Ecological Engineering 97 (2016) 98–105

Contents lists available at ScienceDirect

Ecological Engineering

journal homepage: www.elsevier.com/locate/ecoleng

Review

A review on multifaceted application of nanoparticles in the field of bioremediation of petroleum hydrocarbons

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ARTICLE INFO

Article history:
Received 10 March 2016
Received in revised form 4 August 2016
Accepted 11 August 2016

Keywords:
Nanoparticles (NP)
Bioremediation
PAHs: poly aromatic hydrocarbon
Immobilization
Nanofertilizer

ABSTRACT

Nanotechnology is an emerging field to produce nano-scale products with more efficient reactivity and larger surface area than its bulk phase. These unique attributes of nano-particles offer immense potential for their application to clean up petroleum hydrocarbons, pesticides and metals contaminated sites. As compared to the conventional physico-chemical methods of remediation of contaminated sites, the bioremediation has been drawing an increasing attention due to its economical, eco-friendly and self-propelling attributes. Nanoparticles (NPs) can be either applied directly for removal of organic contaminants through adsorption or chemical modification. It can also serve as facilitator in microbial remediation of contaminants either by enhancing the microbial growth or by immobilizing the remediating agents or through induced production of remediating microbial enzymes. Besides, nanoparticles induced enhanced production of biosurfactants in microorganisms, also contribute to improved solubility of hydrophobic hydrocarbons and thereby, create a conducive environment for microbial degradation of these compounds in environment.

Contents

1. Introduction

Oil spillage occurring during exploration, transportation, storage and refining of crude oil is a frequent phenomenon. The accidental oil spillage of crude oil from tankers leads to spreading of oil over a large surface area of the ocean and has immense impact on flora and fauna of the coastal shoreline and oceanic ecosystem. The oil refineries and off shore drilling operations also discharge large amount of crude oil/sludge which need to be properly disposed off or else they result into large scale contamination of land and water resources. The crude oil/petroleum sludge is a mixture of complex aliphatic, aromatic, asphaltene and resin hydrocarbons, which are well known for their toxicity, and some of them are reported to be mutagenic and carcinogenic in nature. The soil contamination with toxic PAHs results into extensive damage to biodiversity of flora and fauna, leading to reduced productivity on land and contamination of ground water (Thapa et al., 2012). In view of these problems, there is an urgent need to develop a cost effective and eco-friendly

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http://dx.doi.org/10.1016/j.ecoleng.2016.08.006
0925-8574/

technology for effective remediation of petroleum hydrocarbons (Hu et al., 2013). Many conventional physico-chemical methods used for remediation of petroleum hydrocarbons are found to be less economical and, they contribute to generation of secondary byproducts, which are not eco-friendly and also require additional efforts to handle them. While microbes present in our environment have capability to detoxify or complete mineralization of these toxic compounds in CO_2 and H_2O. The bioremediation is, therefore, considered to be more eco-friendly and sustainable state-of-the art, which can be employed under both *in-situ* and *ex-situ* conditions for removal of toxicants (Nkeng et al., 2012).

The natural process of bioremediation of petroleum hydrocarbons is extremely slow as it gets hindered by reduced solubility of the hydrophobic constituents of crude oil and their bioavailability to the microorganisms. Simultaneously, the hydrophobic and toxic nature of hydrocarbons in the crude oil also contributes to poor sustainability of the microbial biomass, particularly at higher concentrations of petroleum hydrocarbon. The present state-of- the art in the field of nanotechnology offers ample avenues for its potential application in the field of bioremediation technology to overcome its perceived limitations. Thus, the present review article intends to highlight the application of nanotechnology that offers a tangible solution to the inherent technological limitations confronted by the process of bioremediation of petroleum hydrocarbons, such as reduced solubility of the hydrophobic substrate and their reduced bioavailability to the microrganisms, kinetic limitations on the crude oil degrading enzymes and poor health of microorganisms. Thus, the nanotechnology in this context is considered as an effective tool to facilitate the bioremediation of persistent hydrophobic toxicants and augment the process.

Nanomaterials are comprised of particles with at least one dimension measuring between 1.0–100 nm. Few specific characteristics like high surface-to-volume ratio, enhanced magnetic and special catalytic properties *etc*. (Gupta et al., 2011) make these nanoparticles (NPs) more advantageous than their bulk phase counterparts in the field of remediation technology. A larger surface area and surface reactivity of nanomaterials than their respective bulk material enables them to remediate the contamination at fast rate with reduced amount of hazardous by-products (Bhattacharya et al., 2013). A diverse form of nanomaterials like carbon nanotubes, nanoscale zeolites, dendrimer enzymes, biometallic particles and metal oxides are now being used for decontamination of the polluted sites (Mehndiratta et al., 2013). Use of multi-walled carbon-nano tube has been employed for removal of organic pollutants like PAHs and Polychlorinated biphenyls (PCB) (Shao et al., 2010; Gotovac et al., 2006). Zhang (2003) has presented a comprehensive list of pollutants which can be potentially remediated by using iron nanoparticles. The use of nanoparticles in enzyme mediated remediation technology is gradually gaining ground due to the reasons that NPs provide a biocompatible and inert microenvironment which is least interfering with the native properties of the enzymes and helps in retaining their biological activities (Ansari and Husain, 2012). In addition, magnetic property of NPs enables an easy separation of immobilized enzymes or proteins from the reaction mixtures by simply applying a magnetic field. Thus, there is no need for centrifugation or filtration which, otherwise, prolongs the procedural trivialities and operational difficulties (Khoshnevisan et al., 2011; Ranjbakhsh et al., 2012). However, a careful selection of nanomaterials for remediation process is a crucial step as they may be toxic to the microorganisms involved in the remediation process (Rizwan et al., 2014). The present review is an attempt to focus on potential application of nanotechnology and its integration with various crucial processes associated with the bioremediation technology such as immobilization of the hydrocarbon degrading microorganisms, substrates and/or the enzymes, solubilization of the hydrophobic compounds and stimulation of degradative properties of microbes.

2. Nanoparticles and enhanced solubility of hydrophobic compounds

Hydrophobicity of the petroleum hydrocarbons reduces its solubility in water and increases the sorption on soil micelle, which acts as limiting factor in bioremediation of these compounds. It has been reported that biosurfactants produced by the microorganisms can reduce the hydrophobicity of the organic compounds by decreasing the surface tension and thereby, enhance the bioavailability of hydrophobic compounds to microorganisms, which is a very critical factor in bioremediaition process (Cameotra and Makkar, 2010). However, production of biosurfactants in microorganisms is limited due to metabolic limitations of the pathways involved in the synthesis of biosurfactants (Desai and Banat, 1997). Kiran et al. (2011) reported that presence of metals is one of the critical factors that regulates the production of biosurfactants in microorganisms. Among the several metal ions, Fe is widely considered as a key nutritional requirement for production of biosurfactants in several microorganisms (Haferburg and Kothe, 2007). The Fe atom is generally used by microorganisms in several crucial metabolic pathways due to low toxicity (Liu et al., 2013) and inherent biocompatibility (Perez et al., 2002). Use of iron nanoparticles in *Nocardiopsis* MSA13A has been shown to stimulate the production of biosurfactant by about 80%, when compared with the control (without Fe). While no observable effect of Fe on the morphology of filamentous structure of *Nocardiopsis* MSA13A was observed under Scanning Electron Microscope that supports non-toxic nature of Fe nanoparticles (Kiran et al., 2014). Similarly, Liu et al. (2013) observed about 57% increase in the bacterial growth and 63% increase in the production of biosurfactants in *Serratia* sp. in the presence of just 1.0 mg L^{-1} of Fe nanoparticle.

In the present scenario, surfactant micelles are generally used to facilitate the solubility of hydrophobic contaminants. But the surfactants are also known to reduce the efficiency of bioremediation as they tend to interact with liposomes of degrading microorganisms, resulting into alterations in the membrane properties (Kaczorek and Olszanowski, 2011). The addition of surfactants in the presence of PAH apparently leads to enhanced PAH toxicity to microorganisms, preferential metabolism of the surfactants, interference with the membrane uptake process and reduced bioavailability of hydrocarbons (Kumari et al., 2014; Mulligan et al., 2001).

Recent advances in the field of nanotechnology have provided promising future to the bioremediation technology, if it is used appropriately at right step. Tungittiplakorn et al. (2005) used Amphiphilic PolyUrethane (APU) nanoparticles to increase the solubilization of phenanthrene—a constituent of crude petroleum. The nanoparticles made out of polyurethane acrylate anionmer or polyethylene glycol precursor chains with hydrophobic interiors showed high affinity for phenanthrene and facilitated the desorption and mobility of PAH from soil micelle. According to Tungittiplakorn et al. (2005), the nanomaterial produced from APU improved the PAH solubility and mobility by forming nanoparticle-PAH suspension which competes with Non-Aqueous Phase Liquids. The main advantage associated with these NPs against the application of the surfactants is that a desired affinity of these nanoparticles towards hydrophobic contaminants can be achieved by simply modifying the hydrophobic segment of the precursor chains and, their mobility can be increased by controlling the charge density on the modified nanoparticles (Tungittiplakorn et al., 2005).