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REVIEW OF NANOMATERIALS FOR REMEDIATION OF PETROLEUM IMPACTED SOIL AND WATER

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ABSTRACT

This paper reviews nanoremediation technologies and potential nanomaterials for oil spill remediation in Nigeria. Oil spillage in the Niger-Delta area of Nigeria emanating from oil and gas operations is a topical issue currently and has resulted in serious environmental degradation leading to loss of human, material and economic resources. Oil polluted soil and water can be remediated with different techniques but the use of existing conventional remediation technologies has limited or partial effectiveness and this has given rise to the integration of these methods in oil spill clean-up. However, more innovative and eco-friendly technologies to enhance clean-up operations especially for major oil spills are also needed. Nanoremediation is one such technology and various nanomaterials can used for the clean-up of oil spill impacted soil and waterbodies due to the advantages it has over conventional technologies. Secondary sources of data (journal papers, textbooks, internet and library) were utilized for the study which analysed the extent of oil spill in the Nigerian environment, causes of oil spill and the current remediation technologies used for oil spill clean-up. The characteristics and potential applications of relevant nanoremediation technologies for oil spill impacted soil and water bodies are presented. These include: nanobioremediation, carbon-based nanomaterials; metal-based nanomaterials (nano-zerovalent iron (nZVI) and Fe_3O_4); nanocomposites (zeolites and magnetic carbon-metal nano-composites). The environmental implications and safeguards for the use of nanomaterials are also considered.

Keywords: Oil spill, clean-up, Niger Delta, nanoremediation

INTRODUCTION

Pollution occurs as a result of the inability of the environment to process and neutralize pollutants. Oil and gas pollution is one of the most serious forms of pollution and long-term damages to the environment has been reported globally as a result oil and gas operations (Jain *et al.*, 2011). Oil and gas industry (OGI) activities include exploration, drilling, production, transportation, processing and storage and incidences like tanker accidents, well blow out, sabotage and accidental rupture of pipelines lead to the discharge of crude and refined petroleum products into the environment (Atlas, 1981; Ebuehi *et al*., 2005). Oils spill is the accidental or intentional release of liquid petroleum hydrocarbons into the environment as a result of human activity (Guidi *et al*., 2015) and depending on the magnitude it can lead to severe consequences for humans, the environment and oil companies. The Deepwater Horizon oil rig exploded in the Gulf of Mexico in 2010 spilling about 5 million barrels of oil leading to serious environmental pollution which has cost British Petroleum, owners of the rig, over $8 billion for the clean-up and is expected to cost a total of $40 billion for complete clean-up. Environmental remediation is the removal or transformation of contaminants in environmental media (groundwater, surface water, sediments, soil and air) to less harmful substances.

Reduction of pollutant concentrations in environmental matrices is limited with conventional remediation technologies (Rickerby and Morrison 2007). An IUCN – NDP study of 2013 concluded that current remediation technologies employed for oil spill sites in the Niger Delta do not measure up to Nigerian or international standards nor lead to the rehabilitation of biodiversity. It is now generally recognized that nanotechnology and the use of nanomaterial is the future of environmental remediation efforts and the oil and gas sector which is responsible for a substantial amount of pollution incidents needs to fully key into this by funding research in the area of Nano remediation (Mahajan, 2014). Nano remediation is a branch of nanotechnology which aims at the use of nanomaterials for the removal or reduction of contaminants in environmental matrices. It is remediation with nanoscale materials (nanomaterials) and involves their application for the degradation or transformation of pollutants. Nanomaterials usually have different or improved properties compared to their macro-scale forms due to their higher surface area to volume ratio (Mansouri *et al.*, 2008). Nanoremediation has the potential to reduce the costs of clean-up of large-scale contaminated sites, reduce clean up time and minimize pollutant concentrations *in situ*. USEPA (2007) states that nanotechnology has stimulated the development of novel and cost effective technologies for pollution detection, monitoring and remediation. Nanoremediation offers faster transformation kinetics, better penetration of contaminated matrices, extension of the spectrum of degradable contaminants and avoidance of remediation intermediates (Elloit, 2016; NanoRem 1, 2016). However, the environmental implications of the use of chemically synthesized nanomaterials are not fully understood and this has sparked interest into environmentally friendly and sustainable nanomaterials namely, biologically synthesized nanomaterial (Cecchin *et al*., 2016).

THE NIGERIAN ENVIRONMENT AND OIL SPILLAGE

Oil spillage in the Nigerian environment, especially in the Niger Delta area, is a topical issue which has led to avoidable human, material and economic losses to governments at all levels, communities and oil/gas companies (Aroh *et al*., 2010). Many sources have reported extensive environmental pollution of water and land in the Niger Delta due to oil spillages with dire consequences for agriculture and water quality (Osuji *et al*., 2010). Oil spill can be categorized into (Egbe and Thompson, 2010):

a) Minor spill – This occurs when spilled oil volume is < 25 barrels in freshwater or < 250 barrels on land, offshore or coastal water provided it poses no threat to public health
b) Medium spill – This occurs when spilled oil volume $\leq$ 250 barrels in freshwaters or has a range 250 – 2500 barrels on offshore and coastal waters
c) Major spill – This occurs when the spilled oil volume is > 250 barrels in freshwaters or >2500 barrels on land, offshore or coastal waters.
d) Catastrophic spill – This occurs when there is uncontrolled discharge of oil and which a looming threat to environmental and public health

The Department of Petroleum Resources reported a total of 4,850 oil spill incidences in Nigeria between 2010 and 2016 resulting in the spillage of 150,875.42 barrels of oil (Table 1).

Table 1: Nigerian Oil Spill Statistics

Years	Number of Spills	Quantity Spilled (Barrels)
2010	537	17,658.10
2011	673	66,906.84
2012	844	17,526.37
2013	522	4,066.20
2014	1,087	10,302.16
2015	753	32,756.87
2016	434	1,658.98
Total		150,875.42

Source: DPR (2016)

The vast majority of oil spill incidences in Nigeria are due to sabotage (NOSDRA, 2018; Shell, 2018). In 2017 NOSDRA, reported 155 oil spill incidences with 145 of them attributed to sabotage while as at 13 May, NOSDRA has reported 29 oil spill incidences in 2018 with 27 attributed to sabotage. Shell has also reported that about 90% oil spill incidences in its operations are due to sabotage. Figure 1 shows the recorded oil spill sites in the Niger Delta in 2015.

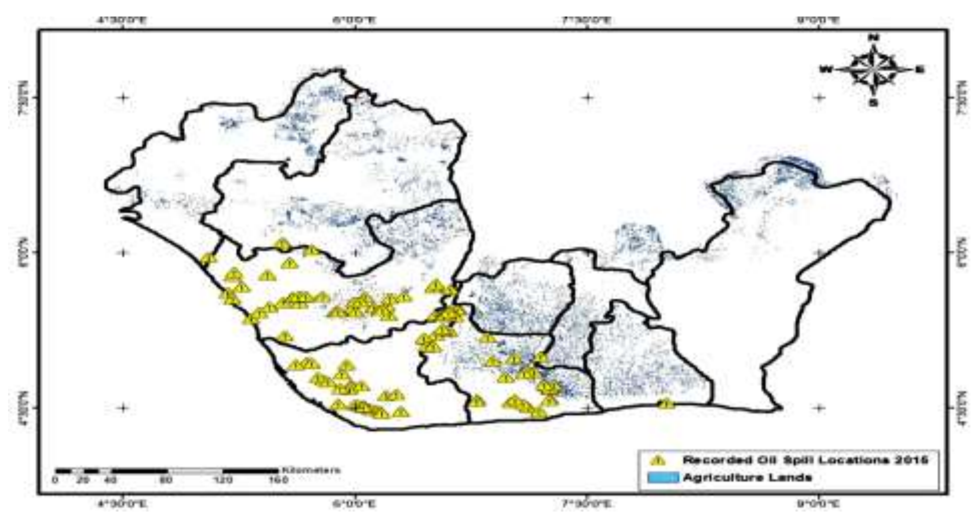

Fig. 1: Oil spill sites in the Niger Delta for 2015
(Source: NOSDRA Oil Spill Monitor Website)

Causes of oil spill

Oil spillages occur due to operational or man-made reasons. Operational oil spills occur as a result of:

a) Soil pore seepage.

b) Shifting of tectonic plates which may rupture or crack underwater pipelines beneath the ocean floor.

c) Formation of petroleum in reservoir rocks

Man-Made causes include:

a) Sabotage (oil bunkering)

b) Siphoning of oil

c) Terrorism

d) Oil facility accidents

STATUS OF REMEDIATION TECHNOLOGIES FOR OIL SPILL ENVIRONMENT

Crude oil is a complex mixture of organic substances including hydrocarbons which are mostly toxic to biota. Nigerian crude oils include Forcados Blend (FB), Bonny Light (BL) and Bonny Medium (BM) which are associated with high toxicity levels (Ezeonu *et al*., 2012). Generally, crude oil contains about 50-98% hydrocarbons while the rest are non-hydrocarbon compounds like sulphur, nitrogen, oxygen and heavy metals (Cerniglia, 1992). Global oil spill management has a market size that was worth \$131.16 billion in 2015. Major offshore oil spill sites are located in the Gulf of Alaska, Gulf of Mexico, North Sea, and the Persian Gulf while countries with major oil spill sites onshore include Nigeria, United States, Russia, Saudi Arabia and Angola (GVR, 2015). The conventional oil spill remediation technologies can be divided into:

(a) Physical/mechanical remediation technologies – Physical remediation is an effective method for short term or immediate response. The aim is to localize and remove as much oil as possible from the oil spill site using booming and skimming, pumps, mechanical separators, wiping with absorbent, washing using low-pressure cold water or hot water at high pressure, relocation of oil polluted materials and burning of oil contaminated sites. Other physical technologies include air-sparging, soil vapour extraction, soil washing etc.

Fig. 2: Use of Boom and Skimmers (Source: Barnes, 2015)

(b) Chemical remediation technologies – Chemical remediation in water bodies is usually done with dispersants. Dispersants are liquids containing surfactants which are dissolved or suspended in solvents and they function by breaking up large oil droplets into tiny droplets which either float or are submerged underwater and are in a form that can be easily degraded by micro-organisms. This method is used routinely in many countries, especially when faced with the constraints of physical remediation. Other methods include the use of demulsifiers to break the oil-water emulsion; solidifiers to promote the polymerization of the oil so that oil becomes stable, minimizing the spread thereby making remediation effective and surface film chemicals to prevent the attraction of oil to the substrate. On land, techniques like hydrogen peroxide oxidation and solidification/stabilization are used.

(c) Bioremediation technologies. The ability of microorganisms (bacteria, fungi, protists or enzyme) to convert pollutants into less toxic substances is Bioremediation (Van Dillewijn *et al.*, 2007). Bioremediation is divided into natural attenuation (the process occurring on its own) and stimulated bioremediation. The common bioremediation technologies are land farming, bioventing, bio-sorption, bioleaching, bio-augmentation, biostimulation and rhizoremediation (Li and Li, 2011; Giwa and Ibitoye, 2017). Rhizoremediation is a process where microorganisms degrade soil contaminants in the rhizosphere. Advantages of bioremediation include safety, cost-effectiveness and ease of administration. For instance, land farming is a simple, inexpensive technology for the remediation of sites polluted by petroleum. Mmom and Deekor (2010) observed that with soil pH at 5.5, 14.5 to 82.2 % degradation of total petroleum hydrocarbon (TPH) can be obtained with land farming in the Niger Delta. However, the effectiveness of bioremediation is hampered at sites which have high levels of pollutants such as hydrocarbons, volatile organic compounds (VOCs), heavy metals and salts that are hazardous for the bioremediating microorganisms (Rizwan *et al*., 2014).

(d) Phytoremediation technologies. This is the use of plants for the extraction and transformation of pollutants in soil and water (Schooner *et al*., 1995). It is regarded as one of the least expensive technologies of remediation for areas whose contamination is low to moderate. It can be used for the removal of contaminants like hydrocarbons, VOCs and heavy metals but it is best suited for areas with low to moderate contaminations due to the toxic effects of high pollutant concentrations on plants used for the treatment.

(e) Thermal remediation technologies. This involves the use of heat for cleaning up contaminated media. It includes thermal desorption, incineration, steam heat injection, thermal conduction etc. In thermal desorption, heat is used to evaporate chemicals (e.g. oil and hydrocarbon) which are consequently collected and destroyed in a treatment system. Thermal treatments have some drawbacks. For instance, incineration is

d)

usually done at high temperatures (750 –1200 °C) and so it is a costly process which may also produce unwanted by - products.

NANO-REMEDIATION TECHNOLOGIES FOR OIL SPILL ENVIRONMENT

Nanoremediation involves the use of reactive nanomaterials for the degradation and transformation of pollutants (Patil *et al*., 2016). Nanomaterials (NMs) are nano-scale materials which have components whose sizes, in terms of structure, are less than 1 µm (1000 nm) in at least one dimension. These include nanoparticles (NPs) that have particles whose structures are nanoscale in at least two dimensions (EPA, 2008; Luoma, 2008). NMs can be classified according to source into:

(a) Natural (b) Incidental and (c) Engineered

In terms of product material, they are classified as: (a) Carbon based (b) metal based (c) Dendrimers and (d) composites

These are developed in a variety of forms such as nanowires, nanotubes, films, particles, quantum dots and colloids (Edelstein and Cammaratra, 1998; Lubick and Betts, 2008). NMs have extraordinary properties such as:

a) Large surface area
b) Quantum effects
c) Electrochemical and magnetic properties
d) Highly active surface bonds and
e) Other size-dependent physical and chemical properties

These properties make nanomaterials more reactive and sensitive to environmental contaminants compared to conventional technologies or to their macro-scale forms (Keiner 2008) and enable their use in technologies like nanoremediation. Processes involved in nanoremediation include oxidation, reduction, sorption or combinations of these and nanoremediation technologies can be divided into:

a) Non-integrated
b) Integrated
c) Hybrid. This combines conventional and nanoremediation technologies. Nanoremediation can be combined with bioremediation resulting in a hybrid technique called nanobioremediation. Nanodispersants can also be used for higher efficiency and less impact on the environment.

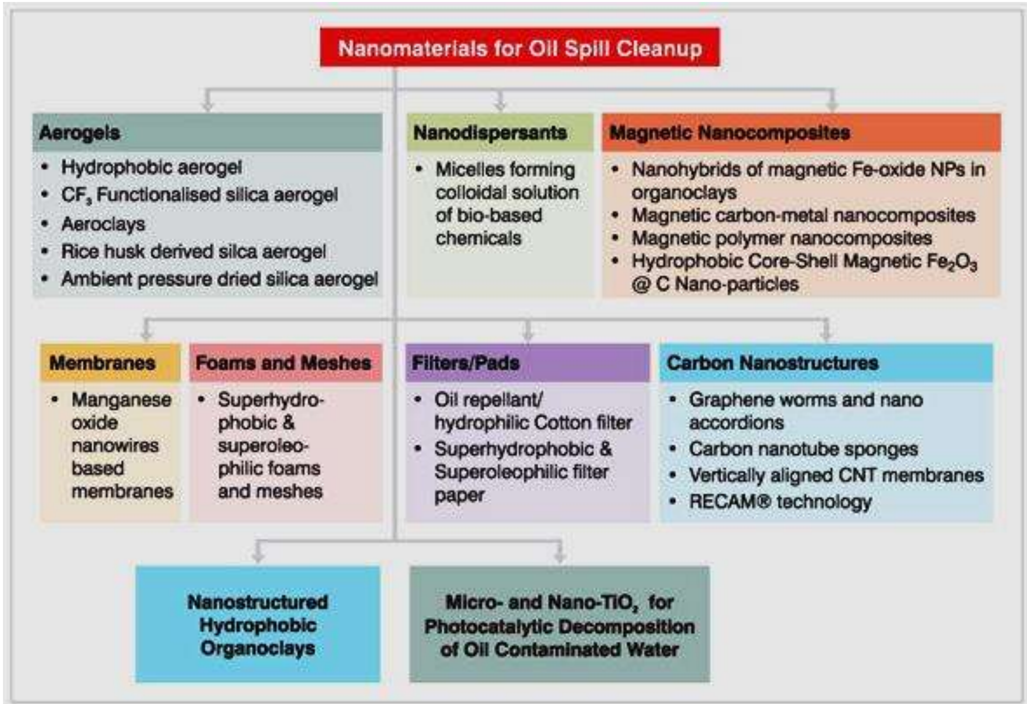

Fig. 3: Types of nanomaterials for oil spill remediation
(Source: Mahajan, 2011; Knapik and Stopa, 2014)

NANOMATERIALS AND OIL SPILL REMEDIATION

Surface Water. Oil spills is cleaned up by containment with barriers and then scooped with skimmers or it can be burned. It can also be solidified, absorbed with adsorbents, dispersed with dispersants or consumed by oil-grazing bacteria. These technologies have limited effectiveness which can be improved by incorporation of nanomaterials. Nanoremediation methods for oil spill impacted water include:

Nanobioremediation – This combines conventional bioremediation and nanoremediation. It has potential application in large scale clean-up at reduced cost and minimum generation of hazardous by - products. Nanoscale metallic particles like Zn, Cu and Fe nanoparticles are now being biosynthesized for remediation. This reduces the costs of the physically and chemically synthesized NPs.

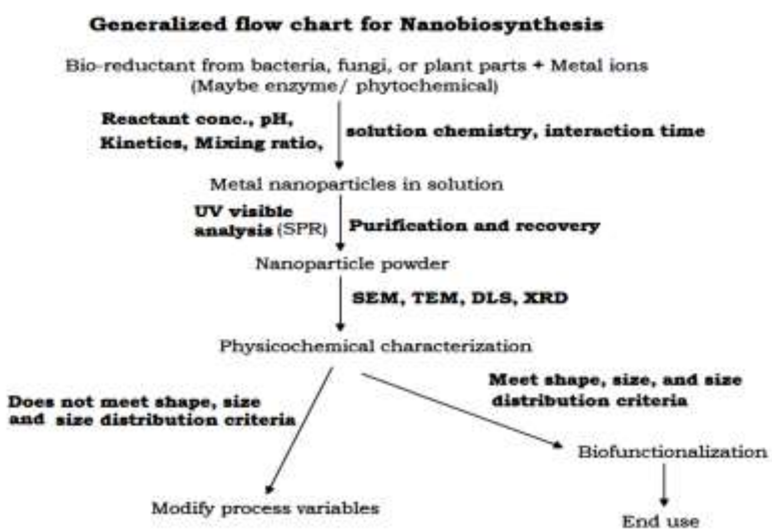

Fig. 4: Flowchart outlining the biosynthesis of nanoparticle
(Source: Yadav *et al*., 2017).

Figure **5.** Studies on the absorption of oil and recycling of CNF aerogels. (a) Sequence for diesel oil absorption by CNF aerogel (Sudan III dye applied) on water surface. (b) Bar chart showing the sorption capacities of CNF aerogels for different organic liquids (c) Scheme for recycling CNF aerogel using heat treatment (d) The recycling capacity of CNF aerogel over ten cycles. (Source: Wy *et al*., 2014).

Nanodispersants. Water based biodegradable nano-dispersants are used to achieve nanoscale encapsulation and emulsification of oil and enhancement of biodegradation. Dispersants are liquids in which surfactants are dissolved or suspended in solvents and they function by breaking up floating oil slicks into tiny droplets which either float or are submerged underwater and are in a form that can be easily degraded by micro-organisms. There are plant derived nanodispersants which breakdown hydrocarbon bonds in oils and suspends them as a nano-colloidal suspension which becomes water soluble. Industry examples of nanodispersants include Gold Crew® oil spill dispersant (OSD) and G-CLEAN OSC 1809.

4.1.2 Groundwater. Nano zero valent iron (nZVI) is used for the removal of chlorinated hydrocarbons (CHCs) like trichloroethylene (TCE) while nano-scale iron oxide is used for the removal of BTEX (benzene, toluene, ethylbenzene and xylenes) from groundwater. 85-95% CHCs reduction has been achieved in Czech Republic (NanoRem, 2017)

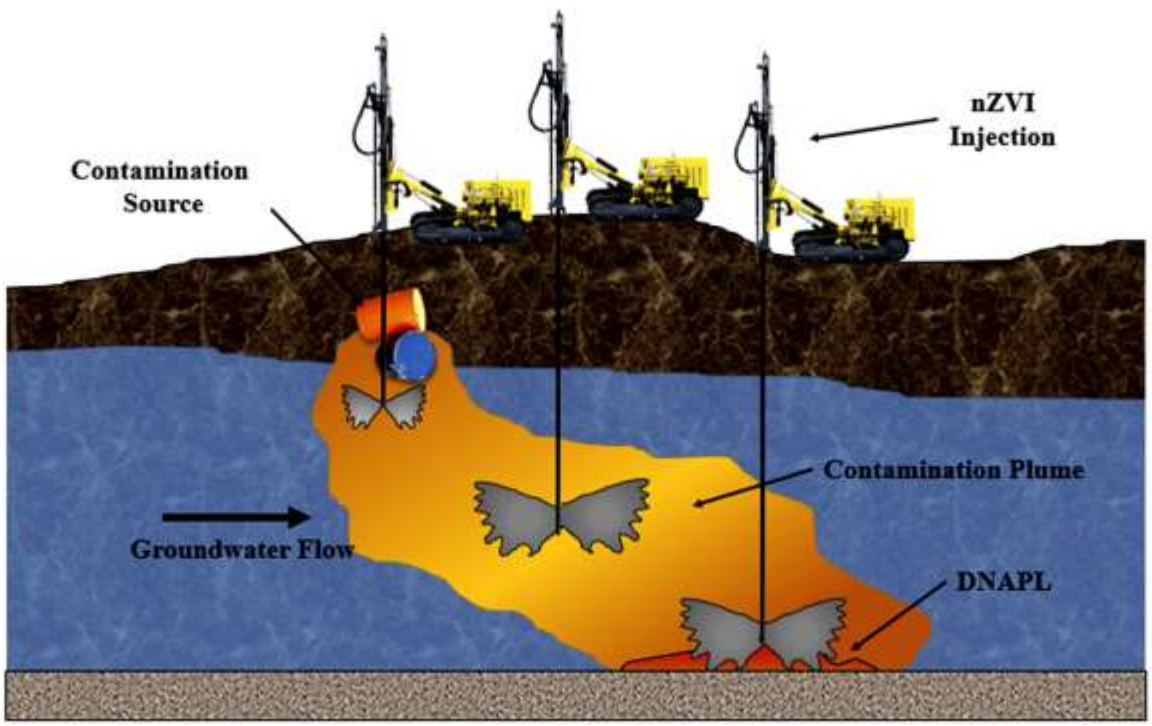

Fig. 5: nZVI injection process for remediation of contaminated Groundwater
(Source: Cecchin *et al*., 2016)

Soil.
Nanomaterials with high potential applications for oil spill impacted soil include zeolites, nZVI, iron oxides nanoparticles and carbon nanotubes. Natural and synthesized zeolites immobilize heavy metals and radioactive nuclides in contaminated soils and sediments. Significant metal reductions of 42% to 72% have been achieved using zeolites. Karthick and Chattopadhyay (2017) obtained a diesel oil removal efficiency of 78% using a non-ionic surfactant foam stabilized with 0.5 wt% hydrophobic silica nanoparticles. Amphiphilic polyurethane nanoparticles have also been used to remove polynuclear aromatic hydrocarbons (PAHs) from the soil. Non-toxic nano-scale surface washing agents are also used to remove oil from solid surfaces like soils, plants and animals not by dispersion or emulsification but via a detergency mechanism.

Table 2: Comparison of Four *In-situ* Remediation Methods: nZVI, Conventional PRB, Thermal Treatment and Chemical Oxidation

	nZVI	**PRB**	**Thermal**	**Chemical oxidation**
Material	nanoscale iron	granular iron	- (energy)	Oxidants e.g. Permanganate, H_2O_2
Material reactivity	High	Low	–	Very high
Material longevity	Months	Years	–	months
Installation costs	moderate	expensive	Moderate	inexpensive
Operation and Maintenance costs	moderate	inexpensive	Expensive	moderate
Requirements regarding site access	Possible for appl. underneath buildings	Open access for machinery necessary	Possible for appl. underneath buildings	Possible for appl. underneath buildings
Major drawbacks	a. Material handling b. Material cost c. Rebound effects (reinjection needed) d. Uncertainties of environmental impact of nanoparticles	a. Plume has to flow through barrier b. Not economical when contaminant at greater depth c. Uncertainty about reactive lifetime of the barrier	a. Possible volatilization of contaminants b. Undefined mobilization and migration of contaminant c. Low efficiency d. Residual contamination	a. Toxicity of substances b. Rebound effects (reinjection needed) c. Low efficiency (about 1%)
Environmental effects	a. possible increase in pH b. dissolved Fe toxicity c. ORP decreases	a. possible increase in pH b. Dissolved Fe toxicity)	a. increase of temperature influences microbiology	a. oxidation of reductive environment → (temporal) chance in microbiology

Source: Mueller and Nowack (2010)

Advantages of nanoremediation

a) Nanomaterials remain active in the soil for many years
b) Nanoremediation can potentially reduce both the time and cost of remediation
c) It can be used in-situ thereby eliminates the need for disposal of polluted soil or water

Environmental implications of the use of Nanomaterials

The use of nanomaterials may lead to emerging toxicological risks due to their higher reactivity and penetration of contaminated matrices and they may negatively impact human health through inhalation, ingestion or dermal absorption. Also, scientific evidence shows that the nanoparticles may travel through the food chain and may also harm important environmental microorganisms. For instance, soil bacteria are of extreme importance for the nutrient cycle in the soil and have been associated with natural degradation of organic contaminants and immobilization of heavy metals (Bokare *et al*., 2012). Fe nanoparticles can exert bactericidal effects leading to reduction of soil bacteria. Some researchers regard nanoparticles as complex and problematic for large-scale use and are worried that accidental nanoparticle releases could damage aquatic life. However, there is no conclusive evidence that this is the case and research is on to replicate in environmental remediation, the success achieved with nanotechnology in other areas like medicine and electronics.

STATUS OF NANO-REMEDIATION IN NIGERIA

Nigeria can be classified as a Nanotechnology dormant nation since it is still largely at the demonstration of interest stage while smaller and less rich countries like Nepal, Bangladesh and Sri Lanka are seriously pursuing research in Nanotechnology (Ejeta *et al*., 2017; Ezema *et al*, 2014). Nigerian government interest in advancing nanotechnology is based on the 2012 Science, Technology and Innovation (ST&I) Policy of the Federal Ministry of Science and Technology which addresses new and emerging technologies like nanotechnology in item 12. However, significant research has not been done in nanotechnology and only a few institutions like the National Agency for Science and Engineering Infrastructure (NASENI), University of Ibadan, University of Nigeria and Obafemi Awolowo University, Ile-Ife are known to engage in appreciable nanotechnology research (Batta *et al*., 2014; Okwuosa, 2014). In the area of nanoremediation, research is few and far between and little information exists on the research and use of nanoremediation by public and private organizations including the Oil and Gas companies. There is a dire need for research funding on nanoremediation techniques if Nigeria is to catch up with global trends. Nigeria and by extension, the Nigerian oil and gas industry (NOGI) should become technology innovators and not merely technology consumers. This will surely improve the technological and socio-economic status of the country and industry.

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