Wasit Journal of Engineering Sciences

Journal homepage: https://ejuow.uowasit.edu.iq

# Treatment Technologies for Removal of Polyaromatic Hydrocarbon Compounds from Wastewater: A Review

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**Accepted**
5-January-2024

**Doi:**
**10.31185/ejuow.Vol12.Iss1.518**

**Abstract**

Industry discharges, home effluents, and/or urban runoff can all introduce polyaromatic hydrocarbon compounds (PAHs) into a treatment center. Regrettably, due to their chemical stability and resistance to biological degradation, PAHs are regarded as organic pollutants that persist. As a consequence, PAHs have indeed been identified as a possible source of skin, lung, and bladder cancer in humans. Due to PAHs' potential to cause cancer and/or mutation, their residues in treated wastewater are now subject to stricter legal regulation. adsorption ,membrane filtration, coagulation, electrocoagulation, flocculation, advanced oxidation reactions, phytoremediation and bioremediation process are some of the techniques used to remove PAHs from wastewater. In order to reduce environmental contamination, it is important to study different treatment technologies to treat wastewater. The aim of this paper is to highlighting and discussing some of treatment technologies for removing PAHs.

**Keywords:** Wastewater, Polyaromatic Hydrocarbon Compounds (PAHs), Treatment technologies, Treated water water.

**الخلاصة:**

تصريفات الصناعة ، والنفايات السائلة المنزلية، و الجريان السطحي في المناطق الحضرية إلى إدخال مركبات هيدروكربونية متعددة العطرية (PAHs) إلى مركز المعالجة. لسوء الحظ، نظرًا لثباتها الكيميائي ومقاومتها للتحلل البيولوجي، تعتبر الهيدروكربونات العطرية متعددة الحلقات من الملوثات العضوية التي لا تزال قائمة. ونتيجة لذلك، حدد الهيدروكربونات العطرية متعددة الحلقات كمصدر محتمل لسرطان الجلد والرئة والمثانة لدى البشر. نظرًا لاحتمال تسبب الهيدروكربونات العطرية متعددة الحلقات في الإصابة بالسرطان و/أو الطفرات، فإن بقاياها في مياه الصرف الصحي المعالجة تخضع الآن لتنظيم قانوني أكثر صرامة. الامتزاز، والترشيح الغشائي، والتخثر، والتخثير الكهربائي، والتلبد، وتفاعلات الأكسدة المتقدمة، والمعالجة النباتية، وعملية المعالجة البيولوجية هي بعض التقنيات المستخدمة لإزالة الهيدروكربونات العطرية متعددة الحلقات من مياه الصرف الصحي. ومن أجل الحد من التلوث البيئي، من المهم دراسة تقنيات المعالجة المختلفة لمعالجة مياه الصرف الصحي. الهدف من هذه الورقة هو تسليط الضوء ومناقشة بعض تقنيات المعالجة لإزالة الهيدروكربونات العطرية متعددة الحلقات.

**الكلمات المفتاحية**: المياه العادمة، المركبات الهيدروكربونية متعددة العطريات (PAHs)، تقنيات المعالجة، المياه المعالجة.

## 1. INTRODUCTION

The environment has been impacted by industrial changes over the past few years. One of the most numerous resources found anywhere on earth, water is used extensively for many industrial processes, yet only around 1.1% of it is safe to consume by humans. Unfortunately, both inorganic and organic contaminants like aromatic polycyclic hydrocarbons (PAHs) pose a persistent danger to the 1% that is now available. Massive amounts and a wide variety of chemicals are produced by industrial production as well as a human activity, seriously polluting the environment. Due to the formation of oxidative DNA lesions and covalent DNA adducts, PAHs have carcinogenic and mutagenic effects [1]. Moreover, they possess negative long-term impacts and are persistent in the environment. As a result, numerous nations have created laws and methods for regulating PAHs. PAHs are cyclic organic pollutants that cause cancer and have two or even more fused aromatic rings. Additional to oil spills, incomplete combustion reactions of many organic compounds, such as coal, gasoline, oil, wood, natural gas, and some other petroleum products, also released PAHs into the atmosphere [2].

PAHs are regarded as organic pollutants (POP) that persist [1]. As a consequence, PAHs have indeed been identified as a possible source of skin, lung, and bladder cancer in humans. Human illnesses including reproductive defects, PAH-DNA adducts in newborns, premature birth, and restrictions on intrauterine growth have all been linked to PAHs in the environment. As a result, even though PAH is related to many serious health issues, cancer still commands the majority of public perception [2,3].

# 2. EXPERIMENTAL WORK

## 2.1 Method: Research Design

Physical, chemical, and biological , such as adsorption ,membrane filtration, coagulation, electrocoagulation, , flocculation, advanced oxidation reactions, phytoremediation and bioremediation process are some of the techniques used to remove PAHs [4,5].

## 2.2 Physical treatment

Large particles, organic and inorganic pollutants, and coarse particles can all be removed from wastewater using physical treatment methods. Before using modern technology, physical treatment is frequently employed as a pretreatment phase. There are several different physical treatment methods, including membrane filtration, adsorption, and flotation [6,7].

### 2.2.1 Adsorption

It is regarded as the greatest technique for treating wastewater since it is economical, widely applicable, and simple to use. Both insoluble and soluble organic contaminants from different sources can be treated with it. Adsorption has therefore been used to remove a number of organic contaminants, including PAH, from various industrial wastewater. The selection of adsorbents, membranes, or filters is crucial for any physical procedure in order to achieve outstanding results [8].

An established method for eliminating PAHs from generated water is adsorption. As adsorbents, carbon-based compounds like biochar, activated carbon, carbon nanotubes, and graphene are employed. A carbonaceous material called biochar is produced from agricultural residues, wastewater, and petroleum sludge [9]. The biochar made during pyrolysis has a high capacity for absorbing organic pollutants and PAHs because it has an aromatic carbon skeleton similar to that of graphene. The honeycomb-like structure of graphene makes it a more effective adsorbent of PAHs like naphthalene and phenanthrene. When utilized as a catalyst, carbon nanoparticles can more effectively remove organic contaminants. Since they are more effective in removing PAHs, nanoparticles are now frequently utilized to purify generated water. Coriandrum sativum, Azadirachta indica, and Aloe barbadensis among other plant extracts have all been utilized to create copper and silver nanoparticles for the adsorption-based elimination of PAHs [10].

## 2.3 Membrane Filtration

Nanofiltration (NF) has a unique appeal in a variety of applications, including drinking water, industrial wastewater, and the reuse of water sectors. A type of reverse osmosis (RO) membrane has been created that is capable of retaining all dissolved salt ions as well as organic solutes without any charges. With pore sizes on the order of 1 nm, which equates to a molecular weight cut-off in the range of 100 to 5000 Da, nanofiltration membranes exhibit separation capabilities that fall between those of reverse osmosis and ultrafiltration membranes [26]. Because of its higher permeation flux and capacity to operate at lower pressures than reverse osmosis, which results in a reduction in energy consumption, nanofiltration can be utilized to create high-quality pollutants in a more sustainable manner than reverse osmosis [27]. Additionally, UF membranes with a particular pore size can effectively be employed for a variety of industrial applications and can refuse any molecular weight greater than 10,000 gm-moles. The predicted pore size of a membrane distinguished by MW removal is connected to the word "NF." What we can refer to as the fourth category of membranes that operate under pressure-driven operation has been introduced by modern membrane technology. Small solutes can be removed from the solution by NF using two different methods.

The first mechanism-known as ionic separation of NF-is the separation of molecules in water according to their charge, and it is widely acknowledged in the scientific world. The second method involves screening uncharged solutes based on their molecular weight [28]. While NF uses are growing dramatically in the industry, RO and ultrafiltration (UF) applications in wastewater and water treatment are rising gradually. Other applications include the production of biomaterials, the medicine industry, tastes, and the separation of solutes or chemicals from solutions. NF membranes are increasingly used in place of RO in a variety of applications, including the extraction of fine and expensive materials and drinking water [17,18]. While NF applications are growing and displacing existing membrane filtration methods, RO and UF have been widely employed for a variety of applications but are still constrained and difficult to expand. The NF membrane is made up of a variety of components, and it may be prepared in a variety of ways by using RO membrane polymers such as cellulose acetate and polyamide as well as other chemical-resistant polymers. NF membranes are currently also constructed from ceramic materials to endure high temperatures. The adaptability of NF preparation and the range of raw materials available will grow and expand its use in many procedures [15,16] A portion of the feed is processed by NF, which involves a semi-permeable membrane. Permeate the stream's filter portion, and retentate or concentrate, the stream's rejected, unfiltered portion, make up the inlet stream (figure 1).

NF has successfully demonstrated excellent removal of organic material. However, the elimination of microbial growth that has been observed in NF distribution networks requires chlorine treatment. NF membranes, which have low inorganic material retention and a high organic material removal rate, may create water of the highest quality while reducing microbial development [29]. NF differs from ion exchange units in that it softens water by removing magnesium and calcium while not adding sodium ions during the filtration process. Because NF does not need further chemical treatment to reduce hardness, the water-softening process can be properly carried out without noticing the presence of sodium resin, as it has been for 50 years. Because NF does not need cooling or heating of the feed like, say, distillation, the cost of separating is effectively reduced. Additionally, the delicate molecular separation will be maintained without the need for mechanical stirring. NF offers the significant advantage of handling a large volume of feed continuously and a constant permeate flow rate [30] However, due to the membrane's restricted nanopore size, NF has a limited range of industrial applications. Since RO and UF can efficiently span the UF range without the cost restriction of NF due to its high startup, operating, and maintenance costs, they are favoured. Since TDS affects when NF membranes need to be replaced, NF membranes need to be replaced more frequently than they actually need to, which drives up the cost of NF [31]. Low-energy systems are a crucial necessity because using NF systems has increased the energy needed for water treatment by 60–150%. Green energy could be a useful tool for reducing energy consumption, but it is more expensive than conventional energy. Using more permeable NF can help reduce the pressure and energy needed for NF, which could have an impact on how well the membrane functions. Therefore, a balance between energy needs and optimal operation is needed [32]. Reverse osmosis and ultra-filtration separation procedures are two well-established technologies, while nanofiltration membranes are a more recent innovation that bridges the gap between them. The ability of Nanofiltration membranes to allow monovalent ions, like sodium chloride, to pass through the membrane while blocking divalent and multivalent ions, like sodium sulfate, is one of their most intriguing features. The establishment of specialty application areas across several sectors is made possible by this flexibility. Since filtration is predominantly utilized in process applications, the development of nanofiltration membranes to expand on the existing wide range of ultrafiltration and microfiltration membranes was a logical step [22].

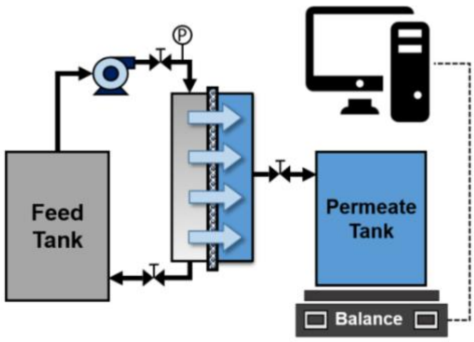


**Figure 1**: Schematic setup of nanofiltration (NF) that removes PAH Compounds from wastewater.

## 2.4 Chemical treatment

Chemical techniques may present certain drawbacks, too, especially since certain byproducts of the decontamination of PAHs are much more harmful than the PAHs themselves. Moreover, the biorecalcitrant, poisonous, and low soluble nature of PAHs in water has restricted their application to biological processes. As a result, the majority of these methods cannot permanently reduce water pollution due to PAHs. Some techniques are particularly expensive on a commercial basis because they need a lot of energy and operating resources [11].

### 2.4.1. Coagulation

Water treatment processes such as coagulation and flocculation are effective at eliminating colloid and suspended materials but not dissolved components. Another study used adsorption in different nano and organo-modified nano-clays, following coagulation-flocculation utilizing alum and poly aluminium chloride (PAC), to remove PAHs from water. The removal of PAHs from the mixture of clay particles, PAC, and alum ranged from 37.8% to 100% [12].

## 2.4.2. Chemical oxidation

Polluting substances are oxidized chemically to lessen their toxicity. PAHs are eliminated from water using substances like modified Fenton (Fe), potassium permanganate, standard Fenton, and sodium persulfate. Nevertheless, chlorine oxidation products are produced during the oxidation of PAHs employing sodium hypochlorite as an oxidant, rendering this reagent inappropriate. With an effectiveness of up to 70%, the chemical oxidant potassium permanganate may efficiently eliminate PAHs [13,14]. There seem to be currently a number of available materials for such removal of PAHs from water samples, including nylon membrane, C18, polyvinylidene fluoride, and porous organoclay composite.

Most stable aromatic compounds, including two or more, make up PAHs. The conjugated pi-electron configuration of PAHs, which is governed by a variety of aromatic rings & their molecular weights, determines their characteristics. They can be categorized according to their molecular mass or composition [15]. The smallest molecular weight PAHs are those that have two or three compressed benzene rings, such as methylnaphthalene, naphthalene, acenaphthylene, and acenaphthene. On the alternative hand, high-molecular-weight PAHs are those that have four or even more fused benzene rings, such as pyrene, benze (a) anthrancene, and chrysene [6]. The solubility of PAHs in water ranges from 0.27 to 31 g $m^{-3}$.

Both natural and anthropogenic processes contribute to the production of PAHs in our environment. After being produced, PAHs are transported into soil and water by wet and dry deposition [16]. The primary sources of PAHs in aquatic ecosystems are anthropogenic, including coal and biomass burning, vehicle emissions, and industrial wastes. Today, a few investigations have shown the presence of PAHs in industrial effluents such as coking and landfill effluent, but it is still unclear what happens to PAHs present in paper-making wastewater (PMWW) and how they change after treatment [17]. Elevated concentrations of total organic carbon and chemical oxygen demand (COD) are characteristics of the complex combinations of organic and inorganic compounds found in PMWW (TOC). The amount of water used in the production of paper varies between 5 and 300 $m^3$ per tonne of pulp products, and an average-sized mill produces roughly 2000 $m^3$ of effluents each day [18,19].

## 2.4.3. Electrochemical technology

Earlier research demonstrated that the breakdown of acids, sugars, phenolic compounds, and lignin derivatives during electrocoagulation (EC) treatments of PMWW is responsible for the elimination of PAHs (Figure 2). However, the various traits of organic pollutants and how they affect the elimination of PAHs have not yet been fully discussed. Full-scale EC treatment applications also show that as organic pollutants break down into compounds with low molecular weights, the efficacy of EC's removal declines, making it impossible for an increase in current density to maintain continuous total organic carbon (TOC) elimination or COD removal efficiency. These results imply that organic transformation matters for how well EC removes organic contaminants [20].

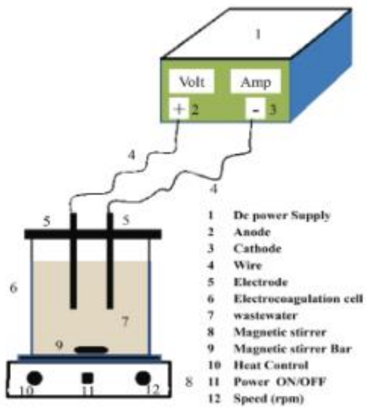


**Figure 2:** Schematic setup of electrocoagulation process that removes PAHs from wastewater.

Wastewater is produced from a variety of sources, including agricultural, industrial, commercial, institutional, and domestic, and contains a variety of pollutants that have an impact on the environment's natural state. Hospitals use a lot of water, and they abstain from discharging wastewater into the environment because it contains toxic pollutants like metal oxides, potentially dangerous liquid waste from different units, radioactive waste, viruses, bacteria, blood, and fluids, as well as varying concentrations of COD and BOD, which have an impact on the environment in various ways [21]. Due to the usage of chemicals and non-biodegradable materials, biological treatment is no longer as effective as it once was.

It is necessary to replace the traditional, pricey, and intensive treatment method employed with an effective, manageable, and simple procedure. Due to the aforementioned factors, the entire globe is searching for innovative, low-cost physical-chemical remedies that can fix the flaws in biological treatments. These technologies must be simple to regulate and apply, inexpensive to build and operate, highly effective, ecologically friendly, and able to be used on tiny amounts of land with little energy use. Electrochemical oxidation, electro-coagulation, electrodialysis, photo-electro-oxidation, electrochemical disinfection and sterilization, membrane separation (ultra-filtration, nano-filtration, and reverse osmosis), as well as nano-adsorbents, are just a few novel technologies that satisfy the previous specifications. These technologies' integrated and hybrid treatment systems offer the appropriate answer for the intended aim [6].

Several advanced treatment technologies, including membrane separation processes, $H_2O_2$-derived oxidation, ozonation, electrochemical oxidation, and sulfate radical-advanced oxidation processes, have indeed been developed and tested for the removal of various emerging contaminants in order to enhance the safety and quality of the reclaimed water and prevent the negative effects that may result from its reuse. Along with many of them, utilizing membrane filtration techniques like nanofiltration, ultrafiltration, and reverse osmosis is currently regarded as a potent solution.

Nanofiltration provides one of the most economically advantageous ways to carry out enhanced wastewater treatment options because it strikes a good balance between both the water quality that is needed and the energy required to generate it [21]. The elimination of biological oxygen demand (BOD5), total nitrogen (COD), and phosphate is maximized in the treatment of municipal wastewater utilizing modern physicochemical techniques. Additionally, electrochemical pretreatment, followed by an ultra-filtration membrane, was used to remove suspended particles. Therefore, cleaned wastewater can be utilized to farm non-edible or woody trees, fish, animals, and sheep. The findings revealed that 93%, 95%, and 100%, respectively, of BOD5, COD, and phosphates were removed using a tiny hydro turbine with a 1.5 k wt/h power output and a fluid velocity of 10-15 L/s [22]. By rotating the tiny hydro-turbine with reject flow, 50 percent of the total energy usage may be recovered. When compared to conventional, inefficient biological treatment plants, the construction and operating expenses of sewage treatment facilities can be lowered by up to 50%. Due to its quality, the sludge produced by the investigation's improved recommended procedure can be utilized as fertilizer, in contrast to the typical treatment technique's sludge, which should not be recycled [23]. With the use of electrocoagulation-electroflotation (ECF) technology, pollutants can be treated and flocculated without the need for coagulants. According to Shammas et al., coagulation happens when current is applied and is capable of eliminating small particles because the direct current applied causes them to move. Electrocoagulation may also lower the amount of coagulant needed to produce sludge [24].

The electrochemical method of electrocoagulation (EC), which removes minute particles from wastewater without the use of chemicals, uses an electric current to treat sewage. Additionally, EC's low initial installation and maintenance costs, the minimal quantity of sludge produced after treatment, a fast settling time, and great contaminant removal effectiveness made it progressive and widely used. Electrodes of the stainless steel (SS), aluminium (Al), and iron (Fe) varieties are employed in the EC process for wastewater treatment [18]. Anodes and cathodes, two pairs of metal plates used in electrocoagulation, are placed in pairs of two. The cathode is oxidized (loses electrons), whereas the water is decreased (gains electrons), by using principles of electrochemistry to improve the treatment of the wastewater. Metal is released into the device whenever the cathode electrode comes into touch with the wastewater. When this occurs, hydroxide complexes are formed in order to neutralize the particles and create agglomerates. These agglomerates can be removed through filtration and start to form toward the tank's bottom. The particles would indeed float to the top of the tank using generated hydrogen bubbles that are produced from the anode when one thinks of an electrocoagulation-flotation setup [19,20].

For a variety of reasons, electrocoagulation-flotation is an alternative to conventional chemical coagulation. Because the electrodes act as the coagulant in ECF, the necessity for chemicals can be decreased. However, many people continue to utilize artificial coagulants in an effort to improve treatment. According to the amount of water being treated, traditional chemical coagulation methods include alum (aluminum sulfate), ferric chloride ($FeCl_3$), or ferrous sulfate ($Fe_2SO_4$), all of which can be very expensive. When the coagulant is used, it serves a similar purpose to that of the electrodes by neutralizing the particle's charge, allowing them to clump together and then sink to the bottom of the tank. Additionally, electrocoagulation-flotation can decrease the amount of trash generated during the treatment of wastewater and the amount of time required [20,21].

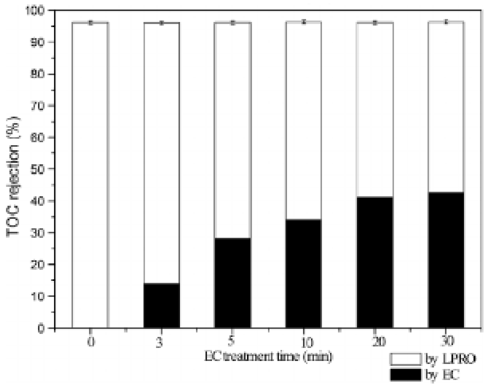


**Figure 3**: Effect of EC treatment time on TOC rejection by EC and downstream LPRO process. EC treatment conditions: initial pH = 7.85, current density = 30 mA cm-2, temperature = 25 ºC; LPRO conditions: trans-membrane pressure = 400 kPa, stirring speed = 400 rpm, temperature =25 ºC. Error bars represent standard deviations obtained in triplicate run.

Figure 3 shows the effect of EC treatment time on TOC rejection by EC and downstream LPRO process. Gong et al in their study stated that when compared to information derived with the LPRO alone, the EC-LPRO technique showed equal extraction efficiency for TOC (Figure 4). The lowering of the organic loading to the LPRO was, in this regard, the principal purpose of the EC pretreatment in the integrated treatment. For instance, EC pretreatment for 30 min decreased the TOC in the PMWW by about 45%, which resulted in a nearly 50% decrease in the TOC loading onto the LPRO membrane. Similar TOC removal as well as significant drops in TOC concentrations is observed [25]. EC studies were carried out at current densities of 10, 20, and 40 mA $cm^{-2}$, with stable initial pH, inter-electrode spacing (1 cm), and area/volume (A/V) ratio (0.16 $cm^{-1}$), according to a study by Gong et al. After 30 minutes of therapy, raising the current density resulted in continuous increase in the TOC extraction efficiency [20].

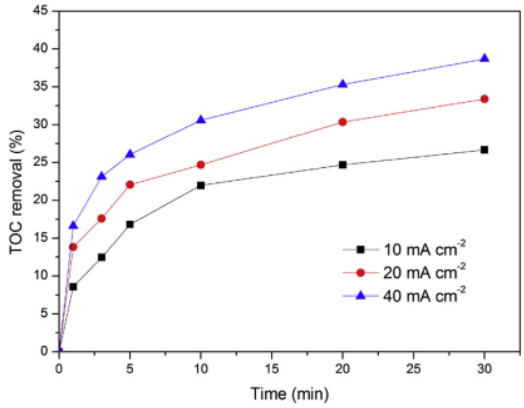


**Figure 4:** TOC removal obtained with different applied current densities for electrocoagulation of the paper-making wastewater. (Initial pH =7.75, area/volume ratio =0.16 $cm^{-1}$, temperature =25 ºC).

## 2.4.4. Advanced oxidation process

In the 20th century, the idea of advanced oxidation processes (AOPs) was invented. Oil and gas production water is treated using AOPs to get rid of organic and inorganic chemicals, as well as colour and odor. Ozone, chlorine, $H_2O_2$, and Fenton reunions; like ultraviolet , hydrogen peroxide, $Fe^{+3}$; are typical oxidants in AOPs. The removal of polycyclic aromatic hydrocarbons can be accomplished with the use of AOPs and catalytic oxidation methods (PAHs). Catalytic oxidation, which is based on this method, is used to remove PAHs from wastewater. AOPs have been found to be the most dependable catalyst-using processes. Hydroxyl radicals, molecular ozone, and other reactive species are used for oxidizing during the elimination of PAHs. Ozone's electrophilic nature allows it to grab aromatic rings, and radicals react to it without discrimination [33].

## 2.5 Biological treatment method

Both aerobic and anaerobic biodegradation of PAHs is possible. Activated sludge processes, sequencing batch reactors, bioreactors, membrane bioreactors, phytoremediation, innovative microbial capacitive desalination cell, and microalgae-based processes are just a few of the biological treatment techniques used to remove PAHs. Sequencing and membrane bioreactors are two of these that are most frequently employed to eliminate organic contaminants. Low molecular weight organic contaminants are more readily destroyed during biological treatment [34].

### 2.5.1. Phytoremediation and bioremediation

Green plants, bacteria, and enzyme are used in phytoremediation and bioremediation to treat contaminated substrates and return them to their natural condition without endangering the environment. Environmental clean-up via bioremediation, also known as green remediation, is thought to be more cost-efficient, environmentally benign, and long-lasting. In order to accelerate the decomposition or removal of inorganic and organic pollutants, plants are cultivated in the proximity of contaminated soil, surface water, or groundwater. This process is known as phytoremediation. For phytoremediation, some trees are more suited and efficient. Via metabolic processes that result in the production of cell biomass, carbon dioxide, and water as byproducts, bioremediation involves the biological restoration of environmental pollutants [33].

## 2.6 Combined treatment methods

Many high molecular-weight PAHs have been successfully degraded, solubilized, and completely removed from water using integrated physical, biological, and chemical methods. The Fenton oxidation process and the biodegradation process are both employed to eliminate PAHs from aqueous solutions, including naphthalene and phenanthrene. A Bacillus fusiformis (BFN) strain from activated sludge is used in the biodegradation process, while tea extract is used in the Fenton oxidation process to create ferrous nanoparticles in a variety of atmospheres, including oxygen, nitrogen, and air [31-34]. Utilizing the nutrition and microbes that were once present in the rivers, biologically active filtration is a combined physical and biological process used to treat oil and gas wastewater. It encourages the development of biofilm that can withstand high or low concentrations of total dissolved solids. The removal of organic contaminants and sediments via biologically active filtration with activated carbon in granular form is a recognized effective technology [33,34].

# 3 CONCLUSION

The release of large quantities of PAHs, organic, and inorganic pollutants into the natural environment produce a significant volume of hazardous produced water which lead to a number of environmental problems. As a result, generated water treatment techniques have recently required consideration. The deep details of the review in this paper showed that the treatment techniques to eliminate PAHs is negatively and positively impacted by their transformation. The conjugated pi-electron configuration of PAHs, which is governed by a variety of aromatic rings & their molecular weights, determines their characteristics. There seem to be currently a number of available effective treatment methods for such removal of PAHs from wastewater. The findings of different treatment technologies give a guidance on selecting the better method for wastewater treatment and reuse.

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