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APPLICATION OF NANOMATERIALS FOR REMOVAL OF OIL WASTE: REVIEW[1]

APLICAÇÃO DOS NANOMATERIAIS PARA REMOÇÃO DE RESÍDUOS DE PETRÓLEO: UMA REVISÃO

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ABSTRACT

Petroleum is a difficult-to-remove chemical compound that leads to harmful environmental impacts when improperly discharged. In fact, this problem generates water, soil and air pollution, requiring advanced treatments such as photocatalysis, bioremediation and adsorption. In addition, nanomaterials make these techniques versatile and with a high percentage of contaminant removal. In this context, we seek to present a literature review on the main nanomaterials that can be used in the removal of petroleum residues. The research was carried out in the Web of Science database, where articles, books, theses and dissertations were selected, using the descriptors "nanoadsorption in Petroleum Waste" and "nanoadsorption in oils waste". The results demonstrate that metallic nanomaterials using adsorption and photocatalysis processes present 50-60% removal of petroleum residue, as well as materials using bioremediation techniques present 50% of the removal of this residue. Based on the results found, it can be concluded that metallic nanomaterials for adsorption and photocatalysis present a high rate of elimination of organic compounds, being promising for the removal of this residue.

Keywords: Ceramics, silicon carbide, nanoparticles, composite.

RESUMO

O petróleo é um composto químico de difícil remoção, que leva a impactos ambientais prejudiciais quando são descarregados inadequadamente. De fato, esse problema gera poluição da água, do solo e do ar, necessitando de tratamentos avançados, como a fotocatálise, biorremediação e adsorção. Além disso, os nanomateriais tornam essas técnicas versáteis e com alta porcentagem de remoção de contaminantes. Neste contexto, busca-se apresentar uma revisão da literatura sobre os principais nanomateriais que podem ser utilizados na remoção de resíduo de petróleo. A pesquisa foi realizada na base de dados do Web of Science, onde foram selecionados artigos, livros, teses e dissertações, utilizando os descritores "nanoadsorption in Petroleum Waste" e "nanoadsorption in oils waste". Os resultados demonstram que nanomateriais metálicos utilizando processos de adsorção e fotocatálise apresentam 50-60% de remoção de resíduo de petróleo, como também materiais utilizando técnicas de biorremediação apresentaram 50 % da remoção deste resíduo. Com base nos resultados encontrados, pode-se concluir que nanomaterias metálicos para adsorção e fotocatálise apresentam alta taxa de eliminação de compostos orgânicos sendo promissores para remoção deste resíduo.

Palavras-chave: *Cerâmicas, carbeto de silício, nanopartículas, compósito.*

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INTRODUCTION

Petroleum is a fossil fuel that has many applications, in addition to being used as a precursor to different non-renewable energy sources. Thus, it has a series of negative aspects, such as the high potential for environmental impacts, causing deaths of animals and plants and compromising the quality of water, air and soil (THOMAS, 2004). For example, in 2019 the amount of oil effluent spilled into the sea varies between 83-240 million gallons of oil (EUZEBIO; RANGEL; MARQUES, 2019).

This fuel has been increasingly studied and explored, being one of the economic divisors of countries like Brazil, as well as a diffuser of negative environmental impacts. In Brazil, there are federal laws such as the National Environmental Council resolution (CONAMA, 2013) and Law nº 6.938 (1981) that regulate the remediation of contaminated soil and water, aiming to convert these situations (THOMAS, 2004).

The fossil hydrocarbon compound, when spilled into the marine environment, prevents the passage of light resulting in the growth of organisms that need light (phytoplankton), negatively affecting the entire food chain (THOMAS, 2004). Studies show that this oil spill can affect mangrove environments, as the oil impregnates the plant's root system, preventing the absorption of oxygen and nutrients (MARTINS *et al*., 2015).

With the growth of nanotechnology came advanced techniques in order to remove and treat these surface waters, aiming at high performance and a lower cost, allowing the reuse of this water. The nanomaterials, due to their greater surface area due to particle size reduction, increase the interaction of the material with the environment, making it feasible to be applied to remove organic and inorganic contaminants (THOMAS, 2004). However, the use of these materials over time can cause toxicity effects where they are applied, such as the death of marine animals and microorganisms (GEANKOPLIS, 2018).

Techniques for removing contaminants such as adsorption, photocatalysis and bioremediation are alternatives to try to solve the generated environmental problems. These techniques differ in the mechanism of action, each specific for each case, however when using nanomaterials these methodologies become more versatile using in several cases and increasing the removal of contaminants content (QU *et al*., 2013).

In this context, the present work aims to present a biological review of nanomaterials for possible application in the removal of oil residues, aiming to increase knowledge of this area and the remediation or mitigation of the area affected by the oil, in the same way that compounds are produced containing two or more nanomaterials for the removal of the petroleum product, the research was carried out on the platforms Science Direct (https://www.sciencedirect.com/) and Web of Science (https://www-periodicos-capes-gov-br) in the period 2010 - August 2021.

BIBLIOGRAPHIC REVIEW

PETROLEUM

Petroleum is defined as a fossil organic compound being a viscous aqueous solution that has in its chemical composition hydrocarbons (such as alkanes), about 10% sulfur, 5% oxygen and 1% nitrogen, in addition to other metallic elements (NASCENTES, 2011). This compound is found underground impregnated in sedimentary rocks at depths of 10 meters to 100 km below the surface on dry land and submerged.

This solution, rich in hydrocarbons, is formed from organic matter that undergoes anaerobic processes when subjected to high pressure or temperature (NASCENTES, 2011). In prehistory, the oil contained more oxygen due to microorganisms having a greater number of fatty acids, over the years the microorganisms showed a decrease in these numbers of fatty acids, resulting in oil rich in hydrocarbons (PELCZAR; CHAN; KRIEG, 1997; ROCHA; ROSA; CARDOSE, 2010).

The fossil compound is not only used as fuel but in several products such as footwear and cosmetics. Due to this great global demand, oil production is carried out in several countries and has been transported by oil tankers across oceans and pipelines, causing damage to the environment. For example, environmental accidents with oil have already been reported in history, such as: (i) Spill in Canada in 1988 of the cargo ship Odyssey, (ii) Guanabara Bay in 1997 and 2000 periods, (iii) Campos Basin in November 2011 and (iv) in 2019, the oil spill off the Brazilian coast (Northeast and Southeast region) (GOMES, 2014; PENA *et al*., 2020).

For Martins et al. (2015) oil is used as one of society's main sources of energy, although it is a non-renewable natural resource. The exploration of this product leads to major environmental impacts, requiring an environmental license, determining requirements to minimize or mitigate these impacts, Table 1 shows the chemical composition and variation of hydrocarbons present in oil.

Table 1 - Representation of the chemical composition and variation of hydrocarbons present in the oil.

PETROLEUM			
ELEMENT (%)		**HYDROCARBONS (%)**	
Carbon	82	Cycloalkanes	30
Hydrogen	12		
Nitrogen	4	Aromatics	40
Oxygen	1		
Salts	0.5	Alkanes	30
Metals	0.5		

Source: Author's construction.

NANOTECHNOLOGY

According to REG European Commission (2019) and Maynard (2011), nanomaterial can be natural or manufactured, being an aggregate or an agglomerate of particles in one or more dimensions (x, y and z) external or internal in the numerical size range of 50% of 1 to 100 (nm), depending on the area such as environment, health and safety, the numerical size range limit must be between 1 to 50 (%) and can be classified by its surface area in a specific volume of 60 m^2 cm^{-3}. In the areas of science and engineering, nanostructured materials must have new properties and different functions compared to the same materials on a macro scale, which are directly linked to new molecular techniques whose objective is to manipulate matter (ROCO, 2001).

Recent advances in nanotechnology offer several growth opportunities for the development of new surface water treatment systems. The various processes used by nanotechnology enable high performance (they do not depend on large structures) and thus allow the use of this source of treated water to expand the water supply. Water treatments involving nanotechnology are the main barrier for large-scale application, indicating the problem that after use it can impact the environment and health (QU; ALVAREZ; LI, 2013).

Some of the applications of these nanomaterials depend on scalable and discontinuous properties, such as: (i) high surface area, (ii) fast dissolution, (iii) high reactivity providing strong adsorption, (iv) superparamagnetism, (v) plasmonic resonance and (vi) quantum confinement effect (MAYNARD, 2011).

Qu et al. (2013) and Kanchi (2014) commented that industries that apply nanotechnology to remove contaminants from effluents can obtain purified water, reducing work, time and costs, in addition to solving several environmental issues. In addition to the treatment, they suggest that before applying this nanomaterial for any elimination involving aqueous solutions, several toxicity tests must be carried out, generating safety data and standard operating procedures (SOPs). Table 2 presents a series of nanomaterials, with their desirable properties for different applications.

Table 2 - Use of nanomaterials for adsorption and photocatalysis processes.

Process	Nanomaterials	Properties of desirable nanomaterials	Applications
Adsorption	Carbon nanotubes	Easy reuse, high surface area and tunable surface chemistry	Detection and adsorption of contaminants
	Metal oxides	High surface area, more adsorption sites and short intermolecular diffusion between particles	Adsorbent media filters
	Nanofibers	Shell surface chemistry	Selective adsorption
Photocatalysis	Nano-TiO_2	Photocatalytic activity in the UV light range and low toxicity	Photocatalytic reactors
	Fullerene	High selectivity	Solar disinfection systems

Source: QU *et al*. (2013).

ADSORPTION

Adsorption consists of the deposition of pollutant molecules (adsorbate) on the surface of a solid material (adsorbent) and is characterized as one of the most effective methods compared to others due to its ease of operation and for being relatively cheap (SIDDIQUI; CHAUDHRY, 2017).

One of the ways to achieve adsorption is through the use of a material with high surface area, high adsorptive capacity and considerable porosity (CAVALCANTI, 2009). However, for it to be applied on a large scale, it is necessary to carry out adsorption tests on a laboratory scale in order to determine the ideal operating conditions (pH effect, adsorbent dosage, contact time, adsorbate concentration) and reduce raw material costs, whether in continuous or batch (GEANKOPLIS, 2018). Adsorbent materials commonly used on an industrial scale are activated silica, alumina, zeolites and different types of activated carbon (HOWE *et al*., 2016). It is worth analyzing that any material that has significant carbon content in its composition or that may have its surface area increased will be considered and used as activated carbon.

Adsorption can be via Van der Waals forces (physical adsorption) or by chemical bonding between the functional groups of the adsorbent and the adsorbate. Physical adsorption is best evaluated by quantifying the surface area, while the other is evaluated by the number of functional groups found on the surface, this characteristic can be favorable for desorption (CAVALCANTI, 2009).

The adsorption process is widely used to remove organic or inorganic contaminants in water treatment. The efficiency of this adsorbent is given by the surface area, active sites and adsorption kinetics, nanoadsorbents are considered an alternative to improve these properties (HOWE *et al*., 2016).

The recovery of nanomaterials after adsorption is called desorption, provided by the addition of magnetic particles (Fe2O3) and acidic (HNO3 and HCl) or basic (NaOH and KOH) reagents (DALLAGO; SMANIOTTO; OLIVEIRA, 2005). Furthermore, after the adsorption and degradation of the adsorbates, the oils are reused in the production processes of fuel and its derivatives through techniques using dispersing agents, surfactants and gelling agents (CARVALHO, 2006; CARDOSO, 2007).

2.3.1 Types of nanoadsorbents

Carbon-based nanoadsorbents are nanomaterials having most of their chemical composition in carbon, namely nanotubes, fullerenes, graphene, activated carbon and quantum dots. They are produced by different techniques, such as laser ablation, solvothermal treatment, hydrothermal carbonization, pyrolysis and chemical vapor deposition. Different methods are sought to obtain materials of different morphologies and porosities (TITIRICI; ANTONIETTI, 2010; ROCHA; ROSA; CARDOSO, 2010).

Metallic nanoadsorbents (Fe_2O_3, TiO_2 and Al_2O_3) are widely used in nanoadsorption, being effective and inexpensive. The adsorption process on metallic oxides involves three mechanisms such as;

(i) complexation between dissolved metals and oxygen, (ii) rapid adsorption of metal ions by the external surface, (iii) diffusion by grain control with limitations due to micropores. The production of these oxides occurs by chemical vapor deposition and solvothermal treatment (TRIVEDI; AXE, 2000).

Dendrimers are used as polymeric nanadsorbents due to their structure having internal shells (hydrophobic) and external branches (functional groups of hydroxyls and amines), being the hydrophobic part for the adsorption of organic compounds while the branches for the adsorption of heavy metals. This adsorption is based on electrostatic interactions, hydrogen bonding interactions, metal complexation and hydrophobic effect (CROOKS *et al*., 2001).

HETEROGENEOUS PHOTOCATALYSIS

The concept of heterogeneous photocatalysis is to separate electrical charges in specific solids through light excitation, with oxidation-reduction processes taking place, promoting the degradation of organic pollutants adsorbed on the surface of the adsorbent (COMPARELLI *et al*., 2005). These materials have two energy regions: one with lower energy (Valencia Band) and another with higher energy (Conduction Band), the region between these energy bands is called the band gap (GOGATE; PANDIT, 2004).

The photocatalysis process begins when the semiconductor absorbs photons with energy equal to or greater than the band gap energy, forming a vacancy in the valence band, these vacancies are responsible for the formation of hydroxyl radicals ($^{\bullet}OH$). In addition to vacancies, they can form other charge-bearing species, resulting in reducing and oxidizing sites, promoting the oxi-reduction process, degrading them in rational environments. The efficiency of this process is directly linked to the competition between these charges that were removed from the surface of the adsorbent (CHATTERJEE; DASGUPTA, 2005).

BIOREMEDIATION

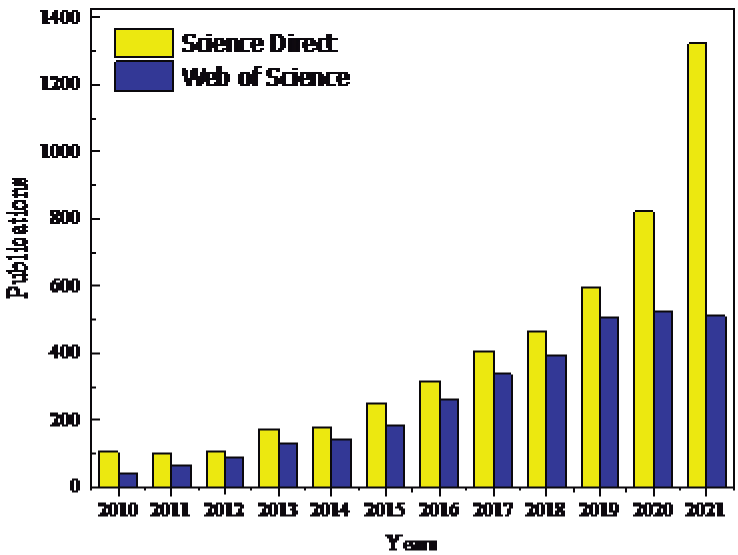
Bioremediation uses microorganisms with the potential to degrade, modify or remediate toxic substances or wastes harmful to the environment (TORTORA; FUNKE; CASE, 2005). For Borém (2005), the bioremediation process is considered the most effective in treatments to clean up soils compared to incineration, washing and landfill processes, in addition to having a lower cost.

The bioremediation process in oil is analyzed for the removal of aromatic hydrocarbons incorporated with xenobiotic compounds that can be degraded by microorganisms through specific catabolic routes (DA-CUNHA, 2008). Furthermore, in bioremediation, environmental factors of a physical, chemical and biological nature influence the ability of a bacterial or fungal system to biodegrade the oil, the main parameters being: temperature, pH, humidity, light, salinity, oxygen content and physical nature of the matrix (soil, water and sediment) (ANDRADE; AUGUSTO; JARDIM, 2010). As well, organic substances (petroleum and oils) are metabolized by co-metabolism, having methane and carbon dioxide as by-products and can be used by other species as growth substrate (WETLER-TONINI; REZENDE; GRATIVOL, 2011).

METHODOLOGY

This work was characterized using a bibliographical review, the accomplishment consisted of analyzing books, scientific articles, theses and dissertations in the Science Direct (https://www.sciencedirect.com/) and Web of Science (https://www-periodicos-capes-gov-br), in the period 2010 - Oct 2021, demonstrating the nanomaterials that were studied to remove petroleum residues using various processes. Figure 1 shows the articles/dissertations and theses found in the Web of Science and Science Direct. The following words were used in the search: Nano Adsorption in Oil Waste and Nano Adsorption in Oil Waste and their respective descriptors in English: “Nano Adsorption in Petroleum Waste”. All works analyzed were those that presented the topic described and in association with oil removal processes, aiming at improvements in the removal of these residues.

Figure 1 - Published papers by year in the Science Direct and Web of Science databases.



Source: Author’s construction.

RESULTS AND DISCUSSIONS

Table 3 presents the materials found for oil residue removal, using different processes and their references.

Table 3 - Nanomaterials for use in the removal of petroleum residue.

Material	Process	Methodological aspects	Reference
Nano-Zirconium@SiO_2	Adsorption	Synthesized using combustion processes	PETKOWICZ *et al*. (2009)
Nano-TiO_2	Photocatalysis	Synthesized using the sol-gel process at low temperature	TIELAS *et al. (*2014)
Carbon Nanotubes and (Pseudomonas)	Bioremediation and Adsorption	Synthesized using the catalyzed chemical vapor deposition technique.	TORTORA *et al*. (2005)
Carbon Nanotubes and (Allochthones)	Bioremediation and Adsorption	Synthesized using the physical deposition technique.	MARIANO (2006)
Nanocomposite (TiO_2 /Fe_3O_4)	Photocatalysis	Synthesized using the hydrolytic sol-gel process.	MOURÃO *et al.* (2019)
Nano-Chitin and Chitosan	Adsorption	The synthesis was by chemical processes	CAMPELO (2013)

Source: Author's construction.

The nanomaterial containing zirconia with silica oxide, reveals to be an excellent material to adsorb stable organic molecules, this composite presents twice as much adsorption as ZrO_2. With an increase in adsorption temperature, SiO_2 exhibited a decrease in adsorption capacity, identifying an endothermic process. Because their similar surface areas have a synergistic effect indicating colloidal stability of excess anionic charges, different from separate materials, this excess charge occurs due to the high zeta potential (they tend to repel particles) decreasing the probability of forming aggregates and resulting in the material of greater stability in suspension, this excess of charges increases the adsorption process. The same nanomaterial was tested for adsorption capacity using photocatalysis processes demonstrating the increased degradation of organic material compared to separate materials (PETKOWICZ *et al*., 2009).

Studies show that the photocatalysis technique using Nano Titanium Oxide (TiO_2) can remove 60% of total organic carbon from a solution, in addition to removing the color of the compound when reached. The advantages of this process are that it can use both UV radiation and sunlight with light sensitivity, reducing electricity costs. This technique can completely degrade all organic compounds and turn them into water or carbon dioxide. Titanium oxide is a photocatalytic semiconductor, has low toxicity, high chemical stability (nanoparticle format is more stable than powder) and prevents aggregation of nanoparticles in the environment. Carbon removal is dependent on parameters used such as: catalytic mass, pH, the addition of the oxidizing agent and temperature (PETKOWICZ *et al*., 2009; QU *et al*., 2013; TIELAS *et al*., 2014).

Due to the difficulty of working with organic contaminants due to their chemical composition and transformation to which they are subject, pseudomas that are capable of degrading petroleum residues (~50%) were used due to their need for carbon and energy and in contact with the oxygen they metabolize two carbons of a large molecule, this degradation depends on environmental factors such as: pH, temperature, oxygen content, light and enzyme activity inhibitors. By adding carbon nanotubes, this nanomaterial adsorbs small molecules due to the large surface area, making the process viable, studies indicate that by making changes to the surface of the nanotubes, the adsorption increases due to the presence of functional groups (TORTORA *et al*., 2005; BONADIMAN, 2007; ROCHA *et al*., 2010; WEBER; SANTOS, 2013).

The literature describes that allochthonous bacteria together with the surfactant Tween 80® can degrade 45.5% of the hydrocarbons present in oil due to the surfactant increasing the bioavailability of hydrocarbons, making it possible to use carbon nanotubes to adsorb fragments of oil hydrocarbons (TORTORA *et al*., 2005; MARIANO, 2006; WEBER; SANTOS, 2013).

The sol-gel technique was used to produce the magnetite nanocomposite (Fe_2O_3) coated with SiO_2, aiming at the magnetic effect of the magnetite. This method proved to be efficient resulting in heterostructures, since the material presented photocatalytic and magnetic properties. They have not been tested for the removal of organic compounds due to the enormous difficulties that can be encountered in the installation of photocatalytic systems (laboratory scale for industrial scale) (MOURÃO *et al*., 2019).

The removal of oil using chitin and chitosan shows that the fiber performs a better removal of this residue compared to natural fibers due to a greater amount of amino and hydroxyl groups in the polymer chains, resulting in chemical effect adsorptions, these chemical groups serve as sites for electrostatic interactions forming stable complexes (BORGES, 2002; CAMPELO, 2013).

CONCLUSION

Nanomaterials containing metallic nanoadsorbents are promising in the removal of petroleum residues by adsorption and photocatalysis processes, having 60% of total removal of hydrocarbons present in petroleum, as well as toxicity studies of the material applied over time, as well as the transposition from laboratory to industrial scale. The Bioremediation/Adsorption Process presents a 50% hydrocarbon removal rate using bacteria to degrade the residue and nanomaterials to adsorb these fragments. It is concluded that materials in manometric scale present an increase in the removal rate compared to the macro scale, being promising for use in oil residue removal.

FUTURE PERSPECTIVES

The topic addressed in the respective work constitutes some areas with intense research activities in waste removal, specifically oil and oils, as possible techniques and the use of nanomaterials to demonstrate the potential for removing these wastes, resulting in their reuse. In addition, it is necessary to mention the importance of advances in the area of nanotechnology, as well as in engineering, contributing to the development of techniques and new nanomaterials with a lower toxicity effect on the environment and potential for removing pollutants.

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