

Review

Review of Techniques for the Removal of Polycyclic Aromatic Hydrocarbons from Produced Water

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Abstract: Polycyclic aromatic hydrocarbons (PAHs), due to their mutagenic, carcinogenic, and teratogenic potential, can lead to numerous chronic and fatal diseases. PAHs have been found in several wastewater streams, including “produced water,” which is wastewater generated during the extraction of oil and gas. The PAHs’ removal from produced water using physical, chemical, biological, and combined methods is crucial. Water is a vital ecosystem component and is extremely vulnerable to PAHs. This article reviews the current PAH situation, including their physical and chemical properties, types, characteristics, and removal methods from produced water. The mechanism of each method of removal of PAHs has been discussed. The current study results show that adsorption by nanoparticles and integrated methods are promising methods to meet the strict authoritarian limit with advanced increase potential in the direction of commercialization for the removal of PAHs and provide opportunities to use produced water as a source of water. The current study results can help the policy/decision makers in the efficient management of water resources.

Keywords: produced water; polycyclic aromatic hydrocarbons; treatment method; nanoparticles



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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are highly dangerous organic compounds that can lead to long-term health effects and are also carcinogenic and persistent pollutants. PAHs consist of two or more fused benzene rings, which give them exceptional stability. Generally, the volatility and solubility of PAHs decline, while their hydrophobicity increases with an increase in the number of benzene-fused rings [1]. The main sources of PAHs in the aquatic environment are due to the incomplete combustion of fossil fuels such as petroleum, diesel, and coal. Other sources include natural forces such as forest fires, volcanic eruptions, and the diagenesis of organic materials, which can also result in the production of PAHs. They are also generated by human activities such as industrial wastewater and waste burning. There are several ways that PAHs can reach the aquatic environment, including deposition or direct release into the water through fuel discharge and oil spills. The accumulation of PAHs can cause severe damage to deoxyribonucleic acid (DNA), which is the main cause of cancer and tumors. Aquatic life can also face severe health problems from inhaling even minute concentrations of PAHs [2].

PAHs are also present in produced water due to its complex composition, which contains many thousands of compounds with different concentrations, such as alkylphenols, dispersed oil, organic matter, and inorganic salts [3]. Produced water is a mixture of formation water and injected water that is produced to surface facilities for gas and oil. The production of produced water is increasing globally and has reached approximately 250 million barrels per day for around 80 million barrels per day of produced oil. Typically, produced water is directly discharged into water bodies without treatment, contaminating

the environment [4]. Due to the increased global demand for oil and gas, the production of produced water is expected to increase in various regions, as shown in Figure 1. The presence of PAHs in produced water can cause mutagenicity and carcinogenicity. In recent years, researchers have been interested in removing PAHs from produced water using various methods. This paper aims to review the different physical, chemical, biological, and combined methods used for the removal of PAHs from produced water.

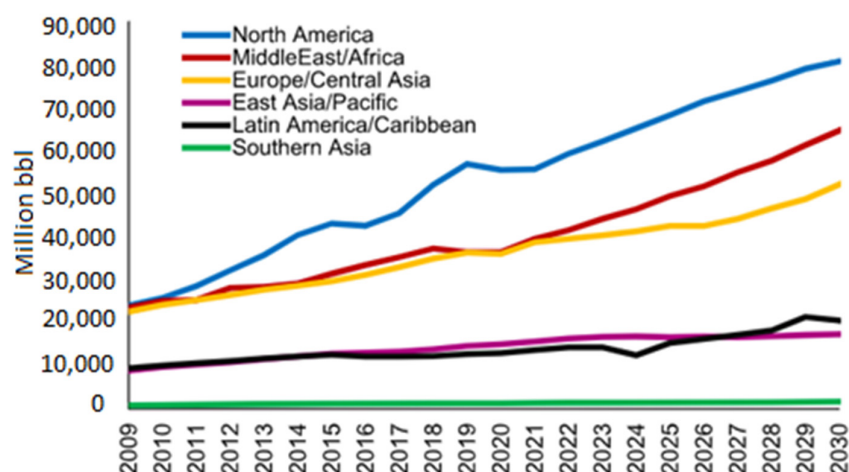


Figure 1. Volume-wise production of produced water by various regions based on literature [5].

2. Classification and Properties of PAHs

PAHs are classified as low molecular weight (LMW) and high molecular weight (HMW) compounds. LMW PAHs contain two or three aromatic rings, while HMW PAHs contain four or more aromatic rings [6]. The U.S. Environmental Protection Agency (EPA) classifies PAHs into sixteen forms based on their hazardous nature [7]. PAHs can also be classified as alternant and non-alternant, where their structure is composed entirely of benzene rings with the involvement of four, five, and six-member non-aromatic rings. The physical and chemical properties of PAHs are determined by their conjugated π -electron structures, which depend on the number of aromatic fused rings and their molecular weight [8]. The types of LMW and HMW PAHs, their formulas, structures, and molecular weights are shown in Tables 1 and 2, as given below.

Table 1. Types of LMW polycyclic aromatics hydrocarbons and their molecular structure.

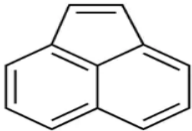
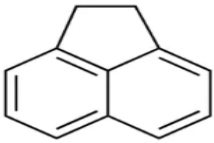
Name of LMW PAHs	Abbreviation	Formula	Structure	Molecular Weight (g/mol)	Source
Acenaphthylene	ACY	$C_{12}H_8$		152.1	[9]
Acenaphthene	ACE	$C_{12}H_{10}$		154.2	[10]

Table 1. *Cont.*

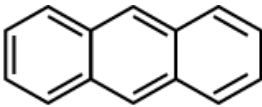
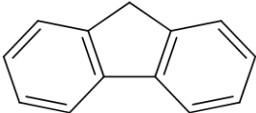
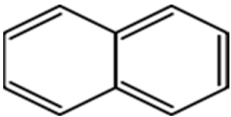
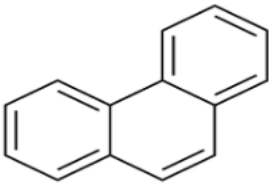
Name of LMW PAHs	Abbreviation	Formula	Structure	Molecular Weight (g/mol)	Source
Anthracene	ANT	C ₁₄ H ₁₀		178.2	[11]
Fluorene	FL	C ₁₃ H ₁₀		166.2	[12]
Naphthalene	NAP	C ₁₀ H ₈		128.91	[13]
Phenanthrene	PHE	C ₁₄ H ₁₀		174.2	[8]

Table 2. Types of HMW polycyclic aromatics hydrocarbons and their molecular structure.

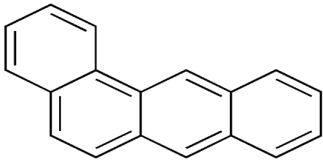
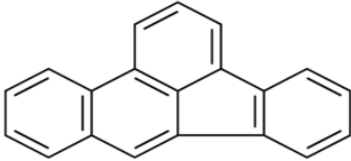
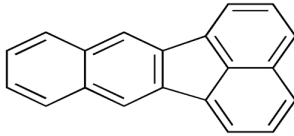
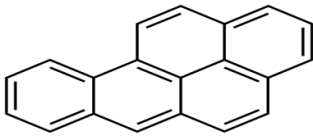
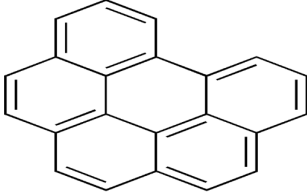
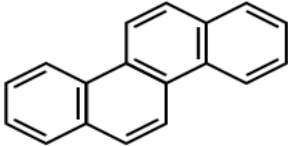
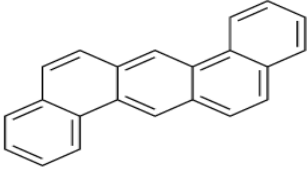
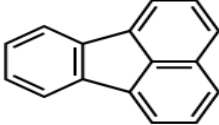
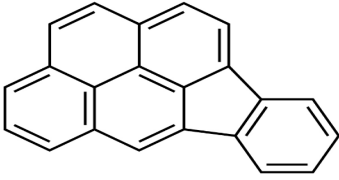
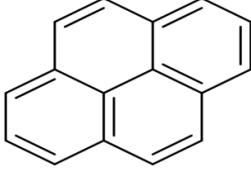
Name of HMW PAHs	Abbreviation	Formula	Structure	Molecular Weight (g/mol)	Source
Benzo (a) anthracene	BaA	C ₁₈ H ₁₂		228.3	[14]
Benzo (b) fluoranthene	BbF	C ₂₀ H ₁₂		252.3	[14]
Benzo (k) fluoranthene	BkF	C ₂₀ H ₁₂		252.3	[13]
Benzo (a) pyrene	BaP	C ₂₀ H ₁₂		252.3	[15]

Table 2. Cont.

Name of HMW PAHs	Abbreviation	Formula	Structure	Molecular Weight (g/mol)	Source
Benzo (ghi) perylene	BhP	C ₂₂ H ₁₂		276.3	[16]
Chrysene	CHY	C ₁₈ H ₁₂		228.2	[17]
Dibenz (a,h) anthracene	DahA	C ₂₂ H ₁₄		278.3	[9]
Fluoranthene	FLU	C ₁₆ H ₁₀		202.2	[11]
Indeno (1,2,3-cd) pyrene	IcdP	C ₂₂ H ₁₂		276.3	[16]
Pyrene	PYR	C ₁₆ H ₁₀		202.2	[18]

The water solubility of PAHs depends on factors such as ion strength, dissolved organic carbon, pH, and temperature. In general, PAHs have low solubility in water but are easily dissolved in oil and fats, and they tend to accumulate in the fatty tissue of living organisms. The water solubility and lipophilicity of PAHs tend to decrease as their molecular weight increases, making them more resistant to removal. LMW PAHs are more water-soluble than HMW PAHs, and due to their increased solubility, volatility, and propensity to serve as a sole carbon source for many microbes, LMW PAHs are eliminated more efficiently by physical, chemical, and biological processes than HMW PAHs. As the molecular weight of PAHs increases, they become more hydrophobic, prone to bioaccumulation, resistant to biodegradation, and environmentally persistent [19]. PAHs have different vapour pressures, and those with lower vapour pressures tend to be associated with particles, while those with higher vapour pressures are found as vapour at ambient temperatures in the air. In general, the solubility of PAHs decreases and hydrophobicity increases as the number of

condensed benzene rings increases. Additionally, volatility decreases as the number of condensed aromatic rings increases [8].

3. Methods of Removal of PAHs

There are various methods used for the removal of PAHs, including physical, chemical, biological, combined, ultrasound treatment [11], activated sludge process [20], coagulation and flocculation, membrane filtration, adsorption [21], advanced oxidation process [22], and Fenton process [23]. Figure 2 shows the different methods for PAHs removal. Table 3 shows the source of a water sample, the efficiency of removal, and various methods for removing PAHs.

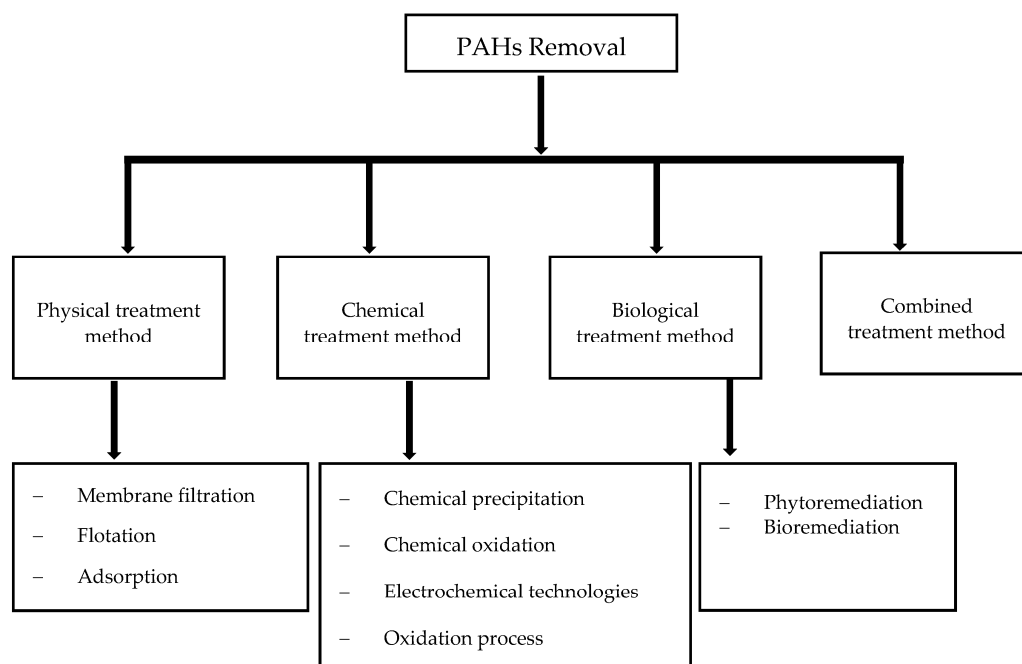


Figure 2. Various methods of removal of PAHs from produced water.

Table 3. Various methods for removal of polycyclic aromatic hydrocarbons.

PAHs	Method	Efficiency of Removal	Sample	Source
Naphthalene	Ultraviolet irradiation	62%	Sea water	[2]
Pyrene & benzo (a) pyrene	Adsorption	40% & 48 %	Synthetic wastewater	[24]
16 PAHs	Photocatalyst ozonation & ultraviolet light-emitting diode irradiation	57%	Offshore produced water	[25]
Naphthalene, phenanthrene, anthracene & pyrene	Bioremediation	100%, 95.4%, 73.8% & 53.4% respectively	Oil fields produced water	[26]
Naphthalene & acenaphthene	Adsorption	100% to 97%	Water treatment plant	[27]
16 PAHs	Ozonation & biological approach	42% to 63%	Urban runoff water	[28]
Anthracene, phenanthrene & fluoranthene	Fenton process	85.47%, 63.16% & 62.95 respectively	Textile dying sludge	[29]

Table 3. Cont.

PAHs	Method	Efficiency of Removal	Sample	Source
Phenanthrene, pyrene, & benzo (a) pyrene	Degradation and sorption	80% to 65%	Oil polluted sediment	[30]
16 PAHs	Phytoremediation	89%	Wastewater	[31]
16 PAHs	Biodegradation	67.27%	River	[32]
Pyrene, benzo (a) pyrene & indenopyrene	Magnetic floatation	89.9%, 66.9% & 78.2 respectively	Sea water	[33]
16 PAHs	Biodegradation	77.38%	Agricultural soil	[34]
Phenanthrene, naphthalene, anthracene & benzo (a) pyrene	Oxidation	90.1%, 97.5%, 55.4% & 26.7% respectively	Soil	[35]
Naphthalene, phenanthrene, anthracene biphenyl & fluorene	Air-assisted liquid-liquid microextraction	82.0% to 116.6%	Water	[36]
Phenanthrene	Adsorption	90%	Wastewater	[37]
Naphthalene & fluorene	Oxidation adsorption	92% to 100%	Produced water	[38]
Pyrene, fluoranthene, chrysene & phenanthrene	Precipitation method	99%, 98%, 87% & 97% respectively	Marine sediments	[39]
Naphthalene, anthracene & fluorene	Oxidations	97%, 95%, & 87% respectively	Landfill leachate	[17]
Benzo (b) fluoranthene	Adsorption	59% to 91%	Water in the treatment cycle	[40]
15 PAHs	Dispersive liquid-liquid microextraction	>90%	Four Rivers and tape water	[41]
PAHs	Electrochemical advanced oxidation	99.9 %	Petroleum contaminated water	[42]
Low & high molecular weight PAHs	Biodegradation	86%	Petroleum wastewater	[43]

3.1. Physical Treatment Method

Physical treatment methods can remove coarse particles and organic and inorganic contaminants from wastewater. Physical treatment is often used as a pretreatment step before applying advanced technologies [44]. Various physical treatment technologies exist, including membrane filtration, flotation, and adsorption. These technologies are discussed in more detail below.

3.2. Membrane Filtration

Membrane filtration is a process in which water and wastewater are treated with a porous filter media. The filter media only allows water to pass and captures impurities through it. Many materials are used as filter media, such as activated carbon, sand, and crushed stone. However, sand is more widely used due to its easy availability, removal efficiency, and low cost. Membrane-based processes have been recognized as 21st-century pretreatment approaches for removing PAHs from produced water. The membrane removes PAHs such as ultrafiltration, microfiltration, nanofiltration, and reverse osmosis. Additionally, the combination of membrane-based processes has been considered a successful operation for removing PAHs [45]. Nanofiltration technology has attracted more attention in the removal of organic micropollutants from water and wastewater. This is because it offers maximum rejections of organic micropollutants but minimum rejections of monovalent salts. However, the size of salt elimination has been widely recognized as the main mechanism of micropollutant removal by nanofiltration [46]. In wastewater, the removal efficiency of the three low-concentration PAHs (phenanthrene, fluoranthene,

and pyrene) was studied in a membrane bioreactor equipped with ultrafiltration membranes. The removal efficiency for all three PAHs was found to be 91%, 82%, and 92%, respectively [47].

A pilot-scale hollow fiber ultrafiltration polytetrafluoroethylene membrane filtration system was studied for the treatment of two types of oily seawater contaminated with light and heavy crude oil. The effect of aeration flow rate and membrane flux on the performance of the membrane and the removal efficiency of various PAHs were studied. The results showed that a removal efficiency of greater than 91% was achieved from both types of oily seawater [48]. Another study was conducted to remove 16 PAHs from the municipal wastewater treatment system. A removal efficiency of 40% to 60% was obtained [49]. The treatment of municipal landfill leachate by reverse osmosis, 59% to 72% removal efficiency was achieved for all 16 PAHs [50]. In another study, the reverse osmosis low-pressure method was used to remove 50%, 85%, 88.3%, and 91% of acenaphthene, phenanthrene, fluorene, and anthracene from wastewater [51]. The ultrafiltration process was used to remove all 16 PAHs, and a removal efficiency of 66.6% to 85.0% was achieved using industrial wastewater [52]. Additionally, naphthalene was removed by 95% using nanofiltration in an acidic solution [53].

Cold plasma surface treatment is used to produce free radicals of oxygen on the surface of the polyethersulfone (PES) membrane. These free radicals serve as indicators to produce a polyacrylic acid (PAA) implant through polymerization, forming a PAA layer on the membrane surface. The PES membrane is then coated with nanoparticles of ZrO_2 through chemical bonding. This results in a super-wetting PES-PAA- ZrO_2 nanofiltration membrane, which can be used to remove PAHs (such as phenanthrene). The removal efficiency of phenanthrene was found to be 96%. After surface modification, the membrane efficiency increased fourfold [54].

3.3. Flotation

The flotation method is widely used due to its high separation rate in short residence periods and low operating costs. In the flotation process, the oil removal efficiency in water effluent is about 99% [55]. The flotation process depends on the relation of gas bubbles to dispersed oil droplets. This relation relies on a multifaceted process involving the oil droplets' surface characteristics and interfaces with gas. The attachment mechanisms involve the relation of the bubble or oil contact, the relations of chemical stabilizers in aiding this contact, and the distribution of the oil around the gas bubble [56]. The process's effectiveness is mainly affected by the impurities to be eliminated, the differences in liquid densities, the temperature, and the size of the oil droplet. The flotation technique can be used to treat generated water with maximum and minimum total organic carbon (TOC) concentrations, as well as water with oil, grease, and nanoparticles that are less than 7% solids. However, it does not function properly with high-temperature feed streams [57].

The dissolved air flotation technique can remove particles larger than 25 μm , and when using coagulation as a pretreatment step, pollutants larger than 3–5 μm can also be eliminated. Utilizing the flotation technique, up to 93% of the oil was removed [58]. Furthermore, when oilfield-generated water with a TDS of 20,000 mg/L was pre-treated using the induced-air flotation method, it was discovered that the COD and TOC levels dropped to 595 mg/L and 115 mg/L, respectively [59]. Therefore, flotation is an efficient method for the removal of oil from produced water. It is robust, durable, and easy to operate. The turbidity of oily produced water can be controlled by the gas flotation method, and oil concentration was also reduced to lower than 40 mg/L [56]. A study was conducted on the removal of PAHs from oil field-produced water by flotation. The researchers examined the significance of the conditioning process and the impact of adding ethanol and Tween 80, which might reduce the flotation time from 30 min to 10 min. It was demonstrated that the effect of natural NaCl on bubble size improved flotation effectiveness [60]. Another study was conducted to create microbubbles using graphene oxide (GO) coating to improve the flotation method's ability to remove PAHs from aqueous solutions. The flotation

approach was then applied to improve the removal of phenanthrene, a low molecular weight PAH, and pyrene, a high molecular weight PAH, from aqueous solutions. To improve the PAH removal effectiveness of GO-coated microbubbles, a closed-loop research lab flotation system was constructed. The elimination efficiencies of phenanthrene and pyrene considerably increased to 64.77% and 74.01%, respectively [61]. Recent studies have shown that high removal efficiency for a variety of contaminants present in aqueous solutions can be achieved through flotation using colloidal gas aphrons (CGAs).

The efficiency of CGAs in eliminating low-ring PAHs, such as naphthalene, phenanthrene, and fluorene, from aqueous solutions was investigated by adjusting several parameters, including surfactant type, pH of the PAHs solution, CGAs flow rate, and temperature of the PAHs and CGAs solution. The effectiveness of PAHs removal was evaluated, and the results revealed that using CGAs (flow rate of 100 mL/min) generated from a cationic surfactant, fluorene, and phenanthrene were completely removed at pH 5.6 and 25 °C, whereas naphthalene was removed to a degree of roughly 88.4% under identical circumstances [62]. Therefore, this method can be used for produced water treatment, and substances such as volatile organic carbons, suspended solids, PAHs, and oil content can be removed without using any further chemicals.

3.3.1. Adsorption

Adsorption is a recognized effective technique for removing PAHs from produced water. Carbon materials such as biochar, activated carbon, graphene, and carbon nanotubes are used as adsorbents. Biochar is a carbonaceous solid derived from petroleum sludge, wastewater sludge, and agricultural waste. Biochar produced during pyrolysis contains an aromatic carbon skeleton like graphene and has a high potential for the absorption of PAHs and organic contaminants [18]. Activated carbon is a porous carbon material mainly generated from agricultural waste such as rice husk, palm or coconut shells, sawdust, and wood. Agricultural waste palm shells and nano magnesium oxide obtained from neem leaf extract are used to prepare nanocomposites that are very effective for removing anthracene, with about 95% removal observed by statistical analysis [63]. Graphene is a nanomaterial with a honeycomb structure in which carbon atoms are arranged in a single graphite layer. Graphene oxide and reduced graphene are the most advanced forms used for pollutant remediation in water [16]. Reduced graphene oxide (rGO) was prepared using graphene synthesis with Terminalia chebula seed extract, and the rGO with oxidized polyphenol was stabilized to form graphene sheets [64]. Graphene and graphene oxide are more efficient at adsorbing PAHs such as naphthalene and phenanthrene [65]. The adsorbent, adsorption model, and adsorption capacity for the removal of PAHs from water sources are shown in Table 4 below.

Table 4. Capacity for adsorption for removal of PAHs from water sources.

PAHs Compound	Adsorbent	Adsorption Model	Adsorption Capacity	Source
Pyrene, naphthalene, phenanthrene, & acenaphthylene	Waste tire crumb rubber, coconut coir fibre & blast furnace slag	Linear, Freundlich, isotherm & Pseudo second order kinematic model	-	[66]
Naphthalene, phenanthrene, & acenaphthene	Rape straw & corn cob	Kinematic model & isotherm model	592.97 mg/g, 480.27 mg/g & 692.27 mg/g	[67]
Benzo (a) pyrene & pyrene	Iron oxide nanoparticles	Pseudo second-order kinematic model	0.96 mg/g & 0.99 mg/g	[68]

Table 4. Cont.

PAHs Compound	Adsorbent	Adsorption Model	Adsorption Capacity	Source
Naphthalene, phenanthrene, acenaphthene & pyrene	Bamboo wood, pine wood, pine needles & pine bark	Pseudo second-order kinematic model	0.008 mg/L to 1 mg/L	[69]
Acenaphthene	Granular activated carbon	Langmuir isotherm	-	[70]

Nowadays, carbon nanoparticles are gaining more importance due to their environmentally friendly and low-cost treatment methods [71]. It has been observed for the past few decades that the concentration of PAHs is increasing daily in water and soil. Carbon nanomaterials used as a catalyst can more efficiently remove organic compounds [72]. Nanoparticles are mostly used in recent times for the treatment of produced water and are more efficient in removing PAHs [73]. The Neem tree is easily available all over the world and has a highly biogenic potential for the synthesis of nanoparticles [72]. Through a literature review, it is observed that a nanocomposite iron oxide of chitosan is more efficient in removing PAHs such as anthracene (95%) and phenanthrene (92%) through adsorption [74]. Silver nanoparticles synthesized with garlic plant extract exhibit a removal efficiency of about 85% for each PAH, such as phenanthrene, anthracene, and pyrene [75]. The magnetic core-shell Fe_3O_4 nanoparticles are shown to be good adsorbents for removing PAHs, such as fluoranthene, benzo(a)pyrene, and pyrene polyaniline, through van der Waals and π - π interactions. The adsorption capacity of PAHs is determined through the Pseudo-second-order kinematic model and Langmuir isotherm model [76].

Various plant extracts, such as *Coriandrum sativum*, *Aloe barbadensis*, and *Azadirachta indica*, have been used to synthesize silver and copper nanoparticles for the removal of PAHs through adsorption. *Azadirachta indica* nanoparticles showed the highest removal efficiency rate for PAHs, with 98.81%, while *Coriandrum sativum* had the lowest rate at approximately 95.29% [77]. Agricultural residues, such as green coconut shell, bagasse of sugar cane, chitosan, and chitin, have also been used as natural, low-cost adsorbents for the removal of PAHs, including anthracenes, naphthene, pyrene, and acenaphthene, from wastewater. The Freundlich model was used to analyze the adsorption of PAHs by these materials [78]. The preparation of nanoparticles using plant extracts and chemical compounds, as well as the size and shape of the nanoparticles, and their application in the removal of PAHs and other metals have been studied. The characterization techniques used include energy-dispersive spectroscopy (EDS), scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier-transform infrared (FTIR) spectroscopy, transmission electron microscopy (TEM), response surface methodology (RSM), Brunauer-Emmett-Teller (BET), vibrating sample magnetometer (VSM), dynamic light scattering (DLS), and X-ray photoelectron spectroscopy (XPS). These techniques are summarized in Table 5.

Table 5. Plant extract with a chemical compound is used in the preparation of nanoparticles.

Plant Name	Chemical Compound Used for Nanoparticles	Nanoparticles Size/Shape	Application	Characterization Technique	Source
Eucalyptus leaves extract	Ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$)	Spheroid with diameter 20–80 nm	Removal of Nitrogen, phosphorus & chemical oxygen demand	EDS, SEM, XRD & FTIR	[79]
Castanea sativa, eucalyptus globulus, ulx europaeus & pinus pinaster extract	Iron (III) nitrate nine hydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) with Chitosan matrix	Beads with a diameter of 01 mm	Removal of Arsenic (V)	-	[80]

Table 5. Cont.

Plant Name	Chemical Compound Used for Nanoparticles	Nanoparticles Size/Shape	Application	Characterization Technique	Source
Mint plant leaves extract	Ferric nitrate (Fe NO_3) ₃	Diameter 20–45 nm with face centre cubic	Removal of Arsenic (III), Arsenic (V)	TEM, EDX, XRD & FTIR	[81]
Green, black & oolong tea leaves extract	FeSO_4 solution	Spherical with particle size 20–40 nm	Removal of Monochloro benzene & chemical oxygen demand	SEM, XRD & EDS	[82]
Citrofortunellamicrocarpa Leaves extract	$\text{Cu (NO}_3)_2 \cdot 3\text{H}_2\text{O}$	Spherical with particles size 54–68 nm	Removal of Rhodamin (B)	XRD, EDS, SEM & FTIR	[83]
Citrus aurantifolia leaves extract	Copper sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$)	Crystalline & average size approximately 22 nm	Removal of Rhodamin (B) & bacteria (<i>S. aureus</i> & <i>E. coli</i>)	XRD, EDS & FTIR	[84]
Pisum sativum peel extract	Ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$)	Spherical & particle size 20–30 nm	Removal of Methyl orange	XRD, FTIR, TEM, BET, & RSM	[85]
Amaranthus dubius leaves extract	FeCl_3 & NaBH_4	Spherical with cubic shape & diameter 43–220 nm	Removal of Methyl orange 1-diphenyl-2-picrylhydrazyl	XRD, SEM & FTIR	[86]
Peltophorumpterocarpum leaves extract	Zinc acetate dehydrate	Average crystalline & size 11.64 nm	Removal of Methylene blue	XRD, EDS, SEM & FTIR	[87]
Pomegranate leaves extract	FeCl_3	Rod shape & average size 45–60 nm	Removal of Congo red	XRD, SEM, FTIR & EDS	[88]
Pomegranate leaves extract	Ammonium ferrous sulfate and ammonium ferric sulfate	Diameter 100–200 nm	Removal of Chromium (V)	XRD, SEM, VSM & FTIR	[89]
Syzygiumcumini seed extract	$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ & sodium acetate	Crystalline & size approximately 14 nm	-	XRD, SEM, EDS, VSM, & FTIR	[90]
Vitex trifolia leaves extract	Zinc nitrate hexahydrate	Spherical & size 15–46 nm	Removal of Methylene blue	XRD, SEM, EDS & FTIR	[91]
Aegle marmelos leaves extract	Nickel nitrate, copper nitrate & chromium nitrate	Nanorods, nanosphere & nanoflower with an average size of 50 nm	Removal of phenol, 2, 4-dinitrophenol & 3-aminophenol	EDS, TEM & SEM	[92]
Eucalyptus leaves extract	Ferric chloride (FeCl_3) & Graphene oxide	Spherical with a diameter of 4–7 nm	Removal of Methylene blue	XPS, EDS, TEM & FTIR	[93]
Green tea extract	Natural Bentonite & ferrous sulfate heptahydrate	Spherical with an average diameter of 40–60 nm	Removal of Phosphorous	TEM, XRD & FTIR	[94]
Cyanometraramiflora leaves extract	Zinc acetate	Hexagonal wurtzite crystalline with a size of 13.33 nm	Removal of Rhodamine (B)	TEM, XRD, FTIR, EDS & BET	[87]
Garlic vine leaf extract	$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	Crystallite with size 13.82–15.45 nm	-	XRD & FTIR	[95]
Terminalia chebula extract	$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ & PdCl_2	Amorphous iron with a size less than 80 nm & cubic palladium with a size less than 100 nm	-	XRD & TEM	[96]
Eucalyptus globulus leaf extract	$\text{FeSO}_4 \cdot 7 \text{H}_2\text{O}$	Spherical with size 50–80 nm	Removal of Chromium (VI)	TEM, XRD & FTIR	[97]
Acalypha Indica leaves extract	Copper Sulphate & Graphene oxide	-	Removal of Methylene blue	TEM, XRD, FTIR & EDX	[98]
Sapindus-mukorossi extract	$\text{K}_4(\text{Fe}(\text{CN})_6)$ & $\text{Fe}(\text{NO}_3)_3$	Hexagonal, spherical & rod with size 10–60 nm	Removal of PAHs	XRD, TEM & SEM	[99]

Table 5. Cont.

Plant Name	Chemical Compound Used for Nanoparticles	Nanoparticles Size/Shape	Application	Characterization Technique	Source
Penicillium expansum	FeCl ₃ ·6 H ₂ O	Spherical with size 15–66 nm	Removal of heavy metals such as Cobalt, lead, chromium, nickel, cadmium, chemical oxygen demand, total dissolved solid, & total suspended solids	TEM, XRD, FTIR, XPS & DLS	[100]
Spent tea leaves	Tea filter bag polyethylene & polypropylene	250–211 µm diameter	Removal of PAHs	EDX, SEM & FTIR	[101]
Amaranthus blitum leaves extract	Fe(NO ₃) ₃ ·9 H ₂ O & AgNO ₃	Spherical with an average size of 92 nm	Removal of caffeine	XPS & SEM	[102]
Oolong tea extract	Ferrous Sulphate	Spherical with a diameter of 40–50 nm	Removal of malachite green	EDX, SEM, XRD & FTIR	[103]
Aloe barbedensis, Azadirachta indica & Coriandrum sativum plant extract	Silver nitrate & copper sulphate	-	Removal of PAHs	FTIR	[77]
Neem leaves extract	Mg(NO ₃) ₂ & palm shells	Average diameter 10 µm	Removal of anthracene	SEM, XRD & FTIR	[63]
Allium tricoccum extract	FeCl ₂ , FeCl ₃ & TiO(OH) ₂	Spherical with size 40–90 nm	Removal of PAHs	EDX, SEM, XRD & FTIR	[104]
Green tea extract	FeSO ₄ ·7 H ₂ O & graphene oxide	Spherical with particles diameter of approximately 4–15 nm	Removal of nitrate	XPS, TEM & FTIR,	[105]
Pomegranate peel extract	FeSO ₄ solution	Amorphous with an average particle size of 2.7 nm	Removal of benzo (a) pyrene & pyrene	EDX, SEM, XRD & FTIR	[68]

3.3.2. Chemical Treatment Method

Various chemical methods are used to treat produced water through chemical reactions, including chemical precipitation, chemical oxidation, electrochemical technologies, and advanced oxidation processes. An appropriate water treatment method for produced water should remove dispersed oil, total dissolved solids, suspended solids, dissolved gases, radioactive materials, chemicals, salts, ammonia, and biological oxygen demand [106]. Since produced water is a complex mixture, a combination of different approaches is often used to achieve maximum water quality [107]. The current advancements in the most common chemical treatment methods, including chemical precipitation, chemical oxidation, electrochemical technologies, and advanced oxidation processes, are discussed in the following subsections.

3.3.3. Chemical Precipitation

Coagulation and flocculation methods are effective in removing colloidal and suspended matter from wastewater, but not dissolved constituents [108]. In these methods, a flock of commonly used aluminum and ferrous is produced by the electro-dissolution of the anode, and hydroxyl and hydrogen ions are generated at the cathode [109]. A study was conducted using coagulation and flocculation methods in semi-synthetic stormwater. Five coagulants were evaluated based on their ability to remove particles, total and dissolved metals, organic carbon, and PAHs. The coagulants were found to remove more than 90% of the organic content and PAHs [110].

In another study, the removal of PAHs from water was achieved using adsorption in various nano and organo-modified nano-clays, followed by coagulation-flocculation using

alum and poly aluminum chloride (PAC). The combination of clay minerals, alum, and PAC resulted in the elimination of PAHs ranging from 37.4% to 100% [111]. Gong et al. [112] investigated the relationship between PAH removal and transformation during electro-coagulation treatment of paper-making wastewater effluent and found that 86% of the PAHs were successfully removed. In a separate study, the influence of the type and dose of coagulants in the coagulation process for the removal of 16 PAHs was investigated. The removal efficiency of benzo(a)pyrene was 83%, while efficiencies for the remaining 15 PAHs ranged from 80% to 91%. Increasing the dose of coagulant enhanced the water purification performance, including the removal of PAHs [113].

3.3.4. Chemical Oxidation

Chemical oxidation processes are used to oxidize pollutants and reduce their toxicity. Substances such as sodium persulfate, potassium permanganate, modified Fenton (Fe), and conventional Fenton are used to remove PAHs from water [12]. However, during the oxidation of PAHs using sodium hypochlorite as an oxidant, chlorine oxidation products are formed, making this reagent unsuitable [114]. Potassium permanganate is a chemical oxidant that can effectively remove PAHs, with an efficiency of up to 70% [115].

In a study, a water sample containing the sum of 16 PAHs was collected from a municipal treatment plant with a concentration of 1.39 g/L, and the chemical oxidation process was used to decrease the PAH concentration in the sample. The addition of hydrogen peroxide caused the level of PAHs in wastewater to drop by 51% to 59%. Different removal efficiencies were observed for mobile hydrocarbons when grouped by the number of rings, ranging from 0% to 64% [116].

The effective liquid oxidants were determined based on favorable reactant dosages during tests against the greatest permissible concentration of 16 PAHs. The removal efficiency of PAHs using KMnO_4 , $\text{Na}_2\text{S}_2\text{O}_8$, conventional Fenton, and modified Fenton reagents ranged from 5.3% to 62.8%, 10.7% to 39.1%, 0% to 30.9%, and 31.5% to 68.3%, respectively [117]. In another study, a catalytic oxidation procedure was used to remove specific PAHs from coking effluent, where dihydrogen dioxide was used in the presence of cobalt and platinum catalysts. The maximum relative efficiency of 63% was observed in the removal of 3-ring PAHs [12].

3.3.5. Electrochemical Technologies

The electro-coagulation process is an effective technique for removing ionic metals from wastewater. A study was conducted using three different anode geometries for the removal of PAHs (naphthol) and investigated using different parameters such as pH, current density, electrolyte concentration, and electrolysis time [118]. Highly efficient procedures, including immersion, painting, and electrophoresis, were used to create dimensionally stable anodes (DSAs). Analysis of the coating's surface and electrochemical composition was performed to determine its quality. It was found to produce hydroxyl radicals and break down hydrocarbons such as naphthalene, fluoranthene, and phenanthrene in aqueous solutions.

The efficiency of such a DSA electrode for electro-oxidation of PAH aqueous solutions is quite encouraging, as it enabled achieving >60% degradation within short durations and >67% mineralization after many hours [119]. Using a Ti/IrO₂ anode, electrochemical oxidation studies were carried out to degrade 16 essential PAHs. Degradation studies were conducted in a batch reactor at a laboratory scale. The electrochemical method was varied on three independent variables: pH level, current densities, and electrolysis time to remove PAHs. The maximum elimination observed was 95.29% within optimal conditions [120]. The surface morphology of Ti/RuO₂ anodes coated with three different concentrations of $\text{RuCl}_3 \cdot \text{H}_2\text{O}$ solutions is shown in this study. Therefore, the more efficient anode was used for the aqueous solution degradation of 16 main PAHs. About 74.82%, 68.94%, and 75.68% of PAHs removal were achieved without the use of electrolytes at pH three, six, and nine, respectively [121].

In another study for the degradation of PAHs, the coated electrodes were used as working electrodes. Although the electrodes made from precursor solutions containing 30% and 100% Ir showed similar deterioration, their service periods were found to be less than 2 h. Only a Ta-containing precursor was used to cover the electrode, and despite this, it was able to remove some pollutants through direct anodic oxidation of PAHs [122]. Similarly, a batch recirculation reactor made up of a Ti/Ir₂ anode and a stainless steel cathode was used in the study to assess the impact of operational parameters on the electrochemical oxidation of naphthalene, phenanthrene, and organic matter in synthetic wastewater. Naphthalene and phenanthrene had the highest removal efficiency, at 91% and 95%, respectively [123].

3.3.6. Advanced Oxidation Process

The concept of advanced oxidation processes (AOPs) was developed in the 20th century. AOPs are used to treat oil and gas-produced water to remove organic and inorganic compounds, as well as color and odor. The common oxidants in AOPs include ozone, chlorine, hydrogen peroxide [124], and Fenton reunions, such as ultraviolet/Hydrogen peroxide/Fe⁺³ [125]. AOPs and catalytic oxidation processes are valuable techniques for removing polycyclic aromatic hydrocarbons (PAHs). Based on this technique, catalytic oxidation is used to treat PAHs from wastewater. The average removal efficiency of municipal wastewater treatment plants is 81% to 92.2%, and AOPs have an efficiency range of 32% to 99%. Overall, it is observed that AOPs are the most reliable process using catalysts [12]. The oxidation of PAHs removal is carried out with molecular ozone, hydroxyl radicals, and other reactive species for oxidizing. Ozone captures the aromatic rings with its electrophilic nature, and the reaction of radicals is non-discriminatory [126]. The Fenton reaction is traditionally a mixture of hydrogen peroxide and salt, which is used as a fast reaction for decomposing contaminated organic matter from wastewater [127]. The methods of oxidation, source of PAHs, and removal efficiency are shown in Table 6.

Table 6. Methods of oxidation for removal of PAHs from water sources.

PAHs	Water Sample	Method of Oxidation	Removal Efficacy	Source
Point source PAHs	Cooking wastewater treatment plant	Ozone & ultraviolet	75%	[128]
8PAHs	Coagulant water, Electro coagulated water & groundwater	Hydrogen peroxide & ultraviolet	76%, 70% & 76%	[129]
Fluorene, dibenzofuran & dibenzothiophene	Treated water	Hydrogen peroxide & ultraviolet	98% to 99%	[130]
15 PAHs	Produced water	Fenton reaction	89.73%	[131]
Hydrocarbons	Synthetic wastewater	Hydrogen peroxide & ultraviolet	90%	[132]
6 PAHs	Water treatment	Xenon & Hydrogen peroxide	100%	[14]
6 PAHs	Landfill leachate	Fenton oxidation & ozone oxidation	70%	[133]

3.4. Biological Treatment Method

PAHs can be biodegraded aerobically and anaerobically. Various biological treatment methods are used for the removal of PAHs, such as activated sludge processes, bioreactors, sequencing batch reactors, membrane bioreactors, phytoremediation, bioremediation, innovative microbial capacitive desalination cell, biological aerated filter, and microalgae-based processes. Among these, sequencing and membrane bioreactors are most commonly used to remove organic pollutants. During biological treatment, low molecular weight organic pollutants are more easily biodegraded [11]. In a study of PAH biodegradation,

an association of *Pseudomonas putida*, *Pseudomonas aeruginosa*, and *Flavobacterium* exhibited maximum removal of less water-soluble PAHs than strains in pure culture [134]. The study also tested a microbial consortium with stimulating phenanthrene and other PAH degradation abilities [135]. The biological method, source of PAHs, and removal efficiency are shown in Table 7.

Table 7. Biological methods for removal of PAHs from water sources.

PAHs	Water Sample	Biological Method	Removal Efficiency	Source
16 PAHs	Wastewater treatment plant	anaerobic-anoxic-oxic biological treatment	99% to 100 %	[20]
Naphthalene, phenanthrene, acenaphthene, fluoranthene & pyrene	Real sample	Green biomaterial sorbent	76.20% to 105.60%	[136]
16 PAHs	Marine sediment	Biodegradation	42% to 77%	[137]

Phytoremediation and Bioremediation

Phytoremediation and bioremediation involve the use of green plants, microorganisms, and enzymes to treat polluted substrates and restore them to their natural state without causing further environmental damage. Bioremediation or green remediation is considered a sustainable, eco-friendly, and more cost-effective technology for environmental cleanup [138]. Phytoremediation is a developing green technology in which plants are grown in the presence of contaminated soil, surface water, or groundwater to enhance the degradation or removal of organic and inorganic contaminants. Some plants are more effective and suitable for phytoremediation [139]. Bioremediation involves the biological restoration of environmental contaminants through metabolic processes that produce cell biomass, carbon dioxide, and water as byproducts. In the mid-20th century, microbial-based bioremediation was reported to remove PAHs in oil regions [140]. The plants used for PAHs removal and the presence of bacteria or substrate are shown in Table 8. A summary of the advantages and disadvantages of all methods for removing PAHs from produced water is presented in Table 9.

Table 8. Plants used for the removal of PAHs.

PAHs	Sample	Plant	Presence of Bacteria/Substrate	Removal Efficiency	Source
5–6 ring PAHs	Gas plant soil	Cucumber	<i>Cucurbita species</i>	85%	[141]
Phenanthrene	Ever bright water treatment plant	<i>Arundo donax</i>	Proteobacteria, Bacteroidetes, Chloroflexi, Actinobacteria & Firmicutes	94.09%	[142]
16 PAHs	Soil	Sudan grass	<i>Mycobacterium vanbaalenii</i> & bacterial consortium	98%	[143]

Table 9. Summary of advantages and disadvantages all methods of PAHs removal from produced water.

Treatment Method	Advantages	Disadvantages	Source
Membrane filtration	Economical, less chances of membrane fouling, condensed modules, suitable for saline water	Mineral scaling, membrane pore wetting and membrane fouling	[144–146]

Table 9. Cont.

Treatment Method	Advantages	Disadvantages	Source
Flotation	Simplicity of operation, amalgamation increase the process efficiency, robust and durable, and has no moving parts	4 to 5 min retention time, maximum amount of air is produced, and skim volume	[108]
Adsorption	Low capital cost, condensed modules, ecofriendly, flexible process and reusable and recoverable adsorbent	Frequent regeneration needed, affected by pH, salinity, high temperature, retention time maximum, expensive adsorbent restoration, and harmful excess adsorbent	[147,148]
Chemical precipitation	Energy saving process, easy to operate, low cost and maximum recovery	Requirement of chemicals, generation of sludge, and secondary waste	[149]
Chemical oxidation	Small treatment time and ecofriendly	Operation and maintained cost maximum	[76]
Electrochemical technologies	Beneficial secondary product, eco-friendly and no chemicals required	Skillful labor necessary and scaling up difficulties	[150]
Advanced oxidation process	Easy operation, high degradation, dissolve oil mineral and compact	Skillful labor required, optimization, monitoring and pretreatment process required	[151]
Bioremediation	Availability of low cost microbes, easy process, whole mineralization leads to production of CO ₂ , H ₂ O and biomass	Lengthy degradation time and time optimization an excessive task	[152]

3.5. Combined Treatment Method

Integrated physical, chemical, and biological approaches have shown promising results in effectively degrading, solubilizing, and comprehensively removing several high molecular weight PAHs from water. Two methods used to remove PAHs, such as naphthalene and phenanthrene, from aqueous solutions are the biodegradation process and the Fenton oxidation process. The biodegradation process employs a *Bacillus fusiformis* (BFN) strain isolated from activated sludge, while the Fenton oxidation process is based on tea extract synthesizing ferrous nanoparticles under different atmospheres, such as oxygen, nitrogen, and air [153]. Biologically active filtration is a collective biological and physical process used for treating oil and gas wastewater, which takes advantage of the nutrients and microorganisms previously present in the rivers while supporting the growth of biofilm that can withstand extreme or variable concentrations of total dissolved solids [154]. Biologically active filtration with granular activated carbon is a recognized effective technology for removing organic pollutants and solids. The spent granular activated carbon with extant biofilm rapidly adapts to produced water. It has been observed that up to 92% of dissolved organic carbon and 81% of chemical oxygen demand are removed within 24 h [155].

4. Conclusions

A large amount of produced water is generated during oil and gas processing, which is toxic and can have negative impacts on the environment and human health. As a result, there has been a recent need for attention to produced water treatment methods. Due to its hazardous nature, the water contains compounds such as various organic, inorganic, and PAHs. Therefore, many physical, chemical, biological, and combined methods are used to remove PAHs from produced water. Adsorption by nanoparticles and integrated methods are promising techniques to meet strict regulatory limits with advanced potential

for commercialization. In the case of global water scarcity, this presents a new source of water that can be used.

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