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Environmental Management

Biodegradation and biotransformation of petroleum hydrocarbons: progress, prospects, and challenges

Biodegradação e biotransformação de hidrocarbonetos de petróleo: progresso, perspectivas e desafios

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ABSTRACT

Oil is considered the main source of energy, essential for the realization of various industrial activities. However, it is detrimental to the environment, since in addition to the release of polluting gases during combustion, it is exploitation involves risks of water contamination, through leaks. Petroleum hydrocarbon pollutants are part of the recalcitrant compounds and their elimination from the environment causes enormous ecological impacts. The restoration of these environments is not a trivial challenge, because the natural degradation, without anthropogenic involvement, depends on the nature, composition, physical and chemical properties of these compounds. Thus, bioremediation appears as alternative in the biodegradation process through the addition of microorganisms, nutrients or other substances that cause and accelerate decontamination. The advantages of these methods involve efficiency and low cost, when compared to other technologies. This work deals with knowledge about the perspectives of application of bioremediation systems in the recovery of environments polluted by petroleum hydrocarbons, discussing progress, perspectives and challenges

Keywords: Petroleum hydrocarbons; Ecological impacts; Bioremediation

RESUMO

O petróleo é considerado a principal fonte de energia, sendo essencial para a realização de diversas atividades industriais. No entanto, é prejudicial ao meio ambiente, pois além da liberação de gases poluentes durante a combustão, sua exploração envolve riscos de contaminação da água, por meio de vazamentos. Poluentes de hidrocarbonetos de petróleo fazem parte dos compostos recalcitrantes e sua

eliminação do meio ambiente causa enormes impactos ecológicos. A restauração desses ambientes não é um desafio trivial, pois a degradação natural, sem envolvimento antropogénico, depende da natureza, composição, propriedades físicas e químicas desses compostos. Assim, a biorremediação surge como alternativa no processo de biodegradação através da adição de microrganismos, nutrientes ou outras substâncias que causam e aceleram a descontaminação. As vantagens desses métodos envolvem eficiência e baixo custo, quando comparados a outras tecnologias. Este trabalho reúne conhecimentos sobre as perspectivas de aplicação de sistemas de biorremediação na recuperação de ambientes poluídos por hidrocarbonetos de petróleo, discutindo avanços, perspectivas e desafios.
Palavras-chave: Hidrocarbonetos de petróleo; Impactos ecológicos; Biorremediação

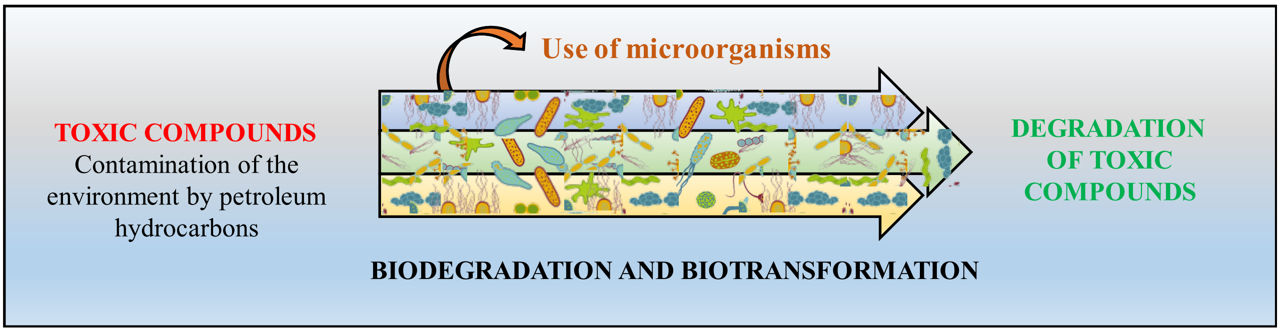
1 INTRODUCTION

Fossil fuels are still one of the fundamental energy sources around the world. However, when it comes to petroleum hydrocarbons, they often contaminate marine environments through accidents (YANTO and TACHIBANA, 2013). These contaminants, mainly polycyclic aromatic compounds (PAHs), are toxic and carcinogenic (PEAKALL *et al*., 1982; KHAN *et al*., 1986). In addition to these, there are aliphatic compounds, resin and asphaltene, considered less toxic than PAHs, but which deserve attention to their effect on ecosystems.

Thus, it is essential to use efficient processes capable of remedying the harmful effects of these compounds in the environment. However, because they are resistant compounds, conventional techniques, such as combustion, adsorption and others, have disadvantages. Among these, the high operating and maintenance costs stand out, in addition to the high energy demand (EL-NAAS, ACIO and TELIB, 2014; GHORBANIAN *et al*., 2014).

Therefore, biological treatment processes appear as competitive alternatives to remedy the pollution of these compounds in the environment. In these processes, petroleum hydrocarbons are used as sources of energy and carbon, making them simple, reliable and ecologically correct (LIU *et al*., 2013). Among these, biodegradation plays an important role. The bioremediation has stood out for being a process of reducing the pollutant's complexity through biodegradation, in which complex compounds are degraded into simple compounds by microorganisms, as shown in Figure 1.

Figure 1 – Biodegradation and biotransformation of petroleum hydrocarbons



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The advantages of biological methods are numerous, including the direct degradation of pollutants preventing increased environmental contamination; reduction of toxic pollutants, due to degradation into less toxic compounds; such as CO_2, water and salts, low operating costs, among others (EL-NAAS, ACIO and TELIB, 2014). Furthermore, it is noteworthy that the success of biodegradation depends, among others, on the composition of the pollutants involved, environmental parameters, the type and number of microorganisms used and the metabolism of these organisms (HARITASH and KAUSHIK, 2009).

Thus, this article aims to present a review of techniques for biodegradation and biotransformation of petroleum hydrocarbons as an alternative to recover contaminated environments. The main contribution of this review reflects on the prospects for the use of these processes, adding important knowledge about the nature and composition of petroleum hydrocarbons, sources of contamination and inherent impacts, in addition to addressing the particularities of biological systems in the decontamination of environments.

This article was organized into four main chapters, including introduction, methodology, development and conclusions. In the first, as discussed earlier, the importance of the subject studied and the work carried out is highlighted. In the second, describes search criteria, selection and inclusion of the works. In the third, mentions to the contamination of environments by petroleum hydrocarbons and the techniques of biodegradation and biotransformation. Finally, in the four, highlight the main contributions of this work, in addition to the perspectives regarding the theme.

2 METHODOLOGY

The present work is composed of a bibliographic review, which was developed using as the scientific databases of Google Scholar, Science Direct and pages of renowned institutions were used. This review was carried out in three main stages, as described below:

Research: in this step, keywords such as petroleum hydrocarbons, environmental impacts, biodegradation and biotransformation were used to identify the works in the aforementioned databases.

Selection: after the research, the most relevant works for the topic addressed were selected.

Inclusion: after a detailed reading of the works and selected information, the main concepts were included, always seeking to bring the main characteristics, innovations and perspectives on the subject of study.

3 DEVELOPMENT: PETROLEUM HYDROCARBONS, NATURE AND COMPOSITION

Hydrocarbons are the most abundant compounds in petroleum and are classified as saturated (single carbon-carbon bonds containing hydrogen) or unsaturated (hydrogen containing at least one carbon-carbon double or triple bond) (SPEIGHT, 2017; IHUNWO *et al*., 2021). These compounds are released into the environment through the decomposition of organic matter and through human activities, such as oil exploration and the burning of fossil fuels (BRUSSEAU *et al*., 2019; IHUNWO *et al*., 2021).

Hydrocarbons can originate from biogeochemical processes in the soil, as well as from the migration of deep oleaginous strata (DOHERTY and OTITOLOJU, 2013; PINEDO *et al*., 2014). Enter the soil after contamination of the surface with petroleum, petroleum products, wastewater and other substances containing hydrocarbons (DOHERTY and OTITOLOJU, 2013; PINEDO *et al*., 2014).

According to GENNADIEV *et al*. (2015), total petroleum hydrocarbons (HPT) or bitumoid hydrocarbon components are petroleum-based hydrocarbons found in crude oil, which reach the ground through natural and anthropogenic sources (ASTDR, 1999).

Petroleum hydrocarbons (HP) are petroleum compounds made up of carbon and hydrogen; and contain hydrocarbons with a wide range of molecular weights. HP is divided into alkanes, cycloalkanes, olefins and aromatics (KUPPUSAMY *et al*., 2020; IHUNWO *et al*., 2021).

Among hydrocarbons, polycyclic aromatic hydrocarbons (PAHs) are classified as carcinogenic and mutagenic environmental pollutants. They arise from the incomplete combustion of organic matter, mainly from the combustion of fossils, the disposal of petroleum and its derivatives, and the post-depositional transformation of biogenic precursors (TOLOSA *et al*., 2004). PAHs are composed of two or more fused benzene rings. They are characterized as colorless, white/pale yellow solids with low water solubility, high melting and boiling points and low vapor pressure (HARITASH and KAUSHIK, 2009).

PAHs are formed during the thermal decomposition of organic molecules and their subsequent recombination. Incomplete combustion at high temperature (500-800 °C) or subjecting organic material at low temperature (100-300 °C) for long periods results in the production of PAH. These hydrocarbons are characterized as colorless, white/pale yellow solids with low water solubility, high melting and boiling points and low vapor pressure (HARITASH and KAUSHIK, 2009). With the increase in molecular weight, there is a decrease in water solubility, an increase in the melting and boiling point and a decrease in the vapor pressure (CLAR, 1964; PATNAIK, 1999).

In the marine environment, petroleum hydrocarbons can originate from biological sources (plankton, plant or animal synthesis) and crude oil or fossil resources. Waxy materials from terrestrial organisms, degradation and burning of biological substances, synthesis of plant hydrocarbons, phytoplankton, bacteria, microscopic and large-scale algae are the main sources of biological entry of hydrocarbons into the sea (TOLOSA *et al*., 2004; ROSTAMI, ABESI and AMINI-RAD, 2019).

Petroleum hydrocarbons, whether of biological activities or fossil origin, once they enter the sea, undergo a series of changes (dissolution, transfer, sedimentation, dispersion, evaporation, weathering, chemical and photo-oxidation). After this diversity of physical, chemical and biological changes due to their low aqueous solubility and hydrophobic nature, hydrocarbons are eventually associated with particulate material and absorbed in deposition environments (TOLOSA *et al*., 2004; ROSTAMI, ABESI and AMINI-RAD, 2019).

3.1 Petroleum hydrocarbon contamination sources

Despite oil being one of the predominant energy sources to maintain the economic and social development of a country (ADIPAH *et al*., 2019). The global use of HP for energy and raw materials in various applications has increased with the extensive release of a wide variety of contaminants into the environment, affecting soil, surface and groundwater (OSSAI *et al*., 2020).

For SINGH and BORTHAKUR (2018), the rapid growth of the oil sector has created serious concerns for the increase in environmental pollution (MOHAMAD SHAHIMIN *et al*., 2016; SINGH *et al*., 2016a; SINGH *et al*., 2016b). HPs contaminants characterize the vast majority of organic compounds and by-products, classified as priority environmental pollutants, such as persistent organic pollutants (POPs) and PAHs due to their persistence and recalcitrant nature. They are mainly durable and stable, so they remain in the environment for a long time and do not degrade easily (ATSDR, 2011; OSSAI *et al*., 2019).

As an example of the pollution of petroleum hydrocarbons in water sediments, which occurred in the Gulf of Mexico in 2010, where high levels of contamination are still detected near point sources, such as oil installations and ports (UDDIN *et al*., 2021). Another major oil spill occurred in mid-October of this year, off the coast of California, in which more than 3000 barrels of oil were thrown from a ruptured pipeline and are covering the Pacific Ocean (CNN, 2021).

This hydrocarbon became one of the most important types of organic pollutants, due to leakage from underground tanks and accidental spills during its transport and disposal (ADIPAH *et al*., 2019). The disposal of waste generated in the crude oil industry, storage and refinery process is a worldwide problem (STASIK *et al*., 2015). Improper disposal of oily sludge leads to water and soil contamination that poses a serious threat to ground and surface water (SHAHIDI *et al*., 2015; FIRMINO *et al*., 2015).

With the expansion of HPs contaminated soil, efforts have been made to remedy the total contamination of these pollutants in the soil. Contaminants can enter the soil from point source (industrial and domestic) and diffuse source (mobilization and transport by floods) which can have long-term implications for the quality and functioning of soils, such as human health and food quality (DEFRA, 2009; ADIPAH *et al*., 2019).

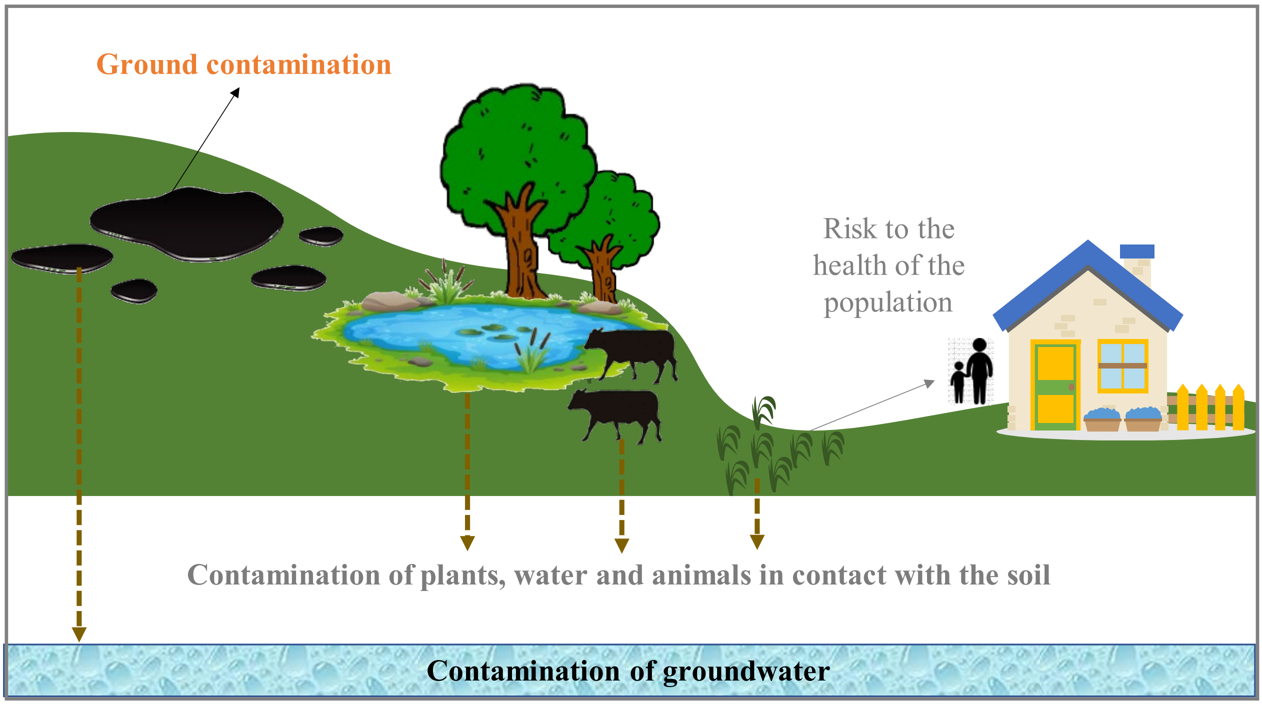
3.2 Impacts on the ecosystem contaminated by petroleum hydrocarbons

In the current scenario, the main sources of HP contamination are highlighted; factors that affect the accumulation of HPs in the soil; absorption mechanisms; translocation and potential toxic effects of HPs on plants (HAIDER *et al*., 2021). The presence of HPs contaminants in soil and water environments causes significant environmental impacts and represents a high risk for humans and other life forms, by contaminating the ecosystem, especially the soil (SAMMARCO *et al*., 2016; OSSAI *et al*., 2019).

According to the Environmental Protection Agency (USEPA, 2007), the release of HPTs into the environment threatens public health and safety by contaminating drinking water, causing fire and explosion risks, decreasing air and water quality, compromising agriculture, destroying recreational areas, habitats and food, and wasting non-renewable resources (USEPA, 1999; ADIPAH *et al*., 2019). Take the recent "potential ecological disaster" that killed fish, birds and algae through the rupture of the pipeline off the coast of California (CNN, 2021).

When HPs contaminate the soil, they begin to interact with its surroundings, including physical (dispersion), physical-chemical (evaporation, dissolution, sorption), chemical (photo-oxidation, auto-oxidation) and biological interactions (plant and microbial catabolism of hydrocarbons) (TRUSKEWYCZ *et al*, 2019). Oil-contaminated soil presents a number of risks (Figure 2), mainly to human health, from direct contact with contaminated soil, contaminant vapors and secondary contamination of water sources within and underlying the soil (THAPA, KC and GHIMIRE, 2012); causes organic pollution of groundwater; causes economic losses; environmental problems; and decreases agricultural soil productivity (WANG *et al*., 2008; THAPA, KC and GHIMIRE, 2012).

Figure 2 – Contamination of the soil and surrounding elements by petroleum hydrocarbons



Source: Authors (2022)

The concern stems from the volatility of HPs, as it can pose a risk of fire or even explosion, especially when steam enters confined spaces. These pollutants can interfere with the transmission of nutrients and water, leading to soil degradation. Oil Waste can become attached to soil particles and remain in the soil for years. HPs can also destroy the aesthetics of the environment by emitting an unpleasant odor, taste or appearance (ADIPAH *et al*., 2019).

In plants, HPs reduce seed germination and nutrient translocation and induce oxidative stress, disturb plant metabolic activity and inhibit plant physiology and morphology, reducing plant yield (HAIDER *et al*., 2021). The toxicity of petroleum hydrocarbons to microorganisms, plants, animals and humans is well established. The toxic effects of hydrocarbons on land plants and their use as herbicides were attributed to the oil that dissolves the lipid portion of the cytoplasmic membrane, allowing the cellular content to escape (CURRIER and PEOPLES, 1954; THAPA, KC and GHIMIRE, 2012).

PAHs are widely distributed contaminants that have harmful biological effects, toxicity, mutagenicity and carcinogenicity. It has also been a significant environmental concern due to its ubiquitous occurrence, recalcitrance, bioaccumulation potential and carcinogenic activity (HARITASH and KAUSHIK, 2009).

The health risks posed by HP pollution have led scientists to research, develop and implement risk-based remediation strategies for restoration and restoration of affected environments. To select the best treatment option for remediation, it is necessary to understand the nature, composition, properties, sources of pollution, type of environment, destination, transport and distribution of pollutants, degradation mechanism, interaction and relationships with microorganisms, intrinsic factors and extrinsic factors that affect remediation (VALENTIN *et al*. 2013; OSSAI *et al*., 2019).

3.3 Biodegradation and bioremediation of petroleum

As discussed, the composition of oil is complex, and may contain several hydrocarbon compounds (CHENG *et al*., 2014; XUE *et al*., 2015). When it contaminates water bodies, the application of cleaning processes is crucial to reduce the potential impacts caused by these compounds. Currently, different methods are being developed, among which bioremediation treatments deserve to be highlighted (BACHMANN, JOHNSON and EDYVEAN, 2014; JIANG *et al*., 2014).

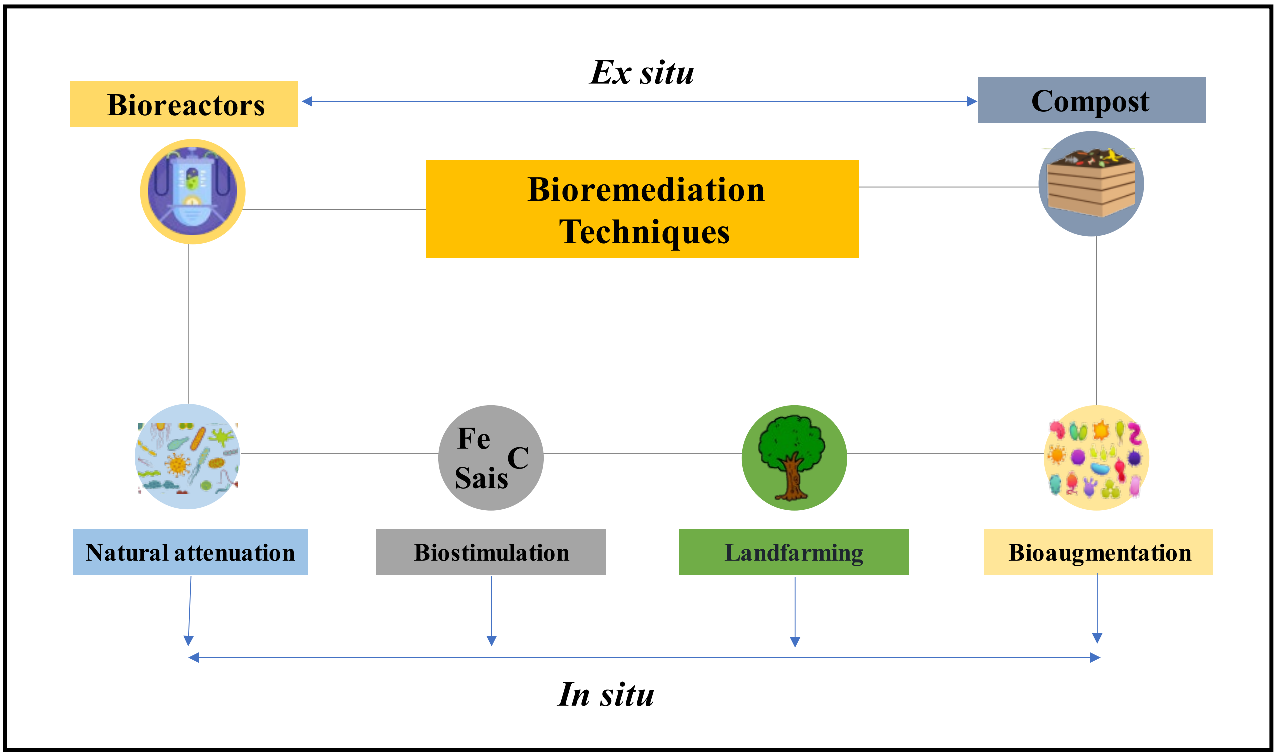
Bioremediation is a process that uses biotechnology as an accelerating method to treat toxic pollutants. In this process, biological agents capable of biodegrading pollutants are used. It is carried out with the use of native or exogenous microorganisms, or even genetically modified organisms, which promote a natural acceleration of degradation (PRINCE, 1996).

In general, the metabolism of microorganisms is used in order to reduce the concentration of pollutants, leaving them less toxic, or even eliminating them completely (YAKUBU, 2007), because these organisms use substances as a source of energy. Thus, bioremediation is seen as a tool to transform hazardous compounds with the minimum addition of chemical elements in the polluted environment (PROVIDENTI, LEE and TREVORS, 1993).

The techniques used for decontamination in these processes can be carried out in situ, that is, at the place where the contaminant leaks, or in an external environment (Ex situ) (MARTINS, 2019). Figure 3 presents a summary of the techniques commonly used in the bioremediation process. Among these, only composting and treatment in bioreactors are Ex situ techniques, the others are in situ.

In the natural attenuation process, biodegradation is carried out by organisms present at the place. In biostimulation, nutrients and salts are added to stimulate the action of these microorganisms in the degradation process. In Landfarming, there is the application of residue on the soil surface for microbial biodegradation, with aeration of the environment, and corrective agents, fertilizers and specific microorganisms can be added. Bioaugmentation involves the addition of selected microorganisms to the polluted site with a high potential for degradation. On the other hand, in the composting process, the contaminated soil, for example, is taken to a specific treatment site where chemical and physical conditions are controlled to optimize the degradation process. While the treatment with bioreactors, the contaminated material is taken to closed containers in which the microorganisms act under controlled conditions (ANDRADE, AUGUSTO and JARDIM, 2010; SUPERBAC, 2019; MARTINS, 2019).

Figure 3 – General technique of the Ex situ and in situ bioremediation process



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In this sense, in the implementation of degradation processes, several aspects must be considered, including the existence of microorganisms with degrading action and capacity, in addition to the availability of the contaminant and favorable environmental conditions (LEAHY and COLWELL, 1990).

3.3.1. Microorganisms with degrading action and capacity

The use of microorganisms with degrading capacity in the remediation of environments polluted with oil has become promising in the treatment of petroleum hydrocarbons due to their efficiency, low cost and environmental compatibility (ZHANG *et al*., 2021; DAI *et al*., 2022).

The presence of these organisms is already common in the primitive environment, however when there is pollution the proportion increases considerably (ZHANG *et al*., 2021). Furthermore, when the amount of microorganisms is not enough to effectively degrade petroleum pollutants, they can be added (SILVA *et al*., 2009). Degradation occurs as microorganisms use petroleum hydrocarbons as sources of energy and carbon. These organisms need to use several enzymes to degrade the compounds, thus, a single species is often not enough to complete the degradation (YANTO and TACHIBANA, 2014).

Several organisms are used in this bioremediation process, due to catabolic activity, such as fungi, algae and bacteria. Among these, bacteria and fungi are the classes of microorganisms actively involved in the degradation of organic pollutants from contaminated sites (HARITASH and KAUSHIK, 2009). The main attributions of the use of these beings, compared to expensive physical equipment and chemical reagents, are due to the price involved in the process and the efficiency in being able to mineralize hydrocarbons into carbon dioxide as the final product with the use of intracellular and extracellular enzymes (MARCHAND *et al*., 2017).

Bacteria generally play important roles in bioremediation, whereas fungi are more useful to degrade high molecular weight pollutants (WANG *et al*., 2012). The enzymatic systems of these beings are different, so the use together in the polluted environment increases the degradation power and the types of degradable substrates (GHORBANNEZHAD *et al*., 2018).

In the literature, there are reports of several genera of bacteria used in the process of bioremediation of petroleum hydrocarbons, including aerobic bacteria, such as Bacillus, Geobacillus, Burkholderia, Mycobacterium and Ralstonia, and also anaerobic bacteria, such as the genera Desulfobulbaceae, Desulfobacteraceae, Desulfarculaceae, Anaerolineaceae, Defirribacteres, Rhodospirillaceae and Magnetovibrio. Fungal species with oil degrading capacity involve ligninolytic and non-ligninolytic fungi, such as Bjerkandera, Irpex, Lentinus, Polyporus, Penicilium, Aspesgillus and Candida (HADIBARATA *et al*., 2009; MATTURRO *et al*., 2017; VARJANI, 2017; GHORBANNEZHAD, MOGHIMI and DASTGHEIB, 2018).

3.3.2. Factors Influencing Biodegradation of Petroleum Hydrocarbons

There are several factors that influence the biodegradation of petroleum hydrocarbons. These factors can be divided depending on the polluted environment. When it comes to the biodegradation of petroleum hydrocarbons in the soil, the main factors can be the oxygen content, pH, water content, temperature and nutrient concentration (WARTELL, BOUFADEL and RODRIGUEZ-FREIRE, 2021). Changes in pH, for example, can affect metabolic and growth rates and can be decisive in ensuring the availability of nutrient acceptors (BRAHMACHARIMAYUM, MOHANTY and GHOSH, 2019).

Temperature influences the biodegradation of oil as it affects the chemical composition of this compound, the rate of metabolism of its constituents by microorganisms and the composition of the microbial community used (ATLAS, 1975; CHENG *et al*., 2014). There are reports in the literature, for example, that the oxidation of alkanes together with the production of methane can occur in the temperature range between 20 and 55°C (GIEG *et al*., 2010; WANG *et al*., 2011; CHENG *et al*., 2013). Furthermore, studies have indicated that when it comes to sulfate-reducing bacteria, the ideal temperature range for the degradation of hydrocarbons is between 28 and 35°C (CHANG, SHIUN and YUAN, 2002; CRAVO-LAUREAU *et al*., 2004). On the other hand, ADELAJA, KESHAVARZ and KYAZZE (2015) when studying the degradation of phenanthrene by anaerobic organisms, concluded that increasing the temperature from 30°C to 40°C improves the biodegradation of this petroleum hydrocarbon.

In addition to these factors, when it comes mainly to aquatic environments, salinity is another aspect that influences the rate of degradation (BANIASADI and MOUSAVI, 2018). Salinity may be responsible for limiting bacterial activity due to the high unfavorable osmotic potential of salts (LI *et al*., 2016).

4 CONCLUSIONS, CHALLENGES AND PERSPECTIVES

In this work, bioremediation was approached as a competitive process in the degradation of petroleum hydrocarbons. This process uses microorganisms to decontaminate oil-polluted environments. It was seen that the main advantages related to this process are the cost and sustainability of the methods.

Advances in hydrocarbon biodegradation techniques are due to the joint use of microorganisms capable of efficiently remediating polluted environments. Recent studies have focused on evaluating the potential of fungi for the degradation of these toxic compounds.

The challenges are focused on the selectivity of degraded compounds being specific to certain types of microorganisms and even though several species of aerobic bacteria have degradation potential, the enzymatic abilities and degradation kinetics still provide limitations for the process. Regarding perspectives, it is believed that many studies are needed to clarify the limitations of biodegradation, especially regarding the factors that can influence the process.

This work is initial research that summarizes the main ideas and concepts for new advances in the bioremediation and biodegradation of petroleum hydrocarbons. Given the above, it is recommended for future work to carry out an approach on the biological techniques mentioned in this work using other contaminants, as well as a comparative study of biological processes with others. Finally, a deeper study on the negative environmental impacts is suggested, citing more examples of environmental impacts in Brazil and in the world. In this way, the field of action is expanded and the role of environmental decontamination techniques is highlighted.

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