



Prospects of nanosorption and photocatalysis in remediation of oil spills

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Abstract

Nanoremediation approaches have been applied to remove oil from surface and ground water as oil spills have been found to have long-term negative consequences for the ecosystem. Nanoremediation via the nanosorption mechanism of different environmental matrices in the world at large is at its formative stages despite the alarming and extensive prevalence of petroleum related environmental pollution. Over 9 million barrels of oil have been leaked in the last five decades, making that ecosystem one of the most deteriorated by oil exploration and extraction activities. The goal of this research is to assess the current status, trends, and future prospects of the nanosorption of surface and ground water in oil spill regions. High surface area of nanomaterials, wide spectrum of treatable contaminants, non-generation of intermediate or secondary products, as well as speed and extent of contaminant destruction give nanoremediation a superior comparative edge over other treatment technologies. Notably, the remediation efficiency of a cleanup is highly dependent on the type of material and treatment routes employed. It is imperative to employ a concerted and practical approach to the development of nanotechnology to combat the bedeviling oil pollution challenges faced in oil producing counties.

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1. Introduction

Oil spills are the major cause of oil pollution and are of the great concern in today's world, especially in the marine environment [1]. Activities such as surface runoff, vessel and pipeline accidents, offshore petroleum exploration and pro-

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duction operations, shipping and illegal bilge water discharges result in marine oil pollution [2]. Research has also shown that for every barrel of oil transported across the world via waterways, the environment may be at risk for spills [3]. In Nigeria, more than 90% of the dilemma caused by oil spills is evident in the Niger Delta communities, an occurrence that has caused significant environmental and socioeconomic degradation in this part of the federation. Since oil exploration and exploitation began, the Niger Delta has seen 1.5 million tons of crude oil spilled with partial or no clean-up carried out as depicted in Figure 1 [4].



Figure 1: Surface and groundwater polluted by crude oil spills sited in Niger Delta [4]

Typically, when an oil spill occurs, several processes that include weathering, evaporation, oxidation, biodegradation and emulsification follow (as shown in Figure 2) [5]. These processes alter the properties of the oil and cause significant changes in viscosity, density and interfacial tension [6]. As oil begins to reach the surface, it is subject to a variety of natural processes. Spreading, evaporation, and photo-oxidation of the oil on the water's surface are examples of these processes. Below the surface, emulsification and dispersion occur, which can aid in the microbial decomposition of the oil. Oil can also settle on the seabed or fall as marine snow [5].

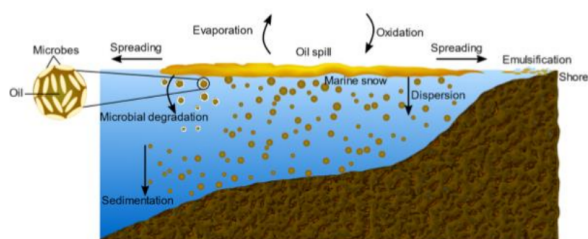


Figure 2: Processes that follow an oil spill [5].

The occurrence of oil spills, despite dating back to many years ago, has seen clean-up techniques remain constant over time. The most common of them employed as a response mechanism include mechanical methods (using booms and

skimmers), thermal methods (burning), chemical methods (dispersion) and natural methods (bioremediation) [7]. Each technique is not without its pros and cons, and its usefulness depends on external factors like viscosity, volume, and composition of the oil, as well as weather conditions and the surrounding environment [8].

1.1. Common Oil Remediation Techniques

The sustainability of the current techniques used for oil spill remediation has been widely critiqued by many authors. The use of chemicals such as dispersants, emulsifiers and oil absorbers, among various other expensive techniques, as treatments for oil spread in marine bodies without recovery of the oil is not a cost-effective method (Atta et al., 2019). Islam et al., (2017) reported an estimate of approximately \$2730 per barrel as the cost of an oil spill clean-up. In addition, a hundred percent cleanup of oil introduced into marine environments may be unattainable, an outcome that further generates a debate on the efficacy of these techniques.

Mechanical methods, otherwise known as physical methods, typically involve the introduction of containment or barriers to prevent the spread of oil spills. A common example of this method is the use of booms which are floating barriers composed of different materials. Although booms are expensive and require utmost care in handling when placing the net below the surface of the contaminated water, they are good for quick oil recovery and are effective in containing different viscosities and thicknesses of oil [5]. They usually exist in three different types – a fence-like structure (fence boom), a non-absorbent structure (curtain boom) or specially designed booms for controlled burning (fire booms) [11]. Other mechanical methods employ the use of skimmers in combination with booms for the prevention of further spread and the recovery of oil from water surfaces [5]. The use of sorbents is also of increasing interest as they help to remove the final traces of oil, or oil closer to shores [7]. They are either adsorbent or absorbent materials which function to bind the oil in a solid or semisolid matrix [5, 12, 13]. They may exist as natural organics like hair, feathers, cotton, straw, kapok, sawdust and vegetable fibers [14], natural inorganics like clay, sand, wool, volcanic ash and glass, and synthetic materials like polypropylene, polyester foam and polystyrene which are the most widely used sorbents [7, 9].

Unlike physical methods, chemical methods change both the chemical and physical properties of the oil. One method involves the use of dispersants/emulsifiers (surfactants) which are surface-active agents that decompose the oil slick into smaller emulsion droplets by reducing the oil-water interfacial tension, thereby making it undergo rapid dilution and easy degradation by microbes [7, 8]. Rather than remove oil, they generally dissolve oil into water [5]. Solidifiers, however, are hydrophobic materials (in the form of dry granules) that form a bond with the liquid oil and convert it to a viscous mass suitable to be removed by physical means. Usually, they

are adapted for relatively small oil spills that allow the boom technology to be applied for recovery [8].

While in-situ burning is a rapid means of thermal remediation requiring simple, non-specialized equipment – usually fire-resistant booms and an igniter [7], bioremediation is the natural process of oil degradation in water and/or soil by autochthonous microorganisms like algae, fungi and bacteria [5]. Bioremediation may be affected by mechanical and chemical remediation methods in that the former involves the aggregation of the oil which can prevent the survival of many microorganisms as a result of a decrease in nutrients such as sunlight and oxygen, and the latter increases toxicity thereby posing difficulty in oil degradation by microbes [5].

Each remediation technique is not all-encompassing, hence, it becomes imperative to start accessing measures that are focused on adopting the most effective oil spill recovery procedures. It is also necessary that the ideal technique is of significant efficiency, productivity, recovery and treatment time, cost, marine life impact, trouble level, the capacity of oil recovery, reliability upon climate conditions, impact on oil properties and level of additional treatment [7]. This further opens up the subject and the most recent attempt at oil spill remediation is in the use of the nanosorption technique as a remedial approach.

The last decade has seen the methods for environmental remediation significantly revamped by the field of nanotechnology as nanoparticles have been found to have diverse merits like large surface area, better and more active adsorption sites, high reactivity, and small size [15, 16, 17]. From the perspective of oil spill remediation, nanomaterials are classified into two categories namely: organic and inorganic, which are further classified into sub-categories based on the origin and stimuli response of their precursors (Figure 3) [5].



Figure 3: Classification of nanomaterials [5]

Advances in nanoscience potentially offer a sustainable solution for enhancing post-oil spill response technology by using nanosized materials for specific purposes such as the formation of Pickering emulsions [19, 20] or magnetically responsive sorbents [21]. Engineered nanoparticles readily adsorb onto the oil-water interface and allow the formation of oil-in-water Pickering emulsions (Figure 4a) while the magnetically responsive inorganic materials are removed with an external magnetic field (Figure 4b).

A Pickering emulsion is an emulsion that is stabilized by solid particles (like nanoparticles or microorganisms) which are adsorbed onto the oil-water interface and prevent coalescence of the oil droplets [5, 22]. Coalescence, otherwise known as

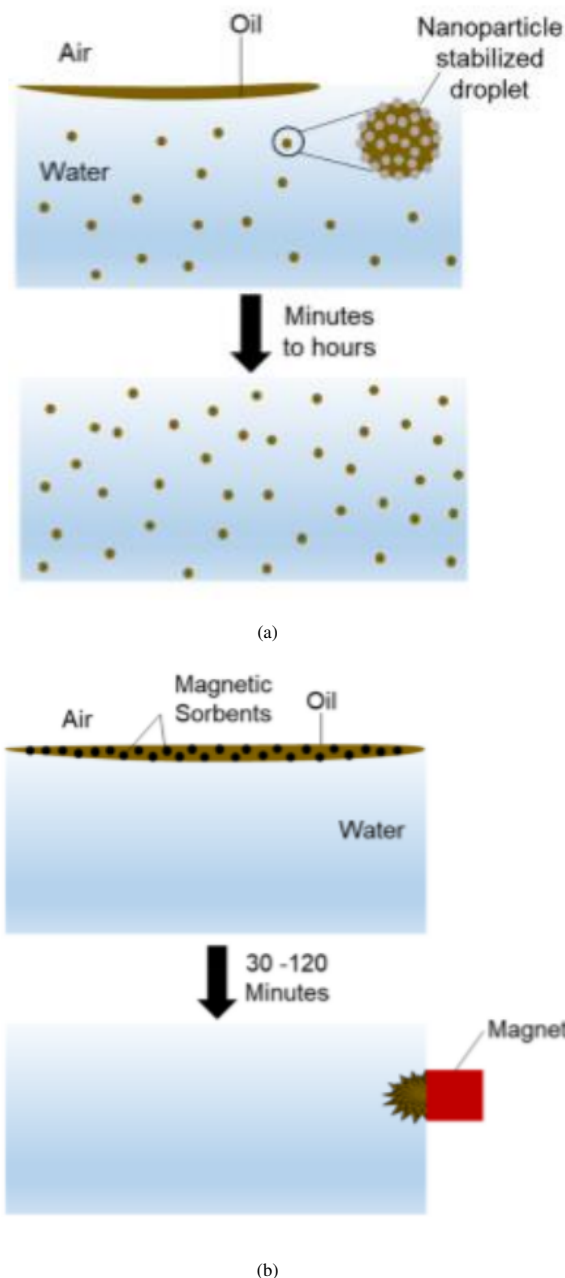


Figure 4: (a) Formation of Pickering Emulsions (b) Removal of inorganic sorbents with magnets [5]

aggregation, is prevented, and stable emulsions are obtained when a monolayer is formed by the irreversible adsorption of particles onto the oil-water interface [23]. Brekke & Solberg (2005) in their studies record the stability of the Pickering oil-in-water emulsion to be primarily dependent upon particle wettability, but stability also depends on many other factors including particle size, shape, surface area, concentration, and interfacial coverage [1, 25].

The efficacy of magnetic sorbent depends on factors such as sorption rate, oil retention capacity, and ease of application and removal. Sorbents are generally known to have low oil

retention and are non-selective, hence they soak up both oil and water [7]. The use of advanced nanomaterials helps in overcoming this inadequacy by developing materials with high selectivity in oil uptake (Figure 4b). Since nanoparticles have a larger surface area to volume ratio, their sorption capacity and oil retention become increased. Nanoparticle-based sorbents can be engineered for their hydrophobicity and selective uptake of oil. The selectivity prevents the sorption of water and hence, the sorbents stay afloat [25].

In this paper, the current trend, status and prospects of nanosorbents for the treatment of surface and groundwater in oil spill areas will be assessed.

2. Nanomaterials for Environmental Remediation

Many definitions of nanomaterials have been publicized in the academic community in recent years but the widely accepted version defines them as materials with a dimension that is less than 100 nm in diameter [26, 27, 28]. Such materials due to their sizes possess distinctive and novel properties lacking in their larger counterparts and find diverse applications in water, effluent treatment, electronics, cosmetics, construction and even in aviation [19] because they are the foundation stone of nanotechnology. Similarly, high surface area, reactivity, rapid dissolution, and strong sorption abilities are some unique properties currently being exploited in many applications as afore listed [30]. Several biological, physical and chemical processes [31, 32] are reported to have and still employ specific nanomaterials for remediation as shown in Table 1 purposes as subsequently highlighted. Nanophotocatalysts, nanomotors, nanomembranes, and nanosorbents, on the other hand, have seen more advances. For the manufacture of nanomaterials, a variety of processing procedures can be used, although it is highly reliant on the precursors used. Solution casting, compression molding, twin-screw extrusion, tape casting, thermal molding and melt blending are regular methods used for the development of biogenic nanocomposites due to their simplicity, flexibility and tunability. Doped metal oxides, doped carbon nanotubes (single and double-walled), nanosorbents, and other nanomaterials are among them. Other nanofabricated materials are processed via physical techniques such as mechanical stirring and melt blending.

Bionanocomposites are groups of nanomaterials gathering attention scientifically due to their eco-friendly nature, mechanical [33], and thermal stability as well as adsorptive properties [34, 35]. However, just like every nanomaterial, their wide application range is hinged on the individual components that make up the eventual product [36]. Because two-thirds of the oil in place is left behind after primary and secondary recovery, and because of the significant increase in oil recovery observed using enhanced oil recovery techniques (EOR), which include chemical injection, thermal recovery, and gas injection, a number of studies have been conducted to improve the various EOR techniques by adding nanomaterials [37]. The properties of a material at the nanoscale differ from the properties

of individual molecules. The surface area and surface energy grow dramatically as the size is reduced. Zinc oxide nanoparticles, phyto-genic magnetic nanoparticles, biopolymer-coated metal nanoparticles, silver-impregnated cyclodextrin nanocomposites, silver nanoparticles in Aloe vera plant extract, and Photocatalytic titania nanoparticles are all examples of biosynthesized nanomaterials [38] [39]. For the procedures outlined in Figure 5, a variety of various materials, such as inorganic, carbon-based, and polymer-based materials, can be employed. The cost of nanomaterials cannot be overstated; a single milligram of gold nanoparticles costs around \$80 at the moment (depending on the size of the nanoparticles). Gold nanoparticles are valued at around \$80,000 per gram, while a gram of pure, raw gold costs only \$50. The cost of treating 1 m³ of textile effluent with Nanoscale zero-valent iron (nZVI) particles was calculated to be \$1.66 USD. The cost of silver nanoparticles is Rs 1 per gram. The pricing of Powder Iron Oxide Nanoparticles for Industrial is Rs 22,000 per kilogram.

The source, composition of oil and nature of polluted water bodies are important features that affect cleaning of an oil spill and recovery. Because of the uniqueness and characteristics of nanoparticles, they have become a preferred choice for remediation. High surface area to volume ratio, reactivity, dispersibility, in-situ treatment capability, and adsorption affinity are just a few of the qualities that set them apart from traditional adsorbents used to clean up oil-contaminated media [42, 43, 44, 45]. Silica nanoparticles coated with polydimethylsiloxane (PDMS) have been used to get gelatin oils out of an oil/water mixture after increasing its surface hydrophobicity and PDMS coating thereby facilitating the separation of the mixture. Similarly, Chen et al., (2013), used polystyrene coated iron oxide nanoparticles for successfully catching floating oil on water and recommending its use in the remediation of oil spills [46]. In a different study, the functionalization of a nanoparticle was reported to significantly alter affinity as reported by Franco [47] where alumina functionalized by petroleum vacuum residue was employed for the treatment of oil contaminated fresh water and the enrichment of the nanosorbent significantly increased the affinity of the functionalized alumina towards non-polar compounds during the adsorption process [47].

Through several chemical modifications methods, properties of nanomaterials undergo remarkable augmentation thereby creating a plethora of application windows. Each modification method imparts distinctive surface features on the starting materials and biomass for instance is systematically enriched via esterification [48], etherification [49]), oxidation [50], carboxymethylation [51], hydroxypropylation and hydroxyethylation [52, 53, 54, 55].

Asphaltenes, an undesirable constituent of heavy oil was reportedly removed via adsorption and oxidation using nanoparticles of NiO, Co₃O₄, and Fe₃O₄ [56, 57, 58]. They opined that a positive correlation exists between adsorption affinity and catalytic activity of the nanoparticles towards

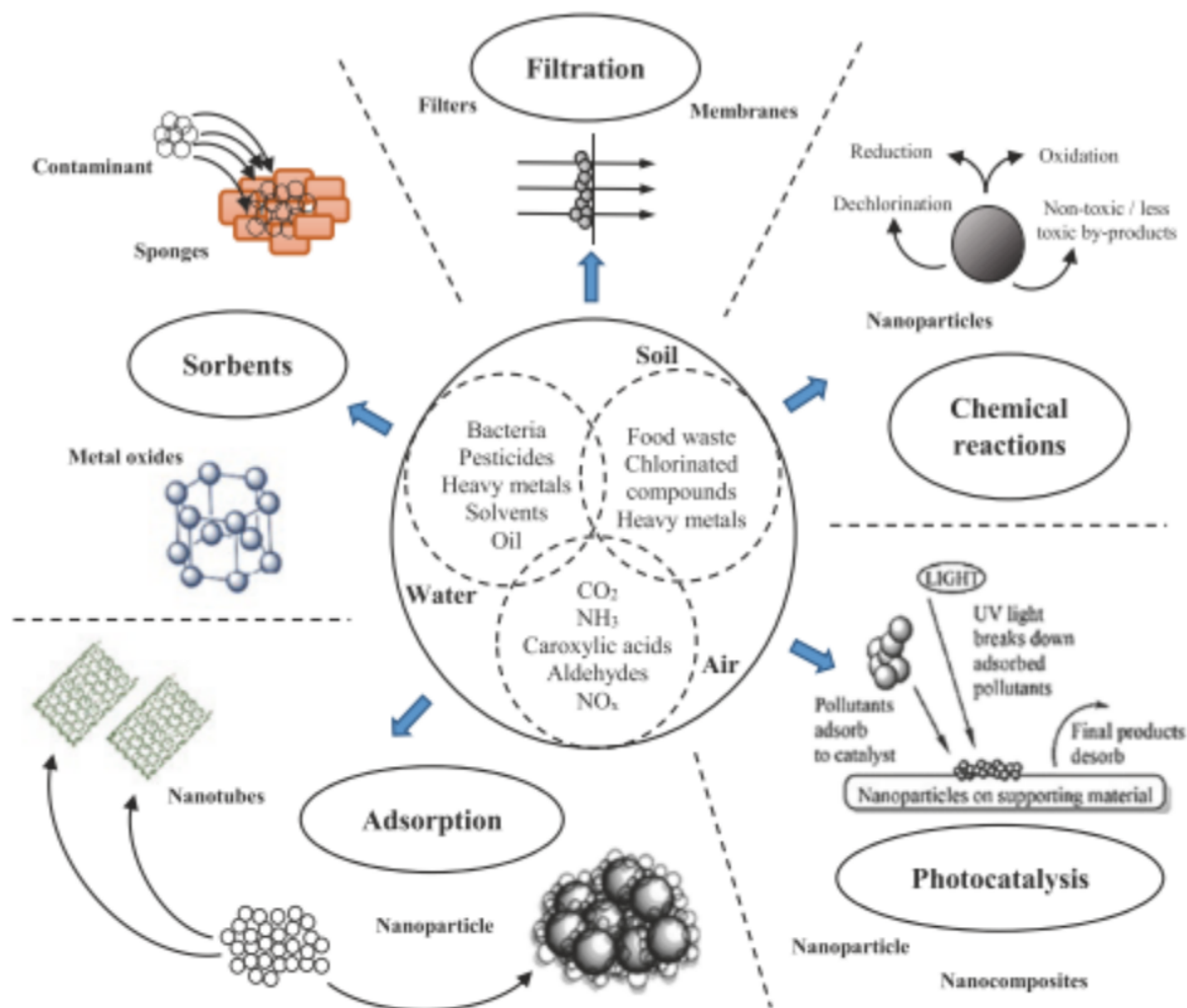


Figure 5: Environmental remediation approaches [40][41]

the immobilization and oxidation of asphaltenes. This was indicative of the good prospect in the application of these nanoparticles in the removal of selected waste hydrocarbons from a mixture of useful compounds [58]. Atta et al. (2015) created hydrophobic magnetite with significant magnetic characteristics that was used as an oil collector in oil spills, both coated and uncoated with amidoxine. Due to its limited hydrophobicity, uncoated magnetite had poor oil spill collecting effectiveness, whereas coated magnetite had remarkable collection capabilities [59]. Silver nanoparticles work as a good antimicrobial agent for the disinfection of water which is fixed to filter materials through in situ reduction of silver nitrate [39]. Ag NPs are placed on the cellulose fibres of an absorbent blotting paper sheet. Filtration is made over paper placed with Ag NPs which serves as a good prospect in the remediation of oil spill water treatment.

Franco et al. (2014) [47] used aluminum nanoparticles functionalized with petroleum vacuum residual (VR) and hydrophobic aluminum nanoparticles to try to lower the amount

of oil in an oil/saltwater emulsion at various pH levels. The results indicated that an increase in the amount of VR on the surface of the alumina led to an improved adsorption capacity but it was highest at neutral pH while the adsorption process was likewise favorable at acidic pH. Similarly, the kinetic adsorption equilibrium of different oils onto silica aerogel from oil water was studied and confirmation that viscous flow through the nanogel pores due to capillary forces was a factor that affects the sorption capacity of the oil onto the nanogel [60, 61]. The adsorption efficiency and amount of surfactant used for the emulsion stabilization have an inverse correlation with each other and that resulted in an adsorption capacity of about 17 mg/L for the three different oils [61]. The adsorption of gasoline and diesel from water using commercial hydrophobic nanosilica, on the other hand, was reported to have removal effectiveness of nearly 100%. The poor removal efficiency was shown to be caused by low pH, and the kinetic data was effectively explained by a first-order model (Aziz et al., 2020). Franco et al., (2014) [47] equally evaluated the adsorption mechanism of oil from oily water and brine studied

at different pH while using silica and silica functionalized with petroleum vacuum residues (VRs) as sorbent. The time to reach equilibrium was higher for freshwater than brine and the functionalized silica was suggested to be a good candidate for remediation of produced water in oil fields. Despite the fact. Although NPs might improve some parameters, they could also negatively affect some other parameters.

2.1. Carbon nanotubes

Carbon nanotubes (CNTs) are allotropes of carbon having cylindrical dimensions. They are used in a variety of fields, including electronics, medicine, and aviation, as well as water treatment, due to their unique characteristics [69]. Many techniques are currently employed for the synthesis of carbon nanotubes as shown in Figure 6 (a, b). Chemical vapors deposition is most commonly used over the years for their synthesis but laser ablation, arc discharge and chemical are the older methods used for the production of carbon nanotubes. Some of these technologies are inefficient because of excessive power consumption, while others produce low yields with carbon impurities, which are often a combination of single and multiwalled carbon nanotubes (SWCNTs) [70]. Carbon nanotubes are divided into three types based on their morphologies: SWCNTs, MWCNTs, and functionalized carbon nanotubes (FCNTs). Furthermore, the adsorption efficacy of functionalized CNTs is dependent on the kind and dose of adsorbents, the type and starting concentration of adsorbates, and the pH and temperature of the crude oil. Surface and groundwater polluted by crude oil spills can be found all over the world as a result of vandalism, sabotage, and equipment failures, among other things, which resulted in the release of various organic compounds into water bodies. Carbon nanotube sponges were synthesized by Gui et al., (2011) [71] and used for adsorption of oil and organics with viscosities above 200 cP. The material's reusability was recorded to be more than 10 cycles and the absorbed oil could be recovered by a simple physical means. In the same vein, they produced magnetic CNT with high reusability, selectivity, and structural strength but with more than 1000 reutilization capacity after adsorption of the oil [70]. In similar research, exfoliated vermiculite/carbon nanotube hybrid was used for diesel oil recovery with an adsorption capacity close to 30 g/g, quite higher than that of undoped exfoliated vermiculite [72].

Adsorption of free oil from oil spills has been studied extensively, with clear oil and water separation achieved [71, 72]. However, little study has been done on separating emulsified oil from water, motivating an attempt to successfully separate emulsified gasoline oil droplets from water using both doped and undoped CNTs with various Fe₂O₃ nanoparticle loadings. In comparison to other adsorbents or systems that adsorb the oil, the adsorption procedure was found to remove both mixed and free oil from water [72].

2.2. Graphene

It is the basic structural element of graphite, charcoal, carbon nanotubes and fullerenes. Currently, graphene is now the thinnest and strongest substance ever measured in the universe, and it has some highly unique features that have led to its use in a variety of industries [74, 75, 76]. The charge carriers of graphene exhibit peculiar properties such as intrinsic mobility, null effective mass and the ability to cover a small distance at ambient temperature without breaking away [74]. Similarly, the atoms near the edges of a graphene sheet have a unique chemical reactivity, with the largest ratio of edge atoms of an allotrope. Exfoliation, chemical deposition, sonication, electrochemical synthesis, hydrothermal self-assembly, and microwave-aided oxidation are examples of graphene synthesis processes (Liu & Kumar, 2014) [70].

Several studies have attempted to apply graphene to the removal of organic pollutants from water surfaces, with varying degrees of success [77, 78, 79]. Dong et al., (2012) [80] capitalized on the superoleophilicity and hydrophobicity of hybrid foam of graphene and carbon nanotube to selectively remove oils and organic solvent from the surface of the water. The monolithic 3D hybrid of graphene and nanotube with excellent adsorption capacity and good recyclability was synthesized using two-step chemical vapor deposition (CVD) procedures (Dong et al., 2012). The method of synthesis is important because it affects the adsorption capacity, however, attenuation of hydrophobicity, poor porosity and non-uniformity of the graphene's surface were attributed to the technique employed for its production (Niu et al., 2012). The sequel to the aforementioned, the adsorption efficiency of a 3D foam of chemically derived graphene (CDG), was studied and a moderate performance in terms of adsorption capacity due to alteration in intrinsic properties and surface morphologies was reported [81].

In contrast, chemical vapor deposition was used to synthesize a 3D monolith of graphene and the resultant material was uniform, extremely thin, macroporous with improved hydrophobicity due to pockets of air trapped underneath the pores of the graphene structure. The integration of CNT into the 3D graphene structure enhanced the hydrophobicity, porosity and nano-roughness of the material [82]. In 2012, Iqbal and Abdala reported the clean-up of oil spills using thermally reduced graphene (TRG) which was synthesized by thermal exfoliation [83]. The oxidation state, surface morphology, size, surface area and composition were ascertained and consequently, evaluation of the sorption capacity, recovery and the recyclability of TRG were methodically carried out. TRG had a better adsorption capability, according to the findings than other carbon-based adsorbents while recovery is simply via filtration. Recyclability of TRG on the other hand was recorded to be up to six cycles [83]. Also, [84]. He et al., [85] fabricated different kinds of graphene oxide (GO) using various freezing methods and subsequently prepared reduced graphene oxide (RGO) foams by thermal reduction approach of the graphene oxide (GO) foams. The RGO foams were said to be hydrophobic

Table 1: Examples of nanomaterials for remediation of oil spill sites

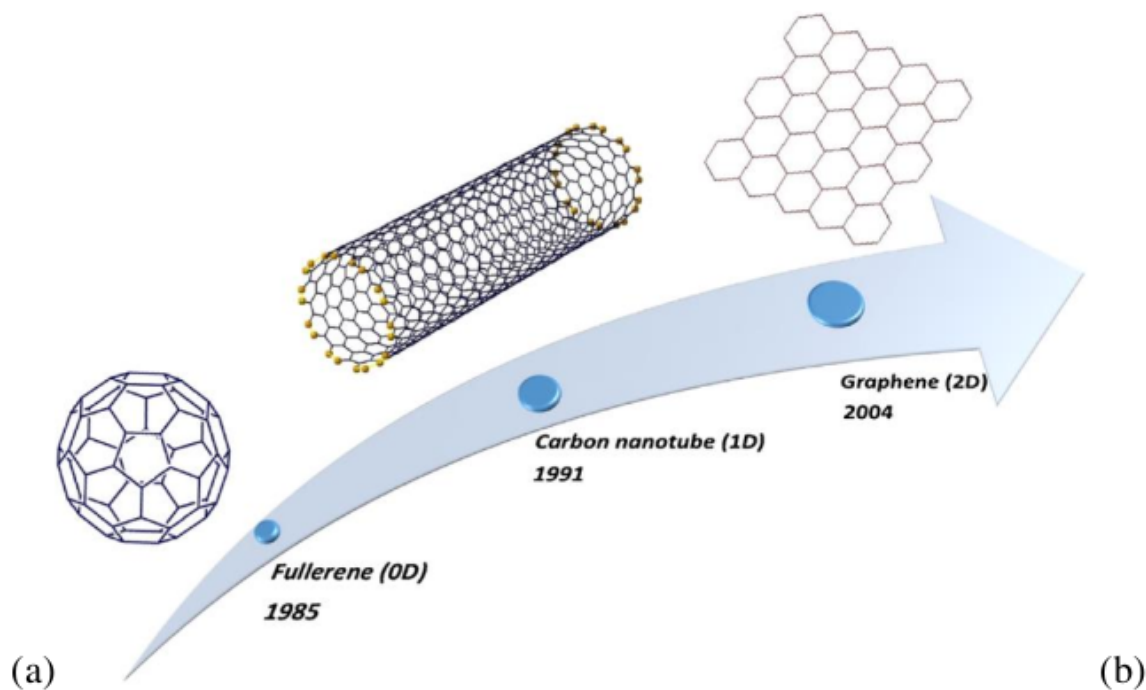
Target contaminant	Nanomaterials employed	Outcomes	References
Crude oil	Polystyrene (PS) nanofibers	Are efficient sorbent material. PS nanomembranes have the super-hydrophobicity needed to prevent exfoliated graphite from absorbing water which shows excellent results.	[64]
Diesel	Zinc oxide nanoparticles (ZnO NPs)	ZnO nanoparticles (NPs) for the effective removal of diesel via adsorption mechanism owing to the high efficiency and benefits of nanoparticles as adsorbents due to their high surface area	[65, 66]
Bitumen	Silicon dioxide (SiO ₂) nanoparticles	To demonstrate enhanced performance (softening point increases as nanosilica content increases, while penetration decreases), as well as a noticeable improvement in storage stability, nanosilica was added to bitumen.	[67, 68]
Gasoline	Polymer-Based Nanomaterials (Amphiphilic polyurethane NPs)	Amphiphilic polyurethane (APU) NPs were created for the removal of phenanthrene from contaminated aquifer sand as well as for the remediation of polynuclear aromatic hydrocarbons (PAHs) from soils.	[42]
Condensates	Calcium carbonate (CaCO ₃) and Silicon dioxide(SiO ₂) nanoparticles	Natural CaCO ₃ nanoparticles containing chitin are cost-effective agents which are for wettability alteration. Nanoparticles of SiO ₂ reducing surfactant adsorption on an oil reservoir's porous medium	[38, 69]

and had a high capacity for absorbing organic liquids. RGO foams had a higher oil adsorption capacity than GO foams for all oils (gasoline, pump oil, diesel oil, lubricating oil, and olive oil), owing to their porosity and less polar functional groups, which resulted in RGO foams having a higher hydrophobicity than GO foams [85].

2.3. Plasmonic-based nanomaterials

A significant and expanding area of research is plasmonic photocatalysis. It makes it possible to drive a variety of chemical reactions with the help of sunlight, a plentiful and sustainable energy source [86]. For More recently, some developing organic contaminants have been selected as the target contaminants since standard water treatment methods are ineffective at removing them, including endocrine disrupting chemicals, pharmaceutical effluents, and personal care products. The environment and water have both been severely contaminated as a result of oil exploration. The creation of TiO₂-coated glass, which is self-cleaning and anti-fogging, is one of the useful uses that has been created [87, 88]. Additionally, photocatalysis's environmental uses have broadened to include the detection of pollutants, inactivation of microorganisms, and self-cleaning of structures. Furthermore, it makes sense to assume that this field will keep growing as more and more serious environmental issues surface on a global scale. The remarkable light absorption efficiency of plasmonic nanostructures, which is reflected in their bright color, is one of their most alluring characteristics. When plasmonic

nanoparticles are exposed to light, the oscillating electromagnetic field causes the free electrons (from the conduction band) to collectively oscillate. The particular wave length at which the oscillation's amplitude reaches its maximum often varies with the types, forms, and dimensions of the metal. The remarkable developments in material science have made it possible to effectively regulate the morphologies of gold or silver nanoparticles [89]. Dye, persistent organic pollutants, and endocrine disrupting compounds can all be successfully eliminated by plasmonic photocatalysts when exposed to visible light. Excellent photo-catalysts should break down pollutants quickly while also maintaining their own stability. However, the poor stability that plasmonic metals typically experience during catalytic processes has restricted the range of their useful applications. Metal-semiconductor core-shell architectures may be able to overcome this drawback. Zhang et al., [90] developed a catalyst with a SiO₂ core, a gold NPs layer, and a doped nanocrystalline TiO₂ shell that was efficient and stable [90]. The capacity of Au to separate charges is suggested by the increased photocatalytic activity found compared to the catalyst without gold NPs. However, this benefit can be lost if too much gold is combined, as this might increase the rate of recombination [86]. Volatile organic molecules include formaldehyde (HCHO), toluene, acetaldehyde, and para-chlorobenzene among other contaminants (VOCs). They could be made of wood, painting, or flooring materials. HCHO has a low molecular weight and is thought to break down quite quickly. HCHO is one of the VOCs present in water bodies that is thought to be the most dangerous. As a result, it was the



(a)

(b)

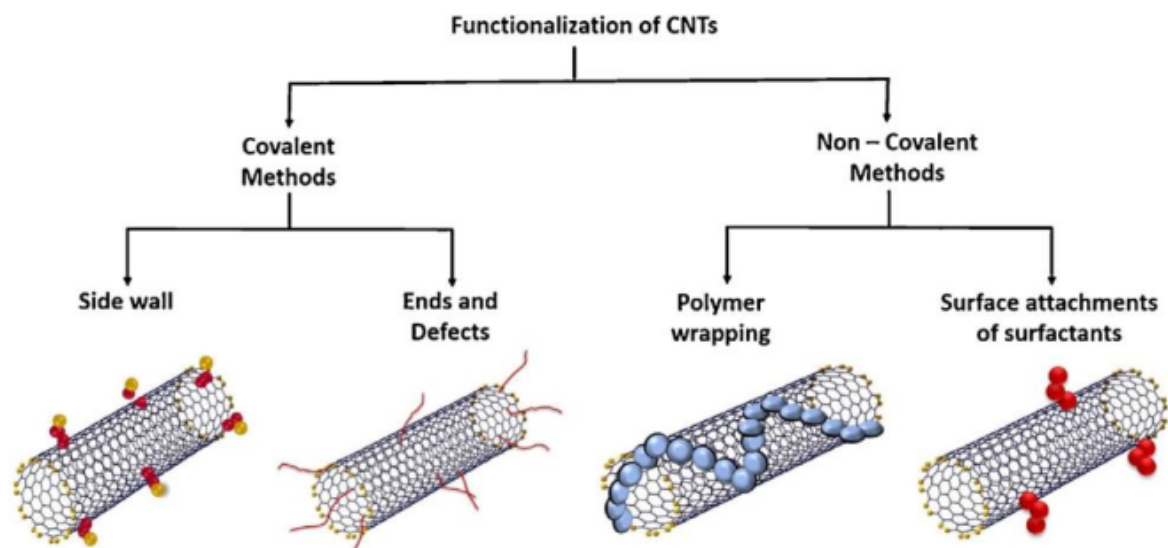


Figure 6: Carbon nanomaterial structures of many types (a); and functionalization methods of carbon nanotubes (b) [73]

target VOC of many investigations that used it to show how well their materials performed as photocatalysts. The potential of several semiconductors for the photocatalytic degradation of HCHO has since been highlighted in numerous investigations. He et al. [85] reported the photocatalytic degradation of HCHO over Pt/ TiO₂ in 2005. They discovered that Pt can considerably increase performance, but more importantly, they discovered that HCHO was completely mineralized into CO₂ and H₂O [91]. Later, other semiconductors, including Al₂O₃, Fe₂O₃, and MnO₂, as well as a number of plasmonic nanoparticles, including Au and Ag, were added to the efficient photocatalytic degradation of HCHO [86, 92].

2.4. Fibrous composites

Utilizing sorbent materials is one of the practical strategies used to mitigate oil spill pollution. In this regard, synthesizing fibrous-porous composites offers a possible substitute for the current sorbent materials. To eliminate pollutants from liquids like oil, water, and blood, high performance filtration and separation media made of polymeric fibers supplemented with magnetic nanoparticles and nanoclays can be utilized. Recently, several techniques, including splitting of bicomponent fibers, have been used to make ultrathin polymeric fibers [93] melt blowing, [94] electrospinning, [95] force spinning, combined spinning, or centrifugal spinning [96, 97]. Among these techniques, electrospinning has proven to be a simple and

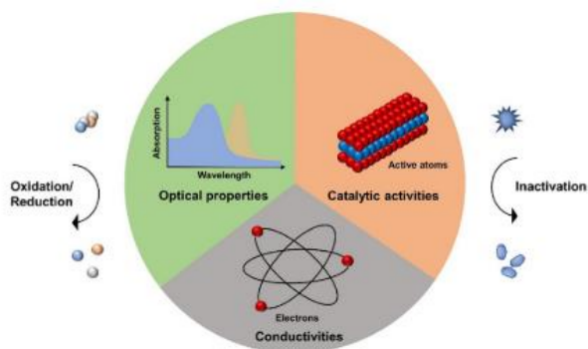


Figure 7: Schematic illustration of the properties of plasmonic nanomaterials and their applications in the photocatalytic oxidation/reduction of pollutants and inactivation of bacteria.

efficient method for creating 1 dimensional nanofibers with a variety of diameters and expansive surface areas. It may be possible to improve the characteristics of conventional composite materials by adding nanoscale fillers to polymer matrices. Polymer nanocomposites, a category of materials that exhibit exceptional adsorption, catalytic, magnetic, and optoelectronic capabilities, are of significant interest to both academia and industry. Different sorbent materials, such as cotton, cellulose acetate, polystyrene, polyurethane, polypropylene, polyester, and others, have been developed in recent years to improve their oil sorption capacities [97, 98, 99]. Due to their excellent separation efficiency, low cost, plentiful sources, and non-toxicity, fibrous porous sorbents appear somewhat promising. PVDF (poly(vinylidene fluoride)) and PSF (polysulfone) are two polymers with a number of beneficial characteristics, including high chemical resistance, good thermal stability, superior mechanical properties, biocompatibility, electroactive characteristics (piezoelectric, pyroelectric, and ferroelectric activities), and low surface energy. The polymeric fibers can be treated with spinel ferrite nanoparticles to create a superior magnetic response. By simply removing the sorbent from the water's surface with an external magnet, the electrospun magnetic fibrous composites provide significant benefits in both controlling oil collection and increasing separation rate. Nevertheless, there have only been a few reports on the creation of magnetic fiber sorbents for the cleanup of oil spills. Liu et al. [100] described how Fe₃O₄/polyacrylonitrile composite nanofibers were made and how they were designed for the adsorption of organic molecules. A straightforward two-step procedure involving electrospinning and solvothermal technique was used to create novel Fe₃O₄/polyacrylonitrile (PAN) composite nanofibers (NFs). The backbone of the PAN nanofiber was characterized and the development of a homogeneous covering of nanoparticles measuring about 20 nm in thickness was demonstrated. Tetracycline (TC) was used as the model antibiotic molecule in batch adsorption tests to determine the viability of Fe₃O₄/PAN composite NFs as a possible adsorbent for antibiotic removal. The results showed that Fe₃O₄/PAN composite NFs removed tetracycline effectively at pH ranges of environmental concern without

causing significant Fe loss (4 to 8). The adsorption of TC onto Fe₃O₄/PAN composite NFs demonstrated a highly efficient and novel adsorbent that can be easily modularized and separated, promising its enormous potential in drinking and waste water treatment for antibiotic removal. This was supported by a better fit of the pseudo-second-order kinetics model. However, Wang et al. [86] found that plasmonic nanoparticles show promise in this field because of their customized features, such as optical, photothermal, conductive, and catalytic properties. These characteristics allow wastewater to be viewed as a valuable resource as opposed to waste, so finding ways to recycle the nutrients, heavy metals, and energy from wastewater is of great interest. The majority of other functional nanomaterials are made for the remediation of water and air pollution, while only a small number of them are used in soil decontamination. Additionally, plasmonic nanoparticles show promise for the detection of a range of contaminants, including organic and inorganic pollutants as well as pathogenic microbes [86].

Advanced oxidation processes (AOPs) have already been reported to be the water treatment technology of the 21st century in terms of their inherent destructive nature, no mass transfer involved and probable mineralization of contaminants. They are also inexpensive, non-toxic and can degrade a wide range of contaminants. AOPs could be homogenous or heterogeneous. The former employs chemical/oxidants while the latter involves using a catalyst to effect degradation [101]. An attempt was made by Hsu et al. [102] for photocatalytic decomposition of several organic molecules found in oil spills. Toluene and 1-hexadecene were used as model aromatic and aliphatic compounds respectively in seawater oil spills. ZnO nanorods were used as a template to make nitrogen-doped TiO₂ nanotube thin films and the results of the degradation efficiencies of the different model compounds under visible light radiation for different present time intervals gave 10 % for toluene polluted seawater and 8 % for 1-hexadecene. The low efficiency observed for the latter was attributed to the length of the compound.

In a similar vein, a double-layer TiO₂ based mesh was used to develop an integrated approach for purifying water and separating oil [78]. The system was created by covering the upper and lower layers of the mesh with TiO₂ micro and nanostructures modified with octadecyl phosphoric acid. Due to the extensive and photocatalytic properties of TiO₂, the photocatalyst became a viable option for the separation of insoluble pollutants in water under UV light due to the integration of superhydrophobic and oleophilic properties as a result of the acid's modification into the intricate chemical network of the mesh [78]. In 2003, transformations in the chemical nature of dissolved fraction of crude oil after photolytic and heterogeneous photocatalytic treatment using TiO₂ utilizing ultraviolet fluorescence spectrophotometry and gas chromatography-mass spectrometry (GC-MS) (UVF) were evaluated [103]. In the presence and absence of photocatalyst, two Brazilian crude oil samples were subjected to UV-Vis irradiation. After short light exposure, complete photodegradation of both aromatic

and long-chain unsaturated hydrocarbons in the water-soluble fractions of both crude oil samples was seen employing photocatalyst, especially under strong solar radiation. However, in the absence of photocatalyst, the aromatic compound was not detected after treatment but the presence of long-chain unsaturated hydrocarbons was indicated by the peaks of the chromatogram. The experiment underlines the efficiency of photocatalytic degradation of water-soluble crude oil fractions in a short period under high solar irradiation [103].

Tony et al., (2009) tested the efficacy of photo-Fenton and heterogeneous photocatalysis for the treatment of a diesel-oil waste water emulsion and found it to be effective. It was attributed to the use of TiO_2 which was believed to increase the reaction time by reducing the chemical oxygen demand by up to 10 %. A note of caution was however given that if the treatment of crude oil impacted affluent is to be attempted a method for the separation of TiO_2 should be considered [104].

3. Nanosorption for Oil Spill Remediation

Nanosorption is a promising and emerging technique, which involves the use of nanomaterials to sequester contaminants from different media [105]. As a superset in which nanosorption is subsumed, adsorption is widely seen as the favorable, economical, simple but effective remediation process for the elimination of contaminants derived from watery media [42, 43, 106, 107, 108]. It involves the transfer of the contaminant from a phase to another phase usually a solid [109]. According to [110], the cost of treating water with adsorption ranges from 5 to 200 US dollars per cubic foot of treated water, compared to 10 to 450 US dollars per cubic foot of treated water for other treatment technologies (reverse osmosis, ion exchange, electro-dialysis, and electrolysis) [110, 111]. Another school of thought contends that adsorption can effectively remove both soluble and insoluble pollutants, with near-complete removal of more than 99 percent possible depending on the adsorbents used and the rest of the experimental environment [112]. Meanwhile, in the development of an adsorbent, the choice of the starting material, method of activation and target contaminants are very important factors to be considered for efficiency due to the roles of the aforementioned experimental dynamics on the surface area, pore sizes, surface charges and functional groups of the resulting adsorbents [113]. Although this technique has its inherent advantages and drawbacks, it can be consolidated by the concomitant application of other procedures. To x-ray the prospects of nanosorbents in the remediation of oil spills, the light will be beamed on different nano-engineered materials (nanosorbents) and their remediation activities in aqueous media. Oil adsorption rate is influenced by oil surface tension, whereas absorption rate is mostly influenced by membrane/exfoliated graphite surface area. Oil absorption rates varied between 2.5 and 40 g/g, while adsorption rates fluctuated between 0.32 g/g/m and 0.80 g/g/min, according to experimental data. T-B systems have also been used as containment barriers and sorbent materials, with positive results, including recyclability. Separation and collection of oil

from the water surface, combining water and oil with dispersing chemicals to aid natural deterioration, and in situ burning of the oil spill are the three types of oil spill remediation methods [63]. When oil and water combine to form an emulsion, the best approach for removing oil from water is to use adsorption techniques. The various materials used for oil adsorption and/or absorption include biomaterials which are the most commonly employed owing to their low cost and availability. To assess the relative effectiveness of dog fur and human hair sorbent products to peat moss and polypropylene sorbent in adsorbing crude oil from various terrestrial surfaces, researchers compared them to peat moss and polypropylene sorbent. Using standardized microcosm experiments, crude oil spills were mimicked, and contaminant absorbency was assessed as a percentage of crude oil eliminated from the initial spilled quantity. Sustainable origin absorbents made from dog fur and human hair were equally effective to polypropylene in extracting crude oil from non- and semi-porous land surfaces, with recycled dog fur products and loose-form hair showing a slight advantage over other sorbent types. Polypropylene sorbent performed much better than all other evaluated items at adsorbing spilled crude oil in a sandy terrestrial environment [114]. Pourmand et al., (2015) investigated the synthesis of nanoporous graphene by chemical vapor deposition (CVD) and its use as a recyclable nanosorbent in oil spill cleanup. Nanoporous graphene which is used as nanosorbent was effectively utilized for the remediation of crude oils and hydrocarbons. Heat treatment, solvent extraction, and filtering under mild suction with an appropriate recovery ratio were used to recover this crude and the hydrocarbons that had bonded to nanoporous graphene. Crude oil can be isolated from nanosorbent and reused after recovery using these recycling processes. It was observed that nanoporous graphene can be employed as a promising option in the removal of oil spills due to its excellent performance and good shape ability. Carbon nanoparticles have been used as adsorbents in recent years as a result of their exceptional surface characteristics and simplicity of modification. Nanosorbents made of carbon have a lot of strength, a lot of electrical conductivity, and a lot of thermal stability. Furthermore, the apparent intermolecular interactions in nanosorbents are effectively useful in the sorption mechanism, allowing it to be biodegradable and nontoxic. When added to typical sorbents, the physical, chemical and mechanical properties also exhibit development [39]. However, zinc ferrite magnetic nanoparticles (ZnFe_2O_4 , ZFO MNPs) were used to sorb oil spills from water surfaces. The magnetic force (40.22 mN) and density (0.5287 g/cm^3) of ZFO MNPs produced using sol-gel techniques were discovered to be sufficient for magnetic separation and flotation. Due to these properties (i.e density and magnetism), ZFO MNPs can be utilized as a sorbent to clean up oil spills on water surfaces [96, 116, 117]. While each technique has benefits and drawbacks, the effectiveness of each method is typically influenced by external factors such as oil composition, viscosity, and volume, as well as location and weather conditions. Also, the final disposal of nanoparticles, nanomaterials, or nanodevices can be a limiting factor when considering a nanosorbent method for the remediation of oil spills[38].

3.1. Negative Impacts of Nanosorbents on the Environment (Nanotoxicity)

The concept of nanotoxicology, according to Roberto & Christofoletti, (2020), presents guidelines for assessing the toxicity induced by nanomaterials. In a more recent review, He et al., (2018) explain the relevance of the nanobioeco interface. Upon introduction into the environment, nanomaterials may undergo modification which can eventually increase or decrease their toxicological profile. Hence, the fate of the nanomaterials becomes dependent on the dynamics of the environment [119].

We have established that industrial manufacturing procedures usually confer toxic properties on nanoparticles. The risk of exposure to these toxic components, however, depends on the likelihood and the extent of their exposure to living cells [120, 121]. Thus, the in vitro and in vivo studies of toxicity assist to ascertain their toxic potential. The toxic potential of several nanomaterials depends upon a set of intricate factors that include the physicochemical properties of the toxicants (size, shape, concentration or dosage and pH of the medium); hence, screening methods posit as the decisive feature [122]. Consequently, this section is a summary of the recent advances in toxicological assessment in the past few years and briefly describes some of them.

3.1.1. Metal Nanoparticles

Because metal nanoparticles are one of the most widely used components for oil spill remediation, the need for toxicity studies to be performed on them becomes highly imperative [122]. In their studies on the influence of solubility in the toxicity impact on living cell type (CHO and HeLa), Studer et al. [123] compared the toxicity of copper NPs having different chemical and physical properties with copper coated with carbon and copper oxide. The carbon-coated copper oxide NPs showed controlled toxicity because of the surface properties, while the Cu NPs showed the Trojan horse-type mechanism and induced significant toxicity. The result obtained from this study proved the relevance of physicochemical property [123]. To evaluate the significance of Trojan horse-type mechanism of toxicity induction, the experiment was tested with less soluble nanomaterials. The study showed that the less soluble copper oxide NPs were absorbed inside the cell by a clathrin-dependent pathway. However, at the low pH environment of lysosomes, they get partially soluble thereby releasing Cu ions, which further contributes to the cytotoxicity [124, 125, 126].

In another study, the influence of media in the cellular uptake and cytotoxicity of Au nanoparticles was evaluated. As soon as the NPs entered the cellular medium, NP-protein complex formed. Hence, a different type of complex forms upon media change. Dulbecco's modified Eagle's medium (DMEM) and Roswell Park Memorial Institute medium (RPMI) were employed in this study. The RPMI formed a complex with the

NP thereby showing larger intake and higher levels of toxicity compared to DMEM [127, 128].

3.1.2. Metal Oxides

Generally, metal oxide nanoparticles like zinc oxide (ZnO) and ferrous oxide (FeO) induce more toxicity than the less soluble ones such as ceria (CeO₂) and titania (TiO₂) [122]. Attarilar et al. [129] conducted an in vitro study of the toxicity influence of a varied range of metal oxides using LDH assay on exposed human epithelial cell lines (A549). The authors observed that an increase in the surface area results in an increased reactive oxygen species (ROS) production, which ultimately increases the toxicity imposed.

The influence of band gap and band edge potential of metal oxides was studied to predict the nature of oxidative stress and pulmonary inflammation. It was observed that the metal oxide particles that were completely soluble in the biological environment and having a band gap value comparable to the cellular redox potential (-4.12 to -4.84 eV) induced high levels of toxicity [130]. Auffan et al. [133] evaluated the influence of the redox state of iron-based nanoparticles and their cytotoxicity on strains of *Escherichia coli*. The materials used for this study are maghemite (γ -Iron), Fe₃O₄, and zero-valent iron. The stable γ -Iron imposed negligible toxicity but the other two ions of iron (Fe²⁺ and FeO) exhibited significant levels of toxicity due to oxidative stress. Iron undergoes a fenton reaction while reacting with the oxygen in the cellular environment and produces ROS.

3.1.3. Carbon Nanotubes (CNT)

Researchers have been persuaded to look into the potential toxicity of CNTs because of their many applications. Several reasons exist for toxicity as assessed in diverse studies, but only a few reports suggest the presence of metal catalyst particles (impurities) on CNT as what informs acute levels of toxicity: yet, it is impossible to synthesize CNTs devoid of metal particles entirely. However, much research has not been focused on ascertaining the major reason [122].

Madannejad et al. [132] studied the significance of the aspect ratio of CNTs in their cytotoxicity potential. The study recorded the cytotoxicity of the materials in the order: carbon black > carbon nano flakes > carbon nanotubes. The filaments were observed to be less toxic than the particles. The morphological alterations of the cells after a few days of exposure to all the materials remained the same but the difference in the viability observed was informed by the variance in the interaction of the nanomaterials with the exposed cells. Moreover, the difference in the interaction of the cells and the nanomaterials was presumably attributed to the presence of dangling bonds. These bonds are highly reactive sites found in high densities on carbon black and least in CNTs. On the other hand, Isobe et al. [133] estimated the cytotoxic potential of the

CNTs and found them to have only low levels of toxicity [134].

Several studies indicating inconsistent results on the cytotoxicity potential of CNTs have been conducted. However, in their attempt to critically examine the cytotoxicity standards for assessing nanomaterials, Rozhina et al., (2021) observed that CNTs show low levels of toxicity and assaying nanomaterials using MTT may not be dependable, since several instances of assay interference exist.

4. Summary, Perspectives and Future Research Needs

A comprehensive report of the different nanoremediation techniques, nanomaterials used and contaminants treated have been discussed. Nanotechnology is emerging and has not been fully accepted and integrated into mainstream research aimed at solving the world problems of oil spill. Studies have demonstrated the effectiveness of nanotechnology for the treatment of contaminated media. However, due to site specificity, geographical disparities and geological make up, results of different researches vary in relation to the aforementioned factors. It is therefore vital to develop and employ promising nanoremediation techniques for cleanup of contaminated sites. There is a dearth of information as regards nanoremediation which underlines the need to gather and harmonize information on the toxicity, reactivity and transport of prospective nanomaterials. Ex-situ and insitu treatments of contaminated surface and ground water in affected areas require methodical, extensive, long-term observation and monitoring for successful cleanup to be achieved.

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