



# Effect of interlayer cations of montmorillonite on the biodegradation and adsorption of crude oil polycyclic aromatic compounds



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## ABSTRACT

Cation exchange capacity, surface acidity and specific surface area are surface properties of clay minerals that make them act as catalysts or supports in most biogeochemical processes hence making them play important roles in environmental control. However, the role of homoionic clay minerals during the biodegradation of polycyclic aromatic compounds is not well reported. In this study, the effect of interlayer cations of montmorillonites in the removal of some crude oil polycyclic aromatic compounds during biodegradation was investigated in aqueous clay/oil microcosm experiments with a hydrocarbon degrading microorganism community. The homoionic montmorillonites were prepared via cation exchange reactions by treating the unmodified montmorillonite with the relevant metallic chloride. The study indicated that potassium-montmorillonite and zinc-montmorillonite did not enhance the biodegradation of the polycyclic aromatic hydrocarbons whereas calcium-montmorillonite, and ferric-montmorillonite enhanced their biodegradation significantly. Adsorption of polycyclic aromatic hydrocarbons was significant during biodegradation with potassium- and zinc-montmorillonite where there was about 45% removal of the polycyclic aromatic compounds by adsorption in the experimental microcosm containing 5:1 ratio (w/w) of clay to oil.

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## 1. Introduction

One of the main negative aspects of the use of petroleum as energy source is the high incident of oil spill arising from exploration, exploitation and transportation activities. Oil spill causes tremendous pollution of the environment and has resulted in large numbers of contaminated sites (Vidali, 2001; Environment Agency, 2006). Ecosystems' vegetation and wildlife are adversely affected by soils contaminated by hydrocarbons. It has been reported that if the contamination is severe, not only would public health be threatened, but also the contaminated area would be rendered unsuitable for human habitation (Sunggyu, 1995). Aromatic compounds especially aromatic hydrocarbons are common constituents of crude oil. Aromatic hydrocarbons contain one or more nuclei of benzene, naphthalene, and phenanthrene and may be linked to paraffinic side chains or naphthenic rings. The aromatic hydrocarbons can be divided into mono-aromatic or polycyclic aromatic hydrocarbon (PAH). Examples of monoaromatic

hydrocarbons are benzene, toluene, ethylbenzene and xylene (BTEX) whereas examples of PAH include phenanthrene, pyrene and chrysene (Harayama and Kanaly, 2000). Aromatic compounds in crude oil generally include both aromatic hydrocarbons and other organic compounds of benzene derivatives that may contain other elements such as N, S, O and metals. Benzothiophenes, indole, carbazole, pyridine, quinoline etc are some of the aromatic compounds in this group. Polycyclic aromatic compounds in addition to being biomagnified via trophic transfers have been reported to have toxic, mutagenic, teratogenic and carcinogenic effects (Cerniglia, 1984; Clements et al., 1994). Hence, United States Environmental Protection Agency (USEPA) has classified some polycyclic aromatic hydrocarbons as priority pollutants. Biodegradation is the principal removal process of crude oil spill in soils and sediments and involves the mineralization of the crude oil hydrocarbons by indigenous or added microbes (Watson et al., 2002; Bogan and Sullivan, 2003; Retz et al., 2008). Clay minerals are abundant in soils and sediments and their properties such as high surface area, interlayer cations and cation exchange capacity (CEC) are known to play important roles in biogeochemical processes (Bergaya et al., 2006). Clay minerals have been reported to affect the biodegradation of oils and saturated fraction of crude oil (Chaerun and Tazaki, 2005; Warr et al., 2009; Ugochukwu et al., 2013). However, there

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have not been reports on the role of interlayer cations of montmorillonites on the simultaneous adsorption and biodegradation of crude oil polycyclic aromatic compounds. It is important to investigate the effect of the different homoionic montmorillonites on the biodegradation and adsorption of the different groups of polycyclic aromatic compounds as this will have a far reaching implication on bioremediation strategies. The homoionic montmorillonites are usually generated by exchanging the interlayer cations of unmodified montmorillonite with excess metallic chloride of the required metallic ion to produce the desired homoionic montmorillonite via cation exchange reactions (Reddy et al., 2007).

Recent research efforts are directed towards improving the strategies that can be employed to effectively return an oil polluted site to its former unpolluted state. This study was aimed at investigating the role of interlayer cations of montmorillonites on the simultaneous adsorption and biodegradation of selected crude oil polycyclic aromatic compounds. The study was based on laboratory study conducted in aqueous clay/oil microcosm experiments with hydrocarbon degrading microorganism community.

## 2. Materials and methods

The clay minerals used in this study were homoionic montmorillonites prepared from unmodified montmorillonite. Bentonite was the source of the unmodified montmorillonite. Bentonite (Berkbent 163) was supplied by Steetley bentonite & absorbent Ltd (now Tolsa UK Ltd; [www.tolsa.com](http://www.tolsa.com)). All the chemicals were supplied by Sigma Aldrich. Microbial communities responsible for biodegradation of the crude oil hydrocarbons were isolated from beach sediment sample consisting of fine sand particles collected in a sterilised glass bottle (Duran) from a site at St Mary's lighthouse near Whitley Bay, Newcastle upon Tyne (National Grid Reference NZ 352 754), United Kingdom and stored at 4 °C in cold room until commencement of the experiment. The Bushnel–Haas (BH) broth and agar were supplied by Sigma Aldrich. The crude oil was an undegraded North Sea crude oil originally supplied by British Petroleum (BP).

### 2.1. Preparation of the homoionic montmorillonites

The homoionic montmorillonites samples were generated from exchanging the interlayer cations of the montmorillonite with Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, Zn<sup>2+</sup>, Al<sup>3+</sup>, Cr<sup>3+</sup> and Fe<sup>3+</sup> to produce, Na-montmorillonite, K-montmorillonite, Mg-montmorillonite, Ca-montmorillonite, Zn-montmorillonite, Al-montmorillonite, Cr-montmorillonite and Fe-montmorillonite. Salt solution concentration of 0.5M of the corresponding metal chloride was prepared and then 200 mL of each of the solution used to disperse 5 g of clay mineral to have a clay suspension (Reddy et al., 2007). The suspensions were shaken for 24 h in a mechanical shaker and then subsequently centrifuged. The supernatant was rejected and the clay mineral washed repeatedly until the chloride level is not measurable.

### 2.2. Characterization of the clay samples

#### 2.2.1. X-ray diffraction (XRD)

The basal spacings of the clay samples were measured by XRD. The samples were prepared for XRD measurement by orienting the clays in a glass slide following standard procedure. The slides were air dried and placed in a desiccator containing silica gel to prevent rehydration. Glycolated and heat treated (at 300 °C) samples were also prepared following standard procedure. The XRD was observed using Cu-K $\alpha$  generated at 40Kv and 40 mA using PANalytical X'Pert Pro MPD fitted with an X'Celerator machine. The data was collected

over a range of 2–70 °2 $\theta$  with a nominal step size of 0.0167 °2 $\theta$  and nominal time per step of 1.00 s. Data were interpreted by reference to X'Pert accompanying software program High Score Plus in conjunction with the ICDD Powder Diffraction File 2 database (1999) and the Crystallography Open Database (October 2010; [www.crystallography.net](http://www.crystallography.net)).

#### 2.2.2. Fourier transform infra red (FTIR)

The clay samples were prepared using potassium bromide (KBr) pellets at sample concentration of 1% and the Fourier transform infrared spectra of the clay samples were recorded on Thermo Nicolet Nexus 870 fitted with a transmission accessory equipped with a DTGS detector. The spectrum of the clay sample was acquired by collecting 100 scans over a wavenumber of 400–4000 cm<sup>-1</sup> at 4 cm<sup>-1</sup> resolution.

#### 2.2.3. Surface area

The surface area of the clay samples were measured by the EGME method following the method of Carter et al. (1965).

#### 2.2.4. Cation exchange capacity (CEC) of the clay samples

The CEC of the clay samples was determined following the standard ammonium acetate method (Lewis, 1949).

#### 2.2.5. pH

The pH of the clay suspension was measured with a pH meter.

#### 2.2.6. GC–MS

The GC–MS used was Agilent 6890 GC instrument with split/splitless injector (280°), linked to a 5975 MSD mass spectrometer. The GC was equipped with a fused silica capillary column (30m × 0.25 mm) coated with 5% phenyl polysiloxane (HP-5) stationary phase. The GC oven was programmed from 40 °C for 5 min and then ramped at 4 °C/min, up to 300 °C where it was held for 20 min. The carrier gas used was helium at a flow rate of about 1 mL/min and initial pressure of 50 kPa while split at 30 mL/min. The sample was injected by an HP7683 autosampler and the split opened after 1 min to vent the solvent. The mass spectrometer used electron ionization energy of 70 eV. The following operating conditions were used: Source temperature of 230 °C; Quadrupole temperature of 150 °C; Multiplier voltage of 2000 V; Interface temperature of 310 °C. Selected ion monitoring (SIM) mode was used to monitor ions of interest such as 156, 170, 178, 192, 206 etc.

#### 2.2.7. GC–FID

The GC instrument used was an HP 5890 series II gas chromatograph equipped with a split/splitless injector and flame ionization detector (FID). The sample was injected using an HP 7673 autosampler. The separation of the crude oil hydrocarbon compounds was carried out on an Agilent HP-5 capillary column (30 m × 0.25 mm) coated with 5% phenylmethyl-polysiloxane (0.25  $\mu$ m thick) stationary phase. The GC oven temperature was programmed from 50 °C for 2 min and then ramped at 4 °C/min up to 300 °C where it was held for 20 min. The carrier gas used was hydrogen at a flow rate of about 2 mL/min at initial pressure of 100 kPa. The GC data was acquired using Atlas software on HP computer desktop.

### 2.3. Biodegradation experiments

The procedure adopted for the enrichment culture and isolation of the microbial cells involved mixing 20 g of Whitley Bay sediments, 100 mL of Bushnel–Haas (BH) medium (prepared by dispersing 327 mg of BH broth in 100 mL of de-ionized water) which was autoclaved and 500 mg of crude oil in a 250 mL conical

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