



## Oilfield-produced water treatment using conventional and membrane-based technologies for beneficial reuse: A critical review

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### ABSTRACT

Oilfield produced water (OPW) is one of the most important by-products, resulting from oil and gas exploration. The water contains a complex mixture of organic and inorganic compounds such as grease, dissolved salt, heavy metals as well as dissolved and dispersed oils, which can be toxic to the environment and public health. This article critically reviews the complex properties of OPW and various technologies for its treatment. They include the physico-chemical treatment process, biological treatment process, and physical treatment process. Their technological strengths and bottlenecks as well as strategies to mitigate their bottlenecks are elaborated. A particular focus is placed on membrane technologies. Finally, further research direction, challenges, and perspectives of treatment technologies for OPW are discussed. It is conclusively evident from 262 published studies (1965–2021) that no single treatment method is highly effective for OPW treatment as a stand-alone process however, conventional membrane-based technologies are frequently used for the treatment of OPW with the ultrafiltration (UF) process being the most used for oil rejection from OPW and oily waste water. After membrane treatment, treated effluents of the OPW could be reused for irrigation, habitat and wildlife watering, microalgae production, and livestock watering. Overall, this implies that target pollutants in the OPW samples could be removed efficiently for subsequent use, despite its complex properties. In general, it is however important to note that feed quality, desired quality of effluent, cost-effectiveness, simplicity of process are key determinants in choosing the most suitable treatment process for OPW treatment.

### 1. Introduction

Water scarcity and its sustainable use have become the world's global challenges, as reflected in the United Nations 2030 Agenda for sustainable development goals (SDG 6 Synthesis Report 2018 on Water and Sanitation, 2018). Therefore, scientists increasingly focus on the treatment of industrial wastewater to increase sources of clean water for safe drinking. With the increasing global demand for energy due to

rising population, fossil-based oil remains the major source of global energy. However, oil exploration brings negative implications on the environment since the exploration activity generates complex oil-produced water (OPW).

This OPW is commonly used to describe wastewater generated as a by-product of crude oil and gas exploration. It accounts for the major waste stream during oil and gas exploration and processing (Al-Ghouti et al., 2019). Global crude oil and gas exploration is estimated at 41

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million m<sup>3</sup>/day (Xu et al., 2018). It is estimated that the production of a barrel of crude oil equivalently produces about three barrels of OPW for a typical reservoir and about nine to ten barrels of OPW for a mature reservoir (Ebrahimi et al., 2018). New reservoir produces more oil and gas than water, however, significantly more OPW is generated as the reservoir becomes more mature, which accounts for the lion share of by-products with oil/gas and water mixture (Alfredo Zendejas et al., 2020).

OPW contains complex mixtures of both inorganic and organic substances (Jiménez et al., 2018a). Its constituents vary with the geographical location of the reservoir, mode of exploration, and age of the reservoir. The major constituent of OPW is oil and grease, which receives attention both in onshore and offshore operations. The OPW also contains salt expressed as salinity, conductivity, or total dissolved solid (TDS), which possesses technical obstacles in onshore operations (Lin et al., 2020). The treatment of OPW involves the de-oiling and de-mineralization process before it is discharged into the environment or before its recycling for subsequent use. Due to the presence of oil, grease, and other complex mixtures in the OPW, an advanced treatment process is required which can completely mineralize these constituents. Unless properly treated before its discharge into the water body, the OPW not only causes serious health implications on the environment and public health but also damages crops during irrigation.

As stated earlier, OPW contains a complex composition depending on various factors. Each oilfield has its specific composition making it difficult to adopt a general technology for the removal of target pollutants present in OPW. This makes OPW difficult to be treated. In most cases, integrated technologies are employed by coupling them together for OPW abatement. In the past, the conventional method for OPW treatment was mainly physical separation between oil-water emulsion via gravity method and then discharged into the environment. This has been shown to have a severe impact on the environment based on the latest standard resulting in serious implications on the environment.

As environmental regulations increasingly become stringent, there is an urgent need to improve the abatement technologies to meet current standards before being discharged into the water body or for reuse. (Witze, 2015). It is expected that before OPW is released into the environment, it should not contain more than 40 ppm of oil-water emulsion. For this purpose, countries have started legislating stringent laws, regulations, and effluent limits for oil and gas companies to comply with before their discharge into the environment (Igunnu and Chen, 2012). Consequently, oil companies all over the world strive to meet “zero-discharge” requirements.

Due to the complex nature of OPW, the available technologies for the treatment of OPW such as adsorption, cyclonic and gravity separators, floatation, coagulation-flocculation, biological treatment, and evaporation are ineffective as a standalone system. They are not only inefficient to separate stabilized oil-water emulsion but also susceptible to fouling (Wei et al., 2019). Nevertheless, the technologies are limited in treating emulsified oil in water with a droplet size of less than 10 µm (Zhu et al., 2014a; Dickhout et al., 2017). To address this limitation, chemical dosage and an external electric field are applied to break down the oil emulsion. However, these two processes are energy-intensive and harmful to the environment (Saad et al., 2019).

To address this bottleneck, membrane filtration has been developed as one of the most promising options for the treatment of OPW due to its ability to meet discharge criteria and cost-effectiveness (Baker, 2004; Mulder, 1996). It has been reported that oils, emulsions, and silts are successfully separated from OPW using polymeric and ceramic membranes (Ebrahimi et al., 2009). Its availability to be integrated into other available technologies make it possible to remove target pollutants in complex OPW.

In addition to its ease of operation, the use of membrane technology for OPW treatment has enhanced the separation of various components in OPW. In terms of purification and separation performance, membrane technology is a better option than the conventional treatment methods

for OPW. It has provided the opportunity not only to use a single treatment unit with little need for pre or post-treatment steps but also provides the opportunity to develop an efficient system of OPW purification for beneficial reuse. Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) are among the membrane-based processes that have been tested. In addition, such membrane-based techniques have been investigated for their technical feasibility to generate treated effluents of OPW that meet the required effluent limit set by local environmental legislation (Macedonio et al., 2014).

Most reviews, on OPW treatment (Dickhout et al., 2017; El-badawy et al., 2022; Fakhru'l-Razi et al., 2009; Igunnu and Chen, 2012; Liu et al., 2021; Munirasu et al., 2016) have mainly focused on the general applications of the various treatment technologies for OPW treatment without emphasis and focus on the real applications of these treated water for beneficial reuse. This review discusses in detail the real application of this treated water for beneficial reuse and presented a qualitative assessment of the various treatment technologies with respect to technical and environmental factors to be considered in the choice of a particular technology for a specific purpose. It is worthy of note that with increase pollution and a corresponding increase in demand for clean water resulting in its scarcity especially in water-stressed regions and also increase in demand for energy due to population increase and increasing industrialization, hydrocarbon sources remains the major source of energy and this is believed to continue for a larger part of this century. In addition, its beneficial reuse after treatment for agricultural and/or livestock as well as other purposes is scarcely reported in the body of knowledge.

To bridge the existing gaps in the body of literature, this article critically reviews the complex properties of OPW and various technologies for its treatment. They include the Physico-chemical treatment process (oxidation, precipitation), biological process (biological aerated filters, activated sludge, etc) physical process (adsorption, filtration, membrane technology). Their technological strengths and bottlenecks as well as strategies to mitigate their bottlenecks are elaborated. A particular focus is placed on membrane technologies which is found to have attracted much attention lately due to the increase in its adoption for OPW treatment. Finally, further research direction, challenges, and perspectives of treatment technologies for OPW are discussed.

## 2. Oilfield produced water (OPW)

Reservoir water, also known as formation water, is usually commonly found with crude oil in the reservoir. This water is highly acidic and located beneath the hydrocarbon in a porous reservoir media (Fig. 1). As oil exploration decreases the pressure within the oil reservoir, the OPW is normally re-injected into the reservoir water bed within

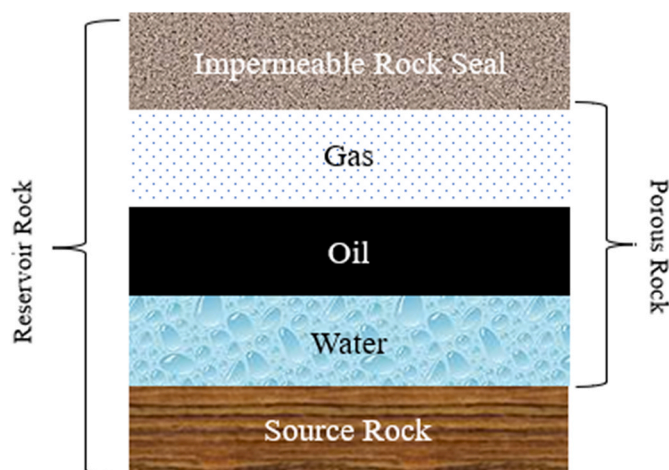


Fig. 1. Schematic diagram of a typical reservoir (Igunnu and Chen, 2012).

the reservoir to improve the recovery of oil from the reservoir (Igunnu and Chen, 2012).

As the exploration takes place, water break outside the reservoir could occur within the reservoir environment in addition to the injection water. As the production continues, a time will reach when natural or reservoir water reaches the well where the production of OPW starts with the hydrocarbon. The OPW accounts for the largest fraction of by-products produced during oil exploration and recovery operations (Alfredo Zendejas et al., 2020). The hydrocarbon mixture, injection water, treating chemicals, and formation water are categorized as OPW (Yousef et al., 2020).

Oilfields constitute over 60% of OPW generated daily worldwide (Hedar and Budiyo, 2018). OPW generation increases as oilfields become mature (Fig. 2). Basic OPW consists of complex mixtures of dissolved gases, which include CO<sub>2</sub> and H<sub>2</sub>S, dispersed and dissolved oil, produced solids, chemicals, and dissolved minerals (Al-Ghouti et al., 2019). The concentration of inorganic and organic compounds varies with the geographical locations of the wells.

## 2.1. Composition of oilfield produced water (OPW)

Although OPW contains a complex mixture, its composition can be categorized into inorganic and organic compounds, which include dispersed and dissolved oils and gases, radionuclides, heavy metals, chemicals, grease, solids, waxes, salts, dissolved oxygen, and microorganisms (Jiménez et al., 2018a). It is estimated that approximately 250 million barrels of OPW are produced daily worldwide resulting from oil exploration. Over 40% is released into the environment (Igunnu and Chen, 2012).

The physico-chemical properties of oil and gas in OPW have significant impacts on its quality. OPW concentrations vary worldwide and their main composition is summarized in Table 1. The Table shows that OPW contains complex constituents, which range from organic, inorganic, and metal constituents with varied concentrations.

### 2.1.1. Dissolved and dispersed hydrocarbon oil components

Dispersed and dissolved oil constituents contain hydrocarbon mixtures, which include polyaromatic hydrocarbons (PAHs), naphthalene, phenanthrene, dibenzothiophene (NPD), benzene, toluene, ethylbenzene, and xylene (BTEX), and phenols. Most oils are dispersed in water as small droplets because all hydrocarbons cannot be dissolved by water (Ekins et al., 2007). The water-soluble organic compounds in the OPW are polar constituents and are referred to as the dissolved oils, while the smaller droplets of oils in the aqueous phase are referred to as dispersed oil (Duraisamy et al., 2013). Phenols, carboxylic acid, aliphatic hydrocarbon, lower weight aromatic hydrocarbons, and BTEX

are categorized as dissolved oil, while heavy alkyl phenols and less soluble PAHs contained in OPW are categorized as dispersed oil as presented (Supplementary Table S1) (Arthur et al., 2011; Veil et al., 2004). The concentrations of dispersed and dissolved oil present in OPW are determined by factors like location of the field, oil density, precipitated oil, and interfacial tension between the water and oil (Liu et al., 2021). Table 2 also provides the concentration of BTEX and phenol in different fields. It shows that the concentrations of these compounds vary from one oilfield to another.

### 2.1.2. Dissolved inorganic minerals

Dissolved inorganic elements/mineral compounds present in high concentrations in OPW are categorized into heavy metals, radioactive materials occurring naturally, cations and anions. The presence of anions and cations in OPW has significant effects on the OPW's physico-chemical properties. Anions such as Cl<sup>-</sup>, CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, and SO<sub>4</sub><sup>2-</sup> are responsible for the buffering capacity of OPW and have effects on the conductivity and scale formation while Cl<sup>-</sup> and Na<sup>+</sup> account for the salinity of OPW as high as approximately 300,000 mg/L (Fakhru'l-Razi et al., 2009). Heavy metals such as Cd, Cr, Cu, Pb, Hg, Ni, Ag, and Zn are present in OPW in trace quantities, the concentrations of which depend on the geographical location and how mature is the well (Hansen and Davies, 1994; Fakhru'l-Razi et al., 2009). The major natural radioactive elements in OPW are <sup>238</sup>Ra and <sup>236</sup>Ra (Utik 2003). The radioactivity level in OPW results from co-precipitation between barium sulfate (scale) or other types of scale and radium. The amount of barium present in OPW can indicate the presence of radium isotope. Radium has been detected up to about 21Bq/l in OPW samples from oilfields (Fakhru'l-Razi et al., 2009). Table 2 provides the composition of dissolved inorganic compounds in different oilfields showing the variation of composition from one oilfield to another in different countries. It can also be seen that heavy metal concentrations also vary from one oilfield to another. The values of concentrations presented in Table 2 show the minimum, maximum, and average values of concentration of the various compounds present in OPW from one field to another.

### 2.1.3. Oilfield production chemicals, polymer surfactants, dissolved gases, and produced solids

During exploration polymer surfactants are added to the oil well to avoid operational problems during oil exploration and to aid the displacement and recovery of oil. The OPW generated also contains various gases and produced solids that accompany this oil during the exploration process. Various chemicals are added as active ingredients after being dissolved in a solvent to be used as a corrosion inhibitor and to prevent hydrate, scale formation, and deposition (Arthur et al., 2011). It is also used for preventing foam production, wax deposition, bacterial growth, emulsion breaking, and gas hydrate formation to increase the oil-water separation efficiency (Hansen and Davies, 1994). Active ingredients such as linear alkylbenzene sulfonate (LAS), 2-alkyl-ethylamine-2-imidazolines are used in the North Sea oilfields and the concentration of this chemical can be as low as 0.1 ppm in the OPW (Veil et al., 2004). The main dissolved gases found in the OPW are H<sub>2</sub>S, CO<sub>2</sub>, and O<sub>2</sub> (Wallace, 2021). They are naturally produced by the chemical reaction and bacterial activities in the OPW. Supplementary Table S2 shows the range of chemicals found in OPW. Produced solids include corrosion products, clays, bacteria, waxes, asphaltenes, sand, silt, scale products, suspended and precipitated solids (Fillo et al., 1992). They possess serious challenges during oil exploration such as oily sludge, emulsion, common scales, and bacterial clogging. Their concentrations vary from one field to another (Neff et al., 2006). The schematic classification diagram of OPW's composition is presented in Supplementary Fig. S1.

## 2.2. Environmental footprint of oilfield produced water (OPW)

Environmental footprints of OPW result in negative implications on

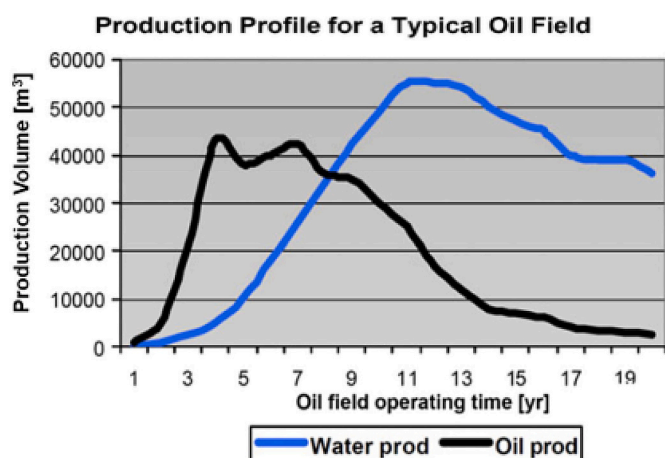


Fig. 2. Typical production profile for an oilfield (Igunnu and Chen, 2012).

**Table 1**  
Range of Organic compounds and Heavy metal Composition of OPW.

Organic compounds composition			Heavy Metal composition		
Parameters	Concentration (mg/l) (Min – Max)	Reference	Parameters (Metals)	Concentration (mg/l) (Min – Max)	Reference
COD	1220–2600	(Ekins et al., 2007; Tibbetts et al., 1992)	Na	0–150000	(Khan et al., 2016; Tibbetts et al., 1992)
TDS	100–400000	(Dolan et al., 2018; Fakhru'l-Razi et al., 2009)	Cr	0.002–1.1	(Cluff et al., 2014; Warner et al., 2014)
TOC	0–1500	(Igunnu and Chen, 2012; Neff et al., 2011b)	Cl <sup>-</sup>	0–270000	(Fillo et al., 1992; Warner et al., 2014)
Total organic acids	0.001–10000	(Igunnu and Chen, 2012; Neff et al., 2011b)	Sr	0–6250	(Fillo et al., 1992; Khan et al., 2016)
TSS	1.2–1000	(Fakhru'l-Razi et al., 2009; Ozgun et al., 2013)	Mn	0.004–175	(Alley et al., 2011; Tibbetts et al., 1992)
BTEX			Li	0.038–64	(Alley et al., 2011; Khan et al., 2016)
Benzene	0.032–778.51	(Dórea et al., 2007; Khan et al., 2016)	Ba	0–850	(Fillo et al., 1992; Tibbetts et al., 1992)
Ethylbenzene	0.026–399.84	(Dórea et al., 2007; Neff et al., 2011b)	Zn	0.01–35	(Alley et al., 2011; Cluff et al., 2014)
Toluene	0.058–5.86	(Dórea et al., 2007; Neff et al., 2011b)	K	24–4300	(Alley et al., 2011; Tibbetts et al., 1992)
Xylene	0.01–1.29	(Dórea et al., 2007; Neff et al., 2011b)	As	0.002–11	(Ekins et al., 2007; Gregory et al., 2011)
Production treatment chemicals			Fe	1.1–1100	(Cluff et al., 2014; Tibbetts et al., 1992)
Corrosion inhibitor	0.3–10	Igunnu and Chen, 2012	Ti	0.01–0.7	(Cluff et al., 2014; Gregory et al., 2011)
Glycol	7.7–2000	Igunnu and Chen, 2012	Al	0.4–410	(Alley et al., 2011; Khan et al., 2016)
Scale inhibitor	0.2–30	Igunnu and Chen, 2012	Pd	0.008–0.88	(Ekins et al., 2007; Tibbetts et al., 1992)
Total BTEX	0.73–24.1	(Orem et al., 2014; Ozgun et al., 2013)	Other ions		
Other pollutants			HCO <sub>3</sub> <sup>-</sup>	0–15000	(Cluff et al., 2014; Gregory et al., 2011)
Phenol	0.001–10000	(Ekins et al., 2007; Tibbetts et al., 1992)	B	5–95	(Fillo et al., 1992; Khan et al., 2016)
Saturated hydrocarbons	17–30	(Neff et al., 2011a)	Mg <sup>2+</sup>	0.9–6000	(Alley et al., 2011; Cluff et al., 2014)
Total oil and grease	2–560	(Fakhru'l-Razi et al., 2009; Ozgun et al., 2013)	Ca <sup>2+</sup>	0–74000	(Alley et al., 2011; Tibbetts et al., 1992)
			SO <sub>4</sub> <sup>2-</sup>	0–15000	(Fillo et al., 1992; McIntosh and Walter, 2005)

the environment due to its improper disposal. The BTEX group has a variety of acute and chronic side effects on public health such as skin diseases, neurological issues, aplastic anemia, leukemia, poisoning, inhalation injuries, headaches, and others (Rahman et al., 2018). Due to their high volatility and quick breakdown in seawater (Neff et al., 2011a; Neff et al., 2006), BTEX are rarely taken into account (Bakke et al., 2013). PAHs have gained considerable attention in environmental pollution regulations and risk assessments of industrial discharges due to their carcinogenic properties and other health-related issues such as oxidative stress (Sturve et al., 2006), deoxyribonucleic acid (DNA) damage (Aas et al., 2000), cardiac function defects (Incardona et al., 2004), or embryotoxicity (Carls et al., 2008). PAHs with higher molecular weight have been linked to toxicity in fish, including disruption of genetic, growth systems, reproductive, and carcinogenicity (Kane Driscoll et al., 2010).

Alkylphenols have been a major concern due to their severe hazardous and endocrine-disrupting effects. With long-chained alkylphenols (C 7–C 9), alkylphenols are found in trace amounts in OPW (Neff et al., 2011b). Moderate and strong alkylated phenols such as nonylphenol have been traced to estrogenic effects in vertebrates (Hylland et al., 2008; Kovarova et al., 2013; Neff et al., 2006; Sundt and Björkblom, 2011), and to a lesser extent in algae, clams, shrimps, and crustaceans (Hylland et al., 2008). The chronic toxicity of alkylated alkylphenols, particularly octyl- and nonyl-phenols, is attributed to their endocrine-disrupting capabilities (Segner et al., 2003; Zha et al., 2007), which is a serious risk because they are rarely found at environmentally

acceptable levels of concentration.

Another component of OPW that has been suggested to act as a xenoestrogen (Thomas et al., 2009) is naphthenic acid. Naphthenic acids have been found in tailings pond water, posing a significant risk to aquatic species and wildlife, particularly birds (Quagraine et al., 2005). The levels of trace metals in fish and shellfish have been reported to be significantly lower than natural concentrations due to dilution and chemical processes when these inorganic elements enter the sea, resulting in a rapid reduction in concentration (Azetsu-Scott et al., 2007).

The environmental impact of OPW is based on the chemical components in OPW due to their intrinsic toxicity potentials. OPW is potentially hazardous to marine life. The discharged OPW has a modest risk of acute toxicity to the marine environment (Ekins et al., 2007; Yeung et al., 2015), with the key risk factor being constituent concentrations (Ekins et al., 2007). As operational discharge associated with production drilling is tied to OPW influent quantities, discharge volumes remain an important issue to address. Because of the large volume of OPW generated from oil exploration, various countries are legislating an acceptable limit of oil that should be contained in the OPW before discharge or reuse. Consequently, oil companies all over the world strive to meet “zero-discharge” requirements. Table 3 also provides guides to limit of acceptable levels of heavy metals and hydrocarbon compounds in OPW before discharge into the environment by Department of Petroleum Resources (DPR, 2002) Nigeria and the United States Environmental Protection Agency (US Environmental Protection Agency, 2016).

**Table 2**  
Concentration of BTEX, Phenol, PAHs, dissolved inorganic compounds, and heavy metal in OPW based on oilfields (µg/L).

Concentration of BTEX, Phenol, and PAHs (µg/L)					Concentration of dissolved inorganic compounds (µg/L)				Concentration ranges of Heavy metals in OPW			
Compound	Samarang Indonesia (Neff et al., 2011a) (Min-Max)	Gulf of Mexico (Neff et al., 2011a) (Min-Max)	Sergipe Brazil (Dórea et al., 2007) (Min-Max)	Norwegian continental shelf Norway (Fillo et al., 1992)	Compound (Dissolved inorganic)	United States of America (USA) (Dwyer and Mcdonald, 2016)	Qatar (Neff et al., 2011a)	Niger Delta, Nigeria (Dórea et al., 2007)	Compound	Brazil (Dórea et al., 2007) (mg/L) (Min-Max)	United Kingdom (UK) (Estrada and Bhamidimarri, 2016) (µg/L) (Min - Max)	Norway (Fillo et al., 1992) (mg/L)
Benzene	0.33–3.64	0.44–2.80	1291–1511	4.0	Barium	N/A	N/A	0.41	Copper	0.001–0.001	<10 - < 50	0.013
Toluene	0.089–0.80	0.34–1.70	1167–1357	2.77	Potassium	90	4900	N/A	Sodium	8800–9600	0.000013	0.029
Ethylbenzene	0.026–0.056	0.026–0.11	136–158	2.3	Magnesium	47	1390	165	Potassium	3100–4900	0.000674	0.01
Xylenes	0.013–0.48	0.16–0.72	89–103	N/A	Sodium	9600	9600	N/A	Lead	0.003–0.003	<2 - 179	0.05
Phenol	0.031–0.2	0.723	194–242	5.1	Calcium	220	504	N/A	Zinc	0.027–0.028	<50 - 411	0.006
					Salinity	N/A	36400	2700	Iron	4310–4770	35.8–106	30
PAHs (µg/L)									Nickel	0.015–0.017	<10 - < 50	0.543
Naphthalene			9.9–10.7						Cobalt	0.003–0.004	<1–5.1	N/A
Acenaphthene			1.6–2.4						Manganese	0.058–0.068	N/A	N/A
Anthracene			0.8–1.7						Chloride	16100–19500	N/A	N/A
Pyrene			0.9–1.0						Arsenic	N/A	<1–5.1	N/A
Chrysene			5.9–9.9						Silver	N/A	4 - < 50	N/A
Perylene			1.6–2.6						Mercury	N/A	<0.01–0.013	N/A
Phenanthrene			2.2–2.4						Aluminium	N/A	<10 - < 500	N/A
Fluoranthene			2.7–6.2									
Benzo [g,h,l] perylene			0.9–3.1									
Benzo [e] pyrene			2.4–2.6									
Benzo [0] anthracene			1.6–1.9									

N/A: Not available.

**Table 3**  
Acceptable limits (µg/L) of heavy metals, hydrocarbon compound and oil-water composition in OPW.

Acceptable limits (µg/L) of heavy metals and hydrocarbon compound in OPW(DPR, 2002; US Environmental Protection Agency, 2016)			Acceptable limits of Oil-in-water for some countries (Kornboonraksa, 2017)		
Compounds	Nigeria (DPR, 2002)	(US Environmental Protection Agency, (US Environmental Protection Agency, 2016)	Country	Legal basis	Average of Oil-in-water limits
Toluene	0.02	520	United Arab Emirate (UAE)	Kuwait convention	40 mg/L/100 mg/L max
Phenol	0.05	4000	Canada	Act RSC 1987	40 mg/L/80 mg/L max
Benzene	0.01	0.58–2.1	United Kingdom (UK)	OSPAR convention	40 mg/L/100 mg/L max
Sodium	ND	20200	United States of America	40 CFR 435	29 mg/L/42 mg/L max
Lead	0.05	0.015	China	GB 4914-85	30–50 mg/L avg./42 mg/L max
Manganese	0.40	100			
Chromium	0.03	0.10			
Calcium	400	N/A			
Chlorine	1050	250			
TDS	5000	250000			
Iron	0.20	2000			
Ethylbenzene	0.02	130			
Potassium	0.30	N/A			
Mercury	N/A	0.4			
Anthracene	N/A	130			
Nickel	0.02	610			
Cadmium	N/A	0.005			
Zinc	1.5	7400			
Barium	0.03	1000			
Copper	1.00	1300			

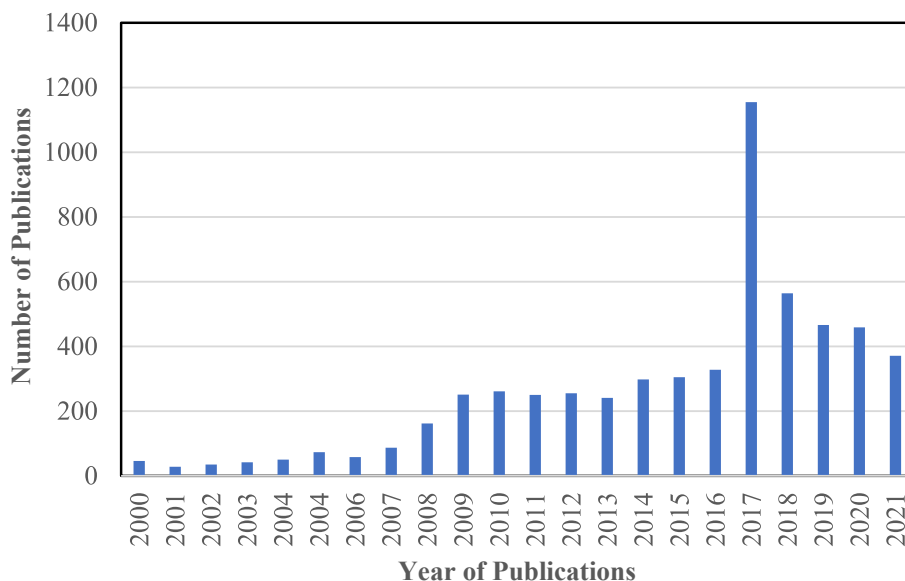
OPW is commonly re-injected into the well to maintain the hydraulic pressure within the well for oil recovery with up to 80% of OPW being reinjected into the well in some cases. This is necessary and economically advantageous in areas, where oil production well is located in water-stressed regions. When properly treated, treated OPW can be reused for irrigation and livestock consumption (Igunnu and Chen, 2012). Considering this requirement, OPW needs to be efficiently treated to meet acceptable standards for its beneficial use.

The growing need for the treatment of OPW is reflected by the increasing published studies in the field of OPW treatment (Fig. 3), with increasing annual publications. This shows that this emerging field of research presents an urgent need to treat OPW for beneficial reuse. Relevant articles in the database were chosen based on a certain keyword such as “produced water (PW)”, - “treatment”.

### 3. Treatment technologies for oilfield produced water (OPW)

Due to the complex composition of OPW, a wide range of treatment technologies for OPW have been studied (Drewes et al., 2009; Estrada and Bhamidimarri, 2016; Fakhru'l-Razi et al., 2009; Gregory et al., 2011; Sheikholeslami et al., 2018). For pollutants and impurities that cannot be eliminated by a single method, the treatment process normally requires a sequence of discrete unit processes. Treatment of OPW can open up other water management alternatives, such as reuse in agriculture and industrial applications.

Arthur et al. (2011) reported that any OPW treatment system should be capable of removing salts, micro-organisms, dissolved gases, naturally occurring radioactive materials, suspended solids, soluble organics, free and dispersed oil, as well as grease. Because of this complex nature,



**Fig. 3.** Annual publication on oilfield produced water treatment (2000–2021). Source: Scopus index, (extracted November 2021)

it is difficult to adopt a specific treatment technique suitable for the removal of all pollutants in OPW simultaneously. Overall, the most cost-effective technique is selected as the most preferable treatment cost of the OPW, depending on the plant's capacity, cost of electricity, influent quality, and the required quality of treated effluent (Dolan et al., 2018).

Furthermore, offshore OPW treatment is complex and challenging because of the lack of space for operation in remote and hostile conditions. Overall, there are three stages to conventional treatment of OPW: primary, secondary, and tertiary (final polishing treatment) steps (Fig. 4) (Dwyer and Mcdonald, 2016). The removal of coarse particles, gas bubbles, and large oil droplets as well as reducing dispersed pollutants and contaminants are completed in the pre-treatment step. Primary treatment removes particles and small oil droplets using the plate pack interceptors, skim tanks, and American petroleum institute (API) separators as the main treatment stage. Using centrifuges, hydro-cyclones, and gas flotation, the secondary treatment removes tinier oil droplets and particles. Using techniques such as cartridge filters, membranes, and dual media filters, the polishing stage is normally applied for ultra-small particles and oil droplets as well as dispersed hydrocarbons (less than 10 mg/L) removal. Finally, tertiary treatment, an optional step, is performed to remove dissolved debris as well as scattered hydrocarbons (less than 5 mg/L) and gases.

Fakhru'l-Razi et al. (2009) reported that combined biological, chemical, and physical treatment techniques can be employed to achieve specific treatment goals. Approaches such as chemical precipitation, membrane filtration, and adsorption have been extensively applied and reported to have achieved over 90% of removal for various OPW contaminants and constituents. As a result, the purpose of treatment (discharge or reuse) and treatment cost with respect to environmental regulations can influence the selection of appropriate technique.

Before being discharged into the environment or recycled for oil stimulation, the treatment method must meet regulatory standards (Piemonte, 2016). Since OPW must be extensively treated due to strict

environmental restrictions, the selected treatment must therefore be robust and capable of removing significant levels of contaminants from OPW, such as suspended solid particles and organic elements to enable it to be reused for the oil and gas industry (Lee et al., 2015).

To comply with the regulatory body, combined physical, chemical, and biological treatments, have been utilized to enhance water quality. Various treatments have been utilized in the oil and gas industry to remove target pollutants from OPW before being re-used (Ali et al., 2018; Andreozzi et al., 2018; Bagheri et al., 2018). The flow diagram of an OPW treatment system is displayed in Fig. 4. The OPW is collected directly from a water storage tank and/or oil/water separator unit (Dwyer and Mcdonald, 2016).

### 3.1. Physico-chemical treatment process

#### 3.1.1. Chemical precipitation

Chemical precipitation is widely used for the treatment of OPW (Li et al., 2000). About 97% of removal could be achieved for colloidal and suspended particles in OPW (Liu et al., 2000). Coagulants and flocculants, which consist of inorganic metals such as aluminum polymers, magnesium, and iron, are commonly used for this purpose (Zhou et al., 2000). Flocculants such as anionic polymer and ferric chloride ( $\text{FeCl}_3$ ) are used in ballasted flocculation to remove carbonaceous compounds, phosphorous, and particulate metals. However, the flocculants were ineffective in removing nitrogen and hydrophilic compounds (Gasperi et al., 2012). However, suspended solids and oil removal of 97% from OPW was achieved by adding coagulation chemicals (Zhou et al., 2000).

#### 3.1.2. Advanced oxidation process (chemical oxidation)

Organics, biochemical oxygen demand (BOD), dyes, chemical oxygen demand (COD), and odor are commonly removed from OPW using chemical oxidation. Because free electrons do not exist in an aqueous solution, this treatment relies on reduction and oxidation which occurs in the OPW (Iggunu and Chen, 2012). Catalysts and powerful oxidants

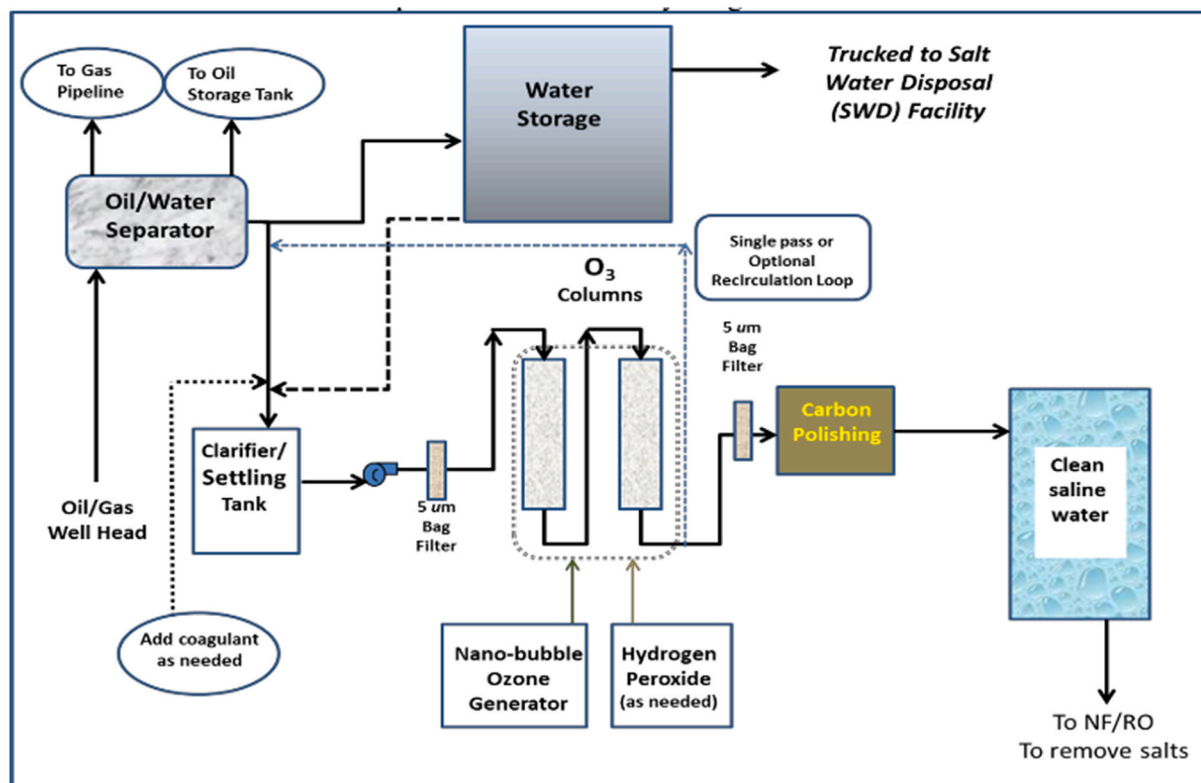


Fig. 4. OPW treatment process (Dwyer and Mcdonald, 2016).

can be employed to decompose organic contaminants in OPW (Huang et al., 1993). Various oxidants such as oxygen, peroxide, ozone, and chlorine can be used to break down a wide range of pollutants.

In addition, several parameters influence the rate of oxidation, including chemical dosage, types of oxidant, quality of raw water, and contact time between oxidant and water (Igunnu and Chen, 2012). This treatment method neither generates waste nor requires any pretreatment process. Despite using minimal equipment requirements, it could attain a maximum water recovery. However, this method has drawbacks such as the high cost of chemicals, the need for regular calibration and maintenance of chemical pumps, and the difficulty in removing by-products from the treatment process (Igunnu and Chen, 2012).

Igunnu and Chen, 2012) also reported that the process requires a final treatment step to remove particulate matter at the end of the oxidation process. Advanced oxidation processes (AOP) is considered as an efficient method for fast oxidation of target contaminants and pollutants by adding oxidants (Huang et al., 1993; Samuel et al., 2021a,b). Chemical oxidation makes use of other oxidants including iron, ZnO, and TiO<sub>2</sub> which are added into the process during treatment (Muruganandham et al., 2014).

### 3.1.3. Electrochemical treatment technology

In spite of its widespread use, this treatment technique is rarely applied for the treatment of OPW. However, it is proposed as a future treatment technique for OPW. The advantages of this method in other wastewater treatment are that it does not require chemicals, produces no secondary waste, and low-cost technology. The method has the potential to remove organic matter, while producing and saving energy without harming the environment (Martínez-Huitle et al., 2014). This treatment can be undertaken by integrating electrochemical techniques such as electrodeposition, electrolysis, and photo-electrochemistry, which combines photocatalysis, photo-electrolysis, and photo-electrocatalysis into a single electro-chemical process (Igunnu and Chen, 2012).

## 3.2. Biological treatment

When anaerobic or aerobic conditions are maintained, this treatment represents one of the most cost-effective strategies for pollutant removal (Günther, 2000). Furthermore, bacteria, fungi, and algae with a diameter of 0.2–10 µm are commonly found in OPW and can be used to treat OPW. Toxins and pollutants are used by the micro-organisms as nutrients for growth (Lu et al., 2009). For biological treatment of OPW, biological aerated filters and sequencing batch reactors can be utilized (Fakhru'l-Razi et al., 2009). This method is efficient with OPW concentration of 60 mg/L, BOD 50 mg/L, COD 400 mg/L (Guerra et al., 2011), and Cl<sup>-</sup> of 6600 mg/L (Ludzack and Noran, 1965).

### 3.2.1. Activated sludge process

Activated sludge is a typical aerobic wastewater treatment method that adsorbs soluble and insoluble pollutants (Fakhru'l-Razi et al., 2009). Activated sludge treatment can remove 98–99% of total petroleum hydrocarbons (TPH) with a solids retention time (SRT) of about 20 days (Tellez et al., 2002). Using the mixture (35% and 45% (v/v)) of sewage and acclimated sewage sludge in a sequence batch reactor (SBR), a COD removal was attained in the range of 30–50% (Freire et al., 2001). Furthermore, they found that the treatment efficiency is not affected by salinity. However, the efficiency of OPW treatment via the biological process is low due to the recalcitrant properties of pollutants in OPW samples (Freire et al., 2001). Fakhru'l-Razi et al. (2009) reported that suspended and trace solids can be removed using this treatment. The activated sludge process is simple and inexpensive; Nevertheless, it requires oxygen, while producing sludge as a waste product. Precipitated solids, dissolved gases, and biomass separation, removal, and filtration require post-treatment.

### 3.2.2. Biological aerated filters (BAF)

This treatment technology helps the biochemical oxidation process by removing organics from contaminated water under aerobic conditions. When sloughing occurs, a BAF filtration system with permeable media has a diameter of 4 inches to prevent pore space clogging. Additionally, downstream and upstream sedimentation is required for the filter bed to be fully utilized (Fakhru'l-Razi et al., 2009). The removal of pollutants from OPW using the BAF treatment was reported by Su et al. (2007), who found out that the technique is effective for OPW with chloride content lower than 6600 mg/L. By employing the BAF process, the removal of BOD, COD, oil, nitrogen, and suspended solids (SS) were 95%, 60%, 80%, 70%, and 85%, respectively (Su et al., 2007). Because the waste generated from the system is removed in solid form, BAF can achieve a 100% of water recovery rate. As a result, the disposal of the solid sludge that accumulates within the sedimentation basins accounts for over 40% of the total treatment cost and this is considered the primary drawback of this treatment method. However, it demonstrates the advantages of no post-treatment and chemicals needed, little maintenance required, having long life cycles, and suitable for water with varying quantity and quality.

### 3.2.3. Microalgae based treatment process

Microalgae-based technology is considered eco-friendly and sustainable for OPW treatment in which a high rate of pollutant removal can be achieved (Comminellis et al., 2008). OPW effluents can be treated via bioremediation using microalgae because of their ability to utilize certain contaminants as a source of nutrients (Mendes et al., 2010).

Takáčová et al. (2015) reported that microalgae like *Parachlorella kessler* use benzene, toluene, ethylbenzene, and xylene (BTEX) as a sole carbon source. To study the toxicity of BTEX, water-soluble fraction (WSF) gasoline was used to understand the effects of BTEX on microalgal growth (Durako et al., 1993; Zieman et al., 1984). With the increasing concentration of BTEX and contact time, a 50% decrease in the growth on cultures of microalgae was observed (Paixão et al., 2007). Microalgae growth was affected by hydrocarbon due to toxicity (Masten et al., 1994). Generally, phosphorus and nitrogen contained in OPW act as limiting factors for the growth of microalgae (Fakhru'l-Razi et al., 2009). Other trace elements, present in OPW, bring impacts on microalgal growth. Consequently, growing microalgae in OPW is promising for increasing the production of microalgae biomass during the treatment. Additionally, energy can be generated by using the cultivated microalgae biomass as feedstock (Chisti, 2007). *Chlorella* sp., *Scenedesmus* sp., and *Monoraphidium* sp. are among various microalgal strains employed for bioremediation of target pollutants in OPW treatment (Johnson et al., 2016; Mendes et al., 2010; Ma and Wang, 2006).

## 3.3. Physical treatment process

### 3.3.1. Media filtration

Filtration is simple for the treatment of contaminated wastewater. This method uses permeable filter media that allows the passage of water only while blocking the passage of any impurity smaller than the pore size of the media. Various porous materials are available as filtration media such as crushed stone, activated carbon, and sand. Although sand is widely used because of its low cost, easy access, and efficiency (Scholz, 2016), Adewumi et al. (1992) proposed metal removal via sand filtration process after a pretreatment.

This involves three steps: (i) reduction-oxidation via pH adjustment (ii) oxygen concentration increase via aeration; and (iii) sufficient retention time for solid separation to settle precipitated materials. After the processes are completed, sand filtration is used to remove any remaining fine solid, not removed during pre-treatment. The pretreated water is routed downwardly via a filter made up of sand layer. Certain characteristics to control water flow with filtration rate ranging between 0.1 and 0.4 m<sup>3</sup>/(m<sup>2</sup>. h) for a slower sand filtration treatment (Scholz, 2016). Before filtration starts, fine grains with a diameter of 0.15–0.35



mm and a depth of 1 m is used to make up the filter beds. The upper part of the treatment traps suspended particles and colloids from the contaminated water. As these particles accumulate, they clog the system and reduce its effectiveness. To remove the clog, it is important to scrape off the top layer of the sand that contains impurities. The filtering process combines adsorption, biological, chemical, and mechanical activity to completely remove contaminants (Huisman and Wood, 1974; Scholz, 2016).

### 3.3.2. Adsorption

Adsorption is recognized as one of the most effective methods for treating contaminated water by reducing pollutant concentrations to the required effluent limit (Daigle, 2012). According to Spellman (2003), the adsorption technique can remove about 85% of heavy metals and recover OPW completely. Although adsorption using commercial activated carbon has a drawback due to its high cost, this can be addressed using a cost-effective medium like low-cost materials, which makes the treatment competitive. Another disadvantage includes the need for disposal of used media and the saturated adsorbent after its regeneration. Several adsorbents can retain a variety of inorganic and organic pollutants. Oil, heavy metals (>80%), TOC, manganese, iron, and BTEX could be removed from OPW using various adsorbents. Granular activated carbon (GAC) has been employed as an efficient adsorbent for this purpose. Because of its efficacy, simplicity, and large surface area, GAC is preferable for water decontamination over other adsorbents.

Surface modification of the adsorbent promotes high surface area and internal pore structure of GAC (Alslaiibi et al., 2012). Extended surface area, microporous structure, and surface reactivity contribute to the high adsorbability of GAC (Kurniawan et al., 2003; Kurniawan et al., 2006). Furthermore, activated carbon possesses functional groups, which contributes to its Physico-chemical properties (Shen et al., 2010). Natural organic matter, mercury, synthetic chemicals such as radionuclides, di (2-Ethylhexyl) phthalate, benzo(a)pyrene, dioxin, and hexachlorobenzene, BTEX, and cadmium are examples of the contaminants that can be eliminated with GAC (Guerra et al., 2011). Several materials, such as agricultural wastes, wood, and fossil fuels, can be utilized to produce GAC, as well as various chemical and physical preparation techniques (Kurniawan et al., 2021a, 2021b). Activated carbon has a removal efficiency ranging from 70 to 85%. Although suspended particles are present in the OPW, this reduces its removal efficiency (Fakhrul-Razi et al., 2009). After treatment, its regeneration is required to sustain the cost-effectiveness of GAC for pollutant removal.

### 3.3.3. Flotation

This technique is used to separate suspended particles, which are not easily removed via sedimentation using fine gas bubbles. This separation is accomplished by gas injection into the water to be treated. Afterwards oil droplets and suspended particles adhere to the air or gas bubbles and they float to the surface, forming foam. This can be carried out with nitrogen, air, or other inert gases. Oil, grease, and volatile organics can be separated and removed from OPW using this method (Çakmakce et al., 2008). Gas flotation techniques include induced and dissolved gas flotation technologies. Their difference mainly lies in the method to generate the bubbles and the size of the resulting bubble. The efficiency and efficacy of this method depend on the impurities and type of pollutants to be removed, as well as temperature, liquid density differences, and oil droplets size. The flotation system does not operate well at high temperatures, but it is suitable for low-temperature feed streams and OPW with low and high concentrations of TOC (Guerra et al., 2011). Particles as small as 25 µm can be removed using the dissolved air flotation (DIF) technique. When coagulation is used as a pretreatment step, pollutants and impurities as small as 3–5 µm can be removed. The disadvantages and technological strengths of the gas flotation approach were reported by Fakhrul-Razi et al. (2009). The limitations include (i) 4–5-min retention time, (ii) skim volume, and (iii) high amount of air generated, while its benefits are (i) durability and robustness, (ii) ease of

operation, (iii) and coalescence. The flotation removed 93% of the oil.

Beyer et al. (1979) and the U.S. Department of Energy and U.S.A., 2005 reported the induced-air flotation method as a pre-treatment step for OPW with a TDS of 20,000 mg/L. They found that the TOC and COD levels reduced to 115 and 595 mg/L, respectively using this technique. This approach can be employed to remove suspended particulates, volatile organic compounds (VOCs), oil content, and other contaminants from OPW without using chemicals. Coagulants can be utilized to improve its effectiveness. However, its disadvantage includes the need for sludge disposal after treatment, which increases treatment costs (Casaday, 1993).

### 3.3.4. Electrodialysis

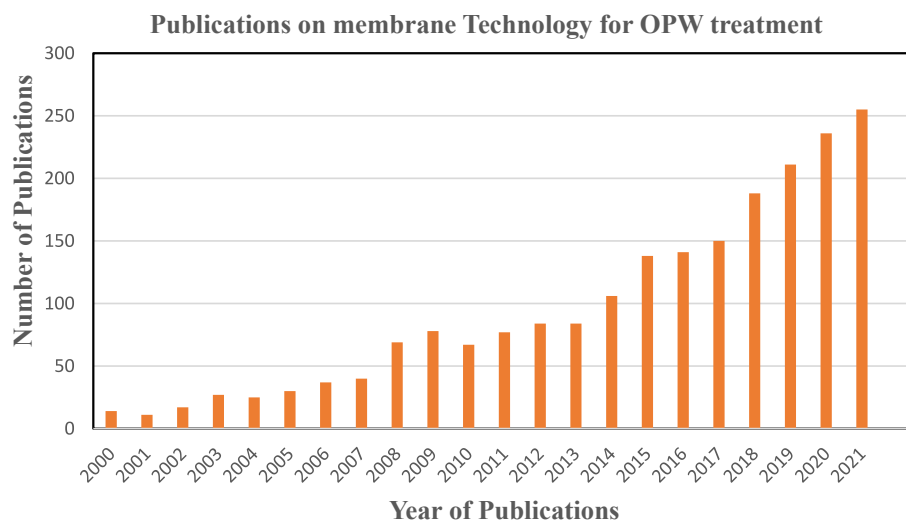
Both electrodialysis reversal (EDR) and electrodialysis (ED) are widely used for brackish water and seawater desalination. Electrochemical charge drives this reaction (Martin, 2014). Using a drift of alternating cation and anion, spacer sheets that separate selective membranes of electrodialysis might be utilized to remove salts from OPW (Hayes, 2004). The process entails the transmission of electrical current into the cell after the water passes through the membrane stack, causing cations and anions to migrate in opposite directions. As the ions migrate and meet the selective permeable membrane, alternate cells of concentrated and dilute solutions form in-between the membranes. This technique requires less energy and operates at a lower pressure than reverse osmosis. This can minimize salt content to less than 200 mg/L. Although this process is commonly used to treat OPW with low TDS levels (IOGCC and ALL, 2006), it could not remove other constituents as oils, heavy metals as well as other impurities. The main benefit of this technology for OPW treatment includes its ability to survive extreme conditions, no requirement of specific infrastructure, the duration of membrane lifetime, which ranges from 4 to 5 years (Igunnu and Chen, 2012). High cost, membrane fouling, and poor capacity to extract non-ionic elements like organic molecules, trained personnel, and regular concentrate dumping represent the disadvantages of this technology (Martin, 2014).

It is important to note that the various treatment methods discussed above have certain drawbacks as highlighted which include high treatment cost due to the disposal of waste during treatment, cost of chemicals, waste generation and its disposal, and the requirement of a large space for operation. With the emergence of membrane-based technology, there is a growing focus on applying the technology for the treatment of OPW due to its easy and simple operation, low cost, no requirement for chemical additives in most cases, and requires small space for operation. The increasing use of membrane technology for OPW and oily waste water treatment can be attested to due to the substantial investment in research by both the industry and academia over the past two decades, as reflected by Fig. 5 by typing the phrase “oil-membrane- treatment “Produced-water- membrane”.

### 3.3.5. Membrane filtration technology and its use in OPW treatment

As global population rapidly increases, the demand for clean water has become a critical issue worldwide. Because the world's supply of portable and clean water is limited, effective and sustainable water treatment and its recycling systems are essential for the Earth's long-term survival. Water reuse could be a solution to tackle the expected water crisis. Advanced membrane technologies are required to treat the OPW to attain this goal. These technologies have gained popularity as a frontrunner in OPW treatment.

Membrane filtration uses pores in a continuous structure to selectively fractionate components in a flowing fluid (Firouzjaei et al., 2018a; Mozafari et al., 2019; Rahimpour et al., 2018a,b; Zirehpour et al., 2016). Membrane separation is classified based on the pore size of applied materials and driving force (Pejman et al., 2020a; Xu et al., 2008). This technology has a wide range of applications in petrochemical (Takht Ravanchi et al., 2009), pharmaceutical (Firouzjaei et al., 2018b; Seyedpour et al., 2020a), and food (Rizvi et al., 2008) industries.



**Fig. 5.** Cumulative annual publication on OPW treatment using membrane technology (2000–2021).  
Source: *Scopus index* (extracted November 2021)

Although various membrane processes and membrane materials are used in a variety of applications, the end goal is separation. Ceramic (Abadi et al., 2011), metal (Li et al., 2020), and polymer are common membrane materials (Seyedpour et al., 2020b).

Membranes can be categorized based on the separation process (Supplementary Fig. S2). The four-basic pressure-driven membrane processes are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). They can also be classified as Porous (primarily MF and UF) and non-porous (RO and NF) membranes (El-Samak et al., 2020; Zhao et al., 2020). Furthermore, forward osmosis (FO) is a membrane mechanism driven by osmotic pressure (Pejman et al., 2020b).

An ideal membrane for a certain separation application should have high flux, high selectivity, appropriate mechanical resistance, and low treatment cost (Judd and Jefferson, 2003). Material and thickness affect mechanical resistance (Elshorbagy and Chowdhury, 2013; Vajner et al., 2016). High porosity and/or large pore diameters are the keys to achieving high flux. Small pores with a narrow range of diameters, can produce outstanding selectivity. As a result, flux and selectivity are frequently traded off (Judd and Jefferson, 2003; Moon et al., 2020; Park et al., 2017) Furthermore, Shirazi et al. (2014) found an inverse correlation between membrane flux and membrane thickness (number of layers), implying a trade-off between flux and mechanical resistance. Membrane-based OPW treatment is designed to remove pollutants from the feed with its size ranging from 1 nm to 1 $\mu$ m (Alkudhiri et al., 2013; Dickhout et al., 2017; Ebrahimi et al., 2010).

Due to the variation in the membrane's layer pore size, the applied operating pressure varies. There is increasing pressure as the layer pore size becomes smaller or decreases. RO filtration process takes place based on solution diffusion (Fick's law), while the UF and MF processes are based on convective pore-flow (Darcy's law) (Munirasu et al., 2016). The NF membrane is also referred to as loose RO membrane because its filtration mechanism lies in-between diffusion and pore-flow mechanism (Baker, 2004; Nagy, 2012, 2019). The MF membrane has a pore size ranging between 0.1 and 10  $\mu$ m, filtering or rejecting particles such as bacteria, suspended solids, etc. However, it does not reject or stop divalent ions or viruses from passing through the membrane.

The UF membrane is made of pore size ranging from 2 to 100 nm, which rejects the passage of viruses and macromolecular colloids. However, it allows most dissolved ionic species to pass through. The change or transition from pore-flow to solution diffusion ranges between 0.5 and 1 nm in diameter. Therefore, the NF membrane pore size should lie within this range (Baker, 2004).

Membranes processes can be used for separation purposes through either dead-end (Supplementary Fig. S3a) or crossflow filtration (Supplementary Fig. S3b). In operating the crossflow membrane separation, permeate exits through the pores and flows over the membrane, while in the dead-end membrane separation process, the retentate concentrates on the membrane surface. Hollow fiber or flat sheet membrane can be used for filtration purposes, which depends on the membrane's operating conditions. Hollow fiber membrane is made of several hundred to thousands of fiber, while flat sheet membranes could be used in a plate/frame set-up rolled into spiral-wound modules (Judd, 2010).

Membranes are classified based on the materials they were fabricated from. This includes ceramic and polymeric membranes (Alami et al., 2018). Inorganic or ceramic membranes are fabricated from materials such as metal oxides, silica, or carbon. They have excellent chemical and thermal stability. Therefore, it is suitable for use in the oil recovery process (Ezugbe and Rathilal, 2020; Deriszadeh et al., 2010; Alpatova et al., 2014; Emani et al., 2014) while on the other hand polymeric membranes are made from a single polymer or mix of polymers such as PVDF, PES, PSF, etc. They are cheap and easy to produce which has made them attractive to researchers for use in various separation processes.

Ceramic membranes are known to be inert to solvent, steam, resistant to strong acid conditions, and durable. These types of membranes have gained popularity because of their high mechanical and thermal stability, and compatibility with harsh cleaning methods such as escalating cleaning cycles (Aziz et al., 2019; Ebrahimi et al., 2020; Fakhru'l-Razi et al., 2009). Periodic cleanings such as cycles of backwashes and chemical soaks are required to eliminate foulants from the surface of the membrane. Backwashing is efficient when ceramic membranes can withstand high pressures (Freeman and Shorney-Darby, 2011). Furthermore, the combination of a high-pressure tolerance and cleaning chemicals results in ceramic membranes with stable permeability. For example, using silicon carbide MF ceramic membranes with OPW from an oilfield in the Persian Gulf lead to a sustained flux of 700 L/m<sup>2</sup>. hr with a permeability of 1400 L/m<sup>2</sup>. hr.bar was reported (Zsirai et al., 2018). Furthermore, using commercial alumina MF ceramic membranes (0.8  $\mu$ m pore size) fed by synthetic OPW that contains 250–1000 ppm concentrations of heavy crude oil droplets of 1–10  $\mu$ m diameter, almost complete removal of oil was achieved (Al-Ghouti et al., 2019; Mueller et al., 1997). The permeate contained less than 2 ppm total hydrocarbons in treated effluents (Mueller et al., 1997).

The main disadvantage of the ceramic membrane is its expensive cost of production and weight. The ceramic membrane works on size

exclusion and its modification with respect to molecular affinity is difficult. Other drawback of this membrane includes fouling, although this can be taken care of by hard cleaning methods. Ceramic membranes are not swollen when being exposed to solvents. They are used for microfiltration (MF) (Barukčić et al., 2014), nanofiltration (NF) Zeidler et al. (2014), ultrafiltration (UF) (Ezugbe and Rathilal, 2020; Murić et al., 2014).

However, polymeric membranes are used in various separations within the oil industry. A broad range of polymer materials includes Polyvinylidenedifluoride (PVDF), polyethylene sulfone (PES), cellulose derivatives, polyacrylonitrile (PAN), polyvinylchloride (PVC), polysulfone (PS), and polytetrafluoroethylene (PTFE). The membranes can be suitable for various separation purposes. In choosing a polymeric membrane, the polymer needs to have relevant affinity and can withstand operating conditions including environmental conditions. Polymeric membranes are prepared from blended polymers, blended with other compounds to enhance their performance (Lalia et al., 2013). Depending on the type of separation, polymeric membranes can be porous or dense. Its surface can be modified to enhance its functionality (Abetz et al., 2021). Polymeric and composite-based membranes are less costly than ceramic-based membranes (Alzahrani and Mohammad, 2014; Dickhout et al., 2017; Lee et al., 2015).

OPW treatment using membrane technology aims at removing oil droplets with a size of less than 10  $\mu\text{m}$  (Dickhout et al., 2017). The principle of separation using membrane technology is anchored on the basis of pollutant's size through a selective media (Wen et al., 2019). Although synthetic polymers make up most of the membranes, inorganic or ceramic membranes are still applied for separation. One of the filtration methods used for oil/water emulsion and oil removal in petroleum wastewater is UF. This process is effective in oil separation in OPW as compared to other membrane filtration. Because of its high oil rejection, UF does not require the addition of chemical additives. In addition, it requires less space, and low operational cost (Igunnu and Chen, 2012). However, UF membranes are prone or susceptible to fouling because of their high permeate flux. Therefore, researches on the UF membrane is aimed at minimizing the fouling on its surface.

From our literature survey, the best way to minimize the fouling problem is by reducing the membrane's surface roughness. This makes the membrane surface hydrophilic (Wang et al., 2021). UF operates between 1 and 30 *psi* transmembrane pressure, which can be used as a pretreatment method for desalination. Li et al. (2006) studied the treatment of oily waste water from an oilfield using a tubular UF membrane equipped with polyvinylidene fluoride and modified with nano-sized inorganic aluminum particles. They reported that nano-sized alumina particles can enhance the anti-fouling ability of membranes. An efficient removal of TOC and COD rate of 98% and 90%, respectively was attained, while the oil rejection was higher than 98% for the system with a feed concentration of the oil, TOC, and COD as 15.5 mg/L, 214.9 mg/L, and 637 mg/L respectively and the high rate of rejection and removal of COD and TOC was attributed to the presence of the nano-sized inorganic aluminium particles which increase the membrane hydrophilicity providing the membrane with an anti-fouling property.

In another study, Asatekin and Mayes (2009) found that polyacrylonitrile-graft-poly (ethylene oxide) (PAN-g-PEO) and an amphiphilic copolymer additive were incorporated into the UF membrane to modify it for OPW treatment. A removal rate of 96% of dispersed oils from the OPW was attained.

Oil and grease (O&G), total suspended solids (TSS), and sulfate could be eliminated in OPW using UF membrane (Al-Ghouthi et al., 2019; Alzahrani and Mohammad, 2014; Mondal and Wickramasinghe, 2008). Using polyamide thin film composite (PA-TFC) NF and low-pressure RO membranes such as BW30 (a RO membrane for brackish water treatment application) or NF270 (a NF membrane for salt rejection application), almost complete removal of the three pollutants has been reported (Al-Ghouthi et al., 2019; Alzahrani and Mohammad, 2014; Mondal and Wickramasinghe, 2008; Ozgun et al., 2013).

Membrane distillation (MD) is a non-pressure-driven membrane separation method (Wang and Chung, 2015). MD uses a vapor pressure gradient across the membrane-based thermally driven membrane process (Macedonio et al., 2014; Wang and Chung, 2015). MD approaches include air gap MD (Meindersma et al., 2006), sweeping gas MD (Shirazi et al., 2014), and permeate gap MD (Shirazi et al., 2014). Direct contact with membrane distillation (DCMD) is widely investigated for OPW treatment (Ali et al., 2018; Anari et al., 2019; Zou et al., 2020). It is a membrane process in which the permeate and aqueous feed are kept at different temperatures, while contact with a hydrophobic membrane allows vapor to pass, but prevents liquid permeation (Ashoor et al., 2016; Lawson and Lloyd, 1996). The difference in temperature causes a difference in vapor pressure, which drives the flux through the membrane from hot feed to cold permeate (Ashoor et al., 2016; Lawson and Lloyd, 1996). TDS may be eliminated by DCMD using hollow fiber membranes with >99% of removal and can operate for 100 h (Ali et al., 2018; Zou et al., 2020).

Pre-treatment methods have been demonstrated to improve membrane performance while lowering expenses (Hilal et al., 2003; Kusworo et al., 2018; Padaki et al., 2015). For example, utilizing adsorbents during pre-treatment can improve membrane longevity, flow, and anti-fouling qualities (Kusworo et al., 2018). Using the bioreactor is an efficient way to reduce chemical oxygen demand (COD) in oil and gas wastewater such as OPW. About 80% of COD removal could be attained with COD concentration ranging from 1,710 to 3,030 mg/L (Huang et al., 2020; Ozgun et al., 2013).

In terms of COD reduction, bioreactors give a better performance than RO pre-treatment. It has however been reported that pre-treatment with UF and MF membranes results in less than 25% of COD removal (Al-Ghouthi et al., 2019; Ozgun et al., 2013). Membrane separation is an effective technique for removing TDS from OPW (Atoufi Hossein and Lampert David, 2021; Çakmakce et al., 2008; Strong et al., 2017; Venkatesan and Wankat, 2017) for the following reasons: (i) low cost and energy consumption, (ii) high rejection rates (Webb and Zodrow, 2020), and (iii) no chemical requirement (Webb and Zodrow, 2020).

UF and MF membranes, remove a significant portion or fraction of solids and can be used as a pre-treatment for FO, RO, and NF process to attain high rejection of TDS (Atoufi Hossein and Lampert David, 2021). Implementing RO could remove 95% of TDS from OPW (Atoufi Hossein and Lampert David, 2021; Funston et al., 2002). FO and MD are promising for the treatment of OPW with high TDS concentrations because they use concentration and temperature gradients, respectively (Coday et al., 2014; Duong et al., 2015; Webb and Zodrow, 2020). However, pressure-driven membrane processes such as RO are impractical for treating high salinity OPW (>35 g/L) (Shaffer et al., 2013); because the hydraulic pressure required to overcome the osmotic pressure of high-salinity waters is higher than the pressure of RO membrane modules for high-salinity OPW with TDS concentrations >35 g/L (Greenlee et al., 2009; Shaffer et al., 2013).

Unlike RO, FO is osmotic pressure-driven rather and is not limited by a high-pressure operating limit associated with TDS concentrations of 70 g/L (Martinetti et al., 2009). Low-pressure NF was reported to be effective in removing sulfate and calcium ions (Fakhru'l-Razi et al., 2009; Webb and Zodrow, 2020). The presence of heavy metals in OPW is a global concern because they possess serious health implications (Atoufi Hossein and Lampert David, 2021; Fakhru'l-Razi et al., 2009; Tchounwou et al., 2012). RO and NF have been effective in the treatment of feed water containing heavy metals. NF and RO have removal of 97% and 99%, respectively (Qdais and Moussa, 2004).

Overall, membrane-based OPW treatment is effective for a variety of pollutants. It can be seen from (Supplementary Table S3) that the use of membrane technology for OPW treatment has an oil rejection rate of over 96% with a flux recovery rate of over 66%. Table 4 presents a comparison between convention OPW treatment and membrane-based technology. It is concluded that membrane technology has better advantages than other treatment technologies, as the former in most cases

**Table 4**

Comparison of some other conventional OPW treatment methods with membrane-based technology (Colorado School of Mines, 2009; Igunnu and Chen, 2012; Liu et al., 2021).

No.	Treatment techniques	Treatment Stage	Principle of operation	F <sub>A</sub> (m·h <sup>-1</sup> ) (Judd et al., 2014)	F <sub>V</sub> (h <sup>-1</sup> ) (Judd et al., 2014)	Merits	Drawbacks	Development trend
1	Media filtration	Advanced Treatment method	1. Filtration 2. Adsorption 3. Coalescence.	27–37 (nutshell filter)	13–24 (nutshell filter)	1. Have separation effects on suspend solids 2. Advanced Purification system.	1. Media replacement is required. 2. Requires regeneration via back washing due to pollution 3. The influent should not have high oil content.	Research on efficient filtration media and innovative backwashing technologies, as well as the development of new materials
2.	Air flotation and compact flotation unit	Standard reaching treatment	1. Reduce the density by making floccules out of gas bubbles and oil drops. 2. Under the influence of gravity there is a density difference between heterogenous phases.	2.4–16 (induced gas flotation)/ 45–90 (CFU)	4–15 (induced gas flotation)	1. Have separation effects on suspend solids. 2. Compact (for CFU).	1. The volume, scale of injected gas affects the separation efficiency therefore will need extra gas. 2. Secondary pollution of scum due to the chemical agents.	Understanding the interaction between gas bubbles and oil drops has led to the modification of the bubble generator and the development of green and efficient chemical agents.
3.	Gravity and enhanced gravity sedimentation	Pretreatment and standard reaching treatment	1. The shallow pool principle is used to reduce the floating distance. 2. Separation efficiency enhancement via coalescence structure 3. Under the influence of gravity there is a density difference between heterogenous phases.	4.4 (API)/ 6.5–14 (CPI)	1.1 (API)/ 3.2 (CPI)	1. Simple equipment with low running costs and minimal maintenance requirements 2. capable of preventing fluctuation of the flow rate, oil content, and gas concentration. 3. Pretreatment equipment that is indispensable and flexible for large scale operation	1. Low separation accuracy (worse for the crude oil with high viscosity and density). 2. Require more space due to split mechanism.	Size reduction based on the anti-fluctuation capability of coupling with multi-physical fields.
No.	Treatment techniques	Treatment Stage	Principle of operation	F <sub>A</sub> (m·h <sup>-1</sup> ) (Judd et al., 2014)	F <sub>V</sub> (h <sup>-1</sup> ) (Judd et al., 2014)	Merits	Drawbacks	Development trend
4.	Hydrocyclone	Standard-reaching treatment	1 Under the influence of gravity there is a density difference between heterogenous phases.	100–440	450	1. Suitable for OPW with high oil content. 2. It's small and doesn't have any moving parts.	1. Separation effects are affected by blockage and wear of one or more cyclone tubes. 2. A small area of high-efficiency operation.	The understanding of the whirling flow field is used to promote operating flexibility.
5.	Combined fibers coalescence	Standard reaching treatment	1. Oil drop coalescence caused by oleophilic fibers. 2. Physical demulsification based on the differing properties of hydrophilic and oleophilic fibers at the crossing point	129	27	1. The suspend solids can penetrate the fiber module without blocking. 2. Capable of handling PW with high oil content	1. The characteristics of fibers have an impact on separation efficiency. The gadget should be built in accordance with OPW's characteristics. 2. Blockage caused by solid particles with millimeter-scale dimensions.	The creation of a general design principle based on material surface properties.
6.	Tubular separation	Pretreatment	1. Under the influence of gravity or a whirling flow field, density varies between heterogeneous phases occur.	It can be installed beneath water or in the space between the platform's components.		1. It's small and can be installed beneath the water.	1. Improvement in anti-fluctuation performance as a pretreatment step is required.	Development of pretreatment technology that can be utilized underwater to improve adaptation to hard environments with fluctuation.
7.	Membrane separation	Advanced treatment method	1. As a chemical agent, catalyst, and microorganism carrier.	312 (crossflow membrane filter)	136 (crossflow membrane filter)	1. Ability to remove dissolved organic	1. With much higher temperature (>50 °C) polymeric material degrades.	Based on material science, new membranes with high flux and

(continued on next page)

Table 4 (continued)

No.	Treatment techniques	Treatment Stage	Principle of operation	F <sub>A</sub> (m·h <sup>-1</sup> ) Judd et al. (2014)	F <sub>V</sub> (h <sup>-1</sup> ) Judd et al. (2014)	Merits	Drawbacks	Development trend
			2. Filtration is pressure-driven.			substance with proper choice of membrane. 2. Have separation effects on suspend solids. 3. Advanced purification process.	(Saththasivam et al., 2016). Generally, when the temperature of the OPW is greater than 50 °C 2. Backwashing is necessary.	pollutant resistance have been developed.

F<sub>A</sub> = Capacity Area, F<sub>V</sub> = Capacity Volume.

does not require pre-treatment steps, is cost-effective, and energy-efficient in comparison to the other technologies.

Jebur et al. (2021) reported the treatment of hydraulic fracturing produced water using combined electrocoagulation-microfiltration membrane distillation and found that the initial concentration of TDS of 245,300mgL<sup>-1</sup> and that of TOC which was 120mgL<sup>-1</sup> were reduced to 44mgL<sup>-1</sup> and 64mgL<sup>-1</sup> respectively after the electrocoagulation-microfiltration process. They further reported that membrane distillation was used to desalinate the pre-treated produced water a high-quality treated water was obtained which gave a reduced concentration of TDS of 56mgL<sup>-1</sup> and TOC of 1mgL<sup>-1</sup>. Other works have also reported the use of various membrane technologies for produced water, shale gas, oil plays and flowback water treatment for the removal of oil, TOC and TDS with varying results of the removal of these pollutants (Chang et al., 2019a, 2019b; Jiménez et al., 2018b) Despite its effectiveness, desalination of OPW with high salinity (>70 g/L) still has potential for improvement (Ahmad et al., 2020). Hybrid treatment, which combines two or more membrane processes, has shown to be feasible and applicable in treating OPW (Ahmad et al., 2020; Murray-Gulde et al., 2003). Atoufi Hossein and Lampert David (2021), proposed a four-step method for salt removal from OPW. The first step, known as feed softening, involves adding calcium hydroxide to the sample to reduce the likelihood of fouling during membrane processes; the second step, sand filtration removes large suspended solids; the third step, the ceramic membranes remove O&G; and the final step, with PA-TFC membrane RO, removes O&G (Atoufi Hossein and Lampert David, 2021). It is critical to evaluate the appropriate combination of pre-, main-, and post-treatments steps to improve its capabilities while saving energy consumption and treatment cost (Ahmad et al., 2020).

Due to the complex nature of OPW, no single method or technique is universally suitable to meet the effluent limit in OPW treatment, Membrane technology could be utilized as a single treatment unit without requiring the need for pre- or post-treatment steps. The various advantages of membrane technology over the conventional methods for OPW treatment are presented in Supplementary Table S4.

However, the challenge of this technology is the complexity of the organic matter and oil-water emulsion. Various cycles of operation during OPW treatment could cover the membrane surface, causing membrane fouling. The presence of total dissolved solid (TDS) also leads to scale and silt formation on the membrane surface.

#### 4. Membrane fouling in OPW treatment

Membrane fouling is a significant drawback in the treatment of OPW using membranes. Due to concentration polarisation (CP) of solute on the surface of the membrane and subsequent fouling, membrane processes are limited in terms of flux reduction with filtering time (Mukherjee and SenGupta, 2006; Seyed Shahabadi and Reyhani, 2014). Membrane fouling reduces membrane permeability while shortening membrane life due to the harsh chemicals used to clean it. The membrane must be replaced when cleaning is no longer effective. Membrane

fouling is one of the most critical technical obstacles in OPW treatment using membrane processes (Tomer et al., 2009). Membrane fouling is a surface phenomenon influenced by the nature of dissolved/dispersed components or compounds in the OPW to be treated, surface morphology, and chemistry of the membrane surface. The complex nature of the OPW is responsible for the fouling of membranes in OPW treatment (Mondal and Wickramasinghe, 2012).

Membrane fouling results from the adsorption and accumulation of rejected oil, suspended particles, and biomolecules on the membrane surface (external fouling) is common in RO and NF membranes while fouling in the membrane pores (internal fouling for porous membranes) is common in MF and UF (Wandera et al., 2012). Membrane fouling decreases or increases the transmembrane pressure (TMP), depending on the constant pressure or constant flux the process uses. Generally, controlling fouling and membrane cleaning strategies are based on trial-and-error techniques. As a result, a better understanding of fouling mechanisms and the materials that causes fouling are required (Mondal and Wickramasinghe, 2012). Foulant-membrane surface interaction is critical for foulant adhesion during the early stages of fouling. Once a small layer of foulant is formed on the surface of the membrane, foulant-foulant interaction becomes the most important factor in foulant accumulation and cake formation.

The thick cake layer on the surface of the membrane has a high specific resistance, leading to high filtration resistance, reducing the effective filtration area, and eventually covering the entire surface of the membrane during filtration (Wang and Wu, 2009). During fouling, Physico-chemical interactions occur, depending on the nature of the foulant molecules and the chemistry and/or morphology of the membrane surface. As a result, the rate of foulant attachment can be controlled by the Physico-chemical parameters of foulant and the surface properties of membranes. Foulant layers can become densely compacted on the membrane surface, reducing water permeability, increasing head loss, and requiring extra chemical cleaning. Membrane fouling caused by interactions between dissolved and suspended solutes and the surface of the membrane can be irrecoverable, irreversible, or reversible, depending on the nature of the foulants (Mondal, 2016).

Physical cleaning strategies for membrane fouling (such as basic hydraulic cleaning) have been demonstrated to be effective in preventing reversible fouling, but less efficient or effective in controlling irreversible fouling (Coday and Cath, 2014; Wandera et al., 2012). The intense physicosorption and/or chemisorption of foulant molecules in the membrane pores and on the membrane surface results in irreversible fouling. The chemisorption of diverse ionic natural organic matter (NOM) happens with the functional group of the surface of the membrane, whereas the physicosorption of colloidal foulants or inorganic/natural NOM occurs through the van der Waals interactions with the membrane surface (Kimura et al., 2004). Irreversible fouling needs a severe chemical and/or heat treatment to remove it, which degrades the surface of the membrane, especially for polymeric membranes, which are vulnerable to these treatments.

Finally, fouling that has accumulated over time is irreversible and

cannot be eliminated using any cleaning methods (Drews, 2010; Mondal, 2013) since membrane fouling occurs due to the interaction between suspended and dissolved solutes in the feed with the surface of the membrane. Therefore, membrane surface wettability such as hydrophilicity/hydrophobicity, charges, and membrane surface smoothness/roughness affect the degree of attachment of dispersed and/or dissolved species (foulants) to the surface of the membrane.

#### 4.1. Types of membrane fouling in OPW treatment

Fouling on membrane surface in OPW treatment can be problematic, and various types of foulant such as biofoulants, colloidal, inorganic, and organic might influence membrane separation. Biofouling, colloidal, inorganic, and organic fouling occur frequently at the same time thereby increasing membrane resistance. Membrane fouling can take many forms, depending on the morphology of the surface of the membrane, the type of foulant, and the type of membrane filtration process used (NF, UF, MF, RO, FO, or MD).

Membrane fouling during OPW treatment can take many forms, depending on the membrane's surface morphology and types of foulants: (a) formation of cake layer on the surface of the membrane due to accumulation of foulants as in the case of NF, UF, and RO, (b) intermediate blocking of the pores of the membrane thereby restricting or preventing the flow of solute through blocked pores however allowing the passage of solvent at a lower rate, (c) complete or total blockage of the pores of the membrane by the foulant preventing the passage of both the solute and the solvent from passing, (d) constriction within the membrane pores which is common in MF and UF restricting or limiting flow by foulants adsorbed onto the pore walls which are smaller than the membrane pores. The fouling layer can be formed by one or more mechanisms working together. In MF and UF, all four mechanisms can work together to generate the foulant layer, however, in RO and NF, the formation of the cake layer is the chief mechanism for membrane fouling (Wang et al., 2011). The most common type of fouling that occurs in membranes during OPW treatment is discussed below.

##### 4.1.1. Organic fouling

This type of fouling is caused by the interaction between the membrane surface and natural organic matter (NOM) (Gao et al., 2011). The petrochemical sector contributes a substantial quantity of TOC in OPW, which varies depending on the kind of OPW, i.e. oil or gas, to the composition of TOC foulants (Alzahrani et al., 2013). Organic foulants are a diverse set of organic compounds found in OPW that are structurally complicated, varied, and poorly characterized (Ke et al., 2013). Because of the unique interactions between foulants and chemical functional groups or foulant molecules on the surface of the membrane, organic fouling particularly on the surface of the polymeric membrane is highly intricate (Shi et al., 2011). Organic molecules with a wide variety of molecular weights are found in OPW. It can also be divided into two groups based on their sizes: (i) dissolved organic carbon (DOC) < 0.45  $\mu\text{m}$  and (ii) particulate organic carbon (POC) > 0.45  $\mu\text{m}$ , (Pandey et al., 2012).

The coexistence of different solid phases, such as colloid, and liquid phases (water and other dissolved organic solute), membrane surface, adsorbent as well as foulant adsorption on the surface of the membrane is complex and based on the competitive interaction between the solvent (primarily water), the solute (impurities in the wastewater and NOM) and the surfaces (solute-membrane interface, solute-water interface, and water-membrane interface) (Gao et al., 2011). In the case of UF, microorganisms, including certain hydrophilic/hydrophobic organics, are absorbed inside the pores of the membrane, while in the case of NF/RO, they are absorbed on the membrane surface. Because the organics do not aggregate well, the resulting cake layer is relatively less dense (Huang et al., 2014). As a result of the synergistic impact of NOM and colloids due to rapid fouling, combined organic-colloidal foulants can cause a faster flux drop than separate foulants (Kim et al., 2014). Organic

fouling can be irreversible or reversible depending on the foulants used and the hydrophobicity/hydrophilicity of the membrane surface.

##### 4.1.2. Inorganic fouling

Concentration polarisation (CP) (Supplementary Fig. S4) is the primary cause of inorganic fouling. This is due to supersaturation which leads to the precipitation of solutes and the formation of scale. The forming of particle layers on the active surface of the membrane and as well the formation of concentration profiles of retained solutes in bulk solutions is called CP. Because more ion molecules are available near the active layer of the membrane surface, CP increases the possibility of fouling, causes a decline in flux in NF and RO due to higher osmotic pressure, and diminishes the quality of the permeate (Mondal, 2016). Spiegler-Kedem model, gel-layer model, resistance-in-series model, osmotic pressure model, film theory, and cake-enhanced CP are some of the models proposed to describe the CP (Shirazi et al., 2010). Due to decreased flux across the membranes, CP increases the concentration of solute in the walls of the membrane, which accelerates fouling. The high concentration of rejected inorganic solutes aggregates dissolved matter at the active layer of the membrane surface into colloid-sized particles, which promotes particle-particle and particle-membrane interactions, allowing colloids to foul on the active layer of the membrane surface (Al-Amoudi, 2010). Various ionic foulants such as magnesium and calcium facilitate the interaction between organic foulants and the surface of the membrane (Alzahrani et al., 2013).

##### 4.1.3. Colloidal fouling

Membrane fouling by colloidal matter occurs when colloidal foulant deposits and adheres to the membrane surface (Peña et al., 2013). Suspended solid (10 mg/L) and turbidity are two colloidal foulants observed in OPW (Alzahrani et al., 2013). The size of colloidal particles in wastewater ranges from a few nanometers to a few micrometers. In the NF membrane, colloids that are close to or smaller than pore size causes pore blockage, but those that are considerably larger than pore size can cause surface coverage or formation of cake layer as demonstrated (Supplementary Fig. S5) (Guo et al., 2012; Huang et al., 2008).

Standard or complete pore blockage appears to be dependent on the relative sizes of foulants, pores of the membrane, as well as foulant surface characteristics. A complete blockage can occur when foulants considerably smaller than the pore size adsorb on the pore walls, whereas standard blocking can occur when foulants much smaller than the pore size is adsorbed on the pore walls. Complete blocking and cake formation occur in NF due to mechanical restriction, while standard blocking happens due to chemical adsorption, which is primarily governed by membrane surface properties such as surface charge (potential), wettability as well as nature of foulant (van der Sman et al., 2012). If the particulate materials in OPW such as bacteria, colloids, or even microscopic flocs are smaller/similar in size to the membrane pores, pore blockage might occur (Lin et al., 2014).

##### 4.1.4. Biofouling

Biofouling is the accumulation of undesirable biological matter on the surface of the membrane which leads to the formation of biofilms by microorganisms. The different parameters that govern the morphology of biofilm include thickness, density, structure, composition, and bioadhesive strength (Bixler and Bhushan, 2012). Biofilm accumulates on the membrane surface as a result of biofouling caused by bacterial aggregation. Apart from the possibility of colloidal, inorganic, and organic fouling, the large population of bacteria in OPW ( $42 \times 10^6$  colony-forming units (cfu)/l) poses major biofouling issues during OPW treatment (Alzahrani et al., 2013). The bacteria's origin could be traced back to the pretreatment process of OPW which included tank settlement and dissolved air flotation, followed by secondary biological treatment (Alzahrani et al., 2013). Biofilms are made up of a matrix of extracellular polymeric substances (EPS) created by bacteria, which is made up of heteropolysaccharides and has a high negative charge density (Peña

et al., 2013). Growth (multiplication) and attachment (bioadhesion and bioadsorption) are the two main phases in biofouling in membrane processes.

Bioadhesion is controlled by microbe qualities, solution parameters, and membrane surface morphology, whereas biofilm multiplication is controlled by micronutrient availability (Guo et al., 2012; Ivnitsky et al., 2005; Subramani et al., 2011). The RO membrane with a rough surface is more prone to biofilm formation during OPW treatment than the NF membrane with a smooth surface similar to colloidal fouling (Alzahrani et al., 2013). Organic foulants are incorporated into biofouling through adsorption to other colloids as aggregates or to the membrane altering biofilm interactions on the surface of the membrane (Alzahrani et al., 2013).

#### 4.1.5. Complex fouling

Various aromatic and aliphatic hydrocarbons (o-xylene, toluene, benzene, dibenzothiophene, phenanthrene, naphthalenes, etc.), as well as inorganic contaminants including heavy metals and silica, are found in OPW. Some of the components are soluble in water, while others are not. The colloidal particles will be formed by the water-insoluble component. The water-soluble component, on the other hand, will be absorbed on the surface of the membrane via physical and/or covalent bonding (e.g. hydrophilic-hydrophilic interaction hydrophobic-hydrophobic contact, and van der Waals force) (Mondal, 2016). Foulant-foulant interactions regulate and govern the build-up of the cake layer once a very thin coating of foulant is generated. In the case of membrane-based OPW treatment, the fouling layer may contain both organic and inorganic components. During the treatment of OPW, the presence of various metallic components may trigger complex formation between inorganic and organic chemicals on the active layer of the surface of the membrane.

#### 4.2. Fouling control in membrane OPW treatment

Membrane fouling in OPW can be mitigated or controlled using various methods including ultrasonic cleaning, chemical cleaning, membrane surface modification or alteration, optimization of operational parameters, and pretreatment of feed. The thick foulant coating on the membrane surface can be broken down by ultrasonic therapy. The fouling of membranes by various contaminants between the foulant molecules and active layer of the membrane surface necessitates thermal and/or harsh chemical treatment to remove, as both oxidation and abrasion cause irreversible damages that make it impossible to restore the original membranes' performance (Drews, 2010; Peña et al., 2013).

Cleaning strategies or methods to minimize or control membrane fouling include proper process design, and operating conditions, ultrasonic membrane cleaning, hydraulic and mechanical cleaning (Agarwal et al., 2013; Ahmad et al., 2014; Chai et al., 2012; Corbatón-Báguena et al., 2014; Hajibabania et al., 2012; Hilal et al., 2005; Luján-Facundo et al., 2013; Naim et al., 2012; Nam et al., 2013; Ramon et al., 2013; Valladares Linares et al., 2012; Wan et al., 2013; Wang et al., 2013).

In conclusion, among the various conventional methods for water treatment and particularly OPW treatment, membrane technology is considered a promising alternative to these methods since it is cost-effective, energy-efficient, does not require pre-treatment step in most cases, requires smaller space for installation, and have high separation efficiency. This technology is mostly considered suitable since the treated OPW is required to meet certain standards for beneficial reuse or discharge into the environment (Syarifah Nazirah et al., 2017).

### 5. Qualitative assessment of OPW treatment technologies

A qualitative assessment is considered as part of this study to determine the suitability of each treatment technology in terms of waste generation, chemical demand, energy requirements, modularity, robustness, flexibility, and reliability. A qualitative performance

assessment based on a ranking scheme as proposed by (Arthur et al., 2005) was adopted in this work as presented in Table 5. This qualitative assessment criterion was adopted as most relevant for OPW treatment based on literature studies, discussion with industry experts and researchers. The basic criterion considered includes:

- (i) Robustness -which refers to an equipment's ability to withstand harsh external conditions while maintaining mechanical stability under treatment.
- (ii) Modularity-which is the capacity of a system to be merged or upgraded in response to changing conditions or as a unit process
- (iii) Flexibility-which refers to a technology's ability to handle a diverse range of OPW during treatment.
- (iv) Reliability-which ensures that the technology will only require little downtime to provide good water quality and that it will not fail.

Table 5 presents the qualitative evaluation of the various treatment technologies used in OPW in terms of the various parameters indicated. This evaluation is aimed at providing a view of the treatment equipment's capacity to withstand some minor technical challenges, environmental concerns, consistency in treatment with good quality of effluent, providing a view of the possibility of integrating two systems or more, the flexibility of the technology in terms of mobility measures, a view of the tendency for the system to fail with minimal downtime and ability of each treatment technologies to accommodate a range of pollutants and contaminants during the treatment process. It can be seen that in terms of flexibility, AOP and adsorption have a higher tendency. In terms of modularity which is the capacity of the system to be merged or upgraded, AOP, electro dialysis, adsorption, NF, UF, and MF show a better tendency. Almost all the technologies presented show a higher degree of robustness except for floatation which has a low degree of robustness. Almost all the treatment technologies also show some degree of reliability except for coagulation/floatation and activated sludge process having an average degree of reliability. In terms of energy requirements, RO shows the tendency of highest energy requirement while other technologies all show the tendency of high energy requirement with API gravity separator showing a low tendency for energy requirement. Coagulation/flocculation activated sludge process, electro dialysis and AOP have an average degree of chemical requirement while all other methods have a low degree of chemical requirement. Coagulation/flocculation, RO, adsorption, and activated sludge have a high degree of waste generation, AOP having the lowest degree of waste generation while other treatment processes have an average degree of waste generation during the treatment process.

Overall this analysis presents a basis for the choice of suitable treatment technology putting the various parameters into consideration to meet the need and purpose for the treatment of the OPW.

### 6. Summary of the various treatment technologies and their effectiveness in OPW treatment

It is evident from the review of literature that most of the technologies widely studied in the literature are membrane filtration, adsorption, and activated sludge which has been shown to have high removal efficiencies (above 90%) for different OPW constituents. Floatation and hydrocyclone are capable of removing oil and particulates from OPW. Adsorption is capable of removing over 80% of heavy metals from OPW and can achieve almost full OPW recovery. Some adsorbents used in adsorption are sometimes capable of removing BTEX in OPW. Electro dialysis can be used to remove heavy metals, BTEX, oil, and grease as well as particulates from OPW. Membrane technology such as NF and UF can remove heavy metals and TSS. UF membranes are mostly used for oil rejection and removal in OPW. It was also found that among the membrane technologies RO will be most suitable for the removal of organic compounds in OPW. However, with the numerous advantages of

**Table 5**  
Qualitative evaluation of the various OPW treatment technologies.

Treatment technology	Flexibility	Modularity	Robustness	Reliability	Energy requirement	Chemical demand	Waste generation
Coagulation/flocculation	Average	Low	High	Average	Average	Average	High
API gravity Separator	Average	Low	Very high	High	Low	Low	Average
Hydrocyclone	Average	Low	Very high	High	Average	Low	Average
Flotation	Low	Low	Low	High	Average	Low	Average
Adsorption	High	High	High	High	Average	Low	High
Activated Sludge process	Low	Average	Average	Average	Average	Average	High
Electrodialysis	Low	High	High	High	High	Average	Average
AOP	High	High	High	High	High	Average	Low
MF	Average	High	High	High	High	Low	Average
UF	Average	High	High	High	High	Low	Average
NF	Average	High	High	High	High	Low	Average
RO	Average	Average	Average	High	Very high	Low	High

membrane technology over other OPW treatment methods, fouling is the major setback of membrane technology. It is therefore important that for efficient removal of various OPW constituents using membrane technologies, a proper understanding of the mechanism of interaction between the OPW constituents (oil, organic compounds, etc) and the membrane surface be properly investigated and understood. This will provide the basis for the design and fabrication of membranes that can be fouling resistant. However, the overall cost of treatment and the purpose (discharge or reuse) influence the choice of treatment technique to be used.

While some of the various technologies discussed are mature in terms of OPW treatment application, others are not designed to meet up with the current requirements of new environmental regulations and legislation, high maintenance and energy cost, as well as potential by-products generation may limit their future application since zero pollution discharge and a substantial reduction in discharge of OPW will be required in the future (Jiménez et al., 2018a). Successful pollutant elimination requires pre-treatment of produced water before it enters the primary environment. Furthermore, because certain technologies are incapable of complying with current or new tighter regulatory standards for OPW discharge due to environmental concerns, post-treatment may be required.

It was also found from this review that the advanced oxidation process (AOP) is an effective technique in the removal and degradation of organic compounds as well as oil in OPW. It has been reported that most of the various traditional technologies for OPW treatment are limited in terms of removal of oil droplets of less than 10 µm (Dickhout et al., 2017; Zhu et al., 2014b), therefore, this review concluded that considering the advantages of the membrane technology and the AOP, the combination of these two technologies (photocatalytic membrane) can provide a treatment technique that can be effective and efficient in achieving a high degree of pollutant removal and degradation and at the same time mitigate the problem of fouling in the membrane since the oxidation process is capable of degradation of the foulants that may attach to the surface of the membrane giving it have an anti-fouling and self-cleaning property.

## 7. Beneficial reuse of treated oilfield produced water (OPW)

Due to the increase in demand for petroleum products globally, production of oil and gas will continue to increase, and as such the negative effects of exploration activity on the environment from the generation of OPW will continue. In addition, as the human population increases rapidly, the demand for fresh and portable water for drinking and other purposes will also increase. Furthermore, as the availability of fresh water becomes scarce OPW could be a critical source of water for various needs upon proper treatment. Lately, researchers have focused attention on reclamation, recycling, and reusing of water normally considered as waste to meet the needs of the global requirements for fresh and portable water for drinking and other purposes (Miller, 2006). Treatment of OPW for beneficial reuse (Supplementary Fig. S6) involves

numerous stages and factors to be put into consideration before adopting a particular treatment technology for the treatment process before the water is obtained for the intended reuse purpose. Various standards have been provided for the reuse of treated water with respect to its intended use. For instance, the US Department of Agriculture Natural Resources Conservation Service provided standards for the reuse of treated water for livestock and irrigation (Natural Resources Conservation Service (NRCS, 1999). The US-EPA also provided standards for the reuse of treated drinking water (United States Environmental Protection Agency (United States Environmental Protection Agency, 2000). Supplementary Table S5 gives the standards (NRSC and USEPA) for water reuse with respect to the various intended purpose. As will be expected, drinking water reuse standards are more stringent requiring a higher cost of treatment for the OPW.

There are various options for the reuse of OPW such as for industrial purposes in dust control as well as in livestock watering, irrigation, drinking water, fire control, oilfield uses such as re-injection into the well, power generation, habitat, and wildlife watering. Due to the complex nature of OPW discussed earlier, proper treatment of OPW is required to meet acceptable standards and quality before reuse. The treatment level will depend on the purpose for which it will be used. For instance, more intensive treatment of OPW is required if it will be used for drinking purposes while a little lower level of treatment is required if it will be used in industrial dust control as well as in oil and gas industries (Council, 2015). In addition, economics or cost also determines the treatment level or degree of treatment and reuse need. For instance, it was found that treatment of OPW suitable for re-injection using conventional methods such as media filters, gravity separation, and hydrocyclone could cost up to \$0.509/m<sup>3</sup> of water. while on the other hand, using advanced desalination methods such as mechanical vapor compression (MVC) combined with other appropriate methods for recycling of OPW to obtain suitable reuse water for the same purpose could cost up to \$3.808/m<sup>3</sup> of water (Bagheri et al., 2018).

### 7.1. Irrigation

When OPW is treated and its quality meets a required standard as well as having a very low value of TDS after removal of all pollutants, we can regard the treated OPW as a suitable beneficial resource for crop irrigation purposes (Sirivedhin et al., 2004). The reuse of OPW for irrigation purposes particularly in dry terrain has been proposed and studied, however, because of high salt concentration (Na = 3–435,000 mg/L, TDS = 35–472,000 mg/L) in some OPW from some oilfields, its use for this purpose remains a challenge (Echchelh et al., 2018). Higher proportions of specific ion toxicity, alkalinity, salinity, and sodicity found in OPW is because of the low quality of the OPW and this is the main challenge to its reuse for irrigation purpose. It is therefore critical to put into consideration the type of crop to be irrigated when utilizing OPW for irrigation (Drewes et al., 2009).

Sirivedhin et al. (2004) studied the treatment of synthetic OPW by varying the concentration of TDS to mimic OPW for beneficial reuse via



electrodialysis and found that all the 3 samples containing lower TDS concentrations met the required standards for beneficial reuse for both livestock watering, irrigation, and drinking purposes. However, they found that the treated water contains higher treatment values of sodium adsorption ratio (SAR) compared to the required irrigation standard values. Although this drawback could be overcome via adding little amounts of  $Mg^{2+}$  and/or  $Ca^{2+}$  into these waters to lower their sodium adsorption ratio (SAR) and make them appropriate for irrigation. They also discovered that water samples having extremely high concentrations of TDS failed to meet up the requisite water reuse standard, as shown in [Supplementary Table S5](#). As a result of their findings, it can be concluded that electrodialysis for water reclamation for beneficial use is an effective technology for OPW reclamation and treatment for this application under experimental settings.

A case study of the reuse of OPW for irrigation of biofuel crops is demonstrated in Texas in the USA where they have several oil wells and reuse OPW is re-injected back into the wells for enhanced oil recovery processes ([Ellsworth, 2013](#); [Keranen et al., 2013](#)). Part of this OPW is reused for irrigation of biofuel crops. Since biofuel production is estimated to increase and being water-intensive, OPW can serve as an excellent means of irrigation of biofuel crops in that region and as well help to minimize the use of fresh water for such purpose but channel purposely for drinking by humans. However, salt content, organic matter, and hydrocarbon present in the OPW are the main parameters to be considered in OPW for reuse in irrigation.

### 7.2. Livestock watering

Because livestock has a higher pollutant tolerance than humans, the water quality consumed by livestock is normally lower than that consumed by humans. Contaminants in animal drinking water, on the other hand, should be kept to a minimum to avoid negative health challenges. Water having less than 1000 mg/L TDS can be used for livestock watering. However, if the amount surpasses 7000 mg/L, it can have some negative implications on the livestock like diarrhea (U.S. Department of Energy, 2005). This concept was used in some projects of coal bed methane (CBM) where livestock watering stations were established for the livestock to use the treated OPW for their consumption ([Veil et al., 2004](#)).

### 7.3. Microalgae production for biofuel generation

Algal growth rate, the yield of biomass, and production of lipid by algal strains cultivated from OPW as nutrient media have only been reported in a few studies. [Godfrey \(2012\)](#) on the other hand, has done the most critical and comprehensive study to date on using OPW as a medium for growing microalgae to create lipid for biofuel conversion. In the OPW, eight (8) strains of microalgae were cultivated successfully, and all produced neutral lipids. He reported from the study that lipid production and *Amphora coffeiformis*' growth were optimal with the addition of 150 mg/L sodium nitrate and no phosphorus added whereas *Chaetoceros gracilis* and *Chlorella* sp growth and high lipid productivities were optimized with 300 mg/L sodium nitrate with no addition of phosphate as well. Furthermore, pretreatment of OPW for hydrocarbon removal using either settling or centrifugation, activated carbon or filtration can boost the yield and growth of lipid. The lipid content of strains cultivated on OPW got to 63% and 63.8 mg biodiesel/l/day, indicating that they make significantly more biodiesel. Therefore, it was determined that substituting OPW enriched just with needed phosphorus and nitrogen nutrients for growth media is more economically and environmentally viable, as equivalent yield of lipid and biomass may be reached at low cost without the use of freshwater.

A case study of the reuse of OPW for microalgae production was investigated in Qatar by Al-Ghouti et al. (unpublished result) where they collected OPW from one of the natural gas fields to establish the heavy metal removal efficiency of this method ([Al-Ghouti et al., 2019](#)). In a

related study [Cai et al. \(2013\)](#), reported that OPW toxicity could increase with a corresponding increase in heavy metal concentration which agrees with the report of the study of [López-Rodas et al. \(2008\)](#) & [Pereira et al. \(2013\)](#) that more elements can be recovered by *Dictyosphaerium* sp. as having better growth rate compared to the other species. It also revealed that *Dictyosphaerium* sp. has the potential to grow in the presence of metal-rich water.

### 7.4. Habitat and wildlife watering

After undergoing semi-intensive treatment and verifying its safety, OPW can be utilized to form an artificial reservoir that will serve as a supply of drinking water for wildlife as well as a habitat for fishes and waterfowl. These impoundments can collect, and store the OPW in large quantity as they have a wide area of many acres ([Sirivedhin et al., 2004](#)).

### 7.5. OPW reuse for internal purpose in the gas industry

China possesses the world's greatest shale gas reserves, including the world's largest shale gas play in the Sichuan Basin ([Rosenblum et al., 2017](#)). Each well produces about 20,000 m<sup>3</sup> of OPW ([Bhadja and Kundu, 2012](#)), which is even more than the volume produced in the United States ([Kondash and Vengosh, 2015](#)). The values of TDS in the OPW ([Guo et al., 2018](#); [Kong et al., 2018](#)) were discovered to be lower compared to those observed in shale plays in the United States ([Barbot et al., 2013](#); [Estrada and Bhamidimarri, 2016](#); [Shaffer et al., 2013](#)). The viability of reusing this OPW within the industry was investigated ([Huifang Chang et al., 2019](#)) due to its increased quality and volume because it can minimize both the wastewater volume and freshwater usage. OPW reuse within the sector such as for fracturing operations is attracting attention ([Jiang et al., 2013](#)). As a result, the integrated treatment methods of coagulation then UF and NF were studied to find out if they could meet the reuse guidelines. In the work by [Jiang et al. \(2013\)](#), they reported the use of two types of coagulants (ferric chloride hexahydrate and aluminum sulfate octadecahydrate). The coagulated water was then injected into the ultra-filtration (UF) process. The molecular weight cut-off (MWCO) of the UF membrane utilized was 100 kDa with a steady flux of 50 L/m<sup>2</sup>/h with backwashing for 5 min per hour. A NF membrane operated at a pressure of 100, 200, 300, and 400 psi was then used to treat the UF-filtered water. Water recovery rates of 50, 70, and 85% were used in the NF system. As a result of the combined coagulation-UF-NF treatment method, turbidity was decreased by 99%, COD by 94.2%, and alkalinity was reduced by 94.1%. Cations like strontium, barium, magnesium, and calcium removal were between 72% and 83%, as seen in [Fig. 6](#). All the treated water parameters obtained from the integrated treatment method were significantly lower when compared to reuse standards ([Supplementary Table S5](#)), showing that it can be reused in the industry.

Thermoelectric cooling for power plants is another area of OPW reuse for internal purposes in the industry. This requires large water withdrawals in once-through cooling systems, but consumption equals only a few percent of withdrawal ([Dieter et al., 2018](#)). They reported that water use for recirculating cooling was ~20% of OPW, highest in the Oklahoma and that OPW would represent 23%–45% of cooling water use in counties with the highest water demand for these plants (Noble and Oklahoma counties).

## 8. Outlook of membrane research

It is increasingly critical to promote the need for the reuse of treated OPW to meet the rising water needs and as well minimize the environmental impacts of oil exploration. OPW reuse minimizes not only the industry's environmental impact but also reduces the strain on freshwater resources. This is critical in water-stressed countries that experience economic and population expansion.

Most of the treatment methods for OPW encounter limitations.

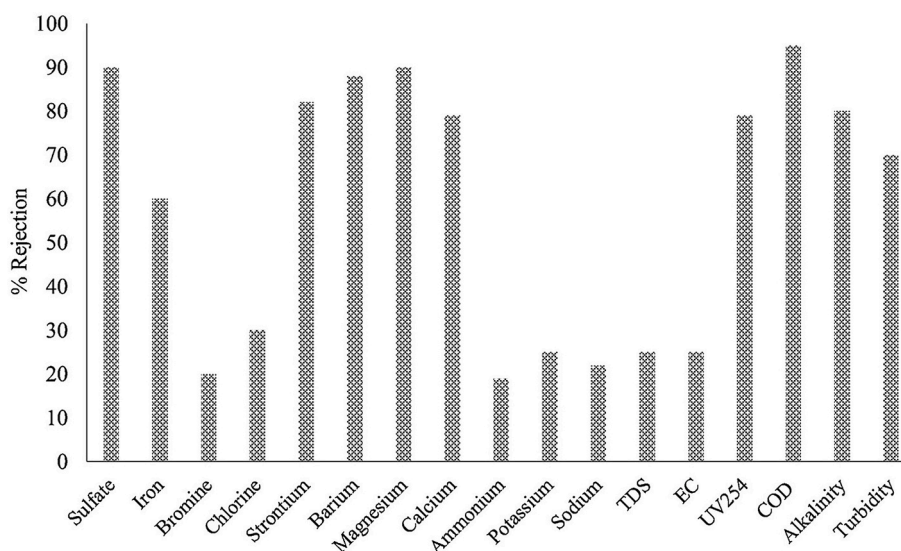


Fig. 6. Combined coagulation-UF-NF treatment system removal efficiency (Jiang et al., 2013).

Membrane technology has gained popularity for OPW treatment due to its ease of operation, low cost, energy efficiency, ability to treat and remove the small size of oil droplets. This makes it suitable to be used on oil offshore exploration facilities. However, its effectiveness is affected by membrane fouling. As a result, it reduces its treatment efficiency.

Considering this reason, the combination of membrane technology and photocatalysis a form of AOP is proposed. It was found from this review that the advanced oxidation process (AOP) is also an effective method for OPW treatment, particularly for oil/water emulsion separation via degradation and mineralization of oil emulsion present in OPW. Therefore, the synergic effect of integrating photocatalysis and conventional membrane-based separation (Photocatalytic membrane) in a single unit enables researchers to address the fouling mechanism, while treating the OPW and degrading organic pollutants simultaneously. With such a carefully developed photocatalytic membrane, treated OPW effluents could generate treated effluents with the desired quality for drinking, irrigation, microalgal production as well as for wildlife and habitat use.

## 9. Concluding remarks

This review has revealed that the nature and qualities of OPW in various oil wells are complex and vary from one region to another, depending on the age of the well. It is conclusively evident from 262 published studies (1965–2021) that no single treatment method is highly effective for OPW treatment as a stand-alone process however, conventional membrane-based technologies are frequently used for the treatment of OPW with ultrafiltration (UF) being the most used filtration process reporting a higher efficiency for oil rejection from OPW and oily waste water. However, it is found from the review that among the various membrane technologies RO will be most suitable for the removal of organic compounds.

After membrane treatment, treated effluents of the OPW could be reused for irrigation, habitat and wildlife watering, microalgae production, and livestock watering. As a result, the selected treatment process must be assessed first, depending on the source of OPW. Treatment cost and quality of treated effluent for reuse influence the selection of treatment methods to be used. Several strategies have been identified in the literature. Overall, this implies that target pollutants in the OPW samples could be removed efficiently for subsequent use, despite its complex properties.

In conclusion therefore, as environmental regulations have become more stringent, requiring an improved quality of treated OPW effluent

with future regulations focusing upon the ability of exploration activities and industrial users to comply with the legal requirements of OPW discharge and reuse, it would be very beneficial to investigate various treatments, which can assist oil exploration companies, oil refineries, industrial users to comply with the environmental laws. This review, therefore, provides the basis for the selection of a suitable method or combination of methods for efficient and effective treatment of OPW.

## Declaration of competing interest

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2022.114556>.

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