

REVIEW ARTICLE

GREEN SYNTHESIS OF NANOPARTICLES FOR THE REMEDIATION OF CRUDE OIL CONTAMINATED SOIL AND WATER- A REVIEW

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ARTICLE DETAILS

Article History:

Received 14 May 2023

Revised 18 June 2023

Accepted 21 July 2023

Available online 28 July 2023

ABSTRACT

Oil exploration and production areas are prone to crude oil contamination and conventional remediation techniques are usually toxic, time consuming and expensive. Nanoparticles have emerged as an effective, rapid, and efficient alternative with the green synthesized ones more environmentally friendly and cost-effective. This review looks at the common sources and constituents of crude oil contamination, available conventional remediation technologies and the dynamics associated with utilizing nanotechnology in the remediation of crude oil contaminated soils and water as an alternative. Chemically synthesized nanoparticles have successfully been used for remediation of both crude oil contaminated soil and water though associated with subsequent environmental pollution. Iron nanoparticles stand out since they are able to remove a number of contaminants. Whereas green synthesis involves the use of either plant extracts or microbes in the reduction of metal salts, plant extracts are more effective and their kinetics equivalent to chemical synthesis. There is need to evaluate the applicability of green synthesized nanoparticles in the field by understanding their fate and potential toxicity in the environment using multimedia fate models for simulation and life cycle assessment.

KEYWORDS

Green synthesis; Nanoremediation; Crude oil; environmental fate.

1. INTRODUCTION

Oil exploration and production zones are prone to contamination with petroleum hydrocarbons which consist of alkanes, alkenes and aromatics (Vu and Mulligan, 2023). The common causes of this oil pollution being accidents involving tankers, barges, pipelines, refineries, drilling rigs and storage facilities (Iskander et al., 2020). Petroleum hydrocarbons are linked to health threats such as outlying neuropathy, dizziness, headaches, nervous system disorder, eyes, lungs and skin irritations (Enuneku et al., 2021). They also cause destruction to the soil fertility and structure thus affecting the germination and growth of plants in soils (Nnaji, 2018).

Remediation of oil contaminated water and soil can be carried out either on the contaminated site (in situ) or away from the contaminated site (ex situ). Some of the ex-situ methods include; pump and treat, dig and dump, incineration, oxidation while in situ methods include soil vapor extraction, solidification/stabilization, chemical oxidation, soil washing, bioventing, steam injection, electrical and radio-frequency heating. Ex situ methods are usually more expensive and more efficient than in situ methods since they require less time, can easily be monitored and achieve more uniformity (Nnaji, 2018).

Remediation technologies such as thermal desorption and soil washing work within minutes or hours and cost between \$39 – 331/t, excluding

excavation, transportation, permitting, samples analysis and equipment rentals. Technologies such as modified electro kinetic remediation, in-vessel composting, and in-situ chemical oxidation achieve high removal performance after several days within a month and cost between \$22 – 131/t, excluding other capital costs. Technologies, such as phytoremediation, bio piling, landfarming, and bioventing work after several months or years and cost between \$8 – 131/t of a contaminated site (Ahmed et al., 2022).

Technologies such as nano bioremediation, carbon-based nanomaterials, metal-based nanomaterials and nanocomposites have recently been adopted for their high reactivity and efficiency (Nnaji et al., 2019). However, production of these nano technologies may have associated environmental concerns depending on the synthesis method employed, hence the recent promotion of green synthesis (Alazaiza et al., 2021; Nnaji and State, 2018; Yarima et al., 2020; Das et al., 2019).

Green synthesis of nanoparticles in particular, involves the use of biological precursors (plants and microbes) to produce environmentally friendly nanoparticles (Jadoun et al., 2021). The process not only eliminates use of expensive chemicals but also consumes less energy. Plants extracts require less incubation time for synthesis when compared to bacteria and fungi (Sarkar et al., 2021). The phytochemicals present in plant extracts not only act as reducing agents for metal salts but also

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DOI:
10.26480/acmy.02.2023.62.67

facilitate the capping and stabilization of the nanoparticles (Ssekatawa et al., 2021).

Green synthesized nanoparticles have only been tested for the remediation of oil contamination at laboratory-scale (Chandraker et al., 2021). Large scale application is feared to face challenges with inconsistent nanoparticle characteristics such as size, shape, and stability as well as selection of suitable green reagents and assembly methods. Other concerns include; availability of raw materials, reaction time, energy requirement, cost of production as well as the quality, stability and reusability of the nanoparticles (Ying et al., 2022).

The purpose of this review is to identify the different methods employed in the remediation of oil contamination with their associated pros and cons. More emphasis on the green synthesis of nanoparticles as an alternative to chemical synthesis is assessed as a nanoremediation technology. The dynamics involved such as the choice of plant raw material, synthesis pH and temperature, concentration of extracts and the protocols used in the synthesis to optimize the process (Xu et al., 2022). The use of multimedia fate models for lifecycle assessment and prediction of exposure concentrations of the nanoparticles in the environment to further assess their viability is also looked at.

2. SOURCES, EFFECTS, TRANSPORT AND WEATHERING OF CRUDE OIL CONTAMINANTS

Oil and gas exploration involves creation of exploration wells to confirm the presence of hydrocarbons, thickness and internal pressure of reservoirs (Devold, 2018). The exploration stage is followed by the appraisal stage which involves drilling more wells to evaluate the size of the field as well as determining the nature of the reservoir and the number of confirming wells (MEMD, 2020). The production stage then follows with crude oil, natural gas and their derivatives undergoing processing into refined products such as diesel, gasoline, petrol and lubricants (Ahmad et al., 2020).

Most of these infrastructures put in place from exploration to production are potential sources of crude oil contamination. Drilling operations, oil well blow outs, underground tank and pipelines leaks, illegal waste disposals and overflows at gathering stations often cause oil spills (Iskander et al., 2020). For instance, the Niger Delta was reported to have a 50% oil spills from corrosion, 28% from sabotage, 21% from oil production operations and the remaining 1% from engineering drills, oil well blowouts, machine failure, loading and offloading vessels and storage (Nuhu et al., 2021).

Crude oil is composed of more than 17,000 hydrocarbons whose classification depends on the most prevalent compound present in it. The hydrocarbon components can be classified as paraffins, naphthenes and polycyclic aromatic hydrocarbons (Ahmad et al., 2020). Crude oil can also be classified as; saturates, aromatics, resins, and asphaltenes (SARA). Saturates are chemical bond hydrocarbons for example straight-chain alkanes, cycloalkanes, and branched alkanes (Iskander et al., 2020). Aromatics include the polycyclic aromatic hydrocarbons (PAHs) which have two or more rings and single ringed aromatics, such as benzene, toluene, ethylbenzene, and xylenes (BTEX) (Atoufi and Lampert, 2020). Resins and asphaltenes are responsible for the oil's black color and contain other elements such as sulfur, nitrogen, and oxygen (Iskander et al., 2020).

The contaminants of concern in crude oil spillage include; Total Petroleum Hydrocarbons (TPH), Polycyclic Aromatic Hydrocarbons (PAHs), and Volatile Organic Compounds (VOCs) (for example benzene, ethylbenzene, toluene, and xylene (BTEX)) due to their toxic properties and negative effects on human health (Ahmed et al., 2022). The movement of crude oil in soil depends on soil saturation level, soil type, specific gravity and solubility of contaminants, velocity and direction of groundwater flow and the infiltration rate from the source (Dago and Justin, 2019). High molecular weight hydrocarbons are either absorbed or concentrated in the surface soil while the lower molecular weight hydrocarbons are self-purified by volatilization (Hu, 2020; Ossai et al., 2019). If the pollution intensity and hydrocarbons content is high, crude oil can migrate to the ground water table (Hu, 2020).

In surface water, crude oil transforms physically, chemically and biologically through spreading, drifting, dispersion, stranding and weathering (Balogun et al., 2020; Lu et al., 2019; Ossai et al., 2019). The low molecular weight hydrocarbons tend to float on the surface and form thin surface films while the higher molecular ones sink to the bottom of the water sometimes forming a viscous substance (Ossai et al., 2019). Degradation by naturally occurring microorganisms may also take place, at a faster rate with the lighter oils when compared with the heavy oils (Iskander et al., 2020).

Oil contamination of less than 1mg/kg may promote plant growth by converting carbon, hydrogen, oxygen, nitrogen in the oil into substances needed for plant growth by lignification. However, depending on the plant species, relatively high oil contents can inhibit plant growth (Hu, 2020). Some crude oil pollutants in soil can accumulate in food thereby changing the quality and leading to subsequent harm to human health. Crude oil soil pollution also has acute effects on microorganisms such as earth worms.

In waterbodies, surface films of floating hydrocarbons prevent oxygen from dissolving in water and the hydrocarbons that sink and penetrate accumulate in aquatic organisms. When consumed by animals and humans, petroleum pollutants can lead to destruction of red blood cells, damage to the immune system, organ failure, skin, mouth and nose allergies among others (Hu, 2020). Additionally, heavy metals do not biodegrade yet they cause cytotoxic, mutagenic and carcinogenic effects which lead to loss of biodiversity of aquatic animals (Enuneku et al., 2021). Polycyclic aromatic hydrocarbons (PAH) are also mutagenic and carcinogenic and their prolonged contact with soil stimulates soil aging which leads to the resistance of soil to remediation (Ahmad et al., 2020).

3. REMEDIATION OF CRUDE OIL CONTAMINATION IN SOIL AND WATER

selecting the best remediation option for oil contamination requires one to comprehend the nature, composition, properties, sources of pollution, type of environment, fate, transport and distribution of the pollutants, mechanism of degradation, interaction and relationships with microorganisms, the intrinsic and extrinsic factors affecting remediation according to (Ossai et al., 2019). It is also important to evaluate and predict the chemical behavior of the pollutants with the short and long-term effects and mitigate the effects of pollution and limit exposure to the pollutants.

Crude oil remediation can be categorized into chemical, biological, thermal and physical methods. Chemical remediation refers to remediation using chemical technologies such as soil vapor extraction, chemical oxidation, chemisorption among others. Despite being highly effective, these technologies produce harmful by products, are expensive and are unable to treat large amounts of pollutants (Ahmed et al., 2022).

Physical remediation involves removal of contaminated media, treatment (soil washing, incineration, adsorption) and return of the media to the original site. Mechanical oil removal using skimmers, booms, pumps and mechanical separators are also physical remediation methods (Nnaji, 2018). Booms are not only expensive and tedious, but also require a secondary method for oil removal. They are commonly used together with skimmers and sorbents while the fire-resistant booms are specifically used with thermal methods (Pete et al., 2021a).

Thermal remediation on the other hand involves the use of heat for cleaning up contaminated media. Some of the thermal techniques include; thermal desorption, incineration, steam heat injection and thermal conduction (Nnaji, 2018). Oil is ignited using kerosene, propane or butane torches, weed burners and absorbents soaked in fuel deployed from either a vessel, land or helicopters. Though in situ surface burning is efficient and rapid, it has a limited time in which burning is effective, leads to massive air pollution, sinkage of burned residue and also has a risk for secondary fires or explosions (Pete et al., 2021b).

Bioremediation involves cleaning up pollutants from the environment using microorganisms. Bioremediation can be through bioaugmentation, bio stimulation and phytoremediation (Yarima et al., 2020). Bioaugmentation involves the use of microbial population supplemented by the oil degrading bacteria whereas bio stimulation refers to the use of growth stimulating substances for indigenous degraders. Phytoremediation involves the use of plants (trees and herbaceous plants) and their associated microorganisms to remove contaminants.

Following a review of large scale and pilot scale remediation technologies, recommended bio stimulation for cleaning polluted sediments, ex-situ trenching for groundwater treatment and bioremediation for contaminated soils in the Niger Delta (Nuhu et al., 2021). A study also studied physicochemical methods, thermal methods, and biological methods and recommended phytoremediation for its cost-effectiveness and applicability in the treatment of a wide variety of soil contaminants (Adesipo et al., 2020).

4. NANOREMEDIATION OF CRUDE OIL CONTAMINATED SOILS AND WATER

Nanotechnology is the manipulation of matter at near atomic dimensions

between 1- 100 nanometers (nanoparticles) (Khan et al., 2019). Nanoparticles can be classified depending on their properties, shapes or sizes for example fullerenes, metal nanoparticles, ceramic nanoparticles and polymeric nanoparticles. Their reactivity, toughness and other properties are dependent on their unique size, shape and structure (Khan et al., 2019).

Nanoparticles are more reactive than the bulk particles because of their high surface area to volume/mass ratio (Das et al., 2019; Jadoun et al., 2021; Khan et al., 2019; Yarima et al., 2020). They have a large surface-to-area ratio when compared to the same volume of larger particles. This facilitates their transportation through small pore spaces and their interaction with adjacent surfaces thus requiring a lower amount of the same material (Alsaba et al., 2020).

Nanoparticles are also believed to be cost effective, less time consuming, efficient, sensitive and selective (Yarima et al., 2020). Nanoremediation can potentially reduce the costs of cleanup for large-scale contaminated sites, reduce clean up time and minimize pollutant concentrations in situ (Nnaji, 2018). Nanotechnology has also been merged with bioremediation through the use of natural pickering emulsifiers (stabilized by nanoparticles) to induce bacterial proliferation on the droplets, coating the bacteria with magnetic nanoparticles for easy removal, and the use of nutrient loaded nanoparticles for targeted release onto oil-water interfaces (Pete et al., 2021b).

Iron nanoparticles have widely been employed for the remediation of organic and inorganic pollutants in polluted water, soil and sediments with various modifications made to overcome the original defects of aggregation or oxidation (Xu et al., 2022). Other nanoparticle types that have been utilized for removal of oil pollutants from contaminated soil include, Carbon nanotubes (CNTs) for removing PAHs and Bentonite clay combined with nZVI to remove PCBs (Vu and Mulligan, 2023). PAHs can also be removed using gold nanoparticles, iron hexacyanoferrate nanoparticles, ZrO₂ nanoparticles, nano Fe³⁺-montmorillonite, nano anatase TiO₂, ZnO nanoparticles, Ti/ZnO-Cr₂O₃ nanocomposite, Fe₃O₄ nanoparticles and TiO₂-graphene nanocomposites (Vu and Mulligan, 2023).

Suspensions of Fe-Cu nanoparticles and biosurfactants showed an oil remediation efficiency of 84% in a study by (Vu and Mulligan, 2022). This efficiency was achieved at rhamnolipid biosurfactant: nanoparticle ratio of 10:1 (wt%: wt%), pH 7, room temperature, and shaking speed of 60 rpm for 60 min. Zero-valent iron nanoparticles (nZVI) have also achieved fast, efficient and safe remediation of crude oil contaminated soils and water (Galdames et al., 2020). They form unique shell-core structures containing iron oxides and iron hydroxide that can effectively adsorb various organic and inorganic pollutants by electro- static interactions and surface complexation (Xu et al., 2022).

The nZVI react with pollutants and electron acceptors and then oxidize to produce secondary minerals and dissolved iron non-distinguishable from natural iron. The reactivity of nZVI is dependent on their surface area, stabilizers, soil pH, soil organic matter and soil type (Xue et al., 2018). However, due to the high surface area, nZVI particles tend to aggregate, leading to the decrease of reaction activity and mobility in soil. Despite the fact that coating nZVI with polyelectrolytes improves their colloidal stability and reduces adhesion to the soil, the number of active sites for contaminant removal is also reduced (Xue et al., 2018).

The remediation mechanism used by nZVI is adsorption and redox reactions that occur as illustrated in the equations 1, 2 and 3 (Galdames et al., 2020). The nZVI reacts with O₂ dissolved in water to produce H₂O₂ (Equation (1)) (Galdames et al., 2020). The hydrogen peroxide formed is either reduced to water by nZVI (Equation (2)) or reacts with Fe²⁺ (Fenton reaction) to produce hydroxyl radicals (Equation (3)). The hydroxyl radicals formed are strong oxidizing agents hence can degrade a considerable amount of organic contaminants (Galdames et al., 2020).



Nanoremediation of oil pollution can also occur through hydrogen reduction. In order to improve the reaction performance of iron nanoparticles, noble metals such as palladium, platinum, nickel, copper are added to form bimetallic iron nanoparticles. The noble metals exhibit catalytic properties by collecting hydrogen produced by iron corrosion. The hydrogen adsorbed on the nanoparticle surface then acts as a reducing agent in the remediation process (Xu et al., 2022).

5. GREEN SYNTHESIS OF NANOPARTICLES

The synthesis method, pH, temperature, pressure, time, particle size, pore size, environment, and proximity influence the quality and quantity of the synthesized nanoparticles, their characterization and applications. Physical, chemical, and biological synthesis routes are commonly used in the formation of nano particles (Sathya et al., 2018).

Physical and chemical synthesis routes tend to cause defective surface formation, have low production rate, high cost of manufacturing, large energy requirement, and use toxic reducing agents (Sathya et al., 2018). The use of green synthesis, microbes and biomarkers with nano materials are currently highly recommended (Nnaji, 2018). Green synthesized FeONPs showed less toxicity than chemically synthesized FeONPs in brine shrimp lethality assay in a study by (Junaid et al., 2021).

When plants and microbes containing polyphenols and proteins are used in place of chemical reagents under suitable conditions to reduce metal ions into lower valence state, this is referred to as green synthesis (Ying et al., 2022). The nanoparticles synthesized are diverse in nature, have greater stability and appropriate dimensions since they are synthesized using a one-step procedure. Undesirable processing conditions are usually eliminated by allowing the synthesis to proceed at physiological temperatures, pH, pressure and at negligible cost.

The size and shape of the nanoparticles synthesized by microbes is dependent on the parameters that influence microorganism growth, such as the substrate concentration, pH, and temperature (Xu et al., 2022). Microorganisms such as genus *Exiguu bacterium*, *E. acetyllicum*, *Actinomyces*, *Fusarium oxysporum*, *Verticillium sp.*, *Aspergillus flavus*, *Aspergillus fumigatus*, *Penicillium fellutanum*, *Pleurotus ostreatus* have been used to synthesize nanoparticles (Pirko et al., 2021).

Green synthesis by plant extracts is currently the most efficient method for short time, large scale production of nanoparticles. It involves synthesizing nanoparticles using roots, leaves, fruits, seeds, flowers, latex extracts through a single step bio-reduction which consumes less energy and is eco-friendly. The kinetics for plant assisted nanoparticle synthesis is higher than other biosynthetic approaches and equivalent to chemical nanoparticle formation (Jadoun et al., 2021).

Plant extracts contain active biomolecules such as phenolic acids, starch, sugars, terpenoids, phytosterols, alkaloids, polyphenol, proteins, enzymes, coenzymes and ferulic acid esters. These biomolecules are useful in the bio-reduction, functionalization and stabilization of nanoparticles (Ssekatawa et al., 2021). Metal ions are reduced to form small nucleation centers that grow in size by sequestering more metal ions and surrounding nucleation sites to form the nanoparticles. This process can take place rapidly and easily at room temperature and pressure in a single step reaction (Xu et al., 2022).

Some of the plant extracts explored for the nanoremediation of oil contamination include *Coriandrum sativum* leaf extract, garlic (*Allium sativum*) extract, *Butea monosperma* (Palash) leaves extract and green mango peel extracts (Das et al., 2019). A group of researchers also used mortiño berry fruit extract to synthesize zerovalent iron nanoparticles for remediation of crude oil contaminated water and soils (Murgueitio et al., 2018).

During green synthesis, the pH, temperature, and reaction time, can influence resultant nanoparticle characteristics. The pH has been reported to influence nanoparticles sizes and shapes. It has also been observed that different plants and different parts of the same plant, can have different pH values. Additionally, the yield of green nanoparticles depends on the reaction time, while the morphology depends on the concentration of precursor materials (Wang et al., 2019).

6. CHARACTERIZATION OF THE GREEN SYNTHESIZED NANOPARTICLES

The key parameters in the characterization of nanoparticles are; size, shape, surface charge, and porosity according to (Mourdikoudis et al., 2018). The size and shape influence functionalization capacity, fluid drag and diffusion, optical properties and uptake into cells. Surface charge controls stability, aggregation and the interaction between nanoparticles and the environment. Additionally, porous or hollow nanoparticles have a vast internal surface area, which can be further functionalized.

The techniques commonly used for characterizing nanoparticles used in crude oil remediation include UV-Visible Spectroscopy, Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), Fourier

Transfer Infrared spectroscopy and X Ray Diffraction microscopy (XRD) (Rajput et al., 2020; Reddy et al., 2018). UV- Spectroscopy is used to confirm the spectrum of nanoparticles and be used to determine the optimum synthesis condition such as pH, temperature and the metal salt concentration (Rajput et al., 2020).

The SEM and TEM scan are used to identify the elemental composition, size, shape and morphology surface of the nanoparticles. Fourier Transfer Infrared investigation is used to identify the organic functional groups such as carbonyl, carboxylic and hydroxyl present on the external surface of nanoparticles and these are compared with the functional groups present in plant extract. The XRD technique is used to reveal peaks associated to various compounds present in the nanoparticles (Alkurdy and Ebrahim, 2020).

7. FATE, TRANSPORT AND LIMITATIONS OF THE GREEN SYNTHESIZED NANOPARTICLES

Multimedia fate models such as MendNano, NanoDUFLOW, RedNano, nanoFate, WASP8 and SimpleBox4nano can be used to understand the behavior of nanoparticles when released into the environment (Meesters et al., 2019). A group researchers combined USEtox2.0 modelling framework with SimpleBox4Nano and successfully used them to carryout life cycle assessment (LCA) for nano-TiO₂ and assessed its impact on freshwater ecotoxicity (Salieri et al., 2018).

MendNano model assumes fixed (time independent) partitioning ratios for the processes of aggregation and attachment while simplebox4nano models mechanistically using first-order rate constants. SimpleBox4nano is used to predict concentrations of nanomaterials in air, water, sediment and soil. The model is operated as a Microsoft Excel spreadsheet supported by numerical computations in R. Mass flow rates are derived using physical and chemical properties as well as characteristics of the environment modelled (Blázquez et al., 2022).

A group of researcher developed RedNano as a simulation tool used to assess response of an environmental system to various release scenarios of engineered nanomaterials (ENMs) (Liu et al., 2016). The tool is based on life cycle assessment, multimedia compartmental modeling and mechanistic intermedia transport processes. The tool also investigates the impact of geographical and meteorological parameters on ENM distribution in the environment, compares impact of ENM production and potential releases on different regions, and estimation of source release rates based on monitored ENM concentrations.

Nanoparticles may undergo sedimentation, agglomeration, adsorption by the microorganisms, surface adhesion, dissolution and transformation when released into the environment. The rate of agglomeration, sedimentation and adsorption is influenced by the concentration of nanoparticles, surface area of interaction and surrounding medium (Samanta and Mandal, 2017).

The magnetic nature of the nanoparticles causes aggregation thus forming larger particles whose soil mobility and reactivity is low (Vu and Mulligan, 2023). Stabilizers such as sodium carboxymethyl cellulose (CMC), potato starch and water-soluble starch have previously been used to minimize aggregation, increase stability, and enhance transport of the nanoparticles (Ibrahim et al., 2020).

Heavy metal nanoparticles of lead, mercury and tin are reported to be so rigid and stable that their degradation is not easily achievable (Khan et al., 2019). Nanoparticles of silver, copper, and zinc have antimicrobial properties and thus their exposure to the environment poses a threat to beneficial soil microbial communities (bacteria, fungi, and archaea) (Rajput et al., 2021). Nanoparticles from gold, silver and Titanium have been reported to directly affect the progeny and reproduction of roundworms (nematodes) and water fleas by penetrating their cell walls and changing the conductivity of the cytoplasm (Samanta and Mandal, 2017).

The application of iron nanoparticles in environmental remediation has significant effect on the environmental pH value and pollutant morphology according to (Xu et al., 2022). The corrosion of iron nanoparticles affects surface and chemical properties of the environment. This in turn affects the interactions between contaminants and iron nanoparticles. Iron nanoparticles also regulate processes in organisms by changing the pH and pollutant speciation. However, coating with organic polymers is believed to reduce their toxicity (Xue et al., 2018).

The impact of the application of different concentrations of nZVI on soil microbial properties in a clay-loam and sandy-loam soil was assessed in a

study by (Gomez-Sagasti et al., 2018). Results indicated that sandy-loam soil was more vulnerable to the presence of nZVI than the clay-loam soil. The toxicity of nZVI was through disruption of cell membrane architecture and enhancement of membrane permeability when in direct contact with the microorganism. The nZVI also indirectly led to molecular and biochemical destruction triggered by oxidative stress. Additionally, nZVI were discovered to cause inorganic phosphate starvation (Wang et al., 2019).

8. CONCLUSION

This review outlines the sources and constituents of crude oil contamination in oil exploration and production areas, the remediation methods commonly employed and the dynamics associated with using nanoremediation. Some of the nano materials utilized in the remediation of hydrocarbons include iron nanoparticles, Fe-Cu nanoparticles, Carbon nanotubes (CNTs), Bentonite clay combined with nZVI, gold nanoparticles, iron hexacyanoferrate nanoparticles, ZnO nanoparticles, nano Fe³⁺-montmorillonite, nano anatase TiO₂, ZnO nanoparticles, Ti/ZnO-Cr₂O₃ nanocomposite, Fe₃O₄ nanoparticles and TiO₂-graphene nanocomposites.

Particular emphasis is put on the modalities required in the green synthesis of nanoparticles for oil pollution remediation as a environmentally friendly nanoremediation technique. Whereas green synthesis involves the use of either plant extracts or microbes for the reduction of metal salts into their nano size, plant extracts have proved to be more effective and their kinetics is equivalent to that of chemical synthesis.

The source of plant extract, concentration of plant extract, temperature, pH, pressure, concentration of precursor will affect the nanoparticle size, shape, morphology and surface charge which in turn affects the efficiency of nanoremediation. Some of the plant extracts previously explored for the green synthesis of nanoparticles for oil contamination remediation include *Coriandrum sativum* leaf extract, garlic (*Allium sativum*) extract, *Butea monosperma* (Palash) leaves extract and green mango peel extracts and mortiño berry fruit extract.

Even though it is well known that the phytochemicals present in plant extracts are responsible for the formation of nanoparticles, the specific bioreducing and stabilizing mediators are not well known. Additionally, the effect of synthesis parameters such as pH, temperature, type and quantity of phytochemicals on nanoparticle characteristics is not well known. The specific characteristics most effective for oil remediations are also not well researched.

The green synthesis of nanoparticles for remediation of crude oil contaminants has mostly been carried out at laboratory scale with limited knowledge on their fate, transport, energy balance, material balance and cost benefit analysis. This is imperative in the assessment of the practicability, effectiveness and possible toxicity associated with using green synthesized nanoparticles. Multimedia fate models can be used to carryout life cycle assessment as well as simulate the behaviour of various nanoparticles when introduced in the environment.

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