OPW is toluene, followed by benzene. Only a small proportion of the amounts of benzene and toluene are present in ethylbenzene and the three xylene isomers. Supplementary Figure S1 and Table S1 present the structure and the physico-chemical properties of BTEX compounds respectively.

3.2. Structure and physical-chemical properties of PAHs

Because of their toxicity and persistence in the marine environment, PAHs, widely known as polynuclear aromatic hydrocarbons, are petroleum hydrocarbons of greatest environmental concern in OPW. They are defined as hydrocarbons with two or more fused aromatic rings, which are difficult to remove from OPW (Samuel et al., 2022b).

Supplementary Figure S2 and Table S2 present the structure and the physico-chemical properties of PAHs compounds, respectively. Total PAH concentrations in OPW typically range between 80 and 3000 μg/L, with the majority of OPW possessing less than 1000 μg/L. The only PAHs that are occasionally found in more than trace amounts are naphthalene and phenanthrene, as well as their alkyl homologs (Zhao et al., 2021). The lower molecular weight PAHs are generally found in larger concentrations in gas well OPW than in oil well OPW (Stephenson, 1992).

Generally, PAHs such as dibenzothiophene (NPD), phenanthrene, naphthalene, as well as BTEX, and phenols are among the hydrocarbon mixes found in dispersed and dissolved oil ingredients. Not all hydrocarbons can be dissolved by water. Most of the oils are spread as minute droplets (Ekins et al., 2007). The soluble organic compounds are dissolved oils, which are polar components, whereas the dispersed oils are the smaller or micro oil droplets in the aqueous phase (Duraisamy et al., 2013). Also organic contaminants with two or more bonded aromatic rings PAHs are difficult to remove from OPW (Shen et al., 2012; Chong

Table 3
Composition of BTEX, PAHs, and Phenol in various oilfields around the world.

Oilfield	BTEX compound (μg/L)					
	Benzene	Toluene	Ethylbenzene	Xylenes	Phenol	Reference
Norwegian continental shelf Norway	4.0	2.77	2.3	N/A	5.1	Fillo et al. (1992)
Sergipe Brazil	1291–1511	1167–1357	136–158	89–103	194–242	Dórea et al. (2007)
Gulf of Mexico	0.44–2.80	0.34–1.70	0.026–0.11	0.16–0.72	0.723	Neff et al. (2011)
Samarang Indonesia	0.33–3.64	0.089–0.80	0.026–0.056	0.013–0.48	0.031–0.2	Neff et al. (2011)

PAHs compound (μg/L)	**Oilfields**				
	Scotian Shelf Canada. (Pampanin and Sydnes, 2013)	**North Sea.** Chowdhury et al. (2009)	**North America Gulf of Mexico.** (Pampanin and Sydnes, 2013)	**Great Britain North Sea.** (Pampanin and Sydnes, 2013)	**Sergipe Brazil.** Dórea et al. (2007)
Naphthalene	1512	194–841	5.3–90.2	237–394	9.9–10.7
Acenaphthene	N/A	0.1–6.1	N/A	N/A	1.6–2.4
Anthracene	0.26	0.1–2.6	0.45	N/A	0.8–1.7
Pyrene	0.36	0.2–7.7	0.01–0.29	0.03–1.9	0.9–1.0
Chrysene	N/A	0.6–.084	N/A	N/A	5.9–9.9
Perylene	N/A	N/A	N/A	N/A	N/A
Fluoranthene	N/A	0.1–3.6	N/A	N/A	2.7–6.2
Benzo[g,h,l] perylene	N/A	0.0–2.7	N/A	N/A	0.9–3.1
Benzo [e] pyrene	N/A	0.0–1.1	N/A	N/A	2.4–2.6
Benzo [0] anthracene	N/A	0.0–0.0–1.2	N/A	N/A	1.6–1.9
Phenanthrene	4.0	9–111	0.11–8.8	1.3–32.0	2.2–2.4
Acenaphthene	N/A	0.3–15.3	N/A	N/A	N/A
Fluorene	13	4.1–66.7	0.06–2.8	2.6–21.7	N/A
Indenol(1,2,3-c, d) pyrene	N/A	0.0–0.4	N/A	N/A	N/A

N/A-Not Available.

et al., 2010).

Even at low concentrations, they are dangerous to the environment and have been categorized as high-risk compounds for environmental protection and public health (DPR, 2002; USEPA, 2020; WHO, 2017). Because PAHs are persistent and dangerous contaminants in the marine ecosystem, they must be carefully treated before being released into the environment (Abdel-Shafy and Mansour, 2016; Munyengabe et al., 2022). The dissolved and dispersed oil concentrations in OPW are determined by parameters such as field location, precipitated oil, oil density, and water-oil interfacial tension (Liu et al., 2021a).

4. Treatment of recalcitrant aromatic hydrocarbon compounds in OPW

Reuse, discharge, and final disposal of OPW require proper treatment and management. In most cases, the combination of multiple treatment technologies is required because of the complexity of the OPW composition and the strict discharge regulations. However, most OPW and wastewater treatment techniques are inadequate for removing hydrocarbon compounds to an acceptable level or threshold for discarge or reuse purposes (Igunnu and Chen, 2014; Samuel et al., 2022b).

Various treatment methods such as bioremediation (Haritash and Kaushik, 2009; Mazzeo et al., 2010), and other physical treatment methods such as adsorption (Mofokeng et al., 2021) are known to remove these contaminants to a certain degree. However, it has been reported that these hydrocarbon compounds remain in the water in trace amounts, which could be harmful to humans and the ecosystem (Abdel-Shafy and Mansour, 2016). As a result, a post-treatment method is required to completely degrade the trace pollutants from the OPW to meet the acceptable standard before discharge or for beneficial reuse. This leads to tremendous research into various treatment methods which could be used to completely remove or degrade these contaminants to an acceptable level for discharge or reuse purposes. Photocatalysis is considered a promising technology to degrade and possibly mineralize these hydrocarbon contaminants to an acceptable level for discharge into the environment or reuse purposes (Byrne et al., 2018).

To categorize the treatment capabilities of the various treatment technologies available for the removal of organic hydrocarbon in OPW, this review adopted the method proposed by Arthur et al. (2005) to give the degree of contaminant removal in percentage. The treatment capacities are rated 1, 2, 3, 4, and 5 representing very low, low, moderate, high, and very high respectively with percentage removal of the pollutant being <40 for very low, 40–60 for low, 60–80 for moderate, 80–95 for high and >95 for very high.

While some of these treatment technologies are well-developed in terms of their application in OPW treatment, others are not built or designed to comply with new environmental laws and regulations. Their potential for producing by-products as well as their high maintenance and energy costs may limit their future use. For example, in the future, a significant reduction in OPW and zero-pollution discharge will be required (Jiménez et al., 2018; Kurniawan et al., 2022c). Successful pollutant removal depends on pre-treating OPW before it enters the environment. Additionally, post-treatment is required to comply with the strict regulatory requirements that are currently in place. Alternatively, they may be introduced in the future for OPW discharge due to environmental concerns, as some technologies are unable to remove hazardous pollutants or even transform these pollutants from one form to another.

The removal of organic hydrocarbon chemical pollutants from OPW using various technologies is assessed based on their capabilities (Table 4). For this purpose, several parameters have been taken into account to summarize their effectiveness. Information from published articles, such as technical papers, website records, and journal articles that have undergone peer review, are taken into account. Comparing the level of removal of organic hydrocarbon pollutants contained in OPW has been done using numbers, which represent the level or grade for percentage removal (Amakiri et al., 2022b). Organic hydrocarbon chemicals eliminated from the OPW include phenol, BTEX, and PAHs.

Table 4 shows that the majority of the treatment methods with over 90% removal efficiency are less effective in the removal of BTEX, PAHs, and phenol from OPW. Treatment technologies such as coagulation and flocculation, floatation, activated sludge process, freeze-thaw/evaporation, API separators, hydrocyclone, ultrafiltration, microfiltration, and nanofiltration have low efficiency in the removal of these

Table 4
Efficiency of various treatment technologies for BTEX, PAHs, and Phenol removal in OPW (Amakiri et al., 2022b; Samuel et al., 2022b).

Treatment technology	Organic hydrocarbon compounds	Indication (level of removal)	Description	Percentage removal from OPW (%)
Membrane Technology				
Nanofiltration	BTEX	2	Low	40–60
	PAHs	3	Moderate	60–80
	Phenol	2	Low	40–60
Reverse Osmosis	BTEX	3	Moderate	60–80
	PAHs	4	High	80–95
	Phenol	3	Moderate	60–80
Microfiltration	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
Ultrafiltration	BTEX	2	Low	40–60
	PAHs	2	Low	40–60
	Phenol	1	Very low	<40
Macro-porous polymer extraction (MPPE)	BTEX	4	High	80–95
	PAHs	3	Moderate	60–80
	Phenol	4	High	80–95
Electrodialysis	BTEX	3	Moderate	60–80
	PAHs	3	Moderate	60–80
	Phenol	3	Moderate	60–80
Adsorption	BTEX	4	High	80–95
	PAHs	3	Moderate	60–80
	Phenol	2	Low	40–60
Coagulation and Flocculation	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
Flotation	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
Activated sludge process	BTEX	2	Low	40–60
	PAHs	2	Low	40–60
	Phenol	1	Very low	<40
Freeze-thaw/ Evaporation	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
API separators	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
Hydrocyclone	BTEX	1	Very low	<40
	PAHs	1	Very low	<40
	Phenol	1	Very low	<40
Heterogeneous Photocatalysis	BTEX	5	Very high	>95
	PAHs	4	High	80–95
	Phenol	5	Very high	>95

hydrocarbon chemical compounds in OPW.

Treatment technologies such as Macro-porous polymer extraction (MPPE) electrodialysis, reverse osmosis, and adsorption have moderate efficiency for the removal of organic hydrocarbon chemical compounds from OPW. Table 4 shows that heterogenous photocatalysis can be efficient in removing organic hydrocarbon compounds from OPW with a very high degree of BTEX and phenol removal as well as a high removal of PAHs from OPW. Other technologies could be more efficient in the removal of oil and grease, heavy metals as well as other contaminants in OPW. However, they are less efficient in removing organic hydrocarbon chemicals such as BTEX, PAHs, and phenol present in OPW.

It is important to note that feed concentration, pH, and other factors could influence the degree of removal of these hydrocarbon chemical compounds in OPW depending on the type of method used or chosen for the treatment process. Heterogenous photocatalysis is considered an environmentally friendly, cost-effective, energy-saving, and easy process for the removal of various contaminants in wastewater. As this treatment method has attracted popularity, research has been intensified to optimize its use for the treatment of various pollutants in OPW, which contains a mixture of complex constituents (Samuel et al., 2022a).

5. Mechanism of heterogeneous photocatalysis for organic pollutant degradation in OPW

Heterogeneous photocatalysis is a chemical process in which a photocatalyst absorbs photons energy ($h\nu$) greater than its bandgap energy (Eg) when light irradiation falls on it, thereby generating e^-/h^+ pairs, which migrate to the surface of the catalyst to initiate a redox reaction (Samuel et al., 2022a; Singh et al., 2020). In the case of OPW, this process involves a chemical transformation of all organic compounds in the OPW in the presence of a photocatalyst, visible, ultraviolet (UV), or solar light (Bharagava, 2020). For photocatalysis to occur, there must be the presence of dissolved oxygen and water, which brings about the formation of the hydroxyl and superoxide reactive radicals, responsible for the degradation of organic compounds in the OPW. This process involves three steps: (i) photoexcitation of semiconductors to generate electron-hole pairs, (ii) separation of e^-/h^+ pairs, and (iii) surface redox reaction (Raizada et al., 2019).

The photocatalyst in heterogeneous photocatalysis is a semiconductor substance that can absorb incoming photons. The photocatalytic reactions, which include the oxidation and reduction of donors and acceptors, respectively, occur either at or near the semiconductor's contact with the fluid (typically aqueous). Various materials can be used as photocatalysts (Curti et al., 2016). Photocatalysis has shown to be a viable and less hazardous method for treating a variety of industrial wastewater (Al-Nuaim et al., 2022).

This process has been considered an effective method for the degradation of recalcitrant hydrocarbon compounds as well as its capability to utilize the UV, solar, and visible-light spectrum (Samuel et al., 2021). Photocatalysis has shown to be an effective and less toxic technology in removing PAHs, BTEX, and phenol (Saien and Nejati, 2007). AOP can be utilized as a pre-or post-treatment technique for highly contaminated water effluents to promote toxic chemical removal, or they can be integrated into existing OPW treatment techniques (Oturan and Aaron, 2014).

Recent advancements in UV/Vis light irradiation and advanced oxidation technology have gained popularity to improve pollutant degradation in water and wastewater treatment (Sadeghpour et al., 2020). As a result, durable, low-cost, and effective methods for water decontamination are required, which do not add to environmental stress or risk human health. With the discovery of new toxins, increased industrial activity, and the dwindling supply of portable water, traditional technologies are becoming less effective. Water contaminated with PAHs can be treated by conventional chemical treatments (chlorination, thermal oxidation, ozonation) or biological treatment plants using activated carbon. However, the techniques are insufficient to meet the legal requirements for purity.

Photolysis is an important transformation mechanism that impacts PAHs' fate in the aquatic environment. The formation of highly reactive intermediates, primarily *OH, a potent non-specific oxidant ($E^o = 2.8$ V) by sunlight photo alteration processes plays an essential role in the breakdown of PAHs and other pollutants in water (Samuel et al., 2018a; Vela et al., 2012). Recent progress in water purification has resulted in the development of oxidation techniques to eliminate persistent organic contaminants from aquatic environments. These approaches, widely known as advanced oxidation processes (AOP), are based on photochemical and catalytic processes to cause significant changes in the chemical structure of pollutants. These can be employed as a stand-alone or hybrid process (Huang et al., 1993).

5.1. Principle of photocatalytic oxidation reaction for organic pollutant degradation in OPW

The photocatalytic oxidation process involves the degradation of organic pollutants, which occur in the company of a photocatalyst (like WO_3, TiO_2, etc), an oxidizing agent like oxygen or in air, and the presence of an energetic light source (Fig. 3) (Samuel et al., 2021). In the

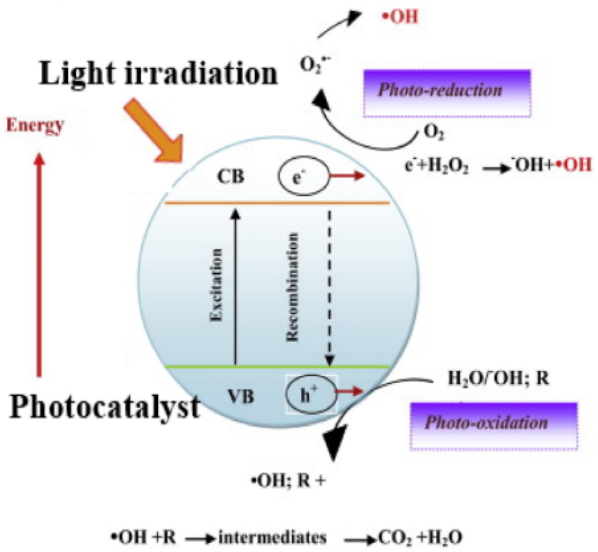

Fig. 3. Schematic diagram illustrating the principle of photocatalysis. Redrawn/Reprinted with permission from (Ahmed et al., 2011), Copyright 2011, Elsevier.

photodegradation of OPW, the photocatalyst absorbs energy above or equal to its bandgap energy when a light source is illuminated or reflected unto the surface of the photocatalyst. This process results in the generation of positive holes (h^+) and electron pairs (e^-) in the valence band (V_b) and the conduction band (C_b). Only photons with energies higher than the band-gap energy (ΔE) can excite the electrons in the valence band (V_b), allowing a reaction with organic contaminants (Fig. 3). Energy dispelling as heat is mainly caused when photons are absorbed with energies less than ΔE or have a longer wavelength (Ahmed et al., 2011). The pollutants are oxidized by the h^+ directly to form the hydroxyl (*OH) radical, while the electrons present in the C_b lower the adsorbed oxygen on the photocatalyst. The following step can be used to describe the activation of a photocatalyst by light irradiation.

The photocatalytic degradation pathway in the treatment of OPW involves a series of steps, which includes: (i) diffusion-based mass movement of reactants from the bulk fluid phase to the external surface of the photocatalyst, (ii) internal mass transport of the reactants to the photocatalyst's surface through its pores, where they can settle on an active site through adsorption, (iii) organic contaminant's adsorption on the photon-activated surface of the photocatalyst, (iv) desorption of photodegradation product, and (v) internal and external diffusion of the final photodegradation products (Amakiri et al., 2022a). The photodegradation performance of any photocatalyst is driven by the characteristics of the photocatalyst such as morphology, surface area, bandgap, and crystal structure (Truong et al., 2021).

$$Photocatalyst + hv \rightarrow e^- + h^+ \quad (1)$$

$$e^- + O_2 \rightarrow O_2^{\bullet-} \quad (2)$$

The reaction's oxidizing and reducing agents are h^+ and e^-, respectively. The following are the oxidative and reductive reaction steps:

Oxidative reaction:

$$h^+ + Organic\ Pollutant \rightarrow Intermediates \rightarrow CO_2 + H_2O \quad (3)$$

$$h^+ + H_2O \rightarrow *OH + H^+ \quad (4)$$

Reductive reaction:

$$*OH + Organic\ pollutants \rightarrow Intermadiate \rightarrow CO_2 + H_2O \quad (5)$$

The photocatalytic oxidation process generates hydroxyl radicals (Eqs. 4-5). The *OH radical formed by the oxidizing of the adsorbed water represents the main oxidant in the degradation of organic contaminants, and the presence of oxygen can inhibit e^-/h^+ pair recombination. Organic molecules, such as chlorinated nitrophenols, aniline, and aromatics are attacked by the *OH, resulting in a variety of intermediates that vary, depending on the compound's nature. The produced intermediates then react with *OH to form the final products like CO_2 and H_2O. When the oxidation of target pollutants and the reduction of oxygen does not proceed at the same time during the photocatalytic degradation process, electron accumulates in the C_b, resulting in an increasing rate of e^- and h^+ recombination (Al-Nuaim et al., 2022). To achieve efficient photocatalytic oxidation, it is critical to avoid electron accumulation.

6. Opportunities, potentials, and performance of photocatalytic degradation process for BTEX, PAHs, and phenol removal in OPW

Various treatment methods have been used in the treatment of OPW over time. However, most of the techniques have their limitations This has led to researchers finding other better, cost-effective, and efficient methods to treat this complex wastewater. Treatment methods such as floatation and hydrocyclones have been reported to remove oil and particulates from OPW. Adsorption is also reported to eliminate more than 80% of heavy metals, resulting in practically total water recovery. BTEX in OPW can occasionally be eliminated using adsorbents. Oil and grease, BTEX, and heavy metal particulates have also been reported to be eliminated using electrodialysis. RO membrane technology is better suited for organic molecules removal in comparison to other membrane technologies.

Heavy metals and TSS can be removed using membrane technologies such as NF and UF. However, effort must be taken to reduce membrane fouling, waste formation, and optimize backwashing for future applications. Consequently, the overall cost and the intended goal have an impact on the treatment technique to be chosen (discharge or reuse).

A relatively new OPW treatment method with the potential for technological advancements is MPPE technology. It has additional advantages over thermal technologies like FTE since it can handle a wide spectrum of pollutants with less energy. Although thermal systems have a lot of benefits, they still need to be improved to reduce their relatively high cost and reliance on temperature (Amakiri et al., 2022b; Veolia, 2015). Biodegradation is known to degrade these compounds to a significant level. Nevertheless, it seems to be inadequate when water contains toxic recalcitrant compounds such as phenol, BTEX, and PAHs (Costa et al., 2012; Nova et al., 2009). Trace amounts of these compounds are known to remain in the OPW after the biodegradation process (Haritash and Kaushik, 2009).

In addition to the limitation of the treatment methods, most of the methods are unable to remove minute-size oil particles completely from OPW when used for the treatment process. Therefore, this necessitates the need to adopt treatment methods such as heterogenous photocatalysis, which has shown to be effective in degrading minute oil particles in OPW (Samuel et al., 2022a).

In recent years, researchers have studied and reported the use of heterogeneous photocatalysis as an OPW treatment technology with TiO_2 being the most widely used catalyst. The pH of the solution does not need to be changed during this operation (Coha et al., 2021). Studies have shown that raising the amount of catalyst increases the removal efficiency of target pollutants. However, at high catalyst loadings, the degradation efficiency typically peaks or even decreases because the suspended catalyst prevents light from penetrating through the solution. Bessa et al. (2001) published their work on the potential use of irradiated TiO_2 for photocatalytic OPW treatment. Subsequently, researchers have published on the photocatalytic treatment of various pollutants in OPW.

For example, Sheikholeslami et al. (2019) synthesized maghemite

nanoparticles (γ-Fe_2O_3) to be used as a semiconductor photocatalyst for photocatalytic degradation of BTEX with 600 mg/L of concentration in synthetic OPW. They studied the photocatalytic degradation of BTEX as well as the effect of pH and light intensity on the rate of photocatalytic degradation of BTEX. The degradation was performed under both visible and ultraviolet irradiation (UV). Their findings show that the photocatalyst exhibited an excellent degradation of the BTEX with a removal efficiency of 95% in 5 days and 97% in 90 min under visible and UV light, respectively. Their findings also show that pH changes significantly affected the degradation of BTEX and that the degradation rate of the BTEX increases with a decreasing pH (3–7) of the OPW. Their findings show that changing the light source from ultraviolet to visible light irradiation increases the reaction time from 90 min to 5 days. It can be deduced from their work that pH and light irradiation has an effect on the photocatalytic efficiency of the photocatalyst for the degradation of the BTEX present in the OPW.

In another study conducted by Liu et al. (2016a), naphthalene was selected to represent PAHs in the OPW due to their abundance and toxic nature. In this work, they immobilized Aeroxide® P25 nano-scale TiO_2 powder onto glass slides for the photodegradation of Naphthalene in the OPW. The findings from the work indicated that the reaction rate constants in the immobilized and homogeneous systems were 0.00305 min^{-1} and 0.00219 min^{-1}, respectively which shows that the immobilized photocatalyst system exhibited better performance in the photocatalytic reaction for the removal of naphthalene. They also reported that the immobilized photocatalysts were resistant to the effect of substrate present in the OPW.

In a separate work, Taghizadeh et al. (2020) synthesized TiO_2/GO photocatalyst and compared its photocatalytic degradation of BTEX to that of bare TiO_2 when immobilized onto a forward osmosis membrane for OPW treatment. Their findings show that the TiO_2/GO immobilized photocatalytic membrane exhibited good ability in desalination and had a better photocatalytic degradation of ethylbenzene. They reported that the enhanced photocatalytic degradation of BTEX by TiO_2/GO was 80% under visible light and 78% under UV light with an initial concentration of 10 mg/L. The TiO_2 photocatalyst showed a degradation efficiency of 62% under UV light. However, the enhanced photocatalytic activity of the TiO_2/GO, resulted from the presence of GO, which extended the photocatalytic activity of TiO_2 into the visible spectrum. This suggests that GO can be used to form a heterojunction with TiO_2 to extend its visible light absorption capacity.

In a seminal work, Bo et al. (2016) also studied the degradation of PAHs and the effect of the water matrix by comparing the TiO_2 induced photocatalytic performance for synthetic OPW and distilled water. They also studied the effects of insoluble particulate matter and salinity on photocatalytic degradation. Their findings show that there was an improvement in the photocatalytic process with the addition of a catalyst. However, the photodegradation of the PAHs with high molecular weight was sensitive to variation in the insoluble matter because of lower solubility and variation in salinity in the OPW. The abundance of aromatic compounds in the OPW affected the photodegradation of the PAHs in OPW. This shows that the water matrix also influenced the rate of degradation of organic compounds in OPW. It is important to note that the concentration of the organic compounds in the OPW can significantly affect the rate of their photodegradation in the OPW.

To study the effectiveness of the photocatalytic degradation of organic compounds in synthetic OPW, Hayati et al. (2018) synthesized ZnO/TiO_2 anchored on *r*GO composite for photodegradation of phenol under visible light. Their study reported that the composite photocatalyst showed improved photocatalytic activity with *r*GO being responsible for reducing the TiO_2 bandgap energy to the visible light region. Their findings showed a complete degradation of phenol in aqueous solutions under visible light irradiation, which could be due to the higher surface area of the photocatalyst provided by the *r*GO and low electron-hole recombination rate. This makes the photocatalytic reaction last longer. They reported that the optimum degradation was achieved at pH 4 with 0.6 g/L of catalyst dosage. They also studied the kinetic and recyclability of the photocatalyst and found that the degradation followed pseudo-first-order kinetic mode with the photocatalyst having good reusability and recyclability. The findings show that modification of the TiO_2 surface with the *r*GO played a key role in improving the photodegradation of the target pollutant.

In separate work, Liu et al. (2021a) combined photocatalysis and ozonation using TiO_2-nanotube arrays to treat real OPW fortified with 16 PAHs compounds and to study the biodegradability and toxicity of the intermediate compounds. They reported that the process removed nearly all the compounds within 1 h of treatment. Higher ozone doses resulted in higher removal of target pollutants. A loading of TiO_2-nanotube arrays greater than 0.1 g/L resulted in increased PAH adsorption on the nano-catalyst. This also leads to low or no traces of PAHs in the treated OPW, but highly concentrated on the photocatalyst's surface. The findings further reveal that ozone could be used to initially oxidize the OPW matrix and then apply UV light to achieve complete degradation of PAHs. Additionally, their findings reveal that the combination of the biodegradation/ozone/UV light treatment could be practically applicable, due to the stability of the photocatalyst and the cost of operation.

Due to the complex nature of OPW, the performance of the various photocatalyst in the degradation and removal of these hydrocarbon compounds may be influenced by various factors such as pH, light intensity, the concentration of the OPW, photocatalyst design and fabrication, catalyst dosage (Kurniawan et al., 2011). Table 5 presents several studies on the photocatalytic degradation of BTEX, PAHs, and phenol present in OPW.

7. Challenges and operational parameters for effective photocatalytic degradation of hydrocarbon compounds and other chemicals in OPW

7.1. Challenges of photocatalytic degradation for organic compounds in OPW

OPW is a complex wastewater, which makes its treatment difficult. Due to its complex nature, no single technique could be capable of completely removing the various pollutants in the OPW. In spite of the effectiveness of the photodegradation and removal of BTEX, PAHs, and phenol, some challenges hinder its overall performance in removing organic carbon and matter from OPW (Liang et al., 2023). Some of the bottlenecks are listed and discussed below.

7.1.1. Decomposition and mineralization of organic compounds

Research on the photocatalytic treatment of OPW over the past decade has been scarce, although the majority of the available studies (Al-Sabahi et al., 2018; Andreozzi et al., 2018a; Hong et al., 2018; Jiménez et al., 2019; Liu et al., 2016b) used synthetic OPW. Target pollutants included xylene, phenol, acetic acid, glutaraldehyde, naphthalene, toluene, and others that are commonly found in OPW. While some of them might quickly decompose, all of the components' mineralization rates remained low. Different AOPs (photocatalysis, ozonation, and Fenton) were examined by Jiménez et al. (2019) for synthetic OPW treatment. Total organic carbon (TOC) removal was determined to be <20% after 4 h of treatment for the best-case scenario, making photocatalysis the least successful of these methods for the treatment of OPW.

It was also observed that with higher P25TiO_2 dosage, TOC removal was not significant. Gas chromatograph-mass spectroscopy (GC-MS) examination verified the complete elimination of smaller and more volatile organic pollutants (such as xylene, naphthalene, and toluene) as well as the reduction in phenol content (up to 99%). Liang et al. (2011) also found that neither TiO_2 photocatalysis nor UV photolysis significantly reduced TOC (for larger molecular organic compounds like natural organic matter, oil, and grease). Andreozzi et al. (2018a) studied the treatment of saline OPW with various compositions of recalcitrant

Table 5
Report of studies on photocatalytic degradation of BTEX, PAHs, and phenol from OPW.

Photocatalyst	Targeted pollutant	Initial concentration of pollutant	Condition	Removal efficiency (%)	Reference
γFe_2O_3	BTEX	600 (mg/L)	pH = 3, γFe_2O_3 amount = 150 mg/l, t = 5 days, Visible light	95%	Sheikholeslami et al. (2019)
Aeroxide® P25 Nano-scale TiO_2 on glass	PAHs (Naphthalene)	0.01 (mg/L)	pH = 6.86, t = 12h, T = ambient temperature, catalyst amount = 0.01 mg/l	Approx. 85%	Liu et al. (2016a)
TiO_2/GO	BTEX	10 (mg/L)	TiO_2 amount = 10 mg/l, t = 180 min, Tempt. = 25 °C, Visible light.	80%	Taghizadeh et al. (2020)
ZnO/TiO_2/rGO	Phenol	60 ppm	pH = 4, ZnO/TiO_2/rGO amount = 0.6 g/l, t = 160 min visible light	100%	Hayati et al. (2018)
TiO_2	PAHs	0.06 (mg/L)	TiO_2 = 100 mg/l, t = 200 min	Approx. 100% after 3 h	Liu et al. (2016b)
TiO_2	BTEX	10 (mg/L)	TiO_2 amount = 10 mg/l, t = 180 min, Tempt. = 25 °C, UV light.	62%	Taghizadeh et al. (2020)
Immobilized TiO_2 on glass plate	PAHs	0.01 (mg/L)	t = 1h, UV light + Ozone	90% of 16 PAH degraded	Liu et al. (2017a)
rGO/TiO_2	BTEX, PAHs, organic acids, and phenols	10 (mg/L) each	rGO (10%)/TiO_2–P25, pH = 3–4, UV light.	% Degradation not reported. But exhibited a high photocatalytic reaction with increasing reaction rates: acetic acid < phenols < naphthalene < xylenes < toluene.	Andreozzi et al. (2018a)
TiO_2-nanotube (TNA)	PAHs	0.02371(mg/L)	TNA amount = 0.2 g/l t = 1 h, UV + Ozone	Approx. 100%, with TNA showing strong photocatalytic capability.	Liu et al. (2021a)
γFe_2O_3	BTEX	600 mg/L	pH = 3, t = 1.5h, γFe_2O_3 amount = 150 mg/l, UV light	97%	Sheikholeslami et al. (2019)
(ZnO/Fe_2O_3 NC)	Phenol	4.5 mg/L	Conc. = 1 mg/L, 2 mg/L, 3 mg/L, t = variable	76.7%, 88.8%, 98.7%	Haiqi et al. (2020)

organic compounds using *r*GO-TiO_2 nanocomposites. They found that the photocatalyst exhibited better photocatalytic activity than bare TiO_2. They found that the increasing rates of photocatalytic reaction was in the order: Acetic acid < phenols < naphthalene < xylenes < toluene, while only 22% of the TOC was removed within 5 h.

Normally for improving oil recovery, partially hydrolyzed polyacrylamide (HPAM) is a frequently utilized polymer. To achieve an efficient solar photocatalytic degradation of HPAM, Al-Sabahi et al. (2018) reported the utilization of zinc oxide nanorods arranged vertically and supported on substrates tailored to enhance their ability to harvest visible light. Following 6 h of treatment, the removal of 25, 50, 100, and 150 mg/L of HPAM was found to be 68, 62, 56, and 45%, respectively. After 7 h and 14 h of reaction time, mineralization was found to be 20 and 37% of TOC reduction respectively. This concludes that photocatalysis could be efficient in breaking down recalcitrant organic molecules, but it is not very effective in achieving mineralization (Levchuk et al., 2020; Lin et al., 2020). It is advised that various technologies need to be employed in a series of operations during OPW treatment to further decrease target organic pollutants and photocatalytic intermediate products.

7.1.2. Toxicity of catalysts

Concerns about possible hazards and toxicological effects on public health and the environment, resulting from the toxicity of catalysts as engineered nanomaterials, have grown in recent years (Liu et al., 2017b). This is related to the widespread usage of photocatalytic nanoparticles, such as TiO_2 nanoparticles, and personal care items like sunscreen and chewing gum. Due to repeated and direct exposure, there are growing concerns about the possible dangers of nanoparticles to human health (Friehs et al., 2016). The difficulties associated with the toxicity assessment of nanomaterials have led to concerns and cautions from toxicologists and researchers around the world (Friehs et al., 2016; Rueda-Marquez et al., 2020). Additionally, not all synthetic materials have been taken into account in toxicology research, and the immaturity of nanotechnology makes it difficult to create realistic exposure scenarios (Lin et al., 2020).

Several works on the toxicity of nanomaterials have been published over the past ten years. TiO_2 nanoparticles have demonstrated some level of toxic effects on a wide range of organisms and cell lines, including human keratinocytes (Simon et al., 2011; Yin et al., 2012), algae (Lee and An, 2013; Miller et al., 2012), fish (Bar-Ilan et al., 2012; Faria et al., 2014), bacteria (Dalai et al., 2012; Tong et al., 2013), Daphnia Magna (Kim et al., 2014; Mansfield et al., 2015), and fungi (Lipovsky et al., 2011). Most of the research found that toxicity was influenced by nanoparticle concentration, irradiation intensity, and exposure time. When exposed to UV-containing light, phototoxicity was also observed in CdSe/ZnSe quantum dots (Kim et al., 2010), ZnO (Lee and An, 2013), and CuO (Dasari et al., 2013). In-depth reviews of photo-catalytically active nanoparticles for several energy-related and environmental applications were conducted by Friehs et al. (2016). They described the generation of reactive oxygen species and the solubility of metal ions as the two primary mechanisms of phototoxic action on living organisms (Garcia-Muñoz et al., 2020a).

However, P25-based TiO_2 is the main focus of the majority of phototoxicity investigations. There has been little species diversity in the study of nanotoxicity (Bour et al., 2015). The materials and the associated coatings, modifications, and doping that alter their physico-chemical properties have not been thoroughly studied so far. Additionally, because there are so many variables that might affect an experiment's results, the results may be inconsistent and contradictory (Bahadar et al., 2016; Lin et al., 2020), making it difficult to meaningfully extrapolate the findings to other nanomaterials. To assure the safe implementation of heterogenous photocatalysis and other AOPs to OPW treatment, the toxicity of catalysts should be thoroughly examined. Despite a surge in photocatalysis toxicity investigations, only a small number of articles have included toxicity testing for OPW. To promote the use of photocatalytic treatment methods, further study is required on the toxicity of OPW and photoinduced toxicity (Lin et al., 2020).

7.1.3. Inhibiting abilities of organics in OPW to the photocatalytic degradation process

One of the main inhibitors for treating OPW is organic compounds.

Since photocatalysis combines adsorption and degradation, the presence of organic compounds in OPW may alter how well these two processes remove target substances. Through hydroxyl scavenging, light absorption, and site blockage, organic compounds can stop the degradation of pollutants (Katz et al., 2015). Toluene and acetic acid are two recalcitrant substances found in OPW that are resistant to decomposition because they react with radicals slowly (Jiménez et al., 2019). There are not many studies addressing components in OPW that are resistant to ·OH, like naphthalene, phenol, and acetic acid, and their removal efficiencies are low (Andreozzi et al., 2018b). It was uncovered that resorcinol hydroquinone, catechol, and dihydroxy benzenes, severely impede photocatalytic processes (Rincón and Pulgarin, 2004).

According to certain reports, organic materials generated from wastewater can scavenge up to 95% of ·OH (Keen et al., 2014). Liu et al. (2016b) assessed the impact of organic content and insoluble particulate matter in OPW on photocatalytic performance. The breakdown of PAHs was found to be more likely to be slowed down by the organic content. Additionally, aromatic molecules were crucial in UV photons absorption due to UV sensitivity and their abundance. Additionally, aromatic compounds may compete with PAHs for active species and adsorption sites in the course of the heterogenous photocatalytic process. As a result, highly concentrated aromatic compounds could dramatically lower the overall photocatalytic activity.

7.2. Operational parameters for effective photocatalytic degradation of organic hydrocarbons in OPW

The rate and efficiency of photodegradation in any photocatalytic system depend on the variety of operational parameters responsible for controlling its performance (Gnanaprakasam et al., 2015; Kumar, 2017).

7.2.1. Effect of pH

In photocatalytic degradation, pH is one of the most important parameters. During heterogeneous photocatalytic degradation, the initial pH of a solution may influence the rate of photodegradation of organic pollutants. The degree of pollutant ionization, the oxidant, the hydrolysis of the pollutant, and the surface characteristics of catalysts are all affected by pH (Hayati et al., 2018). The rate of degradation of BTEX is significantly affected by pH changes. Upon lowering the pH of the solution, the degradation efficiency improves. The catalyst's superficial load or isoelectric point is affected by changes in the operating pH. When the pH of the solution is lower than the pH at the point of zero charges (pH_{pzc}), the photocatalyst's positive charge attracts more anions, increasing the degradation rate (Liu et al., 2017a; Samuel et al., 2016). Sheikholeslami et al. (2019) investigated the effects of pH on the photodegradation of BTEX using maghemite nanoparticles (γ-Fe_2O_3) as photocatalysts by varying the pH of the OPW between 3 and 7. The study reported that pH significantly affects the rate of BTEX degradation. They observed that the degradation efficiency increases with a decreasing pH with a degradation rate of 95% after 90 min with the highest degradation occurring at pH 3 under UV light.

Hayati et al. (2018) studied the effect of pH on the photocatalytic degradation of phenol in petroleum wastewater. The experimental study was carried out at pH ranging from 4 to 10, a phenol concentration of 60 mg/L, and a catalyst dosage of 0.8 g/L, during a 160-min irradiation period. The plot of concentration against time-based on the effect of pH on phenol-degradation in the acidic solution shows a higher pH value than that in alkaline and neutrals, which decreased from 91% to 52% with a rising pH from 4 to 10. A possible explanation for the changes in degradation efficiency at different pH levels could be attributed to hydroxyl radicals with a greater ability to oxidize under acidic conditions. However, under alkaline conditions, the presence of CO_3^{2-} and HCO_3^- interferes with the reactivity of hydroxyl radicals, lowering their oxidation potential. This proves that photo-induced radicals degrade phenol faster in acidic than in alkaline solutions. In addition, low Ph-degradation rates result from limited adsorption of negatively charged system components and CO_3^{2-} and HCO_3^- interference (at pH = 10). As a result, an effective phenol degradation might have taken place at pH ranging from 4 to 6.5. (Lower than pH_{ZPC} in the acidic area). The result from the photodegradation process showed a degradation rate of 91% at pH 4.

7.2.2. Effect of chloride ions

Because of its capacity to change •OH into a less reactive transitory species $Cl_2^{\bullet-}$ at pH less than 5, chloride is considered an interfering agent in numerous AOP. Chloride's effect on •OH reactions could be more complicated than previously imagined. Depending on the reactivity of the target pollutant and other substrates with $Cl_2^{\bullet-}$ and •OH, chloride may inhibit or even accelerate the degradation process when a specific pollutant is targeted during AOP. Phenol degradation is enhanced by Cl^- in the presence of methanol and 2-propanol because phenol competes better in the presence of alcohol for reaction with Cl_2*, as compared to its reaction with •OH. However, in the presence of 4-methoxyphenyl and hydroquinone, Cl^- hinders degradation since the compounds react faster with Cl_2*than with phenol, unlike their reaction with •OH (Coha et al., 2021; Mamah et al., 2022).

Liu et al. (2016b) studied the effect of salinity on the photocatalytic degradation of PAHs using TiO_2. They reported that the rate of degradation of low molecular weight PAHs increases with increasing ionic strength. They found that PAHs with higher molecular weight were sensitive to variation in the insoluble matter because of lower solubility and variation in salinity in the OPW media. This shows that the presence of Cl^- due to the salinity of OPW could increase or decrease the rate of PAHs degradation, depending on the specific compound during its treatment.

7.2.3. Effect of light intensity and wavelength

The extent to which a semiconductor photocatalyst absorbs light at a given wavelength is dependent on the light intensity. The rate of generation of electrons and holes during a photocatalytic reaction is influenced by light intensity (Cassano and Alfano, 2000). The overall efficiency of the process of converting and degrading pollutants is determined by the distribution of light intensity within the reactor (Pareek et al., 2008).

As a result, the degradation of various organic pollutants has been undertaken to determine the relationship between light intensity and the rate of pollutant's degradation. While some studies found a square root relationship between the reaction rate and light intensity, others found a linear relationship (Ollis et al., 1991; Samuel et al., 2018b; Terzian and Serpone, 1995). Al-Sayyed et al. (1991), Herrmann, (1999) observed that the rate was proportional to the radiant flux ϕ for $\phi < 25$ mW/cm^2. However, above this value, the rate varied as $\phi^{1/2}$, indicating a too-high flux and an increase in the rate of e^-/h^+ recombination. The reaction rate is unaffected by the light intensity at high intensities (Ollis et al., 1991). This could be explained due to the fact that e^-/h^+ generation reactions predominate at low intensities, while e^-/h^+ recombination is negligible.

Sheikholeslami et al. (2019) studied the effect of light intensity on the degradation rate of BTEX in OPW by varying the light intensity between 0 and 100W under UV light and 0–225W under visible light. Their report showed that light intensity affected the rate of degradation of BTEX. Their study reported that light intensity had a direct influence on the degradation. As the intensity increases, the degradation rate also increases to a degradation rate of 95% after 5 days under 225 W of light intensity. They concluded that the e^-/h^+ formation depends on the light intensity. The highest degradation was found to be at 225 W of light intensity.

7.2.4. Effect of catalyst loading

The rate of reaction speed is directly related to the catalyst loading or concentration. However, up to a certain amount of catalyst loading, the reaction speed is independent of the amount of catalyst loading. A

typical catalyst area is always exposed to light, creating more active sites for reaction (Ahmed et al., 2011). A higher concentration of photocatalyst nanoparticles prevents light from reaching other parts of the nanoparticles during the photocatalytic treatment, lowering removal rate or degradation efficiency (Momoh et al., 2022; Shahrezaei et al., 2012).

Pardeshi and Patil (2008) investigated the effects of photocatalyst dosage on the photodegradation of phenol. They varied ZnO photocatalyst from 0.5 to 3.5 g/L and found that at 2.5 g/L of photocatalyst concentration under sunlight, complete removal of phenol with 75 mg/L of concentration was degraded after 8 h. As the photocatalyst concentration increased, they found that the phenol photodegradation became fast and more effective to an optimum photocatalyst concentration of 2.5 g/L. With the photocatalyst higher than this optimum dose, the photodegradation efficiency of the photocatalyst decreases. The increasing photodegradation, accompanied by the increasing photocatalyst loading, is attributed to the fact that the number of absorbed phenol molecules improved due to the increasing number of ZnO particles.

Sheikholeslami et al. (2019) studied the effects of photocatalyst loading on the degradation of BTEX in OPW. They found that the rate of BTEX degradation into intermediate products was increased or decreased by varying the photocatalyst concentration during the photodegradation process. They varied the photocatalyst's concentration from 1 to 2.5 g/L and obtained the maximum degradation rate at 1.5 g/L of catalyst concentration.

7.2.5. Concentration and nature of pollutant

The concentration and nature of pollutants play roles in the efficient photodegradation of pollutants in OPW. Hayati et al. (2018) reported phenol degradation from petrochemical wastewater using ZnO/TiO_2 decorated on reduced graphene oxide nanocomposite. They studied the effects of the phenol concentration on its photodegradation rate by varying its concentration from 60 to 90 mg/L. They found that at 90 mg/L of phenol concentration, its degradation rate was 76%. The rate of degradation increased significantly as phenol concentration decreased from 90 to 60 mg/L. This is attributed to the fact that an increasing phenol concentration and its intermediate by-products lead to the deactivation of active sites while reducing the penetration of light to the active sites on the photocatalyst's surface which generates oxidative radicals.

In a separate study, Liu et al. (2016b) investigated the photocatalytic degradation of PAHs in OPW using TiO_2. They found that the photocatalyst strongly improved the photodegradation of PAHs in the OPW. However, the photocatalytic process was hindered by the concentration and nature of the OPW. They reported that the PAHs with higher molecular weight showed sensitivity to insoluble matter and salinity content of the OPW because of their solubility. In general, organic constituents, particularly the aromatic compounds present in the OPW hinder the photodegradation of PAHs because of their role as UV-photon absorbents. This makes them compete during the photocatalytic degradation of the PAHs in the OPW. This is attributed to the higher concentration in the OPW. This shows that the nature and concentration of the pollutants in the OPW can affect the hydrocarbon's rate of degradation in the OPW media. Table 6 provides a summary of various factors affecting the efficiency of the photocatalytic process. The table shows the optimum range of pertinent parameters to be considered during the treatment for efficient photocatalytic degradation of various pollutants in water/wastewater.

Table 6
Factors influencing photocatalytic degradation performance.

Factor	Optimum range	Remarks	Reference
Temperature	20 °C < T < 80 °C	High temperature: Water or the target molecule will volatilize, which will accelerate the recombination of charge carriers. Low temperature: Poor desorption of the final product	Malato et al. (2009); Rauf and Ashraf (2009)
Light intensity and wavelength	≤50 W/m^2 (in 300–400 nm range)	Until the mass transfer limitation is achieved, the increased light intensity accelerates the reaction rate.	Ahmed et al. (2010); Malato et al. (2009)
Catalyst loading	–	An increase in the opacity and the phenomenon of light scattering due to a high number of immobilized coating layers can cause blockage of the inner layers.	Henderson (2011); Qiu et al. (2000)
pH	The point of zero charge should be avoided	Since it affects particle agglomeration and adsorption, an optimal value is required.	Mozia (2010); Zangeneh et al. (2015)
Concentration and nature of pollutant	–	An increase in an initial concentration above the ideal value has negative effects.	Zainal et al. (2005); Zangeneh et al. (2015)
Oxygen contents	≥Po_2 = 0.21 atm.	The oxygen contents may hinder or enhance the degradation rate depending on the rate of degradation.	Kabra et al. (2004); Malato et al. (2009)

8. Pilot and commercial scale application of photocatalytic degradation in OPW and other wastewater treatment

Recently, the development of multi-dopant metal oxides and composites for use as new photocatalysts has increased. However, the use of photocatalysis in practical applications for environmental needs is still elusive. Investments in commercial-scale photocatalysis applications have so far been limited due to worries concerning the unknowable toxicity and fate of nanoparticles, insufficient performance in practical settings, durability factors, and mass transfer restrictions (Alalm et al., 2021). Despite the numerous laboratory-scale studies demonstrating the modern photocatalysts' ability to degrade a variety of water contaminants, there is still growing doubt regarding the widespread commercial use of photocatalysis for various environmental applications (Hodges et al., 2018; Loeb et al., 2019).

To demonstrate the pilot scale application of photocatalysis in water treatment applications, Hoang et al. (2021) used a pilot plant continuous flow reactor. They immobilized gel-derived Ag–TiO_2–SiO_2 photocatalytic nanomaterial onto glass beads and applied compound parabolic collectors as solar radiation collector for an efficient treatment of drinking water. They reported that from the bactericidal test, total coliform in the inlet water was inactivated within 6.67 and 10 min of retention time for the groundwater and river, respectively. They found that the effectiveness of coliform deactivation depends on the irradiation intensity at a lower retention time. They also reported that Ti and Ag^+ were detected to have been released in lower concentrations in the treated water. However, their concentration is lower than the World Health Organization (WHO) standard for drinking water. This implies that the pilot-scale treatment system could be a promising system for the treatment and inactivation of bacteria in both river and groundwater under sunlight or solar radiation and could be explored for large-scale application.

Colina-Márquez et al. (2009) utilized a solar pilot plant compound parabolic collector (CPC) photoreactor to investigate the degradation of a commercial herbicide mixture (ametrine, 2,4-D, and diuron) using TiO_2 as the photocatalyst. They reported that the fluid-dynamic models, and reaction kinetics combined with the six-flux absorption scattering

model (SFM) of the radiation field in the photoreactor demonstrated the degradation of water pollutants in heterogenous photocatalytic reactors with accuracy. They concluded that the overall model obtained fitted well with the experimental data obtained for the mineralization of the herbicide in the photoreactor for both sunny and cloudy days. It could be said that the pilot scale photoreactor demonstrated that it could be adopted for large-scale applications in water treatment by heterogeneous photocatalysis.

Agüera et al. (2000) used a preindustrial pilot plant photocatalytic reactor to study the degradation of commercially formulated (CF) and technical grade product (TP) pyrimethanil of 40% (*w/v*) and 98% of purity, respectively, under sunlight using TiO_2 as the photocatalyst. They found a complete degradation of both the TP and CF pyrimethanil after 4 h of sunlight irradiation. However, total mineralization did not take place as the TOC analysis indicated a value of 3–4 mg/L after the degradation process. Intermediate compounds were formed during the degradation process, and this was demonstrated by the GC-MS analysis. It could be concluded that the preindustrial pilot plant has demonstrated the ability to upscale the photocatalytic process to a larger scale or industrial scale when other factors are taken into consideration.

In another study, Boyjoo et al. (2012) demonstrated the use of a photocatalytic pilot scale plant with a continuous slurry recirculation mode reactor to study the photodegradation of pollutants in real shower water for reuse purposes using TiO_2 as the photocatalyst. They studied various operational parameters including airflow rate, catalyst concentration, initial pH of the slurry, and the rate of slurry recirculation. They reported 57% of TOC removal after 6 h of the photocatalytic process. They concluded that the ease of operation of the photocatalytic process coupled with the potential to use solar irradiation makes the photocatalytic process promising. This study demonstrates that the photocatalytic degradation process can be successfully upscaled from laboratory to pilot plant scale.

Overall, the photocatalytic degradation process for polluted water treatment can be scaled up to the commercial scale. However, other factors need to be considered before a successful and efficient process can be achieved. It is important to note that with the increasing awareness and interest in heterogenous photocatalysis for the treatment of wastewater, commercial-scale applications are still limited. The primary challenges which limit the large-scale or commercial application of heterogenous photocatalytic processes in wastewater treatment are elaborated in the following section.

8.1. Bottlenecks in pilot and commercial scale application of photocatalytic process in OPW and other wastewater treatment

Despite numerous research on the use of heterogenous photocatalysis for wastewater treatment, this technology is still a long way from being fully commercialized in the market. The cost estimation and life cycle assessment (LCA) analysis of the processes and the developed materials need to be given further consideration for the photocatalytic technology to be both economically viable and environmentally sustainable (Loeb et al., 2019). The remediation of municipal wastewater by photocatalytic treatment has so far been constrained by cost and scale-up issues. This makes the process uncompetitive with other AOPs and conventional technologies (Kurniawan et al., 2022c).

However, specialized uses of photocatalytic technology, such as treating isolated water sources or hydroponic and aquaculture waste streams, are more likely to succeed in the future. To revive industrial interest in the technology, pilot-scale investigations and tests on actual samples are required. A discussion on the bottlenecks of the photocatalytic process's pilot and commercial deployment is presented in the following subsections.

8.1.1. Reactor configuration and design

Photocatalytic reactors' primary purpose is to bring contaminants and the illuminated catalyst into contact. Dispersing the photocatalyst in an agitated vessel with a proper source of irradiation will quickly and effectively achieve this purpose. Most research reporting photocatalytic degradation or disinfection of micropollutants used this approach (Garcia-Muñoz et al., 2020b). Despite dispersed catalysts' strong photon absorption and effective reactive oxygen species (ROS) interaction with contaminants, it is challenging to recover nanostructured powder after the process, which hinders photocatalyst reuse. After the initial photocatalytic cycle, researchers introduced drops of concentrated pollutants to demonstrate the reusability and stability of the photocatalysts (Gar Alalm et al., 2016).

The constraint of this approach is that it is not applicable in real life. As a result, this limits its application. Furthermore, as the toxicity and effects of the nanoparticles on the environment are still not well understood, a possible release of photocatalyst powder is another disadvantage of the system setups using dispersed catalysts. It is recommended that for pilot and commercial scale wastewater treatment applications, photocatalysts used in retain form in the reactors could facilitate its reusability, removing the need to recover the photocatalyst powder, which typically requires ultrafiltration, as well minimize the risks of the release of nanoparticles into the environment (Alalm et al., 2021).

A photocatalytic reactor system that maintains the photocatalyst inside its confines is the fluidized-bed reactor. To adsorb and degrade bisphenol A in a fluidized-bed reactor, Fang et al. (2019) synthesized a composite macrostructure comprising graphene and TiO_2 nanotubes. With a flow rate of 1 mL/min, this system successfully removed 50 and 86% of the pollutant from solutions with 0.5 and 0.05 mg/L of Bisphenol A, respectively. The Bisphenol A degradation efficiency was maintained in the course of the process, which lasted 16.5 h.

8.1.2. Cost and environmental life-cycle assessment

Numerous researchers assert the synthesis of cutting-edge photocatalysts, but offer no details on their lifecycle, cost, or toxicity (Loeb et al., 2019). For any photocatalytic technology to be fully applied, the challenges are of utmost significance. Electrical energy per order (EEO) estimates are commonly used to discuss the long-term economic viability of large-scale photocatalytic wastewater treatment (Benotti et al., 2009; Keen et al., 2018). The median EEO values for several AOP, such as UV/chlorine, ozonation, UV/persulfate, and UV/H_2O_2, are less than 1 kWh/m^3, making them competitive for drinking water applications (Miklos et al., 2018; Sadik et al., 2019). In contrast, the EEO values for UV-activated photocatalysis are typically greater than 10 kWh/m^3 and this value must be reduced for photocatalytic water treatment to be a competitive strategy for treating municipal wastewater (Miklos et al., 2018; Sadik et al., 2019).

Although assessing the process' energy efficiency by EEO is suggested, comparing several technologies using just one criterion might be misleading. The integrated energy costs of consumable chemicals are another issue that needs attention (Kurniawan et al., 2022d; Loeb et al., 2019). In this way, a remediation process' overall sustainability can be better understood using life-cycle assessment (LCA). LCA is an integrated technique that uses a large number of databases to collect the environmental effects of a process related to energy use, materials, and emissions from the cradle to the grave (Imtiaz et al., 2023; Mu et al., 2016). Despite the abundance of literature devoted to the development of new photocatalysts, the environmental effects associated with their manufacture and applications are frequently disregarded.

Few studies compared the effects of TiO_2 photocatalysis for micropollutant removal to those of other treatment methods. According to Magdy et al. (2021), the photo-Fenton process in the same compound parabolic collectors (CPCs) reactor had a lower effect on the environment than activated carbon adsorption and electro-Fenton methods. It was more eco-friendly when used to degrade phenol. The generation of TiO_2, power utilized to circulate the reactor's water, and leftover transformation products were the key environmental impact factors. Similar findings were reported by Muñoz et al. (2006), who contrasted

solar-based heterogeneous photocatalysis with the solar Fenton process.

On the other hand, Pesqueira et al. (2021) discovered that the photo-Fenton process for the degradation of pharmaceuticals had a higher environmental impact than TiO_2 photocatalysis. This variation could be explained by the variance in chemical doses and reaction times. It should be emphasized that there are few reports of LCA assessments of photocatalytic processes in the literature. As the majority of novel complex photocatalysts are yet to be added to the LCA databases, researchers need to expand the LCA inventory to accommodate the energy consumption and the precursors for the catalyst synthesis process. This makes an exhaustive LCA analysis difficult. Additionally, there are still significant knowledge gaps on new nanostructure's toxicity and environmental effects.

The paucity of knowledge regarding the market prices of innovative catalysts has made it difficult to estimate the costs of large-scale photocatalytic wastewater treatment. Most of the contemporary nanocomposites used to improve the photocatalytic process in visible light are not manufactured on a large scale for commercial applications. Furthermore, there is not much research on actual wastewater, making it difficult to determine the amounts needed for industrial wastewater treatment. As a result, only a few research (Gar Alalm et al., 2015; Navarro et al., 2009; Samy et al., 2021) have provided estimates of the costs associated with photocatalytic wastewater treatment. Because the pollutants and photocatalysts in the few studies were designated for unique circumstances, they were unable to demonstrate significant generic costs (Li et al., 2019, 2021).

8.1.3. Photocatalytic activity in real samples and complex water matrices

The water matrix's role is another challenge in relation to reactor configuration and design. The majority of research has described photocatalytic degradation processes in ideal laboratory settings, such as solutions of pure water with a single pollutant of which the concentration can be quantified (from a few mg L^{-1} to even maximum solubility) (Ateia et al., 2020; Kurniawan et al., 2022e). Even though strict control conditions are used to characterize new photocatalysts, they are not at all like the actual cases. Streams of industrial wastewater normally comprise a mixture of contaminants in complicated matrices or compositions with natural organic matter and a variety of electrolytes. Additionally, the micropollutant's level on the surface, as well as in drinking water, are typically in the range of ng L^{-1} to μgL^{-1}, which is significantly lower than in the majority of research in the literature on photocatalytic treatment (Alalm et al., 2021).

8.1.3.1. Pollutants degradation at low concentration. The precision and detection limit of the analytical technique affects studies on the micropollutant's removal in the range of ng L^{-1} to μg L^{-1} to mimic natural and groundwater streams. Chromatographic and photometric techniques are frequently employed in target analysis to assess photocatalytic degradation. Since neither accuracy nor detection limit can reliably operate at 0.1 mg/L, these approaches are typically suitable for mg L^{-1}scale.

The concentration of emerging micropollutants in the environment typically falls below this range, and threshold limitations are becoming increasingly stringent for key pollutants. For instance, the average maximum annual perfluorosulfonic acid and its derivatives concentration is 3.5 ng L^{-1}, while the maximum permissible pesticide concentration in drinking water in the European Union is 0.1 μg L^{-1} (Schmidt, 2018). Recently, micropollutant analysis with an accuracy of a few ng L^{-1} and the screening for the identification of transformation products were made possible by liquid or gas chromatography coupled with tandem mass spectrometry (MS/MS) (Bletsou et al., 2015). However, due to high costs, the technologies are not accessible. When choosing model contaminants for conventional liquid or gas chromatography, researchers without access to this equipment frequently chose pollutants with large concentrations (Liang et al., 2023). It is not practical to employ specific analysis to verify the acceptability of water for specific uses since water streams typically contain hundreds of specified organic components (Schmidt, 2018).

Nontarget analysis, which allows the relative detection of thousands of organic compounds without the need for reference standards, may provide a solution to this problem (Schymanski et al., 2015). For instance, computer-aided data processing techniques may precisely identify organic molecules using high-resolution mass spectrometry (MS), which is capable of detecting the mass-to-charge ratios (m/z) with an accuracy of 10^{-5} Da (Bletsou et al., 2015). We anticipate having photocatalysis studies using actual water and wastewater treatment based on non-specific analyses and much lower concentrations later in the future due to the expansion of these technologies.

Another strategy used to assess photocatalytic degradation is the removal of chemical oxygen demand (COD), dissolved organic carbon (DOC), or detoxification. Arcanjo et al. (2018) employed hydrotalcite and iron oxide to TiO_2 modified for the treatment of wastewater from the textile industry. The DOC and COD removals were not high (<10% and <20% for the DOC and COD in 360 min, respectively), in spite of color removal reaching 96%. The meager elimination was attributed to the production of recalcitrant by-products, which can take longer to degrade. The 48-h immobilization test of *Daphnia* similes used to measure toxicity showed a reduction in toxicity (from EC_{50} = 70% to EC_{50} = 79% by modified TiO_2 and to EC_{50} = 95% by pristine TiO_2) with respect to EC_{50} (dilution causing 50% immobility). Similar case studies of municipal and industrial wastewater that had undergone photocatalytic detoxification have been reviewed previously (Antonopoulou et al., 2021; Rueda-Marquez et al., 2020). In a separate application, Ru-WO_3/ZrO_2 was explored to disinfect real surface waters that were sampled from the intake source of water treatment facilities in the Nile Delta (Fouad et al., 2021).

8.1.3.2. Real water matrices with the presence of interfering species. To prevent interference, especially from unidentified organic species, most literature investigations have used photocatalytic tests using one contaminant at a time in a synthetic water matrix. To study the interaction between the pollutant and catalyst as well as the role of various reactive oxygen species (ROS) in oxidation, laboratory experiments on photocatalytic degradation of contaminants in transparent, background-free water are appropriate. However, this is not the case in real-world applications like the treatment of groundwater, industrial wastewater, or surface water which can contain a wide range of compounds. These are some examples of real-world applications where this is not the case (Maiurova et al., 2022; Ren et al., 2018). Other organic or inorganic species may prevent photocatalytic degradation from taking place or necessitate the use of additional catalysts (Zhao et al., 2019). Additionally, photocatalysts could exhibit varied responses to complicated water matrices, underscoring the limited relevance of comparing photocatalytic performance in ultra-pure water (Meroni et al., 2020).

As radical scavengers, inorganic electrolytes like chlorides and bicarbonates might negatively impact photocatalytic activity (Rimoldi et al., 2017). The overall effect is dependent on the relative concentration of the species and the pollutant. The photocatalytic degradation of bisphenol A by Rh–TiO_2 was studied by Repousi et al. (2017) using secondary treated wastewater with humic acid and inorganic salt solutions and ultra-pure water found in bottle. They found that humic acid presence accelerated the breakdown of bisphenol A due to Rh–TiO_2 particles. The particles formed fewer aggregates due to the repulsive forces created by the build-up of humic acid on their surface. Despite the total organic content (TOC) of 6.2 mg/L is considered low in the treated wastewater matrix than humic acid organic content of 9.2 mg/L, the photodegradation process in the treated wastewater was 30 times slower. It was anticipated that treated wastewater with recalcitrant organics would survive the biological treatment process as well as scavenge the ROS during the photodegradation process, even though the organic composition of the wastewater was unknown.

Ren et al. (2018) studied the restrictive effects of a synthetic mixture of citric acid, sodium alginate, and lake water on the photodegradation of clofibric acid. They found that the molecular weight of natural organic matter (NOM) had a greater impact on the suppression of photocatalytic activity than its concentration. While adjusting the pH did not influence the rate of degradation, increasing the amount of dissolved oxygen can reduce the inhibitory effect due to the increased formation of superoxide radicals. Depending on the type and quantity of organic compounds, NOM can hinder the micropollutant's photocatalytic degradation by various concurrent mechanisms.

Separately, Awfa et al. (2020) studied the photodegradation of carbamazepine using carbon nanotubes (CNTs) and TiO_2 composite in two stages of river water, treated domestic wastewater as well as three commercially available synthetic NOM solutions namely, Suwannee River fulvic acid (SRFA), Suwannee River humic acid (SRHA) and Suwannee River reverse-osmosis isolates (SRNOM). They found out that the photocatalytic degradation of carbamazepine was reduced from 93 to 40% in river water and from 93 to 87% in ultra-pure water. Additionally, commercial NOM surrogates inhibited the photocatalytic degradation in the following order: SRHA, SRFA, and SRNOM, showing that the inhibitory impact is dependent on NOM properties such as light absorption capacity, molecular weight, and aromaticity (the proportion of aromatic content to total organic content) (Zhu et al., 2020).

The photocatalytic degradation process can be negatively impacted by the inner filter effect because of low effective irradiation by the photocatalyst as a result of light absorption by organic compounds (Awfa et al., 2020). This effect is strongly influenced by the NOM's molecular structure because complex organic structures absorb light over a wider spectrum than simple organic structures and secondary by-products (Liu et al., 2010). Therefore, it is anticipated that water matrices with higher specific UV absorbance (SUVA), need enough light to activate the photocatalyst's surface to make up for what NOM would absorb (Zhang et al., 2021).

NOM accumulation on the surface of the photocatalyst can also inhibit the photocatalytic degradation process, resulting in the blockage of the active sites. This reduces the illuminated surfaces and minimizes the rate of ROS production even with a well-illuminated catalyst (Fouad et al., 2021). The presence of aromatic NOM also results in the scavenging of the generated ROS in wastewater, which prevents the oxidation of the target pollutant (Drosos et al., 2015). With numerous studies on the impact of NOM on the photodegradation of various pollutants, there is a growing need to better understand the effect of photodegradation on the NOM composition. This could be explained by the difficulties in characterizing the different NOM molecules' molecular structures, which restricts research to the analysis of aromaticity and fluorescence (Zhang et al., 2021).

8.2. Modification and design of photocatalyst for pilot and commercial scale applications

To scale up the photocatalytic degradation process for pilot and commercial scale applications, there is a growing need to consider the development and design of new photocatalysts with enhanced photocatalytic degradation efficiency. To achieve this purpose, the surface modification of this photocatalyst is important to improve its e^-/h^+ pair separation and charge transfer efficiency, porous structure, and specific surface area (Yan et al., 2013). To overcome the constraints of the photocatalytic material that limits its efficient utilization for pilot and commercial scale applications, surface modification is necessary by involving the addition of external components to the photocatalyst's matrix. This leads to the shifting of the band-gap absorption wavelengths and boosting its adsorption and degradation efficiency for organic pollutant's degradation (Lee et al., 2021). Photocatalyst surface modification has been considered a viable option to enhance photocatalysts' selectivity toward target contaminants (Phuangburee et al., 2020; Zhang et al., 2020).

It is therefore imperative to note that designing highly effective photocatalytic systems with stability over the long run for the simultaneous adsorption and degradation of organic pollutants is necessary (Guan et al., 2021). Although modification techniques such as doping, metal deposition, and forming heterojunction for improving the photodegradation of organic pollutants in wastewater have been investigated, most of the work needs to be further studied with respect to cost and complexity in the modification techniques to improve their effective deployment for pilot and commercial scale applications (Das et al., 2020; He et al., 2017; Liu et al., 2021c)

Further research needs to focus on investigating more cost-effective and less complex surface modification methods or a combination of various methods. To achieve an efficient degradation of organic pollutants, we need to explore the possibility of scaling up to pilot or commercial scale applications (Guan et al., 2021; Lee et al., 2021). So far, surface modification of photocatalysts has been able to increase the photocatalyst adsorption wavelength as well as create highly reactive species to enhance the degradation of the photocatalyst for organic compound degradation (Low et al., 2017).

8.3. Photocatalyst recovery, stability, and reuse

Two crucial factors to take into account for the practical implementation of photocatalyst materials are the catalyst's stability and reusability (Hao et al., 2021). A photocatalyst's stability is defined as its capacity to maintain the structural, morphological, and chemical properties necessary to sustain its photocatalytic activity for an extended period. A major challenge in the design and development of an efficient photocatalyst for commercial-scale applications is ensuring its long-term stability (Kurniawan et al., 2022a). It is well known that the build-up of partly oxidized organic matter on photocatalyst active surfaces or sites can quickly render the photocatalyst surfaces inactive (Jeong et al., 2013; Pavel et al., 2023). However, long-term exposure to UV light irradiation on the photocatalytic surface in the absence of pollutants may help its regeneration (Jeong et al., 2013). Due to the inadequate hydrophilicity of TiO_2 photocatalyst, oily pollutants exposed to its surface are typically difficult to be removed using water. The synthesis processes, calcination temperature, defects on the catalyst surface, and other factors play a critical role in the stability of photocatalysts (Kongsuebchart et al., 2006; Li et al., 2019, 2021).

For example, Koli et al. (2020), synthesized a boron-doped TiO_2-CNT nanocomposite photocatalyst and studied the regeneration of the photocatalyst for the photodegradation of toluene. They reported that the photodegradation performance was only reduced by 5% after four cycles of degradation. This reduction was attributed to the loss of the photocatalyst due to transfer from one place to another during regeneration. Several methods such as surface modification and heterojunction formation between various semiconductors have been developed for the fabrication of novel photocatalytic materials to prevent deactivation. However, before they can be used, these high-performance photocatalysts need to be further enhanced due to their high cost and challenging synthesis conditions (Hao et al., 2021).

A photocatalyst's capacity to be used repeatedly is referred to as its reusability. Environmental applications encounter a significant challenge due to the loss of photocatalytic activity over repeated reuse processes (Jeong et al., 2013; Pavel et al., 2023). The photocatalyst's stability and reusability determine the cost and the treatment efficiency of photocatalytic processes (Karim et al., 2022). The catalyst's ability to be reused may be hampered by the loss of photoactive sites, reduction in surface area, irregular morphology, and poor crystallinity (Ahmed et al., 2017). The ability to reuse photocatalysts without losing their photocatalytic activity is a promising development for organic pollutant degradation in OPW for pilot and commercial scale applications. To resolve the challenge of photocatalyst deactivation, Chen et al. (2020) designed for the first time a novel fixed bed dual-stage circulating TiO_2-based photocatalytic reactor for the degradation of toluene. This

design allowed the reactor to recycle and regenerate inactive photocatalysts during the degradation process.

9. Future research perspective on photocatalytic technology for OPW

Based on the literature review, the routes of intermediates formed during photocatalytic oxidation, as well as the biodegradability and toxicity of treated OPW are not known (Samuel et al., 2022b; Lin et al., 2020). Hence, future research may focus on (i) investigating the behaviors of the basic components in OPW during photodegradation; (ii) studying the OPW's matrix effects on catalysts (both immobilized and suspended) to fill knowledge gaps in future works to enhance current industrial applications; (iii) evaluating and optimizing novel techniques that can incorporate UV-light-emitted diode (UV-LED)/photocatalyst, (photocatalysis/ozonation) treatment process for OPW treatment, where the OPW is complex; and (iv) determining the photodegradability and toxicity of the treated OPW by examining the intermediates produced during integrated photocatalytic treatment (Kurniawan et al., 2022b). Overall, heterogeneous photocatalysis is emerging as the most promising method for the treatment of wastewater, which could degrade a wide range of pollutants indiscriminately in water and wastewater within a short period.

10. Conclusions

With the increasing generation and discharge of OPW, global environmental concerns have been raised about its toxicity to the marine ecosystem. The soluble organics in OPW, particularly PAHs and BTEX, are highly hazardous and potentially carcinogenic, while the existing treatment methods do not efficiently remove the compounds. Photocatalytic oxidation has demonstrated its ability to be a promising technology for addressing the growing demand for OPW treatment. It is considered to be an environmentally friendly process that requires less energy, especially when being used under sunlight or visible light irradiation. This technique can degrade even the smallest oil compounds and remove hydrocarbon compounds in the OPW.

Various studies that investigated the use of photocatalysis for the removal of hydrocarbon compounds have reported good performance of the various photocatalyst in terms of degradation efficiency of target contaminants. However, due to the complex nature of the OPW, few studies have reported that its efficiency could be influenced. It was conclusively evident from a literature survey of 239 published articles (1988–2022) that novel methods such as the use of advanced photocatalysts, low-energy light sources, and combining other AOPs technologies are being studied to potentially increase effectiveness. It was found that heterogenous photocatalysis could be an excellent post-treatment technique in a situation where a particular technique has been used and could not remove target hydrocarbons to a satisfactory level. Therefore, using heterogenous photocatalysis at this stage gives a superior degradation efficiency.

This review has revealed that various factors are responsible for the treatment efficiency of the photocatalytic system. Although there is no available commercial plant for the treatment of OPW using photocatalysis, this can be achieved through proper design and synthesis of advanced photocatalyst and improvement in the design of the photocatalytic reactors, while considering factors such as pH, concentration of target pollutant, light intensity, as well as treatment cost and environmental factors.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2023.137567.

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