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BIOREMEDIATION OF CRUDE OIL CONTAMINATED SOIL USING ORGANIC AND INORGANIC PARTICULATES

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ABSTRACT

The potential of organic and inorganic agents for remediating crude oil contaminated soils was investigated in this study. Kinetic models were also developed to predict the bioremediation rates for these agents. The bioremediating agents (organic and inorganic particulates) used in this study were CB-1, PS-1, AL-1, CA-1, SD-1 and OC-1. The percentage (%) degradation of the bioremediating agents calculated from the total hydrocarbon content (THC) obtained over eight weeks were 78.17%, 80.97%, 95.98%, 96.58%, 97.19% and 95.51% for CB-1, PS-1, AL-1, CA-1, SD-1 and OC-1 respectively. The presence of crude oil degrading micro-organisms in these bioremediating agents (particulates) were found to have contributed to the degradation rates obtained. The particulates which are good bulking agents also served as conducive hosts for these micro-organisms. The bioremediation reactions were mostly of the first order and the rate constants (k) obtained were 0.029 day^{-1}, 0.031 day^{-1}, 0.059 day^{-1}, 0.063 day^{-1}, 0.067 day^{-1} and 0.057 day^{-1} for CB-1, PS-1, AL-1, CA-1, SD-1 and OC-1 respectively. It was however deduced that plant organics (AL-1, CA-1 and SD-1) were better bioremediating agents compared to animal organics (CB-1 and PS-1) and competed favorably with the synthetic bioremediating agent. (OC-1).

1. INTRODUCTION

The contamination of soil by crude oil and petroleum products has become a serious problem to the environment because of the potential consequences on the eco system and human health (Onwurah et al., 2007). Oil spillage and effluents from petrochemical and petroleum industries are a major source of environmental pollution which have degraded the environment and impacted negatively on people living in these areas. The most rational way of decontamination of the environment is by application of methods based mainly on metabolic activities of micro-organisms (Obahiagbon and Aluyor, 2009). The short term effects of these oil spills include animal and plant fatality and disruption of the food chain and ecosystem, while the long term effects include birth defects and alterations in genetics, (Nwilo and Badejo, 2005*)*.

Bioremediation is a treatment process that uses naturally occurring micro-organisms to degrade organic substances into carbon dioxide (CO_2) and water, while bioaugumentation and biostimulation involves specific application of microbes and nutrients respectively to increase degradation rates (Wilson and Jones, 1993). Frankenberger (1992) suggested that bioremediation was a more attractive approach for remediation of crude oil-contaminated soils because it is simple to maintain, applicable over large areas, cost effective and leads to complete destruction of contaminants. However, a major limitation of biodegradation of hydrocarbons is often the availability of nitrogen and phosphorous sources (Prince, 1993). The application of these particulates (bioaugumentation) in remediation of crude oil contaminated soils is preferred to other methods because it is time and cost effective and requires little or no energy requirement. Also, it does not require intensive or skilled labour and it is an environment friendly method as it is a purely biological process.

Biostimulation involves addition of nutrients to balance the carbon/nitrogen (C/N) ratio which boosts the activities of microorganisms. This is usually employed where there is deficiency in competent indigenous microbes in the polluted medium to degrade the contaminants in the soil. Biostimulation by urea and NPK fertilizers (biostimulants) is effective in reducing organic matter and also enhancing bioremediation capabilities of the indigenous microbes (Amenaghawon et al., 2013*)*. Bioremediation can accelerate naturally occurring biodegradation under optimized condition in the presence of suitable microbial population and nutrients (Trindade et al., 2005). Micro-organisms involved in the degradation of crude oil include *Pseudomonas spp* for alkenes/mono-aromatic hydrocarbons and *Mycobacterium spp* for poly-aromatic hydrocarbons (Berry et al., 2006). Adams et al. (2017) studied bioremediation of crude oil contaminated soil using agricultural wastes (rice husks and chicken dung) with results indicating that rice husks removed more petroleum hydrocarbons compared to chicken dung under same conditions. Ibiene et al. (2011) also studied bioremediation of hydrocarbon contaminated soil in the Niger Delta using spent mushroom compost and other organic wastes (cow and chicken droppings). The spent mushroom recorded the highest degradation rate at 99.9% compared to cow dung and poultry droppings with 97.83% and 98.21% degradation rates respectively.

This work seeks to establish various plant and animal particulates as viable bioremediating agents. Experimental results will be used to assess the suitability of a first-order, second order or nth order empirical models in predicting changes in total hydrocarbon concentrations during the treatment process.

2. MATERIALS AND METHODS

2.1. Materials

Crude oil (with specific gravity of 0.86 and viscosity of 4.99 at 40^o C) was obtained from the Nigerian Petroleum Development Company (NPDC) flow station at Ologbo near Benin City, Edo State, Nigeria. CB-1, PS-1, AL-1, CA-1 and SD-1 (organic particulates) were sourced locally in Benin City, while OC-1 (a synthetic absorbent), n-hexane, filter paper (Whatmann no 2), methylene blue solution and NPK fertilizer (50:50:50) were obtained from the Chemical Engineering Laboratory of University of Benin, Benin City, Edo State, Nigeria.

2.2. Bioremediation Process

Loam soil (1 kg) was weighed into seven (7) different buckets and then polluted with 200 g of crude per kg of soil and mixed thoroughly. One of the samples was set aside to serve as the control containing only soil mixed with crude oil. The samples were left for one week to enable the micro-organisms adapt to the new contaminated environment. The particulates (250 g each of CB-1, PS-1, AL-1, CA-1, SD-1 and OC-1) were added to sample buckets 2, 3, 4, 5, 6 and 7 respectively. NPK fertilizer (100 g) was also added to these buckets.

2.3. Analytical Procedure

The total hydrocarbon content (THC) of the samples were determined using a UV spectrophotometer (Jenway 6715 UV/Vis) according to the method of Namkoong et al. (2002). The total microbial count in the samples was recorded using a colony counting chamber according to the method of Mrayyan and Baltikhi (2004).

3. RESULTS AND DISCUSSION

3.1. Effect of Bioremediating Agents on THC of Crude Oil-Contaminated Soil

The efficiency of different plant and animal organics as bioremediating agents was investigated with results showing significant reduction in total hydrocarbon content as seen in Figure 1. Contaminated soil amended with plant particulates (SD-1 and CA-1) had the highest degradation rates of 97.19% and 96.58% respectively. This shows that they are better bioremediating agents compared to CB-1 and PS-1 (animal particulates) with degradation rates of 78.16% and 80.97% respectively. From Figure 1, it was observed that there was a general gradual reduction in total hydrocarbon content over the first seven days owing to the acclimatization time for the micro-organisms. This was followed by a steep decrease in total hydrocarbon content over the next four weeks which coincided with an increase in the microbial counts and thus it could be deduced that the decrease in total hydrocarbon content (THC) was proportional to increase in microbial loads. The plant particulates were better bioremediation agents with degradation rates ranging from 95-97% compared to animal particulates with degradation rates ranging from 50-78% . This is possibly due to the presence of cellulose in plant particulates. Cellulose contains a high energy content which serves as food/host for these micro-organisms. The higher number of micro-organisms in the plant particulates due to the cellulose content thus resulted in higher bioremediation rates. A higher surface area per gram for particulates of plant origin also increased the potency of the plant particulates as bioremediating agents by improving aeration which aids bioremediation reactions (Rhykerd et al., 1999). These particulates also contain different enzymes which act as catalysts by helping in the alteration of bioremediation reactions. The number of enzymes involved in bioremediation reactions has an effect on the rate and duration of bioremediation reactions (Ezeonu et al., 2012).

3.2. Effect of Bioremediating Agents on Total Microbial Count (TMC) of Crude Oil Contaminated Soil

Figure 2 shows a plot of total microbial loads of the bioremediating agents against time. Total microbial counts in soils amended with plant particulates were high in the range of 140,000-160,000 colony-forming units/gram (cfu/g) compared to those with animal particulates which range from 1500-24000 cfu/g as seen in Figure 2. It was observed that higher microbial loads aid faster bioremediation rates. The high microbial load in the plant particulates could be associated with their high cellulose content which have a convenient C:N (carbon/nitrogen) ratio required for crude oil degrading micro-organisms to thrive. (Teng et al., 2010).

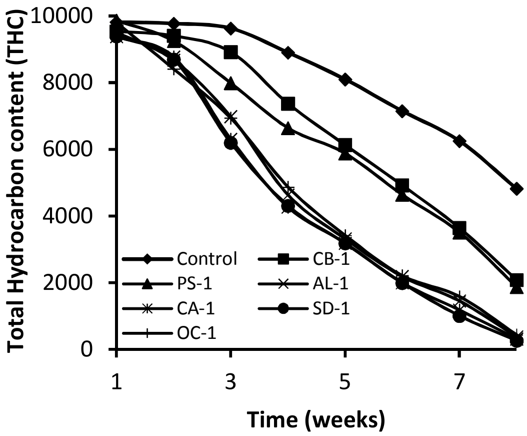

Figure 1: Plot of total hydrocarbon content (THC) against time

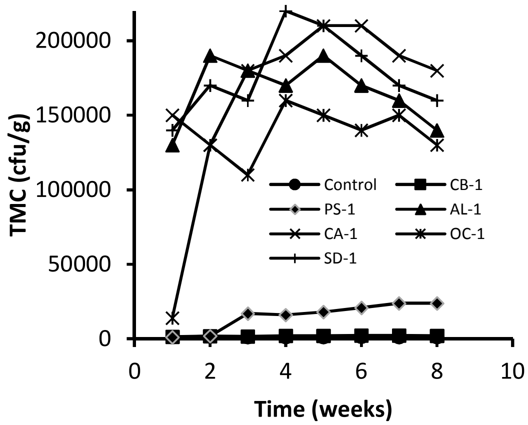

Figure 2: Plot of total microbial count against time

3.3. Kinetic Modeling of Bioremediation Rates

A 1st, 2nd and nth order of reactions were assessed using integral method of analysis to predict the bioremediation rate. In the first order prediction (Equation 1), a plot of $\ln CAo/CA$ vs t as seen in Figure 3 was made, with slope k representing the rate constant.

$$\ln\frac{CA0}{CA} = kt \tag{1}$$

Where C_{A0} and C_A are initial and final THC concentrations and t is the remediation time.

A plot of $1/C_A$ vs t was also made for 2nd order reactions from Equation 2. However, this was a poor fit as low R^2 values were obtained as seen in Table 1.

$$\frac{1}{CA} = \frac{1}{CA0} + kt \tag{2}$$

Making an nth order prediction (Equation 3), a log-log plot of rate of reaction ($-r_A$) vs C_A yielded a straight line graph with the slope representing order of reaction (n) and intercept representing the rate constant (k). From the nth order prediction, negative orders and low R^2 values obtained also indicated they were not good fits as seen in Figure 5.

$$\log(-rA) = \log k + n\log CA \tag{3}$$

The bioremediation reactions were mostly first order reactions as first order models had the highest R^2 values as seen it Table 1. The rate constants (k) obtained from the first order prediction were; 0.029 day^{-1}, 0.031 day^{-1}, 0.059 day^{-1}, 0.063 day^{-1}, 0.067 day^{-1} and 0.057 day^{-1} for CB-1, PS-1, AL-1, CA-1, SD-1 and OC-1 respectively.

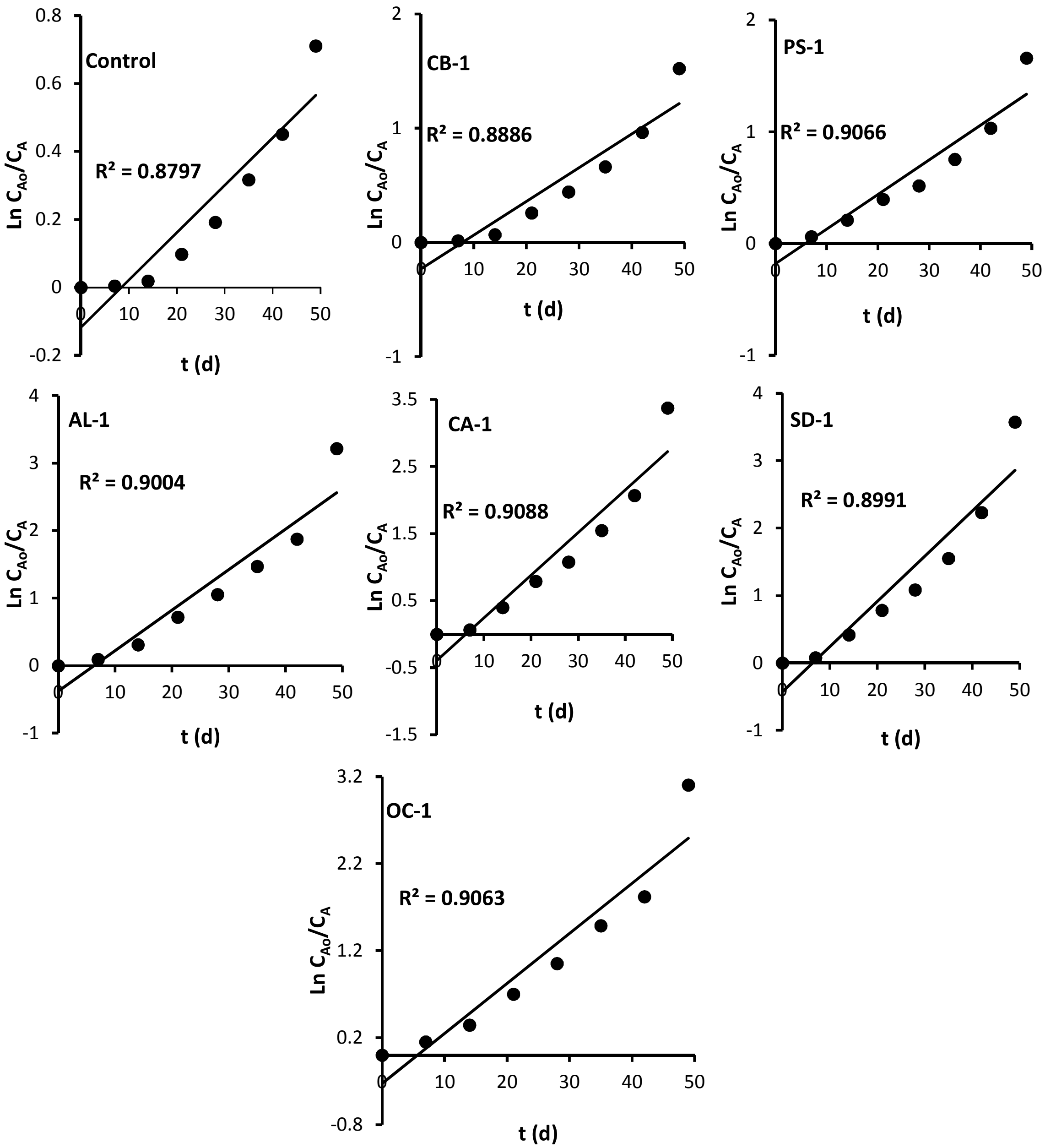

Figure 3: Plots of first order predictions for bioremediation rates

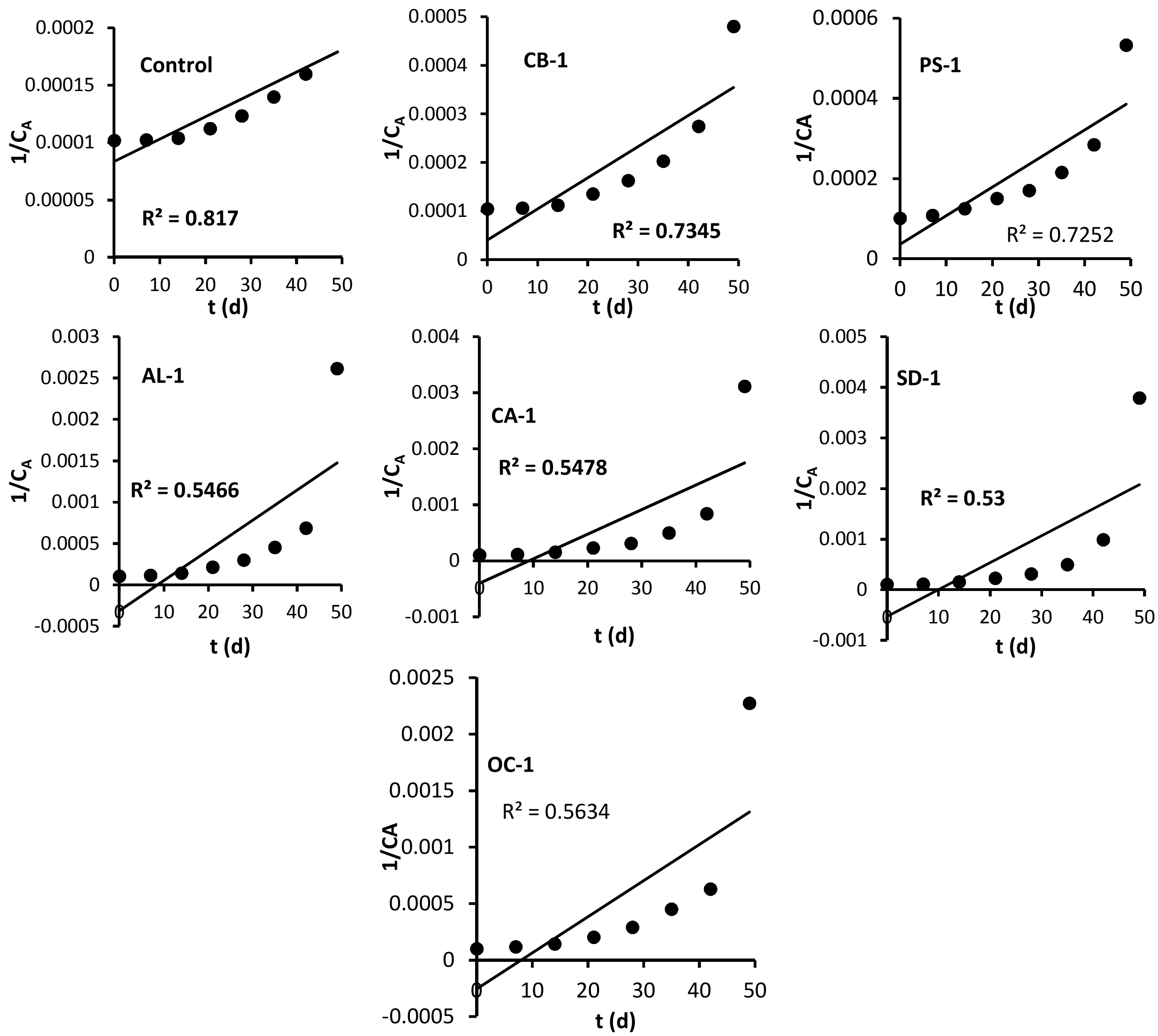

Figure 4: Plots of second order predictions for bioremediation rates

Table 1: R^2 values for 1st, 2nd and nth order predictions

Sample	Reaction order		
	1st	2nd	nth
Control	0.8797	0.817	0.45
CB-1	0.8886	0.7345	0.76
PS-1	0.9066	0.7252	0.87
AL-1	0.9004	0.5466	0.95
CA-1	0.9088	0.5478	0.97
SD-1	0.8991	0.53	0.964
OC-1	0.9063	0.5634	0.956

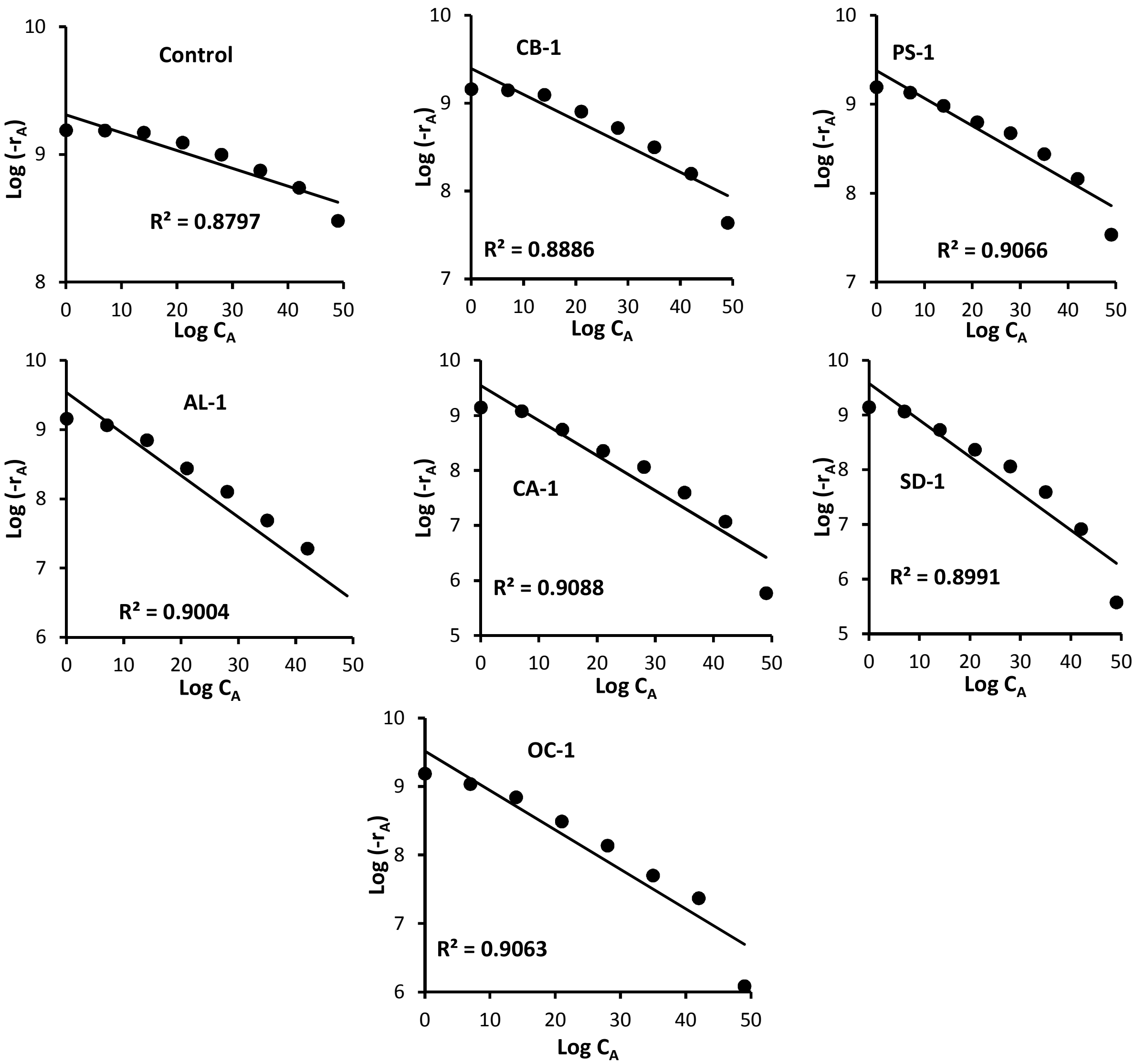

Figure 5: Plots of nth order predictions for bioremediation rates

4. CONCLUSION

In this study, organic and synthetic particulates were used in bioremediation of crude oil contaminated soil and kinetic models proposed for prediction of biodegredation rates. It was deduced from the investigations carried out that crude oil contaminated soils could be remediated using these plant and animal amendments which have varied hydrocarbon degrading abilities proportional to their microbial loads. The total hydrocarbon content reductions were: SD-1(97%), CA-1(96%), AL-1(95%), PS-1(80%), and CB-1(78%) showing favourable competition with OC-1, an inorganic particulate which yielded a 95% reduction. The bioremediation reactions were found to be mostly first order reactions. Prediction of total hydrocarbon content can also be made using proposed kinetic models which help in forecasting the time required for these bioremediation reactions to occur. The use of these particulates for bioremediation is highly beneficial because of its eco-friendly nature as it not only remediates the contaminated soil but also serves as a convenient waste disposal technique.

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6. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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