REVIEW ARTICLE



Removal of Petroleum Contaminants Through Bioremediation with Integrated Concepts of Resource Recovery: A Review

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Abstract There is an upsurge in industrial production to meet the rising demands of the rapidly growing population globally. The enormous energy demand of the growing economies still depends upon petroleum. It has also resulted in environmental pollution due to the release of petroleum origin pollutants. Soil and aquifers, especially in the direct impact zones of petroleum refineries, are the worst hit. The integrated concept of bioremediation and resource recovery offers a sustainable solution to mitigate environmental pollution. It involves biodegradation, a benign utilization of toxic wastes, and the recycling of natural resources. Bioremediation is considered an integral contributor to the emerging concepts of bio-economy and sustainable development goals. This review article aims to provide an updated overview of bioremediation involving petroleum-based contaminants. Microbial degradation is discussed as a promising strategy for petroleum refinery effluent and sludge treatment. The review also provides an insight into resource reuse and recovery as a holistic approach towards sustainable refinery waste treatment. Furthermore, the integrated technologies that deserve indepth exploration for future study in the refinery sector are highlighted in the present study.

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Introduction

Petroleum-derived products such as fuels and petrochemicals are essential for economic development and constitute a vital role in our everyday lives. Steadily increasing demand since the late nineteenth century has elevated crude oil consumption globally. Petroleum is the fossilized fuel source that is an indispensable source of energy but adversely impacts our environment and living beings' health. Given this, bio-derived fuels' significance is highlighted, which also ensures to minimize our dependence on fossil fuels [1-4]. The main routes for petroleum-derived pollutants entering the environment include refinery emissions, automobile exhaust, industrial combustion facilities, accidental spills and leaks during exploration, transportation, refining, and storage [5]. The enormous anthropogenic activity in the oil and gas sector is a prominent contributor to air, soil, and terrestrial water pollution, including greenhouse gas emissions like methane and carbon dioxide. Methane is known to cause 20-fold higher global warming effects than carbon dioxide and is generated at around 331 teragrams per year globally, which is very alarming [6, 7]. The primary route of soil and water contamination is refinery discharge, which mainly comprises petroleum products like total petroleum hydrocarbons (TPH), polycyclic aromatic hydrocarbons (PAHs), and other organic compounds like benzene, ethylbenzene, toluene, xylene (BTEX). Among these, PAHs have been enlisted as priority pollutants by the United States Environmental Protection Agency based on their extreme toxicity, carcinogenicity,

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and mutagenicity. Industrial wastewater, mainly from petrochemical industries, contains high concentrations of toxic chemicals is considered a significant source of water pollution. Therefore the treatment of refinery wastewater must be strictly enforced before its legal discharge. It is imperative to sustainably utilize these pollutants from the refinery wastewater and contaminated soil. Compared to traditional chemical effluent treatment, biological methods are considered more versatile and eco-friendly. These generally involve microbes, plants, or biocatalysts derived from them (from enzyme pathways) to degrade complex chemical pollutants to simpler forms. Additionally, they have the advantages of high conversion rates, selectivity, and economy over chemical methods [8]. An overview of bioremediation is presented (Fig. 1).

The removal of petroleum toxicants can be accomplished either by reviving the growth of native microbes in an impacted ecosystem or through the external addition of hydrocarbon-degrading microbes and plants. It is applicable for various environmental matrices, including oil reservoirs, oil spill sites, and other contaminated water bodies. However, the application of single microbial species has limitations in its effectiveness for specific hydrocarbons only. This problem can be overcome using a mixed population which acts due to synchronized influence and diverse enzymatic action in the mineralization of a broad range of petroleum hydrocarbons. Their better adaptability, performance over broad physiological conditions, tolerance towards various waste matrices, and enhancement of biocatalytic potential make them superior [9, 10]. Another sustainable approach for refinery waste remediation is based on the adsorptive potential of bio-sorbents derived from bio-derived wastes like microbial and lignocellulosic biomass. This philosophy towards valorization of refinery wastes opens a gateway to achieve a cradle-to-cradle approach. This article provides an overview of removing petroleum contaminants through the bioremediation approach integrated with resource recovery and reuse by combining ecological and economic drivers. It also narrates the applications of biological products like natural fibers, bio-composites, and biocatalysts for remediation of refinery wastes.

Petroleum Refinery Effluents

Hydrocarbons, PAHs, phenols, inorganic complexes, sulfur, and nitrogen-containing organic chemicals are the major environmental pollutants in the petroleum refinery effluents and sludge. According to a Transportation Research Board and National Research Council report, global annual hydrocarbon and PAHs releases are 4,988,699 and 6,319 tonnes per year, respectively, through land-based sources [11]. Nearly 85% of hydrocarbon release in the environment is attributed to petroleum processing industries [12]. PAHs, possessing high-toxicity and carcinogenicity, are reported to alter aquifers' community structure and soil microflora [12]. Petroleum refineries

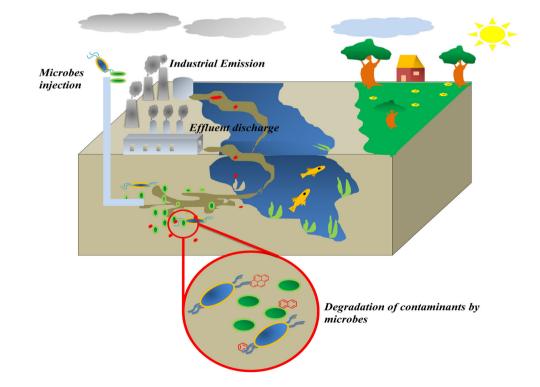


Fig. 1 An overview of terrestrial pollution caused by industries and role of bioremediation cause soil contamination due to oil spills either during transportation or exploration, tank leakage, or improper dumping of petroleum waste. Refinery contaminants that cause soil pollution was sub-categorized (Fig. 2).

Petroleum refineries are also significant sources of water pollution. A large quantity of water is required in petroleum refining, typically in the range of 30–50% v/v of the input crude processed [13]. While the composition of petroleum refinery waste depends upon the complexity of the refining process, refinery wastewater broadly contains a range of organic, inorganic compounds, especially aromatic hydrocarbons and heavy metals, found in various effluent streams from the refining process, which include dissolved oil, minerals, gases, and solid compounds (Fig. 3). More efficient management and reuse of this wastewater are required in petroleum industries to minimize the freshwater intake and meet strict regulatory requirements.

Methods for Remediation of Petroleum Refinery Waste

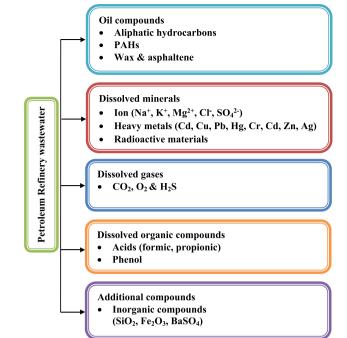


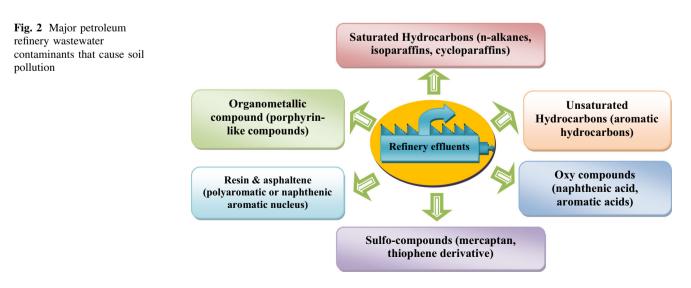
Fig. 3 Pollutants present in petroleum refinery wastewater

Bioaugmentation

The toxicity, carcinogenicity, teratogenicity of petroleum refinery wastes poses a threat to both humans and the environment. Removal of these hazardous contaminants can be effectively achieved by augmentation with native or exogenous microbes. The aforesaid approach can be implanted either alone or in microbial consortia to treat refinery waste [14] and hence considered more economical and feasible due to the application of microbes capable of mineralizing diverse pollutants [15]. Musa et al. reported the degradation of refinery wastewater effluents using bioaugmentation with native microbes, and the result

reported 100% removal of naphthalene and phenanthrene and almost six times higher pyrene removal in case of a mixed microbial consortium due to secretion of bioemulsifier when compared to native microbes [17]. The advantage of the mixed microbial consortium over indigenous microbes is the diversity in substrate-directed activity and inhibitor tolerance across consortium members. It is an essential prerequisite for detoxification as bioremediation efficiency directly correlates with individual microbes' metabolic potential [1, 2]. This bioaugmentation approach can also be made to remove various organic contaminants from wastewater through an

showed 66% hydrocarbon removal [16]. Isaac et al.



activated sludge process before its discharge. This treatment process offers a sustainable solution to overcome the scarcity of water through reuse and recycle. As the activated sludge process could become limited to a few contaminants due to the complexity of refinery waste, bioaugmentation of activated sludge can counterbalance such limitations. It involves the addition of potent hydrocarbon-degrading microorganisms for enhanced treatment.

In a recent study, Jamal et al. reported the PAH contaminated refinery wastewater in higher saline conditions using a continuous stirred tank reactor. The study resulted in the complete removal of phenanthrene and fluorene and 90% degradation of pyrene using halophilic microbial consortia [18]. Similarly, in another study, the addition of hydrocarbon-degrading microbes was investigated for PAH degradation in aged contaminated soil, which resulted in a 99% reduction in PAH concentration, signifying the process's effectiveness at PAH contaminated site [19]. Despite various reported examples of this approach for removing priority pollutants like PAH from soil and water, a few limitations associated with the bioaugmentation process include low inoculums size or competition with the existing indigenous microbes under natural conditions [20]. Furthermore, the introduction of a new strain under natural conditions may fail to deliver desired growth rates due to bacteriophages' presence or lack of acclimatization to the hostile environmental conditions [21]. Additionally, studies also reported retardation of bioaugmentation under extreme environmental conditions such as starvation periods, pH, and low temperatures [22].

Biostimulation

Unlike bio-augmentation, which requires the addition of external microbes to accelerate contaminant degradation by complementing indigenous microbiota, biostimulation focuses on inhabitant microbes in the affected environment and seeks to strengthen their metabolic potential. However, adding bacteria with the desirable catabolic capabilities into a particular environment does not always guarantee enhanced degradation of petroleum toxicants. Therefore the average contribution of inoculated microbial diversity to the total richness of species to tolerate the targeted environment is essential for survival. Even after employing native microbial populations, the success of on-site bioremediation is primarily constrained by imbalanced nutrients and/or unfavorable factors, which include: adequate nutrients, pH, ambient temperature, moisture, oxygen, and contaminant concentration [23]. Among these factors, nutrient deficiency severely retards inhabitant microbes' catalytic activity and limits the rate of intrinsic bioremediation [24]. In a study, Sun et al. reported the retardation during crude oil degradation due to ammonium and phosphorous deficiency [25]. In general, the addition of nutrients also plays a pivotal role, and therefore the adequate concentration of nutrients is essential for efficient degradation; the deficiency thereof may be counterproductive. The aforesaid process also includes the addition of other stimulants like surfactants for the availability of hydrocarbons since increased mobility of any hydrocarbon molecule, enabled by surfactant-induced emulsification dispersion, makes it more susceptible to microbial action. Borah et al. reported a biosurfactant-producing strain Bacillus cereus for the remediation of a hydrocarbon-contaminated subsurface, signifying applicability in enhanced oil recovery [26], as did Marchut-Mikolajczyk et al. [27]. Gharibzadeh et al. reported removal of phenanthrene via sequential washing of phenanthrene contaminated soil with biosurfactant. The process showed 97% removal efficiency of a real PAH contaminated site over seven cycles, suggesting the effectiveness of reuse and potential economic viability [28]. Roy et al. demonstrated enhanced hydrocarbon reduction from 57 to 75% by simultaneous application of nutrient and biosurfactant producing microbes [23]. Therefore the application of biosurfactant-mediated removal of petroleum contaminants is considered promising due to its low toxicity, biodegradability, broader applicability over diverse substrate range.

Another recently reported interesting approach for biostimulation involves accelerated PAH degradation through the addition of sawdust and wheat straw biochar; simultaneous adsorption of PAH on the biochar surface while the biochar provides a better habitat for consortium survival enhanced microbial degradation significantly [18]. Additionally, other biostimulation agents like electron acceptors can play a significant role during anaerobic treatment since, in the absence of oxygen, anaerobic microbes may grow on an alternative substrate such as nitrate, sulfate, iron, and CO₂, which link to the anaerobic processes of denitrification, desulphurization, iron reduction and methanogenesis [29]. The combination of bioaugmentation, biostimulation, and biosurfactant addition, depending upon the defiled site's attributes, might be a promising strategy to accelerate bioremediation [30].

Composting

In composting, often a cost-effective strategy, contaminated soil is mixed with a bulking agent (primarily agricultural residue) to create pores and make the process aerobic. The bulking agent is utilized as a carbon source by microbes and makes degradation co-metabolic. Advantage of using composting as a bioremediation technique is the generation of mature compost that can be used in land restoration [31]. Composting increases the metabolic diversity of microbes and has been referred to as "super bioaugmentation" [32]. Apart from the degradation of organic components, heavy metal contamination can also be addressed effectively by composting. Cadmium, which is considered the most mobile metal in an oil-contaminated surface, is removed through the combined composting action of cellulolytic bacteria and organic-degrading species [33]. Removal of PAH was also reported through the co-composting action of cattle manure and wheat straw [34]. Abtahi et al. investigated the composting process's efficiency with indigenous compost microbes and petroleum degrading microbes in a composting bioreactor, which suggested a decline in the effectiveness of petroleum degrading microbes due to competition among them [35]. During composting, the optimizations of rate-limiting factors like pollutant concentration, soil: compost ratio, and compost stability significantly affects composting efficiency. Therefore, the ratio of oily sludge and amendments must be balanced. Given this, Koolivand et al. performed two-stage composting (windrow composting and in-vessel composting) of storage tank bottom sludge in which the effect of the various mixing ratio of C/N/P and nutrients addition was investigated for the removal of TPH, which resulted in the removal of 93.72% of TPH [36].

Enzymatic Bioremediation

The degradation of petroleum hydrocarbons can be accomplished by microbes or their extracted cellular components, i.e., enzymes. Compared to microbes, enzymatic bioremediation is a rapid method for the removal of petroleum toxicants. The enzymes have broad substrate specificity and helpful in the bioremediation of various petroleum-based toxic compounds like phenols, PAHs, etc. [37, 38]. The mechanism of degradation in both cases is similar since, in whole cell-mediated degradation, the organism secretes enzymes that either cleave the aromatic structure of hydrocarbons or substitute it with different functional groups that make the degradation products less harmful. Although whole cells are also used for remediation purposes, the process is slow, and competition with native microflora is generally a concern. Sometimes, the degradation efficiency or the viability of the chosen microbe is impaired due to the unavailability of appropriate conditions at the contaminated site as compared to in-vivo. The enzymatic approach provides alternative ways to improve xenobiotic bioremediation approaches. Since enzymes are biocatalysts, they increase the reaction rate by lowering the activation energy, and therefore, degradation can be achieved quickly [39]. Several enzymes, like oxidoreductases, laccase, peroxidase, have been used in the bioremediation of petroleum toxicants [40–42].

A special significance of enzymatic bioremediation is that it can be applied in a nutrient-deprived contaminated site. The biocatalyst size is another critical parameter that should be considered, as the small size of the biocatalyst increases the diffusion rate of enzymes and thus accelerates the degradation process. Unlike whole cell-mediated degradation, no toxic by-products are generated, and the cost of operation is also less [43]. Enzymes can be used directly in crude or semi-purified forms in many cases, depending upon the prevailing requirements. Different microorganisms produce biocatalysts with different catalytic efficiencies for various contaminants. Biocatalysts used for the degradation of petroleum hydrocarbons are listed (Table 1).

Bio-Materials and Bio-Composites for Refinery Waste Treatment

Microbial biomass can be utilized for the treatment of petroleum effluents either directly or in immobilized form. Various microbial biomass immobilization approaches on biomaterials and biocomposites, including adsorption, encapsulation, and covalent methods, have been demonstrated to improve bioprocess stability and waste effluents treatment [48–51]. Banerjee et al. deployed immobilized microbial biomass to treat petroleum spills, delivering the removal of more than 95% of phenolic content from refinery water [52]. Cost et al. reported plant-derived biomass—peat and angico hardwood sawdust—as adsorbent material for removing prominent refinery contaminant family BTEX from waste discharge [53]. Recently, Imam et al. utilized rice straw biochar as an adsorbent for anthracene removal [54].

The utilization of natural fibers and bio-composites is another potentially cost-effective approach for treating liquid refinery waste streams. Limited research has yet been done in this direction. Akhbarizadeh et al. engineered a low-cost bio-composite derived from shrimp shell and acid-activated montmorillonite for simultaneous removal of 93 and 87% for metals crude oil, respectively [55].

Bio-sorbents can thus be considered desirable choices for the treatment of refinery waste due to their low cost, density, strength, and eco-friendly nature. Moreover, they help in the mitigation of greenhouse gases (GHGs).

Table 1 Biocatalyst derived from the microbes for the degradation of petroleum hydrocarbons

S.No	Microbe	Enzyme	Contaminant	Degradation (%)	References
1	Pycnoporus sanguineus	Laccase,	Anthracene, Pyrene	67.5%	[44]
		Cytochrome- P450		31.1%	
2	Shewanalla chilikensis,	Lipase,	TPH	96%	[45]
	Bacillus firmus,	Catalase & oxido-reductase			
	Halomonas hamiltonii				
3	Ganoderma lucidum	Laccase,	Phenanthrene,	99.65%	[46]
		Lignin peroxidase,	Pyrene	99.58%	
		Manganese peroxidase			
4	Trametes versicolor	Laccase	Anthracene	60%	[47]

Integrated Refinery Waste Treatment and Resource recovery

Energy Recovery

The integrated refinery waste treatment and simultaneous energy recovery (usable heat, electricity, or bioenergy production) using microbial action could be the significant breakthrough in waste to energy research in the coming days. This integrated process using the microbial route can simultaneously have the potential for environmental, economic benefit, and value addition. A recent study reported simultaneous hydrogen and electricity generation from biodiesel effluents [56]. Like refinery waste, other bioderived waste can also be minimized via microbial action through the integrative approach with simultaneous value addition [57, 58]. Electricity generation using microbial fuel cells (MFC), innovative and sustainable technology for treating organic pollutants from wastewater, is considered promising since no external source of energy is required for its operation; this ensures dual benefits of waste treatment and simultaneous energy recovery through biochemical reaction. MFCs can remediate a wide range of contaminants, such as biological wastes, heavy metals, petroleum products, dyes, phenolic compounds, quinolone, pyridine derivatives from wastewater. However, not much has been reported on MFCs in the general treatment of refinery effluents like refinery wastewater, petroleum sludge, and oily waste.

Guo et al. set-up both single and doubled-chambered MFC configurations for recycling refinery wastewater as fuel. A dual-chambered MFC showed the highest power density output of 310.08 mW/m³ and the pollutant removal efficiency of 83.6%, comparatively higher than a corresponding single-chambered MFC [59]. Mohanakrishna et al. also demonstrated the treatment of refinery wastewater and simultaneous electricity generation using single chamber air–cathode MFC technology, eliminating

aeration during operation and improving sustainability. A maximum power density of 132 mW/m^2 and substrate removal efficiency of 48% was reported in the study [60].

The MFC approach is considered regenerative as the organism is generally self-replicating and cost-effective compared to energy-intensive conventional treatment. Moreover, MFCs enable the recovery of sustainable energy from the wastewater with the simultaneous assimilation of effluents while limiting both the energy input and the excess sludge production. Apart from these additional advantages, MFCs are mostly insensitive to the operational environment. The key challenges remain scale-up to refinery volume requirements, which need to be addressed through engineering design.

Similar to electricity generation via MFC, anaerobic digestion through microbial action is another integrated approach for refinery waste treatment and simultaneous resource recovery in the form of renewable energy, namely biogas [61]. Compared to conventional wastewater pretreatment approaches, anaerobic digestion has significant advantages like low nutrient requirement, little or no external input of energy, low sludge production, and installation ease. Despite several advantages, there remains a significant limitation of hydraulic retention time and influent feedstock concentration during the operation of anaerobic treatment, and therefore a proper optimization is required for the operation of the process. However, Upflow anaerobic sludge blanket digestion (UASB) could be a practical solution for the treatment of refinery wastewater because of its improved effectiveness relative to other digester configurations in terms of flexibility for a vast concentration of feedstock due to a wide range of hydraulic retention times [62]. Unlike aerobic bioremediation processes, anaerobic digestion requires no external energy supply is required and also helps in recovering bio-energy as biogas (a mixture of methane and CO_2), which reduces net dependence on fossil fuel within a petroleum refinery. Additionally, obtained methane can also be utilized as a

substrate for electricity generation through MFC or an alternate feed for value-added products to minimize their emissions via methanotrophic microbial action [63, 64].

Water Recovery

Water recovery through Constructed Wetland (CW) is another exciting development for resource recovery from refinery waste. CW removes contaminants while generating fresh water for irrigation, water reuse in the refinery, fodder for livestock, and some energy. CW uses shallow beds or channels, marsh plants, soil, sand and gravels, and various microorganisms for the treatment of typical refinery wastewater streams [65]. Compared to traditional wastewater treatment methods, CW is considered to have a low maintenance cost in operations and appears visually attractive, mimicking the aesthetics and functionality of a natural wetland for the improvement of water quality to enable its productive reuse. Additionally, CW also reduces GHG emissions via carbon capture and can serve as a renewable energy source via the produced biomass.

Several recent studies have sought to treat refinery effluents using the CW approach. Information about the wastewater characteristics, the efficiency of treatment, and installation location has been presented (Table 2). Plant species like *Typha latifolia*, *Pragmites Australis, Eichhornia crassipes* have treated refinery effluent in low maintenance operations. These plants also serve as habitats for diverse microbial communities, which also enhance degradation. Several limiting factors include retention time, effluent concentration, type of wastewater, water depth, type of plant species, microbes, and climatic conditions that can constrain pollutant removal using CW [66]. However, this approach is beneficial due to the low set-up cost for effluent remediation at a large field-scale study.

Hybrid MFC-CW systems are now being explored, which combine the benefit and leverage synergies of both methods for simultaneous waste treatment and energy generation. The combined bio-cum-electrochemical approach using MFC-CW may be even more promising for removing refinery contaminants than the standalone bioremediation or phytoremediation process CW.

The MFC-CW approach works on a principle similar to that of a standalone MFC. However, in MFC-CW, the potential difference is generated across the rhizosphere's oxic zone and the anoxic zone of CW, thereby making the process sustainable. These unique characteristics make the integrated MFC-CW an ideal approach for waste treatment and recovery of resources in electrical energy while also mitigating methane emissions [70].

Wei et al. implemented integrated microbial electrochemical technology-CW in-situ to remove major refinery contaminants benzene, methyl-tert-butyl ether, and ammonium from groundwater. This study reported the complete removal of contaminants with a significantly low power density of 1.74 mW m^{-2} due to limited substrate concentration [71]. Yang et al. did the comparative analysis between MFC and MFC-CW and investigated the improved power generation in MFC-CW due to the enhanced redox activity in the rhizosphere due to photosynthesis or due to increased accumulation of active microbes [70]. CW and MFC are compatible and complementary technologies since both are dependent on microbes or plants' actions to remove contaminants from wastewater and recover energy. Though combined MFC-CW retains the best features of both the subcomponent

Type of CW	Wastewater type	Pollutant removal (%)	Location	References
Horizontal sub-surface flow	Petrochemical waste	BOD*, COD**, TSS [#] : 95% Phenolic compound: 90%	Tamil Nadu, India	[66]
Vertical surface flow	Refinery wastewater	BOD: 94.6%, COD: 80.2%, TPH***: 92.6%, oil and grease: 90.4%, Cd: 94%, Pb: 92.5%, Cr: 93%, Fe: 94.8%, Ni: 92.2%, Cl: 57.7%	Oleh, Nigeria	[67]
Vertical subsurface flow constructed wetlands	Secondary refinery wastewater	Cd: 96%, Cr: 85%, Cu: 87%, Zn: 83%, Fe: 74%, Pb: 78%	Kaduna, Nigeria	[68]
Horizontal sub-surface flow CW	Petrochemical industries	BOD & COD: 95–97%,	Tamil Nadu, India	[69]
Bacterially augmented floating treatment wetlands	Oil field- produced wastewater	Hydrocarbon: 95%, COD: 90%, BOD: 93%	Chakwal, Pakistan	[65]

 Table 2 Refinery waste treatment through the Constructed Wetland (CW) method

*BOD, Biological Oxygen Demand; **COD, Chemical Oxygen Demand; ***TPH Total Petroleum Hydrocarbons; #TSS Total Suspended Solids

technologies, bioelectricity generation at the present power level is not yet significant for real-world applications. Therefore, the optimized use of such hybrid systems for the treatment of petroleum effluents appears to be a gap that can be addressed in further research.

Co-product Recovery

Another approach of resource recovery from refinery waste involves product recovery either from sludge or refinery waste. Oil sludge comprises a viscous mixture of sediment, water, oil, a combination of complex hydrocarbons like aliphatic and aromatic hydrocarbons. Besides, oil and hydrocarbons sludge also comprise heavy metals, asphaltenes, etc., that are a potential threat to the environment and health and are generated in substantial amounts during refining operations, cleaning of oil storage tanks, and even as a residue from conventional refinery wastewater treatment plants themselves [72].

The first step of oily sludge treatment involves oil recovery since it constitutes 80% of oil and 20% solids. Incorporating a microbial-derived biosurfactant is an alternative to increase the available reactive surface area of hydrophobic compounds. Surfactants accumulate at the hydrocarbon and water interphase and stabilize smaller droplets of the dispersed oil phase by reducing the interfacial tension. It increases the bioavailability of the oily contaminants for remediation [73]. Biosurfactant-mediated degradation of aged contaminated petroleum hydrocarbons is gaining rapid currency due to the resilient and versatile nature, low toxicity, eco-friendliness, biodegradability, and applicability of these biosurfactants even under extreme conditions of temperature, pH, and salinity. In a recent study, biosurfactant (rhamnolipid) application was demonstrated for oil recovery from oil tank bottom sludge [74]. The use of biosurfactants enables compliance with legislative requirements and environmental considerations. A deeper investigation and technology scale-up of ex-situ oil recovery after biosurfactant treatment appears to be warranted. Apart from oil recovery, biosurfactants have also been used to obtain petrochemical products such as emulsifying agents, biocides for sulfate-reducing bacteria against biocorrosion, and bitumen from coal tar.

Recovery of metal from spent catalysts is another opportunity for resource recovery in the petroleum processing sector. These catalysts, used in several process units involved in converting crude oil to fuels and petrochemicals, essentially contain valuable metals like Ni, V, Mo, Co, Cu, Pb, Zn, and Cr, which require regeneration once they lose their activity with time. However, due to such spent catalysts' toxic and hazardous nature, there exist stringent environmental regulations for their disposal. Recovery of these valuable metals is hence considered a beneficial solution from both economic and ecological standpoints. Biological processes (bioleaching) for metal extraction of refining spent catalysts are reported in several studies. Vemic et al. reported a comparative evaluation of chemical and biological leaching to recover molybdenum from spent catalyst, wherein 90% leaching efficiency in the chemical process and 70% in the biological process was observed; however, bioleaching is a simple and cost-effective technology for the extraction of metal from lowgrade ores and minerals concentrates [75]. Srichandan et al. showed 79 and 90% recovery of nickel and vanadium in 240 h through sequential biological leaching from decoked spent refinery catalyst [76]. In another study, Srichandan et al. reported the highest leaching of 97 and 92% for nickel and vanadium using a thermophilic consortium [77]. Microbe-mediated metal recovery results in enhanced metal recovery; such secondary raw material generation reduces the primary raw material dependence from environmentally unsustainable mining activities. Bioleaching of spent catalyst from bench to pilot scale-up to commercial deployment is yet to be industrially realized and offers an opportunity for exploration.

Challenges, Knowledge Gaps, and Perspectives

Waste generated from petroleum refineries comprises a complex mixture of hydrocarbons, a few of which are classified as priority pollutants, along with toxic metals. Although various approaches have been reported for the degradation of petroleum contaminants from soil and aquifers, no single technique to date is effective for the complete removal of these contaminants. The search continues, therefore, for scalable, affordable, and sustainable technologies. A core failure mode arises in transferring microbes from labs where they are cultured to the actual site of contamination. Incomplete understanding of microbial physiology under field conditions and bioavailability of hydrocarbons to the microbes contribute to the deployment challenge.

Scientists worldwide are also looking for cost-effective bio-stimulating agents for enhancing the growth of microbes under natural conditions. Methanotrophs are potential microbes for this purpose due to substantial susceptibility to unfavorable conditions and broad substrate specificity. In keeping with the increasing global focus on circular economy practices, novel materials derived from various wastes are sought to be reutilized in the form of bio-composites to remove petroleum hydrocarbons and metals from refinery wastewater. A deep understanding of the critical factors limiting the biodegradation of priority pollutants like PAH, biphenyls, etc. can help develop superior microbial consortia and treatment processes. Apart from developing feasible techniques for bioremediation, estimation of degradation using analytical techniques is also an important step. It helps determine the efficacy of microbes and offers insights into degradation kinetics under various conditions, and helps to understand the degradation products of such bioremediation at contaminated sites and their environmental implications.

This paradigm shift in refinery waste management by incorporating a waste-to-resource approach is an innovative step towards addressing sustainability goals. The integrated concept of bioremediation and resource recovery in the form of energy, water, and other valuable products can, in principle, enable companies that operate such refineries to make investments in environmentally sound emerging technologies in anticipation of multi-pronged benefits to the triple bottom line. However, significant scale-up challenges (such as for MFCs and microbial metals recovery), field application-oriented architecture design for large capacities of wastewater treatment, and appropriate sustainability assessments need to be addressed at all development and deployment stages. Similarly, in the constructed wetland approach, climatic conditions, widearea occupancy, plant species choice, and non-standard design are some illustrative challenges to be overcome. As product recovery is key to this integrated approach's economic viability, greater effort is predicated in this direction.

Conclusions/Opinion

Petroleum refinery waste comprises various hydrocarbons that are detrimental to health, biodiversity, and the overall environment. While bioremediation overcomes the wellrecognized limitations of physico-chemical treatment methods, the integration of multiple processes generally appears to deliver superior outcomes compared to individual component approaches. This review on the treatment of refinery waste through microbial or plant-based interventions suggests an emerging and exciting gateway for waste valorization and resource recovery while addressing the environmental impact and pushes the existing boundaries of bioremediation. Process intensification and bioengineering aspects of integrated refinery waste treatment and resource recovery shall also be explored for future research.

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