

## Recent advances on the treatment of oilfield-produced water by advanced oxidation processes: A review

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### ABSTRACT

Despite the advancements in alternative fuels and energy sources, there continues to be a significant global dependence on oil production and extraction. A substantial volume of oilfield-produced water (OPW) is generated during the production and extraction processes of oil fields. Recurrent OPW treatments encountered significant challenges in addressing this particular type of wastewater. Advanced oxidation processes (AOPs) are regarded as a promising alternative approach for the degradation of recalcitrant organic compounds in the OPW. This review focuses on the characterization of OPW. The treatment of organic pollutants in wastewater using AOPs, such as ozonation, Fenton oxidation-based processes, heterogeneous photocatalysis, and persulfate oxidation, is comprehensively reviewed in terms of their efficiency for pollutant degradation. The primary challenges in this field and the future directions for development are proposed, with the aim of providing a valuable reference for achieving highly effective treatment of OPW.

**Key words:** Fenton oxidation-based processes, heterogeneous photocatalysis, oil field-produced water, ozonation oxidation, persulfate oxidation

### HIGHLIGHTS

- Characteristics of oilfield-produced water were analyzed.
- Treatment of oilfield-produced water using advanced oxidation processes was discussed.
- Recommendation and future research are proposed.

## GRAPHICAL ABSTRACT



## 1. INTRODUCTION

The utilization of oil plays a pivotal role in meeting the global demand for power generation (Scafutto & de Souza Filho 2016). The advancement of global industry has spurred an increasing demand for oil, resulting in extensive extraction of crude oil through artificial means (Jia *et al.* 2019). The majority of oil fields in China are developed through water injection, particularly during the middle and late stages of development. The water content in the produced liquid typically reaches 70–80% at this stage and may even exceed 90%. During the process of oil and gas exploitation, a substantial volume of formation water and injection water is discharged, commonly referred to as oilfield-produced water (OPW). The OPW is a complex and multifaceted mixture. The composition of OPW is typically influenced by geological conditions, formation depth, oil production techniques, crude oil transportation conditions, and dehydration technologies. Direct discharge of untreated produced water (PW) may result in irreversible harm to organism survival (Lu *et al.* 2021), human health, and the ecological environment (Zhou *et al.* 2021). Relevant treatment measures should therefore be implemented to ensure the proper management of substance discharge or disposal, thereby effectively mitigating potential environmental impacts.

The petroleum industry has yet to fully explore advanced oxidation processes (AOPs), which are widely recognized as environmentally friendly technologies capable of minimizing the generation of secondary waste (Hasan *et al.* 2011). The exceptional reactivity of AOP technology enables effective interaction with a diverse range of organic compounds through the generation of hydroxyl radicals ( $HO\cdot$ ) and other reactive species during the process. These processes facilitate the mineralization of recalcitrant organic compounds into carbon dioxide and water or at least their degradation into innocuous substances (Ferrarese *et al.* 2008; Boczkaj & Fernandes 2017). The application of AOPs, such as ozonation, Fenton oxidation-based processes, heterogeneous photocatalysis, and persulfate (PS) oxidation, has demonstrated significant efficacy in the removal of soluble compounds in OPW (Scott *et al.* 2008; Afzal *et al.* 2015; Leshuk *et al.* 2016; Liu *et al.* 2016c; Jiménez *et al.* 2019; Fang *et al.* 2020). Liu *et al.* (2021) employed the ozonation process for the treatment of OPW. They found that the ozonation system significantly augmented the rates of oxidation and effectively eliminated the polycyclic aromatic hydrocarbons (PAHs) within 30 min. Finally, the sequential oxidation of the OPW matrix by ozonation is highly recommended. Mario *et al.* (2018) studied the degradation of high hydrophobic organics in saline-PW through photocatalysis using reduced graphene oxide (rGO)– $TiO_2$  nanocomposites. The photocatalytic degradation rates exhibit the following increasing order: acetic acid < phenols < naphthalene < xylenes < toluene. However, the degradation of acetic acid is almost not degraded under the same experimental conditions. The findings of this study can contribute to the exploration of novel perspectives in designing high-performance photocatalysts based on graphene-like  $TiO_2$  for the efficient removal of hydrophobic bio-recalcitrant pollutants from saline water. Dong *et al.* (2024) investigated the utilization of  $Cu^0$  as a catalyst

to activate PS for the degradation of hydroxypropyl guar gum (HPG) in oilfield water. They found that the degradation rate of HPG reached up to 81% at a temperature of 45 °C, pH value of 7, oxidant ratio of 2:3 ( $\text{H}_2\text{O}_2:\text{Na}_2\text{S}_2\text{O}_8$ ), total oxidant dosage of 10%, and  $\text{Cu}^0$  dosage of 0.3 g/L. The results demonstrated that the combined degradation of HPG pollutants by  $\text{Cu}^0$ -activated  $\text{H}_2\text{O}_2/\text{Na}_2\text{S}_2\text{O}_8$  exhibits a broad spectrum of applications, offering novel strategies and mechanistic insights for the advanced oxidation of oilfield pollutants.

The aim of this review is to explore the treatment and removal of dissolved organic matter in OPW through AOPs (including ozonation, Fenton oxidation-based methods, heterogeneous photocatalysis, and oxidation) as documented in scientific literature. The mechanism of oxidation was discussed. Finally, the practical application of these processes in real-world scenarios and the potential challenges that may arise are taken into consideration.

## 2. OPW CHARACTERISTICS

The composition of OPW consists of a combination of particulate matter as well as dissolved organic and inorganic compounds. Its physicochemical properties vary with the production region and the predominant hydrocarbon types present in the crude oil. The variability of OPW is influenced by factors such as geographical location, oil and gas composition, reservoir characteristics, and history of water injection. Table 1 summarizes the main components of OPW. The key components in OPW can be broadly categorized into the following groups:

### 2.1. Dissolved organic compounds

The natural organic matter in OPW is closely associated with the composition of crude oil, as it originates from the process of crude oil extraction. It contains a rich variety of dissolved organic compounds including PAHs, organic acids, BTEX (benzene, toluene, ethylbenzene, and xylenes), and phenol in the OPW (Binet *et al.* 2011). These aromatic compounds have been classified as high-priority chemicals (Neff *et al.* 2011a; WHO 2019). They are defined as polyaromatic hydrocarbons with two or more fused aromatic rings that exhibit resistance to removal from OPW. The presence of BTEX can be detected in OPW water from various sources. Table 1 illustrates PAH concentrations ranging from 0.04 to approximately 3 mg/L, BTEX concentrations ranging from 0.73 to about 24.1 mg/L, and phenol concentrations ranging from 0.4 to around 23 mg/L present in OPW.

### 2.2. Dissolved inorganic compounds

The reservoir's significant burial depth results in OPW being characterized by a high concentration of inorganic ions, typically ranging from 1,000 to 40,000 mg/L. The types and concentrations of these ions vary considerably under different geological conditions. Inorganic ions commonly found in OPW include  $\text{Fe}^{2+}$ ,  $\text{Na}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Ba}^{2+}$ ,  $\text{Sr}^+$ ,  $\text{Al}^{3+}$ ,  $\text{Li}^+$ ,  $\text{Cl}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{OH}^-$ , and  $\text{GN}^-$  (Neff *et al.* 2011a). Among them, the abundance of inorganic anions follows the order:  $\text{Cl}^- > \text{SO}_4^{2-} > \text{Br}^- > \text{HCO}_3^- > \text{I}^-$ . Similarly, the abundance of inorganic cations obeys the order:  $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ . These compounds significantly enhance conductivity, salinity, and the scale formation (Jiménez *et al.* 2018). In addition, OPW may contain a range of heavy metals such as mercury, lead, cadmium, chromium, zinc, and nickel (Igunnu 2014; Dwyer & McDonald 2016). The composition and concentration of toxic metals present can vary across different oilfields due to factors such as well longevity and geological formation (Gregory *et al.* 2011).

### 2.3. Oil and grease

Oil and grease, whether emulsified, dispersed, or free, are of utmost concern as OPW components in both onshore and offshore fields. They have emerged as a pivotal control parameter for legislators (Islam *et al.* 2013). The concentration of oil and grease ranges from 2 to 654 mg/L.

### 2.4. Produced solids

The oil production wastewater contains two types of solids: natural and artificial. Natural solids, such as plankton, silt, algae, clay, sediment, and sand, are inherent reservoir materials that exhibit upward mobility toward the surface. Artificial solids introduced into the system through external interventions include waxes, proppants, corrosive debris, gravel pack particles, drilling mud, and cement fine particles. The presence of these solids leads to obstructions in the wellbore, resulting in reduced flow rates and subsequent production decline (Amakiri *et al.* 2022).

**Table 1** | Characteristics and composition of OPW

Constituent	Concentration range	Reference
Phenol (mg/L)	0.4–23	Utvik & Hasle (2015); Jiménez <i>et al.</i> (2017)
PAHs (mg/L)	0.04–3	Utvik & Hasle (2015); Jiménez <i>et al.</i> (2017)
Total BTEX (mg/L)	0.73–24.1	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Benzene (mg/L)	0.032–14.97	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Toluene (mg/L)	0.058–5.86	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Ethylbenzene (mg/L)	0.086–0.57	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Naphthalene (mg/L)	0.194–0.841	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Organic acids (mg/L)	0.001–1 × 10 <sup>4</sup>	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Aliphatic hydrocarbons (mg/L)	17–30	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Ketones (mg/L)	1–2	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Xylene (mg/L)	0.01–1.29	Dórea <i>et al.</i> (2007); Neff <i>et al.</i> (2011b)
Potassium (mg/L)	44–2162	Tibbetts <i>et al.</i> (1992); Janson <i>et al.</i> (2015)
S <sup>2-</sup> (mg/L)	828	Janson <i>et al.</i> (2015)
HCO <sub>3</sub> <sup>-</sup> (mg/L)	0–1.5 × 10 <sup>4</sup>	Benko & Drewes (2008); Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020); Abou <i>et al.</i> (2021)
Na (mg/L)	0–1.5 × 10 <sup>4</sup>	Akmakce <i>et al.</i> (2008); Benko & Drewes (2008); Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Cl (mg/L)	0–2.5 × 10 <sup>4</sup>	Akmakce <i>et al.</i> (2008); Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020); Abou <i>et al.</i> (2021)
Ba (mg/L)	0–850	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Sr (mg/L)	0–6250	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
SO <sub>4</sub> <sup>2-</sup> (mg/L)	0–1.5 × 10 <sup>4</sup>	Tibbetts <i>et al.</i> (1992); Benko & Drewes (2008); Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Ca <sup>2+</sup> (mg/L)	0–7.4 × 10 <sup>4</sup>	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
K <sup>+</sup> (mg/L)	24–4300	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
SO <sub>3</sub> <sup>2-</sup> (mg/L)	10	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
NO <sub>3</sub> <sup>-</sup> (mg/L)	2.15–9.492	Veil <i>et al.</i> (2005); Abou <i>et al.</i> (2021)
S <sub>2</sub> O <sub>3</sub> <sup>2-</sup> (mg/L)	14	Janson <i>et al.</i> (2015)
Mg (mg/L)	4.7943–12341	Tibbetts <i>et al.</i> (1992); Udeagbara <i>et al.</i> (2021)
Fe (mg/L)	0.1–100	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Al (mg/L)	310–410	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
B (mg/L)	5–95	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Cr (mg/L)	0.02–1.1	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Li (mg/L)	3–50	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Mn (mg/L)	0.004–175	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Ti (mg/L)	0.01–0.7	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Zn (mg/L)	0.01–35	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
As (mg/L)	0.005–0.3	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Pd (mg/L)	0.008–0.88	Fakhru’L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Br <sup>-</sup> (mg/L)	51	Janson <i>et al.</i> (2015)
C <sub>2</sub> H <sub>3</sub> O <sub>2</sub> <sup>-</sup> (mg/L)	347	Janson <i>et al.</i> (2015)
NH <sub>4</sub> <sup>+</sup> (mg/L)	11–14.54	Janson <i>et al.</i> (2015); Abou <i>et al.</i> (2021)

(Continued.)

**Table 1** | Continued

Constituent	Concentration range	Reference
Oil and grease (mg/L)	2–654	Tibbetts <i>et al.</i> (1992); Benko & Drewes (2008); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Corrosion inhibitor	0.3–10	Igunnu & Chen (2014)
Scale inhibitor	0.2–30	Igunnu & Chen (2014)
Glycol (mg/L)	7.7–2000	Igunnu & Chen (2014)
Ammonia (mg/L)	9.66–74	Veil <i>et al.</i> (2005); Abou <i>et al.</i> (2021)
Total phosphorous (mg/L)	0.71	Veil <i>et al.</i> (2005)
TOC (mg/L)	491–1700	Veil <i>et al.</i> (2005); Benko & Drewes (2008)
BOD (mg/L)	750–957	Tibbetts <i>et al.</i> (1992); Veil <i>et al.</i> (2005)
COD (mg/L)	1220–1910	Veil <i>et al.</i> (2005); Akmakce <i>et al.</i> (2008)
Salinity (mg/L)	$5 \times 10^3$ – $3 \times 10^8$	Fakhru'L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)
Conductivity ( $\mu$ S/cm)	7200–87542	Janson <i>et al.</i> (2015); Abou <i>et al.</i> (2021)
pH	4.3–7.5	Veil <i>et al.</i> (2005); Janson <i>et al.</i> (2015)
Total suspended solids (mg/L)	1.2–7820	Benko & Drewes (2008); Fakhru'L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020); Abou <i>et al.</i> (2021)
TDS (mg/L)	$1 \times 10^2$ – $4 \times 10^5$	Fakhru'L-Razi <i>et al.</i> (2014); Jiménez <i>et al.</i> (2018); Kabyl <i>et al.</i> (2020)

## 2.5. Treatment chemicals

To ensure the stable production of oil fields, it is imperative to incorporate specific chemical additives during the reproduction process. These chemicals, encompassing gas treatment agents, oil displacement agents, biocides, corrosion inhibitors, coagulants and flocculants, scale inhibitors, emulsion breakers, wax removal agents, defoaming agents and demulsifiers, and so on, effectively mitigate operational issues encountered in the oil and gas production such as pipe corrosion, scaling, wax accumulation in production tubing and enhanced oil recovery (Al-Ghouti *et al.* 2019; Pedro Da Silva Almeida 2019). However, the addition of chemicals may potentially introduce undesirable side effects or potentiate the toxicity of OPW (Beyer *et al.* 2020).

## 2.6. Dissolved gases

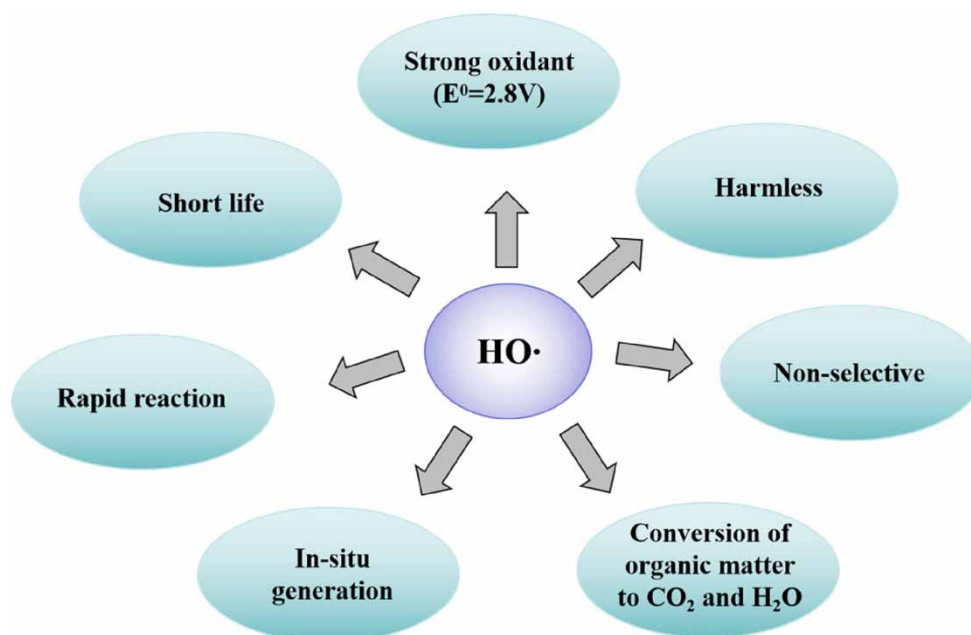
The OPW solution is characterized by a high concentration of dissolved gases, including natural gas components (methane, ethane, propane, and butane), carbon dioxide, nitrogen, oxygen, and hydrogen sulfide. The presence of significant quantities of these gases can pose a potential hazard due to their toxic and corrosive nature. Notably, hydrogen sulfide has garnered significant attention (Amakiri *et al.* 2022).

## 3. TREATMENT OF OPW BY AOPS

AOPs encompass a wide range of methodologies for generating oxide species, including HO·, superoxide radicals ( $O_2^-$ ), sulfate radicals ( $SO_4^-$ ), and others. The active substances mentioned herein exhibit rapid reactivity towards various organic pollutants, leading to the formation of environmentally friendly by-products, predominantly carbon dioxide ( $CO_2$ ), and water ( $H_2O$ ) (Glaze *et al.* 1987; Boczkaj & Fernandes 2017). HO· is a common oxidizing substance in all AOPs and possesses the highest standard oxidation potential compared to other oxidants ( $E^0 = 2.8$  V) (Pignatello *et al.* 2006). Its characteristics are illustrated in Figure 1.

AOPs represent an oxidation technology that exhibits the capability of achieving the complete mineralization of a wide range of organic pollutants. The fundamental steps involve (1) generation of potent oxidants, such as HO·; (2) reaction between these oxidants and organic pollutants in wastewater, leading to their conversion into biodegradable compounds; (3) complete mineralization of the intermediate products from the second step into water, carbon dioxide, and inorganic salts. The utilization of AOPs for the removal of organic pollutants offers several advantages, including rapid reaction rates, effective treatment at low pollutant concentrations, absence of sludge formation, and potential for complete





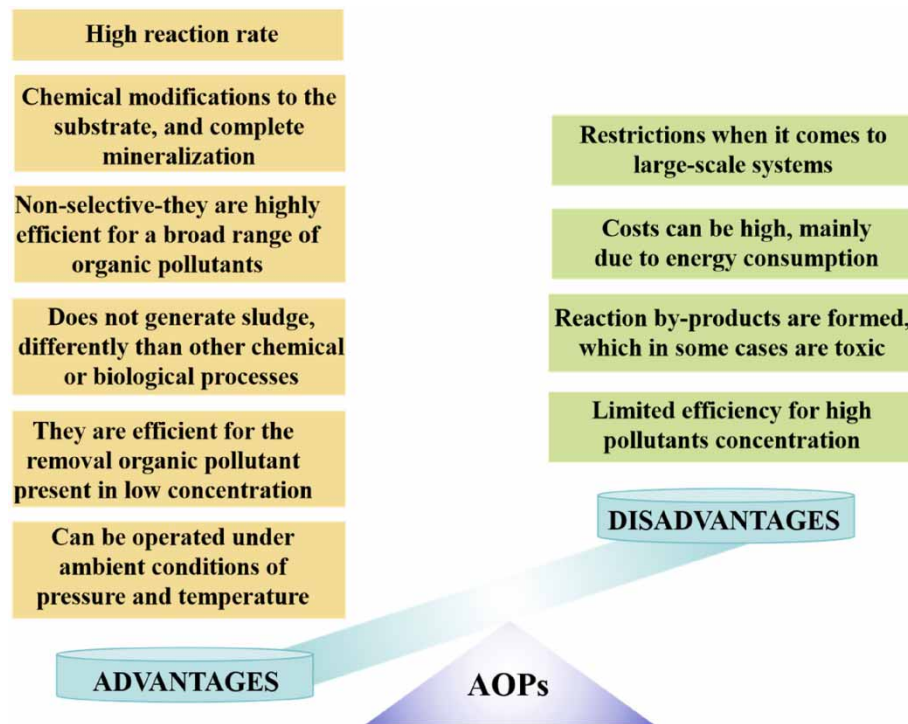
**Figure 1** | Characteristics of hydroxyl radical.

mineralization. Nonetheless, further research is needed to address existing technical challenges associated with these processes. Figure 2 provides a summary of the advantages and disadvantages associated with the application of AOPs for aqueous wastewater treatment.

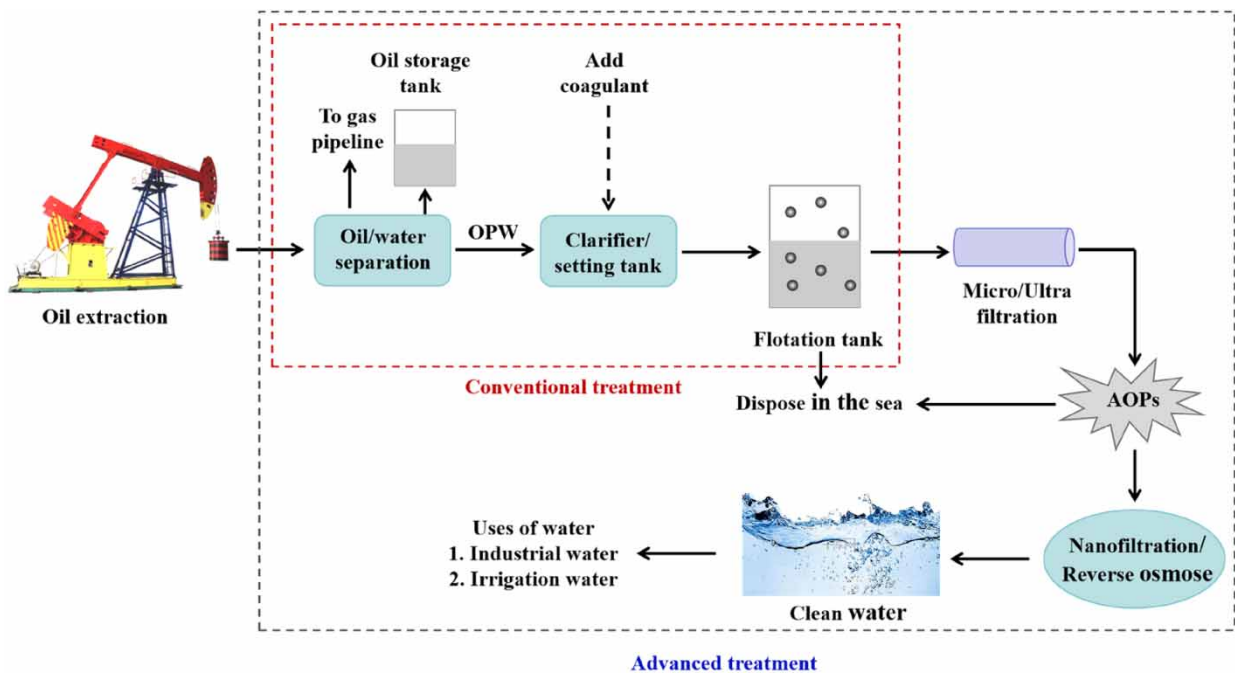
The primary processes of AOPs can be classified into electrochemical, ozone, UV, catalytic processes, and physical AOPs (Miklos *et al.* 2018). However, these classifications are not rigid as each technique may involve multiple processes. Extensive research has been conducted on these processes for the removal of organic compounds generated by petroleum activities. This article focuses specifically on ozonation, Fenton oxidation-based processes, heterogeneous photocatalysis, and PS oxidation (Scott *et al.* 2008; Garcia-Garcia *et al.* 2011; Wang *et al.* 2013; Chaplin 2014; Islam *et al.* 2014; Liu *et al.* 2016c). Figure 3 presents a general flow diagram illustrating the treatment of OPW using AOPs. The conventional treatment process primarily aims at removing solid particles and separating dispersed oil. In addition, flotation can remove remaining dispersed oil and some dissolved compost. AOPs facilitate complete mineralization of recalcitrant pollutants such as naphthenic acid. Consequently, the treated OPW can therefore meet the maximum value specified by current regulations and be discharged directly into the sea.

### 3.1. Ozonation oxidation

Ozone is a widely used potent oxidizing agent, and its initial application in water treatment primarily focused on disinfection by eliminating bacteria in wastewater and eliminating chroma. Ozonation technology has been extensively employed for the removal of organic matter from wastewater. In comparison to traditional treatment methods, ozonation technology offers several advantages: (1) The AOP does not involve biological reactions, making it a relatively straightforward physicochemical process that could be easily automated and commonly utilized for pretreatment or subsequent advanced treatment (Herndandez-Leal *et al.* 2011). (2) It exhibits rapid reaction rates with most pollutants within the range of rate constants between  $10^6$  and  $10^9 \text{ M}^{-1}\cdot\text{s}^{-1}$  (Yang *et al.* 2021). (3) Ozonation reactions typically require milder operating conditions, resulting in reduced energy consumption. (4) The hydroxyl radicals play a crucial role as intermediate products and reactants, while also serving as initiators for subsequent chain reactions. This makes them highly suitable for the treatment of recalcitrant pollutants that are difficult to degrade using conventional methods. (5) Hydroxyl radicals exhibit nonselective reactivity toward most pollutants and intermediates present in wastewater, leading to their degradation into smaller molecular products such as  $\text{H}_2\text{O}$  and  $\text{CO}_2$ . This significantly enhances the degradation rate of COD in wastewater.

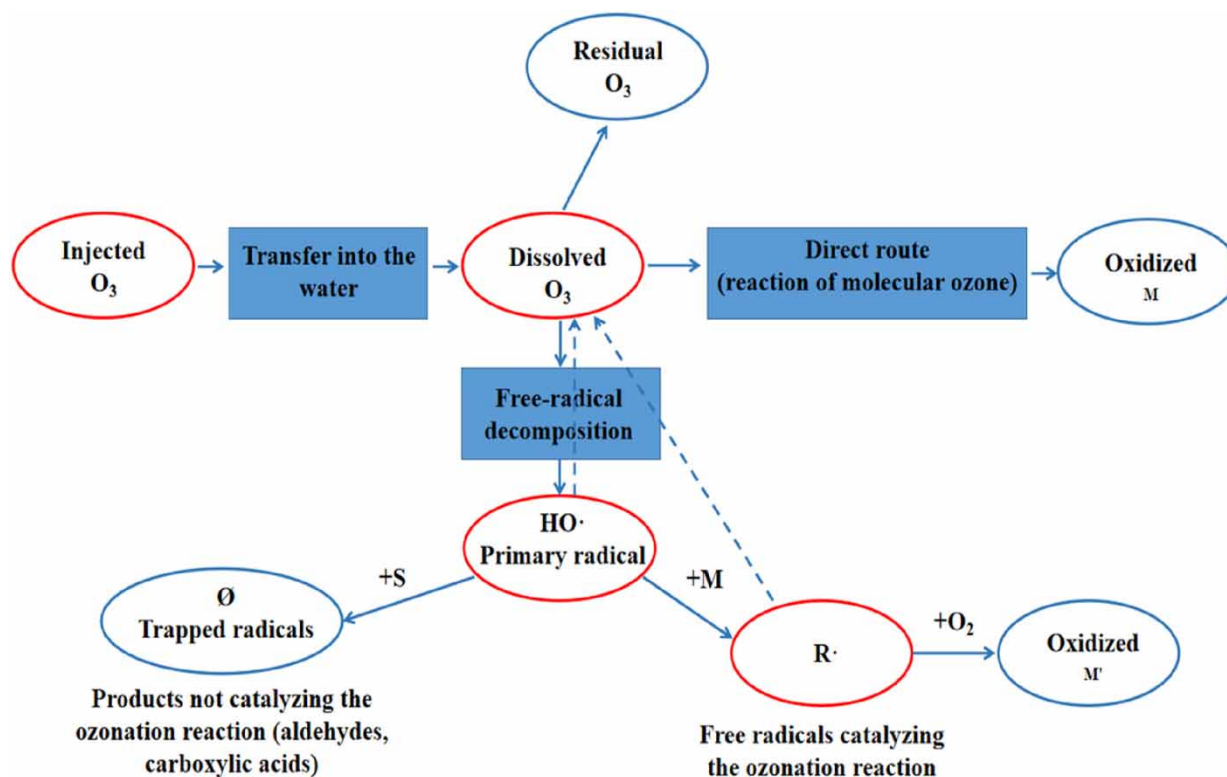


**Figure 2** | Schematic of the advantages and disadvantages of AOPs (Gogate & Pandit 2004; Pignatello *et al.* 2006).



**Figure 3** | Schematic diagram of treatment of oilfield-produced water (Olajire 2020).

The reaction of ozone with organic matter occurs through two distinct pathways, namely, indirect and direct reactions (Kurniawan *et al.* 2006). Figure 4 presents an overview of the complex interactions between the indirect pathway (involving HO· radical reaction) and the direct pathway (involving direct reaction of ozone molecules). These reaction routes could be



**Figure 4** | Ozonation reaction in water during oxidation of a pollutant M.

influenced by various experimental parameters, including solution pH, rate of reaction between molecular ozone, substrate, and solute, presence of substances promoting HO· formation, as well as the nature and concentration of chemicals present (Mahmoud & Freire 2007). The decomposition of ozone in the aqueous solution into free radicals is more pronounced at higher pH values, and the process could be described as follows (Equations (1)–(7)):



The efficiency of contaminant removal through ozonation is marginally inferior compared to other AOPs. Enhancing the efficacy of organic matter elimination becomes imperative. The degradation of pollutants in OPW treatment using ozone as an oxidizing agent generally demonstrates a slightly lower performance compared to other reported AOPs, as indicated by the existing literature (Coha *et al.* 2021).

Messele *et al.* (2020) employed carbon-based metal-free materials as catalysts for the ozonation of 1,3-adamantanedicarboxylic acid (ADA). They observed a remarkable removal efficiency of 65% for ADA in the catalytic ozonation process, which was nearly twice as high as the 33% achieved through single ozonation. In addition, the first-order rate constant for the catalytic ozonation reaction ( $0.0631 \text{ min}^{-1}$ ) was found to be double that of the ozonation reaction alone



(0.0258 min<sup>-1</sup>). The degradation mechanism of ADA was determined to be governed by hydroxyl radicals. These findings highlighted the exceptional efficiency and promising potential catalytic ozonation as an approach for environmental treatment.

The study conducted by Jiménez *et al.* (2019) employed ozonation as a treatment method for PW. They found that all the organic components were removed, including a high percentage of acetic acid, in the ozonation combined with the H<sub>2</sub>O<sub>2</sub> system. The recalcitrant nature of acetic acid in PW was well recognized. Under the conditions of an O<sub>3</sub> dosage of 4 g/L, H<sub>2</sub>O<sub>2</sub> concentration of 1,500 mg/L, pH value at 10, and a reaction time of 2 h, removal efficiencies for total organic carbon (TOC) and acetic acid were found to be 74 and 78%, respectively. In addition, in terms of TOC, ozonation system had the lowest energy consumption per target compound.

The study conducted by Vaiopoulou *et al.* (2015) demonstrated that the ozonation of a mixture of naphthenic acids (NAs) followed pseudo-first-order kinetics. In this investigation, ozone exhibited a preferential reactivity toward NAs with high cyclic and molecular weight structures. The reactivity of ozone toward acyclic/unicyclic model NAs was found to be dependent on their molecular structure. Notably, the introduction of branching, tertiary, and quaternary carbons in NAs resulted in reduced ozone reactivity. By combining ozonation with biodegradation, 89% removal of NA and a 15-fold reduction in toxicity were achieved. Therefore, the combined treatment method involving ozonation and biodegradation proved to be highly effective.

Wang *et al.* (2016a) reported that ozonation emerged as the most efficacious technique for treating oil sands process-affected water. By utilizing a dosage of 2.0 mmol/L O<sub>3</sub>, ozonation exhibited nearly complete removal of fluorescent aromatics, sulfur-containing naphthenic acids, and classical naphthenic acids. The reaction mechanism of ozonation was found to involve molecular ozone reaction followed by HO· reaction. Qualitative and quantitative confirmation of the removal of aromatics and naphthenic acids was further obtained through the 1H NMR analysis. Furthermore, the ozonation process demonstrated a reduction in acute toxicity toward *Vibrio fischeri* and goldfish primary kidney macrophages.

Scott *et al.* (2008) employed ozonation as a treatment method for precipitation-free oil sands process water. A 50-min ozonation process resulted in the production of nontoxic wastewater and led to an approximate 70% reduction in naphthenic acid concentrations. Following 130 min of ozonation, the residual concentration of naphthenic acids in filtered oil sands process water was measured at 2 mg/L. The TOC remained constant throughout the entire 130-min ozonation process; however, there was a significant decrease of approximately 50% observed in chemical oxygen demand (COD) levels. Gas chromatography-mass spectrometry analysis revealed that ozonation caused a reduction in the proportion of high-molecular-weight naphthenic acids.

### 3.2. Fenton oxidation-based processes

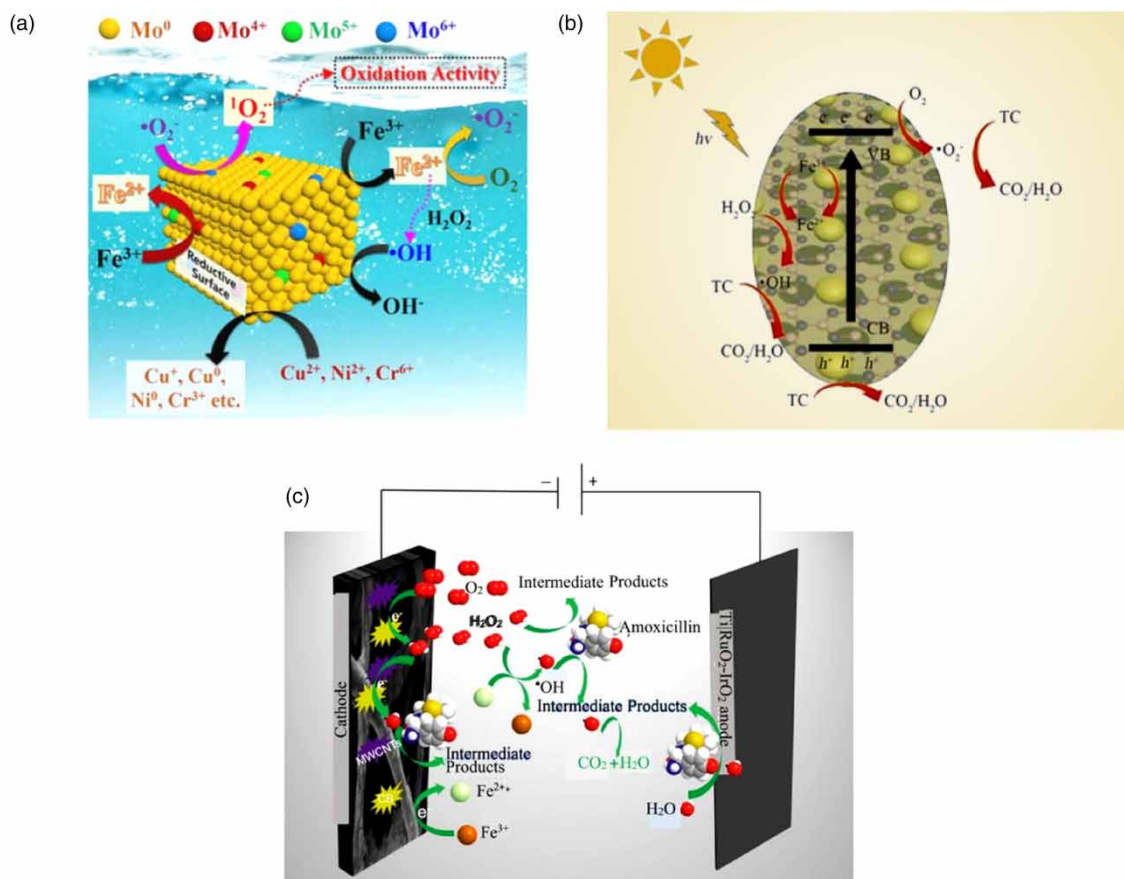
The Fenton process is widely recognized as one of the most prominent AOPs due to its utilization of simple reactants and rapid degradation kinetics for various pollutants. Moreover, it exhibits versatile applicability and demonstrates a remarkable capacity to withstand interference (Wang *et al.* 2016b; Zhang *et al.* 2019).

Fenton reagent comprises a combination of H<sub>2</sub>O<sub>2</sub> and ferrous ions (Fe<sup>2+</sup>). Through the Fenton reaction, Equation (8) produces HO· in the aqueous solution (Lucas & Peres 2006; García-Montoya *et al.* 2015). This reaction was discovered by Fenton (1894) and was later clarified by Haber & Weiss (1934). The reaction mechanism involves the oxidation of ferrous ions to iron ions, while excess H<sub>2</sub>O<sub>2</sub> may undergo further reduction to Fe<sup>2+</sup> (Equation (9)). The Fe<sup>3+</sup> undergoes a reaction with the generated hydroxyl radical, resulting in the production of oxygen (Equation (10)). Furthermore, our observations indicate that Fe<sup>2+</sup> not only acts as a scavenger for free radicals but also facilitates the generation of HO· (Equations (11) and (12)) (Ali *et al.* 2018; Wang & Shizong 2020; Chauhan *et al.* 2021).



The conventional Fenton process, which involves the addition of Fenton reagents to the solution, presents certain limitations in terms of cost and  $\text{H}_2\text{O}_2$  handling risks, sludge formation resulting from excessive iron usage, as well as the necessity for acidification due to its operation at an acidic pH (Malato *et al.* 2009; Moreira *et al.* 2017). To enhance the efficiency of the Fenton process, novel systems such as heterogeneous Fenton-like processes, photo-Fenton utilizing UV-visible radiation, and electro-Fenton employing electrochemical reactions have been developed. Catalytic oxidation refers to a reaction analogous to Fenton's classical reaction, wherein a solid catalyst is employed to expedite the degradation of organic compounds. The iron ions are typically impregnated using natural minerals (such as magnetite, hematite, pyrite) or appropriate structures, resulting in a catalyst with exceptional catalytic activity and stability (Figure 5(a)). The photo-Fenton process involves the synergistic combination of the classical Fenton reaction,  $\text{Fe}^{3+}$  hydroxyl complex, and UV-vis radiation to facilitate the regeneration of  $\text{Fe}^{2+}$  ions. This process generates a mixture of highly reactive  $\text{HO}\cdot$  radicals, which play a crucial role in oxidizing organic compounds (Figure 5(b)) (Khan *et al.* 2013; Silva *et al.* 2015). The Electro-Fenton (EF) process was first described by Brillas *et al.* (2000) and Oturan (2000). The procedure involves the following steps: (i) continuous *in situ* generation of  $\text{H}_2\text{O}_2$  through a carbon cathode under acidic conditions in the presence of oxygen or air; (ii) introduction of a small quantity of  $\text{Fe}^{2+}$  into the solution to facilitate the reaction; (iii) retrieval of  $\text{Fe}^{2+}$  through cathodic reduction to  $\text{Fe}^{3+}$  and subsequent production of  $\text{HO}\cdot$  via Fenton reaction (Brillas *et al.* 2009; Vasudevan & Oturan 2014; Jiao *et al.* 2020). Its mechanism is illustrated in Figure 5(c).

The generation of PW represents the largest wastewater flow in the context of oil and gas exploration. PW is the largest wastewater flow generated during oil and gas exploration. Afzal *et al.* (2018) investigated the application of Fenton oxidation for the degradation of organic matter in PW. The experiments focused on the removal of COD and involved varying concentrations of hydrogen peroxide ( $0.12 \times 10^{-3}$  mol/L to 3 mol/L), molar ratios of  $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$  (2 to 75), and reaction times



**Figure 5** | (a) Possible Fenton-like degradation mechanism (Yi *et al.* 2019); (b) photo-Fenton degradation mechanism (Zhang *et al.* 2022); and (c) electro-Fenton degradation mechanism (Pan *et al.* 2020).

ranging from 30 to 200 min. The Fenton reaction was initiated using ferric sulfate heptahydrate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) and  $\text{H}_2\text{O}_2$ . The enhancement in COD removal was significantly amplified with an increase in the molar ratio of  $[\text{H}_2\text{O}_2]$  to  $[\text{Fe}^{2+}]$ . The removal of COD was observed to increase with an elevation in the concentration of  $\text{H}_2\text{O}_2$  from 0.001 mol/L up to an optimal value of 0.01 mol/L, beyond which the degradation efficiency decreased at higher concentrations of  $\text{H}_2\text{O}_2$ . The COD removal exceeded 90% under the optimal reaction conditions. The Fenton oxidation process exhibited excellent efficacy in the removal of organic matter from PW.

Jin & Davarpanah (2020) demonstrated that photo-Fenton treatment exhibited significant efficacy in the removal of both solids and chemicals from flow-back water. The implementation of chemical-enhanced oil recovery techniques results in a remarkable 75% reduction in water consumption. Moreover, this translates to an annual conservation of approximately  $10^4 \text{ MM m}^3$  of water, which was sufficient to sustain the livelihoods of 1,000 individuals. Furthermore, these methods obviated the necessity for substantial utilization of freshwater resources during hydrocarbon production.

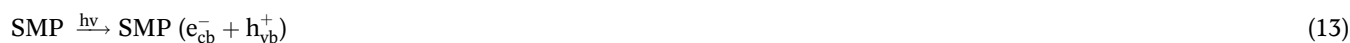
The removal of benzene (B), toluene (T), and p-xylene (X) in aqueous solution was investigated using the EF method under various electrode conditions, as reported by Radwan *et al.* (2019). The complete degradation of BTX can be achieved within 30 min under optimal conditions. The removal efficiencies of TOC were 95.4, 88.7, and 87.6% when sacrificial stainless steel, carbon, and nickel anodes were applied, respectively. The  $\text{Fe}^{2+}$  ions were continuously generated in the stainless steel anode through sacrificial electrode mode in the solution. The cost of EF treatment for one cubic meter of BTX-contaminated water was estimated to be \$1.40 under specific conditions.

### 3.3. Heterogeneous photocatalysis

The polyphase photocatalysis technique, pioneered by Fujishima and Honda in the 1970s, represents a highly promising technology for wastewater treatment aimed at effectively eliminating recalcitrant organic compounds such as PAHs, phenolic compounds, sulfides, and other substances originating from OPW. The process is based on the utilization of metal oxides as catalysts and oxygen as electron acceptors to facilitate the generation of oxidizing species, such as  $\text{HO}\cdot$ ,  $\text{O}_2^-$ , and  $\text{HO}_2\cdot$  (Hasan *et al.* 2011).

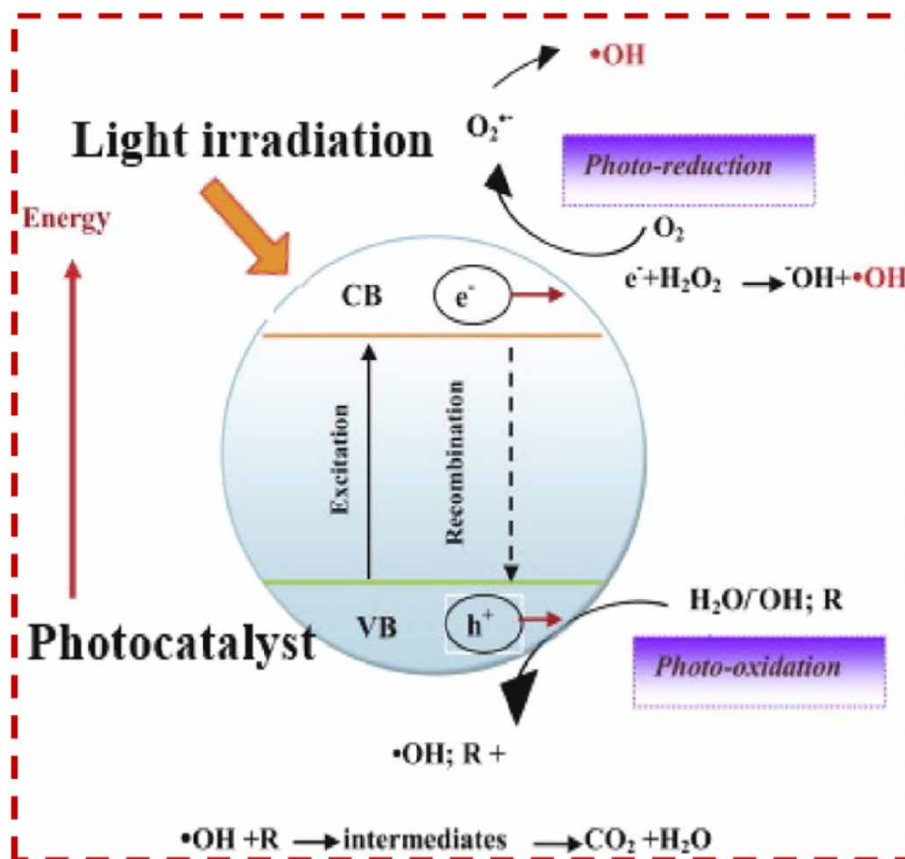
The process of photocatalysis involves the excitation of semiconductor particles (SMP) through irradiation with specific wavelengths. As depicted in Equation (13), upon irradiation of the SMP and subsequent energy absorption, the electrons undergo a transition from the valence band (VB) to the conduction band. The energy required for the movement of electrons across different bands is referred to as the band gap. When this energy is reached and electron transfer occurs, a hole ( $\text{h}^+$ ) is created in VB. This pore serves as an oxidation site, capable of catalyzing chemical reactions (Hoffmann *et al.* 1995; Lasa *et al.* 2005) (Figure 6).

$\text{h}^+$  species can undergo direct reactions with organic molecules or react with  $\text{H}_2\text{O}$  to generate  $\text{HO}\cdot$ , as shown in Equation (14):



The photocatalytic degradation pathway for treating OPW involves a series of sequential steps: (i) diffusion-driven mass transfer of reactants from the bulk fluid phase to the outer surface of the photocatalyst, (ii) internal mass transport of reactants through pores to reach the active sites on the surface where they could be adsorbed, (iii) adsorption of organic pollutants onto the photoactivated surface of the photocatalyst, (iv) desorption of photodegradation products, and (v) internal and external diffusion of the final photodegradation products.

$\gamma\text{-Fe}_2\text{O}_3$  nanoparticles were synthesized by Sheikholeslami *et al.* (2019) using co-precipitation methods for the purpose of photocatalysis. The degradation of BTEX was employed as an indicator for assessing the efficiency of PW treatment. The impact of varying parameters, including pH (3–7), catalyst concentration (0–250 mg/L), UV light intensity (0–100 W), and visible light intensity (0–225 W), was investigated. The results demonstrated that the removal efficiency of BTEX was 95% within a span of 5 days and reached 97% in just 90 min when employing maghemite nanoparticles under both visible and ultraviolet light conditions. In addition, the adsorption (approximately 30%) and photolysis processes (approximately 20%) of maghemite nanoparticles significantly contributed to the removal efficiency observed in the photocatalytic process.



**Figure 6** | Schematic diagram illustrating the principle of photocatalysis (Ahmed *et al.* 2011).

The photocatalytic treatment of naphthalene in real offshore PW was conducted by Liu *et al.* (2016a, 2016b) using UV irradiation and nanoscale  $TiO_2$  powder (aerioxide<sup>®</sup>P25) as the catalyst. Furthermore, it was observed that the presence of bivalent cations induced catalyst agglomeration. Thus, the reduction of active sites on the catalyst surface leads to a decrease in the photocatalytic rate of the analytes. The rate constants of the photocatalytic reaction in the immobilized systems reached  $0.00305 \text{ min}^{-1}$ , surpassing that of the homogeneous system ( $0.00219 \text{ min}^{-1}$ ). The immobilized catalyst exhibited excellent photo-oxidation performance.

Liu *et al.* (2016a, 2016b) employed UV and  $TiO_2$  to investigate the impact of the OPW matrix on the photodegradation of PAHs. The  $TiO_2$  nanocatalyst with an average particle size of approximately 12–17 nm was synthesized using an enhanced sol-gel method. The photocatalytic experiments conducted on degrading PAHs in distilled water samples revealed that a degradation rate is 10–100 times higher than that observed in OPW. The photocatalytic performance was hindered by the complex composition of OPW. The impacts of various operational parameters, namely, salinity, insoluble particulate matter, and organic composition, on the degradation of PAHs were investigated. The removal rates of PAHs with lower molecular weights increased as the ion strength increased. The salinity variation and presence of insoluble matter played a pivotal role in the efficient removal of high-molecular-weight PAHs, primarily due to their limited solubility caused by the ‘salting out’ effect and the competitive adsorption between PAHs, insoluble matter, and  $TiO_2$ . The attenuation of ultraviolet light primarily stems from organic components. The present study offered an opportunity for further investigation into the influence of these organic substances and halogen ions on the efficacy of photocatalytic techniques in treating OPW.

Hayati *et al.* (2018) investigated the catalytic efficiency of a ternary nanocomposite heterojunction catalyst consisting of  $ZnO/TiO_2$  anchored on rGO for the degradation of phenol in aqueous solutions. The effect of various operational conditions, including pH, catalyst dosage, phenol concentration, and light illumination, on the photocatalytic performance was investigated. The experimental findings demonstrated that a complete degradation rate of phenol was achieved at an initial



concentration of 60 mg/L, with a catalyst dosage of 0.6 g/L, light intensity set at 150 W, pH maintained at 4, and a reaction time of 160 min. The introduction of radical scavengers, namely, isopropyl alcohol, p-benzoquinone, and ethylenediaminetetraacetic acid, into the reaction system indicated the presence of hydroxyl radicals for photocatalytic oxidation. The removal ratios of  $\text{TOC}_t/\text{TOC}_0$  and  $\text{COD}_t/\text{COD}_0$  were observed to decrease to 14 and 8%, respectively, after treatment duration of 440 min for the real petrochemical effluent.

### 3.4. PS oxidation

PS oxidation is a highly efficient advanced oxidation technique that employs thermally activated PS, ultraviolet light, and/or acidic pH conditions to generate free radicals for the degradation of various contaminants. The electron transfer from transition metals, such as iron, manganese, and cobalt, along with the potential mechanisms are illustrated by Equations (15)–(19):

Mechanism of PS oxidation (Liang *et al.* 2009):



where R represents an organic compound and Me represents a transition metal.

The oil industry generates significant volumes of PW, which is characterized by the presence of organic pollutants such as NA and high concentrations of inorganic salts. Aher *et al.* (2017) employed membranes functionalized with iron oxide ( $\text{Fe}_x\text{O}_y$  nanoparticles) as catalysts and PS as an oxidizing agent for the oxidation of NA from water containing high concentrations of total dissolved solids (TDS). The results indicated that the concentration of NA decreased by up to 8 mg/L (from 34 mg/L) after the second passage through a  $\text{Fe}_x\text{O}_y$  functionalized membrane with the addition of PS at 40 °C. They reported that this system had demonstrated promising potential as an alternative to NA treatment.

The role of temperature in PS oxidation is of significant importance. The effect of temperature on oxidant consumption was investigated by Ferreira *et al.* (2022), revealing an inverse relationship between temperature and oxidant consumption. The reasons for this phenomenon could be attributed to two factors. First, the presence of sulfate radicals led to the oxidation of organic compounds in solution. Second, an increase in the levels of sulfate anions, protons, and oxygen was observed as the oxidant decomposition progressed (Domínguez *et al.* 2021). The reduction of COD and Biochemical oxygen demand (BOD) was achieved through heat-activated PS oxidation; however, the biodegradability of the PW remained unchanged due to the formation of by-products.

Gholami *et al.* (2021) conducted experiments on the electro-activation of persulfate (EC/PS) for real PW treatment and investigated the influence of operational conditions on COD and ammonia removal. The results obtained indicated that a maximum COD removal efficiency of 71% was observed. The ammonia removal efficiency of 69% was observed at a pH of 10, which can be attributed to the physicochemical adsorption facilitated by air stripping and the thermal activation of PS. The removal of contaminants was predominantly governed by the hydroxyl radical mechanism, accounting for 67% of the overall contribution, while sulfate radicals contributed 33%. Compared to other AOPs, the EC/PS process demonstrated superior cost-effectiveness and efficiency.

Interactions among chemical constituents can induce alterations in the composition of hydraulic fracturing fluids. Manz *et al.* (Manz & Carter 2017) investigated the application of activated PS oxidation for the degradation and transformation of 3-furfuraldehyde (or furfural). The utilization of two chemicals as additives in hydraulic fracturing was prevalent. A study was conducted to investigate the impact of elevated temperatures, varying PS dosing and Fe (III) concentration, initial pH, and the addition of other chemical additives on furfural degradation. The reaction was found to exhibit pseudo-first-order kinetics with respect to the furfural concentration. The inclusion of radical scavengers, methanol, and tert-butyl alcohol in the reaction supports the involvement of a  $\text{HO}\cdot$  mechanism. The addition of iron sulfate to a solution with a pH of 5.4 did not



exert any influence on the activation energy of furfural oxidation, which remained at 107 kJ/mol. However, when the pH was reduced to 2.54, the activation energy decreases to 75 kJ/mol.

#### 4. CONCLUSIONS AND PERSPECTIVES

This article provides a comprehensive review of the application of AOPs for pollutant removal in PW treatment. Specifically, ozonation, Fenton oxidation-based processes, heterogeneous photocatalysis, and PS oxidation are critically examined. However, it should be noted that the utilization of these AOPs for treating OPW still presents significant challenges, and further research is warranted to overcome current barriers. Several potential areas for future investigation include:

- (1) The combined ozonation process is considered as an alternative to address the limitations of conventional ozonation, including low ozone dissolution and slow mass transfer from the gas phase to the liquid phase, which results in high-energy consumption.
- (2) The utilization of photocatalysis in industrial water treatment facilities is limited due to the inherent constraints of heterogeneous photocatalysis, including challenges associated with the separation of colloidal catalysts from the treated solution and mass transfer limitations imposed by fixed catalyst surfaces on substrates.
- (3) Given that OPW exhibits a natural pH in close proximity to 8, which holds significant importance for the oil industry, subsequent research endeavors have been directed toward the development of treatment methods under neutral pH conditions.
- (4) The consideration of the influence exerted by diverse cations and anions present in the PW on the oxidation process is imperative.
- (5) The investigation of diverse water quality systems at various well sites is necessary to provide robust data support, propose effective technical solutions for practical water quality issues, and offer insights into the scalability and commercial viability of this technology.
- (6) The treatment of aromatics compounds in micro concentration is not addressed in previous studies, which is a persistent issue.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

#### CONFLICT OF INTEREST

The authors declare there is no conflict.

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