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Stabilisation/solidification and bioaugmentation treatment of petroleum drill cuttings

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ABSTRACT

Petroleum drill cuttings are usually treated by techniques suitable for particular contaminant groups. The significance of this study consists in the development of a treatment technology that can simultaneously handle the hydrocarbon and metal constituents of drill cuttings. Bioaugmentation is combined with stabilisation/solidification (S/S), within S/S monoliths and in granulated S/S monoliths. Portland cement was used for S/S treatment at 30% binder dosage. Bioaugmentation treatment involved two bacterial densities of a mixed culture bio-preparation. The effects of inclusion of compost, fertiliser and activated carbon were also evaluated. After 28 days, the combined S/S and bioaugmentation treatments recorded up to 15% higher total petroleum hydrocarbon (TPH) loss than control S/S treatment without bioaugmentation. Embedding fertiliser, activated carbon and higher bacterial density within S/S monoliths resulted in the highest (99%) TPH reduction but higher concentrations of metals. The addition of compost and lower bacterial density to granulated S/S monoliths led to similar (98%) TPH degradation and lower amounts of metals. The results suggest that with better mixture optimisation, combining S/S and bioaugmentation could engender more sustainable treatment of drill cuttings.

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1. Introduction

Drill cuttings are soil, rock fragments, and pulverised material that is removed from a borehole drilled for oil and gas extraction. They may include a small amount of fluid that results from the drilling process ([Mauger et al., 2014\)](#page--1-0). This particular liquid, called drilling mud, is used to lubricate and cool the drill bit and transport the drill cuttings to the surface. It is also used to balance down-hole formation pressures to prevent blowouts of oil and gas ([Ball et al.,](#page--1-0) [2012\)](#page--1-0). It is then separated from the drill cuttings at the surface. Due to the drilling fluids used and the geologic formation penetrated, drill cuttings are usually co-contaminated with total petroleum hydrocarbons (TPH) and metals. Elevated concentrations of metals such as As, Cr, Cu, Pb, Ni, Zn, among other metals have been reported [\(Balgobin et al., 2012; Johnson and Graney, 2015\)](#page--1-0). Thus, drill cuttings from oil- and synthetic-based fluids are categorised as hazardous waste by the European Waste Directive

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([Technical Guidance WM3, 2015\)](#page--1-0). However, drill cuttings from water-based fluids are not classified as special waste unless they contain oil ([Ball et al., 2012](#page--1-0)). In contrast, drill cuttings are exempted from being classified as hazardous waste by the United States Environmental Protection Agency (US EPA). However, to prevent health and environmental hazards, proper management practices are suggested [\(USEPA, 2002](#page--1-0)). Thus, it is essential that drill cuttings be properly treated before disposal or reuse. Potential reuse options include as road aggregate, construction material and engineered fill ([Mauger et al., 2014\)](#page--1-0).

Drill cuttings can be managed by disposal, either at sea or to landfill, burial in pits, thermal treatment, stabilisation/solidification (S/S) and bioremediation ([Ball et al., 2012\)](#page--1-0). Thermal treatment is commonly used but resulting fugitive emissions makes it less ecofriendly. Biological treatment can, however, convert contaminated matrices into stable and reusable products in a more eco-friendly manner ([Ball et al., 2012\)](#page--1-0). Nevertheless, there would be incomplete degradation of the hydrocarbons. The presence of high concentrations of metals may also limit the effectiveness of biological treatment. S/S is however very efficient in chemical fixation and physical encapsulation of contaminants ([Conner and Hoeffner,](#page--1-0)

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[1998](#page--1-0)). It is most suitable for the immobilisation of metals due to the high pH of cement and, to a lesser extent, organic contaminants. Organic contaminants often have a deleterious effect on the hydration of Portland cement ([Kogbara, 2014](#page--1-0)). There is much uncertainty regarding the long-term performance of S/S technology. Contaminants are merely immobilised within the high pH cement matrix, which may pose a further risk in the future. Several degradation mechanisms that could affect the long-term performance of a stabilised/solidified material has been identified ([Perera](#page--1-0) [et al., 2005\)](#page--1-0). These include a progressive reduction of the initial alkalinity of such materials through carbonation by $CO₂$ uptake ([Kogbara et al., 2013\)](#page--1-0). The initial alkalinity of stabilised/solidified materials could also be progressively reduced by natural leachants such as rainwater or landfill leachate with slightly acidic pH ([Kogbara et al., 2012\)](#page--1-0). Hence, S/S treatment may become unsustainable in the eventual breakdown of the treated materials. Therefore, it is important to combine S/S with biological treatment to bring about some form of contaminant attenuation over time.

Portland cement is the most widely used binder for S/S treatment. Cement is preferred as it can chemically bind free liquids, and encapsulate waste particles surrounding them with an impermeable coating. It can also chemically fix hazardous constituents by reducing their solubility and facilitating the reduction in toxicity of metals and TPH ([Conner, 1997; Kogbara et al., 2012](#page--1-0)). Initial attempts at combining S/S with biological treatment entailed adding a range of additives to Portland cement. These included compost as a source of microbes and nutrients and air-entraining and waterretaining agents. Compost showed superior performance compared to other additives ([Harbottle and Al-Tabbaa, 2006, 2008\)](#page--1-0). The numerous and diverse microbial population associated with composts could be possibly responsible for the observation. The high levels of substrates in compost can also lead to cometabolism of organic contaminants [\(Barker and Bryson, 2002; Kogbara, 2013\)](#page--1-0). Subsequent efforts utilised magnesium phosphate cement(s) with relatively lower pH that could favour microbial survival than highly alkaline Portland cement [\(Kogbara et al., 2011](#page--1-0)).

The previously mentioned studies on soils artificially contaminated with organic and metallic contaminants suggested the possibility of biodegradation of two organic compounds, 2 chlorobenzoic acid and phenol, within the cementitious systems. However, different abiotic factors were implicated in the contaminant losses recorded. Especially, as microbial activity tests did not corroborate contaminant losses in some cases. The possible abiotic factors include irreversible sorption of contaminants to cement matrices, volatilisation and reductive dechlorination of chlorinated organics in the presence of Fe(II) ([Kogbara, 2013](#page--1-0)). It was also shown that biological activity within the cementitious systems did not increase the leachability of metals such as Pb and Zn. Engineering properties such as compressive strength, elastic stiffness and hydraulic conductivity were also not affected [\(Kogbara et al., 2011\)](#page--1-0). Another study in a similar direction indicated the capacity of microbial cells such as Saccharomyces cerevisiae (yeast) and Rhodococcus ruber embedded in magnesium phosphate cement to degrade spiked phenol ([Soltmann et al., 2011\)](#page--1-0). These studies set the stage for this work, which considered bioaugmentation within S/S monoliths and in granulated S/S monoliths for drill cuttings treatment.

S/S and bioremediation (including the addition of microbes with known ability for contaminant degradation $-$ bioaugmentation) has been used individually for the treatment of drill cuttings in several studies ([Ball et al., 2012](#page--1-0)). The reactions of enzymes involved in the biodegradation of lignin (an additive in drilling fluids) have also been identified ([Chen et al., 2011, 2015b](#page--1-0)). However, the combination of S/S and bioremediation for drill cuttings treatment is not well documented. The effect of weathered contaminants in drill cuttings is likely to present a different scenario compared to freshly spiked contaminants used in previous related studies. This work considers the use of NPK fertiliser as an additive since it can potentially serve as a nutrient source alongside compost. Especially since biostimulation of autochthonous microbes through nutrient supplementation has proven to be very effective in TPH reduction in oil-contaminated soils and sludges [\(Kogbara, 2008; Ayotamuno](#page--1-0) [et al., 2009, 2010](#page--1-0)). Activated carbon is also used due to its potential to sorb hydrocarbons and make them amenable to microbial degradation [\(Bakhaeva et al., 2001\)](#page--1-0). These additives are readily available and the technique considered here only requires them in small quantities. Hence, the cycling of nitrogen, phosphorus and carbon from the additives is unlikely to cause serious environmental problems ([Chen et al., 2015c](#page--1-0)).

As opposed to previous related studies, this work considers deployment of a microbial consortium and a Portland cement system to facilitate TPH degradation. It was aimed at investigating the possibility of TPH degradation by microbes embedded within, and mixed with granulated stabilised/solidified drill cuttings. The study also sought to evaluate the effect of the additives mentioned above on TPH biodegradation.

2. Experimental

2.1. Materials for drill cuttings treatment

Drill cuttings: The drill cuttings used were obtained from a private treatment, storage, and disposal facility in the Niger Delta region of Nigeria. It was then characterised for initial concentrations of TPH and metals.

Uncontaminated soil: A silty clay (13% sand, 40% silt and 47% sand) uncontaminated soil was mixed with the drill cuttings since the treatments involved incorporating microbes in a cementitious system. The soil served as a carrier material for the microbes. Its key properties have been reported in a related study ([Kogbara et al.,](#page--1-0) [2016\)](#page--1-0). These include a total heterotrophic bacterial (THB) count of 7.0×10^5 colony-forming units per gramme (CFU/g) and a pH of 4.27.

Compost: Compost prepared from poultry manure was used as additional carrier material for microorganisms, and as a source of nutrients and microbes. It was obtained from the Research Farm of the Rivers State University of Science & Technology, Port Harcourt, Nigeria. The compost had an initial THB count of 5.3 \times 10⁵ CFU/g. The chemical properties of the compost are detailed elsewhere ([Ogbonna et al., 2012](#page--1-0)).

Inorganic fertiliser: A commercially available 20-10-10 NPK fertiliser replaced compost as a source of nutrients in some treatment options.

Activated carbon: Granular activated charcoal (Swanson, USA) served as the source of activated carbon in line with the previously mentioned use.

Cement: An all-purpose 42.5 grade ordinary Portland cement (Dangote Cement Plc, Nigeria) was employed as the binder for S/S treatment of the drill cuttings.

Mixed culture biopreparation: A biopreparation was formulated by mixing equal proportions of pure bacterial cultures ([Ghazali](#page--1-0) [et al., 2004\)](#page--1-0). These were isolated from a mixture of the drill cuttings and uncontaminated soil to ensure they were hydrocarbon degraders. The mixed culture bio-preparation consisted of Pseudomonas spp., Bacillus spp., Enterobacter spp., and Micrococcus spp. It was employed as an inoculum at 10% (v/w) using two levels, visà-vis, 7.6 \times 10¹¹ and 1.52 \times 10¹² CFU/g-drill-cuttings-soil-mixture. The methods for bacterial cultivation, enumeration and identification are described subsequently.

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