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Kinetics and physicochemical studies of surfactant enhanced remediation of hydrocarbons contaminated groundwater

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ABSTRACT

Kinetics and physicochemical studies of surfactant enhanced remediation of hydrocarbons contaminated groundwater were investigated for efficiency and effectiveness. 10% pollution was simulated in the laboratory by contaminating groundwater samples with crude oil, automatic gasoline oil (diesel) and domestic purpose kerosene (DPK) in replicates of five. Physicochemical properties of the hydrocarbons contaminated groundwater samples and a control sample were investigated before and after treatments. Total petroleum (TPH) hydrocarbon as target contaminant was monitored periodically to assess the extent of the remediation process. TPH was determined by molecular spectrophotometry technique. Other physicochemical parameters such as pH, turbidity, alkalinity, dissolved oxygen (DO), biochemical oxygen demand (BOD), chemical oxygen demand (COD), conductivity, ammonia, nitrate, phosphate, salinity, total dissolved solids (TDS), total suspended solids (TSS) and total solids (TS) were obtained using standard methods while heavy metals levels were determined by atomic absorption spectrophotometry. Different kinetics models were tested to determine the appropriate kinetics model. The pseudo-first order kinetics is established with rate constant as 1.80×10^4 ; 1.78×10^4 ; $1.53 \times 10^4 \text{ mg}^{-1} \text{ L h}^{-1}$ for crude oil, diesel and kerosene respectively at 30 °C. At the end of the remediation after 6 h there was 89.11%; 93.21%; 87.76% reduction in TPH as crude oil, diesel and kerosene for the treated samples in that order. The application of surfactant enhanced remediation using sodium dodecyl sulphate is found to be very efficient, effective and rapid in reducing total petroleum hydrocarbon as crude oil, kerosene and diesel as target contaminants. There is the need for post-treatments after remediation for most of the physicochemical parameters are impaired and do not meet the Guideline and Standards for Environmental Pollution Control in Nigeria set by Federal Ministry of Environment and World Health Organization for drinking water and agricultural uses in order to make them fit for these purposes.

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1. Introduction

Surfactants can be classified according to the nature of the hydrophilic portion of the molecule: anionic, cationic, nonionic, zwitterionic. Zwitterionic surfactants have both positive and negative charge in the head group. Non-ionic surfactants generally have smaller critical micelle concentration (CMC) values than ionic surfactants and are known to be good solubilizers of hydrophobic substances [1,2]. Cationic surfactants are often toxic in the mg/L range to a wide variety of aquatic organisms [3]. Skrtic, et al. [4] reported that cationic surfactants are unsuitable as extracting agents due to

poor extraction efficiency. Surfactants may aid in remediation of soil and water contaminated with hydrophobic organic compounds (HOC) [5,6]. These studies showed that aqueous surfactants solutions significantly enhanced the removal of HOC from soil and water. Extractive efficiencies of surfactant solution for HOC were seven to ten times greater than those which could be obtained by flushing with water alone. Surfactants can be used to vastly increase the solubility of the HOC in water and also lower the interfacial tension at the water - HOC interface [6,7].

Choosing the best surfactant solution for desorbing and solubilising petro hydrocarbon can be a daunting task since over 700 different types of surfactants are commercially available [8]. Sodium dodecyl sulphate as a surfactant used in this remediation work is cheap, non-odorous and soluble in water at room temperature.

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Material Safety Data Sheet (MSDS) information on the surfactant indicated low toxicity and favourable biodegradability.

Although surfactant technology in remediation works are widely cited in literature yet the application of sodium dodecyl sulphate is scantily reported in most literature. The present work is poised at looking at kinetics and physicochemical studies of surfactant enhanced remediation of hydrocarbons contaminated groundwater for efficiency and effectiveness with view of contributing to the existing body of knowledge on chemical remediation technology.

2. Materials and methods

2.1. Materials

The petroleum contaminants used in the study were (i) Brent crude oil. It was obtained from Shell Petroleum Development Company (SPDC) Flow Station, Kokori, Delta State, Nigeria. (ii) Automatic gasoline oil (diesel) and (iii) Domestic purpose kerosene (DPK). The latter two were obtained from Petroleum Marketers at Agbor, Delta State, Nigeria. All plastics and glass ware utilized were pre-washed with detergent water solution, rinsed with tap water and soaked for 48 h in 50% HNO₃, then rinsed thoroughly with distilled water and air-dried in the laboratory.

2.2. Preparation of samples

Fifteen 30 mL vials were each filled with 14 mL groundwater. 1.4 mL of kerosene, diesel and crude oil were respectively added to each of the three sets groundwater containing vials and stirred thoroughly using a magnetic stirrer to obtain 10% contamination of each contaminant type. These preparations were carried out in replicates of five for each contaminant.

2.3. Control groundwater samples

Groundwater control samples were investigated for background contamination with any of the three hydrocarbons.

2.4. Preparation of sodium dodecyl sulphate treatment solution for surfactant method

Several concentrations of 10,000–90,000 mg/L of sodium dodecyl sulphate were prepared by dissolving 1–9 g of sodium dodecyl sulphate in nine different 100 mL volumetric flasks; thoroughly shaken and made up to marks using distilled water. The solutions were left overnight to properly dissolve.

2.5. Optimisation studies

Optimization of concentration and pH of treatment solutions were investigated to ascertain the optimum conditions for TPH removal.

2.5.1. Optimization of concentration of sodium dodecyl sulphate on surfactant method

TPH as crude oil was determined by molecular spectrophotometry following standard procedure adopted by Wang, et al. [9] with slight novel modification of drying the wet extracts with anhydrous sodium sulphate and also to avoid unnecessary interference of impurities. 4.6 mL of the several prepared sodium dodecyl sulphate solutions (10,000–90,000 mg/L) were taken in 30 mL vials to which 14 mL groundwater and 1.4 mL of crude oil were added. The vials were properly sealed with Teflon films and shaken for 1 h using a mechanical shaker at 200 revolutions per minute (r.p.m.).

The crude oil in the water layers were separated from that in the sodium dodecyl matrix using separating funnels. Crude oil in the water layers was extracted using hexane. The wet extracts were passed through 5 g of anhydrous sodium sulphate on Number 42 Whatmann filter paper in different filtration set ups in order to obtain clean and dry extracts. TPH as crude oil was read off by UV/Visible spectrophotometer (made in the United Kingdom, 2007 model) at wavelength of 460 nm. The procedure was repeated using the other four replicate samples. The experiment was repeated using diesel and kerosene at wavelength of 350 and 310 nm respectively.

2.5.2. Effect of pH on surfactant method

The effect of pH on surfactant method was investigated by adjusting the pH of two sets of crude oil contaminated groundwater samples in 30 mL vials to pH = 3 and pH = 8 using 1 M H₂SO₄ and 1 M NaOH. The samples were then treated with the optimum treatment concentration obtained in the same study. The vials were then properly sealed with Teflon film and shaken with a mechanical shaker for an hour followed by extraction and analysis. Crude oil in the water layers were extracted using hexane and TPH as crude oil were determined by UV/Visible spectrophotometer at wavelength of 460 nm using standard procedure adopted by Wang, et al. [9] and also described in the same work. Procedure was repeated using the other four replicate samples. The experiment was repeated using diesel and kerosene at wavelength of 350 and 310 nm respectively.

2.6. Kinetics studies

Kinetics experiment was carried out to establish the optimum time and efficiencies of the remediation method employed in the study. Several solutions of 4.6 mL of sodium dodecyl sulphate of concentration of 50,000 mg/L obtained from the same work were prepared and added to 14 mL groundwater and 1.4 mL of crude oil in 30 mL vials. The pH of the solutions in the vials were adjusted to pH = 3, optimum pH obtained in this same study. The vials were properly sealed with Teflon films and shaken for 1 h using a mechanical shaker at 200 rpm (r.p.m.). The crude oil in the water layers were separated from that in the sodium dodecyl matrix using separating funnels. Crude oil in the water layers were extracted using hexane and TPH as crude oil were determined by UV/Visible spectrophotometer at wavelength of 460 nm [9].

The concentrations of TPH as crude oil left were plotted against time. The kinetics model for the remediation process was tested by plotting $\ln[B]_0 - \ln[B]_t$ against time using the experimental kinetics data. The slope of the graph is the rate constant, k . The actual rate constant was obtained by multiplying the value of the slope from the graph with the optimum concentration of the treatment solution that was used in excess, $[A]_0$ in the optimisation experiment in this same work. The kinetics experiment was repeated using diesel and kerosene at wavelength of 350 and 310 nm respectively.

2.7. Determination of physicochemical properties of samples

Physicochemical parameters such as pH, turbidity, alkalinity as CaCO₃, dissolved oxygen (DO), biochemical oxygen demand (BOD), chemical oxygen demand (COD), conductivity, ammonia, nitrate, phosphate, salinity as chloride, total dissolved solids, total suspended solids and total solids were obtained using standard methods adopted by APA, [10].

Heavy metal contents were determined using atomic absorption spectrophotometer (AAS) following procedures adopted by APA, [10]. Sample was first digested using the standard method proposed by Nabil and Barbara [11]. 50 mL of the sample was

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