Diffusion study of municipal solid waste contaminants in compacted lateritic soil treated with bacillus *coagulans*

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**ABSTRACT**

The diffusion of municipal solid waste (MSW) contaminants in compacted lateritic soil-Bacillus *coagulans* (B. *coagulans*) mixture was studied. Diffusion test for single reservoir, decreasing source was adopted. Soil samples were treated with B. *coagulans* at one-third (1/3) pore volume in stepped suspension density of 0, 1.5 × 10^5, 6 × 10^6, 1.2 × 10^7, 1.8 × 10^7 and 2.4 × 10^7 cells/ml, respectively, before compaction. Specimens were prepared at optimum moisture content (OMC) of British Standard heavy (BSH) compaction energy. Cementation reagent containing 3 g Nutrient broth, 20 g urea, 10 g NH_4Cl, 2.12 g NaHCO_3 and 2.8 g CaCl_2 per litre of distilled water was injected by gravity in three (3) cycles of 6 hours interval into the compacted specimens. Thereafter, the specimens were sealed at the top with plastic sheets with small openings for the sequential introduction of water and leachate for 21 days and 69 days, respectively, to simulate field condition. Results obtained show that diffusion testing water content, pH and electrical conductivity within the soil column decreased with depth. Generally, all the cations considered had diffusion coefficient (*D*) and tortuosity factors (τ) values that increased with increase in B. *coagulans* suspension density. However, Mg had value that initially decreased from 5.98 × 10^9 at 0 cells/ml to a minimum value of -4.30 × 10^8 at 1.5 × 10^5 cells/ml and thereafter increased to 1.64 × 10^9 at B. *coagulans* suspension density of 2.4 × 10^9 cells/ml. The apparent *D* and τ values for the anions considered initially decreased to a minimum and thereafter increased. The pore fluid concentration profile for the numerous chemical classes tested showed that the compacted lateritic soil - B. *coagulans* mixture can attenuate K^+ and Cl^- ions in MSW containment application.

**Keywords:** B. *coagulans*, diffusion, diffusion coefficient, lateritic soil, leachate, tortuosity factors

1 INTRODUCTION

Movement of contaminants through waste containment system affect human life when they come in contact with groundwater use for human consumption. The need to study the mechanism of flow of contaminants in a waste containment system is of great significance. Movement of contaminant can be by diffusion, advection, dispersion and sorption. Movement of contaminants by diffusion, which is the focus of the study encompasses the migration of contaminant beginning from points of great chemical potential to points of little or small chemical potential (Shackelford 1988; Eberemu et al., 2013). The use of compacted clay liner and cover system has been reported to aid in mitigating the contamination of ground water in landfill systems (Amadi and Eberemu, 2012; Eberemu et al., 2013; Osinubi et al., 2017, 2019a).

Compacted lateritic soil modified with pozzolanic materials such as bagasse ash, waste wood ash; cement kiln dust, blast furnace slag for waste containment application have been shown to be suitable and have recorded positive results (Eberemu, 2008; Osinubi and Eberemu, 2009; Osinubi et al., 2017; Osim, 2017). Osinubi and Umar (2003) reported that utilization of industrial/agricultural wastes as additives for soil modification is one among several suitable methods of soil modification for different engineering applications such as roads, dam or landfill systems. However, open burning of agricultural wastes and the manufacturing process of industrially manufactured additives such as cement and lime has led to increased generation of carbon (IV) oxide which has negative environmental effect on human life (Osinubi et al., 2019b). Thus, study on sustainable and more eco-friendly mechanism for soil modification such as microbial induced calcite precipitation (MICP) is very important.

MICP is a new technique which utilises bacterial activity for the improvement of the geotechnical properties of soil. This approach makes use of a cementation method that connects natural subsurface
soils using a process called ureolysis. Ureolysis is urea hydrolysis that facilitates calcite precipitation at particle-particle contact in the soil environment (Dejong et al., 2006). Mitchell and Santamarina (2005) reported the relative abundance of microbes in the soil considered that favoured their suitability for use as a sustainable and eco-friendly mechanism for soil improvement. The MICP technique involves urea hydrolysis that facilitates the effective precipitation of calcite in the soil thereby improving the strength and stiffness as well as decreasing the water permeability (Nemati and Voordouw, 2003; Whiffin et al., 2007; Dejong et al., 2010; Harkes et al., 2010; van Paassen et al., 2010; Burbank et al., 2012; Choi et al., 2016; Chi et al., 2017; Osinubi et al., 2019a, 2020a).

The investigation of the performance of lateritic soil-B. coagulans mixture when use as hydraulic barrier material in MSW application was reported by Osinubi et al. (2020b). Compatibility test results recorded reduction in long-term hydraulic conductivity values for the different permeation procedures considered showed the ability of MICP technique improved soil to minimise fluid flow through the compacted soil. Therefore, this study focused on diffusion in B. coagulans treated lateritic soil.

1.1 Theoretical background

Shackelford (1988) defined diffusion as a transport process of chemical species in form of solutions in response to its concentration gradient. The concept of diffusion is well defined by Fick’s first law for one dimensional flow transport defined as

$$J = D_s \frac{\partial C}{\partial x}$$

(1)

Where: $J$ = mass flux (mass transported per unit time per unit cross sectional area) (m/s/mg/l).

$C$ = concentration of the solutes in solution (mg/l).

$x$ = direction of transport.

$D_s$ = constant called free solution diffusion coefficient.

In Fick’s first law, migration of contaminants does not need a hydraulic gradient. Different forms of diffusion are reported in the literature (Eberemu et al., 2013).

Fick’s law for saturated soils is expressed as

$$J = -D^* \theta \frac{\partial C}{\partial x}$$

(2)

Where: $\theta$ = volumetric water content.

The volumetric water content $\theta$ is defined in terms of total porosity $n$ and the soil degree of saturation $S_r$.

$$\theta = nS_r$$

(3)

Also, effective diffusion coefficient $D^*$ defined as

$$D^* = D_s \tau_a$$

(4)

Where: $\tau_a$ = apparent tortuosity factor.

From the laboratory test results, using equation (2), the effective diffusion coefficients $D^*$ can be evaluated mathematically.

From equation (4), the apparent tortuosity factor ($\tau_a$) can be evaluated using their corresponding values of free solution diffusion coefficient.

Shackelford and Daniel (1991) defined the Fick’s second law for time related movement of non-reactive solutes in soil as

$$\frac{\partial C}{\partial t} = D^* \frac{\partial^2 C}{\partial x^2}$$

(5)

The movement of solutes that are dependent on chemical/biochemical reactions are known as reactive solutes, otherwise called non-reactive solutes (Shackelford and Daniel, 1991).

The expression for non-decaying, reactive solutes which depend on reversible sorption reactions during diffusive transport in soil is given as (Bear, 1972; Freeze and Cherry, 1979):

$$\frac{\partial C}{\partial t} = D^* \frac{\partial^2 C}{\partial x^2} \frac{\partial q'}{\partial t}$$

(6)

Where: $q'$ = sorbed concentration of the chemical species defined as

$$q' = \frac{\rho_d}{\theta} q$$

(7)

Where:

$q$ = sorbed concentration expressed as the mass of solute sorbed per mass of soil.

$\rho_d$ = dry density of the soil.

By differentiation of equation (7) in relation to time and replacement in equation (6), Fick’s second law is expressed for reactive solutes as:

$$\frac{\partial C}{\partial t} = D^* \frac{\partial^2 C}{\partial x^2} \frac{\partial q'}{\partial t}$$

(8)

Where: $R_d$ = retardation factor.

This is dependent on reversible sorption reactions through diffusive transport in soil as reported by Bear (1972) as well as Freeze and Cherry (1979).

The retardation factor signifies the comparative rate of fluid flow to the transport rate of a reactive solute is expressed as (Freeze and Cherry, 1979; Shackelford and Daniel, 1991; Shackelford, 1994):

$$R_p = 1 + \frac{\rho_d}{\theta} K_r$$

(9)

$$K_r = \frac{dq}{dC}$$

(10)

Where:

$K_r$ = partition coefficient. When the $q$ against $C$ relationship is linear, $K_r$ is called the distribution coefficient and it depends on the equilibrium concentration in the pore water of the soil.
One analytical solution to Fick’s second law for one dimensional diffusion through saturated soils for a case of decreasing source concentration and for a finite cell length (Crank, 1975; Shackelford et al., 1997) is defined as:

\[
\frac{C(x, t)}{C_0} = \frac{\alpha}{1 + \alpha} \sum_{n=1}^{\infty} \left(1 + \alpha + q_m \alpha^2 \right) \times \exp \left( -\frac{Dq_m^2 \alpha t}{R_s L^2} \right) \cos \left( q_m \left(1 - \frac{x}{L}\right)\right) + \alpha q_m \frac{C_0}{L} \tan(q_m) = -\alpha q_m 
\]

Where: all \(x\) is depth of chemical species in soil specimen \(C_0\) is initial concentration of

\[
\tan(q_m) = -\alpha q_m
\]

Where: \(\alpha\) in the above equation is expressed as:

\[
\alpha = \frac{H_i}{nR_s L}
\]

Where:

\(H_i\) and \(L\) = height of the liquid and the source reservoir and length of the soil specimen, respectively \(n\) = total porosity of the soil specimen.

2 MATERIALS AND METHODS

2.1 Materials

2.1.1 Soil sample

Soil sample was sourced using disturbed sample method from Abagana (68°24’31’’N and 27°52′11″E), Anambra state, Nigeria. The collected sample was air-dried, pulverized and then sieved through several standard sieves for different types of tests with the largest sieve being BS No. 4 sieve (4.76 mm aperture).

2.1.2 Microorganism

The urease positive bacteria used in this research is B. coagulans. American Type Culture Collection (ATCC) classified the micro-organism as ATCC 8038 (ATCC, 2013), rod shaped and spore forming.

2.1.3 Cementation reagent

Cementation reagent composed of 3 g Nutrient broth, 20 g urea, 10 g NH4Cl, 2.12 g NaHCO3 and 2.8 g CaCl2 per litre of distilled water reported by Stocks-Fischer et al. (1999) was adopted in the study.

2.1.4 Bacteria suspension

The inoculated B. coagulans suspension contained 3 g Nutrient broth and 20 g urea per litre of distilled water.

2.2 Methods

2.2.1 Isolation of the bacterium species

Isolation of the microbes from the soil was carried out by means of serial dilution. It entailed the addition of 1 ml of the original bacteria culture (i.e., medium) to 9 ml of sterile water to make a 1:10 dilution; addition of 1 ml of the 1:10 dilution to 9 ml of sterile water produced a 1:100 dilution; etc. The bacteria suspension density/ml decreased by 9/10 for each dilution. Further dilutions were made in ratio of 1:1000, 1:10000 and 1:100000, etc. The isolates were stored at 4°C in nutrient medium prior to classification and characterization.

2.2.2 The culture medium and growth conditions

The method reported by Stocks-Fischer et al. (1999) was adopted for the study.

2.2.3 Diffusion test

Laboratory diffusion test was carried out using the single reservoir decreasing source method (Shackelford, 1989; Shackelford and Daniel, 1991). Polyvinyl chloride (PVC) pipes were used for the experiment. Soil samples were treated before compaction with B. coagulans at one-third (1/3) pore volume (as recommended by Rowshanbakhta et al., 2016) in stepped suspension density of 0, 1.5 × 10^-3, 6 × 10^-3, 1.2 × 10^-3, 1.8 × 10^-3 and 2.4 × 10^-3 cells/ml, respectively. Soil samples were prepared at OMC of British Standard Heavy (BSH) compaction energy to a height of about 10 cm. Cementation reagent was injected into the compacted soil by gravity in three cycles at 6 hours interval. The specimens were sealed at the top with plastic sheet and small opening created for the introduction of the permeant. Water and leachate were sequential used as permeants for 21 days and 69 days, respectively, to simulate field conditions. Diffusion took place under concentration gradient. The soil specimens were extruded and sliced at the end of the test and the concentrations of the pore water of the soil were measured to provide the soil concentration profile. All diffusion tests were performed at laboratory temperature of 25 ± 2°C. At the end of the 90-day diffusion test, the soil specimens were carefully extruded from the PVC pipes and sectioned at regular interval into slices approximately 2.5 cm in thickness. This procedure was carried out to provide data for the concentration profile of specified ions used to determine the effective diffusion coefficient. The concentrations of ions in the pore fluid of the soil were measured using AA500 Atomic Absorption Spectrophotometer.

3 RESULTS AND DISCUSSION

3.1 Characteristics of natural lateritic soil

The natural soil is fine-grained, with natural moisture content of 11.3%. The soil is classified as A-4(2) soil based on American Association of State Highway and Transportation Officials (AASHTO) classification system (AASHTO, 1986) and SC soil based on Unified Soil Classification System (USCS) (ASTM, 1992). A summary of the properties of the natural soil is provided in Table 1. The chemical composition of leachate sample used is presented in
Table 1. Characteristics of natural lateritic soil.

| Property                               | Quantity |
|----------------------------------------|----------|
| Percentage passing No. 200 Sieve       | 35.4     |
| Natural Moisture Content (%)           | 11.3     |
| Liquid Limit (%)                       | 37.5     |
| Plastic Limit (%)                      | 19.3     |
| Plasticity Index (%)                   | 18.2     |
| Specific Gravity                       | 2.62     |
| AASHTO Classification                  | A-4(2)   |
| USC S                                  | SC       |
| Dominant clay mineral                  | kaolinite|
| Colour                                 | Reddish brown |

Table 2. Chemical composition of leachate sample used.

| Measured parameters            | Value  |
|--------------------------------|--------|
| Potassium (mg/l)               | 335    |
| Sodium (mg/l)                  | 60850  |
| Calcium (mg/l)                 | 655    |
| Magnesium (mg/l)               | 25.36  |
| Nitrate (mg/l)                 | 0.154  |
| Sulphate (mg/l)                | 42.35  |
| Chlorine (mg/l)                | 8350   |
| Iron (mg/l)                    | 0.6383 |
| Manganese ((mg/l)              | 0.0093 |
| Copper (mg/l)                  | 0.0109 |
| Zinc (mg/l)                    | 0.0253 |
| Chromium (mg/l)                | 0.0432 |
| Lead (mg/l)                    | 0.0205 |
| pH                             | 6.91   |
| Electrical conductivity, EC (dS/m) | 1.5   |
| Total Dissolve Solids (mg/l)    | 1793   |
| Total Hardness (mg/l)          | 445    |
| Biological Oxygen Demand (mg/l) | 666    |
| Chemical Oxygen Demand (mg/l)   | 1252.08|
| Turbidity (NTU)                | 44.8   |
| Dissolved Oxygen (mg/l)        | 2.0    |

3.2 Diffusion analysis

Diffusion is a transfer mechanism for fluid flow from area of higher concentration or gradient to lower concentration or gradient. The flow of contaminant in a landfill system most times is based on advective-dispersive transport alongside the fractures or micropores as reported by past investigations (Rowe and Booker, 1989; Fitkus et al., 1999; Neretnieks, 2002; Sivakumar, 2011). Also, transport of contaminant may be by diffusive transport from the fractures or micro pores into the adjoining porous soil matrix (Barone et al., 1990; Shackelford, 1991; Liu et al., 2002; Sivakumar, 2011). Monovalent cations (Sodium, Na⁺ and Potassium, K⁺) and divalent cations (Calcium, Ca²⁺ and Magnesium, Mg²⁺) were chosen for the evaluation of the adsorption potential of lateritic soil-B. coagulans mixtures. Tests were carried out to examine their effect in landfill leachate diffusion through the treated soil matrix. Also, anions tested include Chloride (Cl⁻) and Sulphate (SO₄²⁻) commonly found in high concentration in natural aqueous systems and polluted groundwater as well as landfill leachates were chosen. The B. coagulans treated soil specimens were sufficiently saturated with water for 21 days before being replaced with leachate to curtail the solutes mass transport which might arise in the unsaturated state and downward diffusion was used throughout the 90 days period of diffusion.

3.2.1 Effect of pore water content

The effect of pore water content with depth of soil slices is shown in Fig. 1. It was observed that the water content generally decreased with depth of slice which may perhaps be connected with reduction in the degree of saturation of the treated soil with depth. The water content of the first slice is relatively higher than the subsequent slices due to its direct contact with the surface leachate. Although the water content decreased with depth, no difference was observed with increase in B. coagulans suspension density up to 2.4 × 10⁹ cells/ml. However, the lowest or bottom slice in the column recorded marginally higher water content than the other slices apart from the top slice. The recorded increase could be due to the fact that the bottom slice served as a reservoir for all the diffused leachate from the slices above it. Similar trend was reported by Oluremi (2015).

3.2.2 Effect of pore solution pH

The effect of pH with depth of soil slices is shown in Fig. 2. The pH initially increased from 0 to 20 mm depth and thereafter decreased significantly with depth for all B. coagulans suspension density with the exception of the natural soil which showed a decreasing trend from 0 to 100 mm depth of slice. The initial increase in alkalinity of the treated soil with depth could probably be associated with urea hydrolysis produced from the reaction pathway of urease enzymes released by the microbes. Generally, the effect of B. coagulans suspension density on the treated soil tends to cause a reduction in the pH of the soil with exception of few cases. The decrease in pH values with depth is probably due to the replacement of protons H⁺ up on the surfaces of the clay minerals with the metallic cations in the leachate. The final pH values for soil-leachate mixtures are 5.12, 4.93, 5.02, 5.24, 5.09 and 4.99 for 0, 1.5 × 10⁸, 6 × 10⁸, 1.2 × 10⁹, 1.8 × 10⁹ and 2.4 × 10⁹ cells/ml, respectively, which suggest an acid environment. Similar trend was observed by Interno et al. (2015) and Osim (2017). The reduction in
alkalinity of the soil (marked by decrease in pH) with depth for lateritic soil - B. coagulans mixture favours its use as liner in MSW system. This is consistent with findings reported by Mitchell (1993) that pH is a factor that controls the adsorption of cations in soil for use in any engineering system. Also, Abollino et al. (2003) and Angelica et al. (2015) reported that adsorption of metal decreases with increase in the pH of the solution.

For Na⁺, apparent $D^*$ increased linearly. Values increased from $-8.6 \times 10^8$ m²/s at 0 cells/ml to peak value of $1.3 \times 10^9$ m²/s at 2.4 × 10⁹ cells/ml. The $\tau_0$ values also increased with increase in B. coagulans suspension. The $\tau_0$ values of -0.644, -0.597,-0.455, 0.535,0.696 and 0.850 were recorded for B. coagulans suspension density of 0, 1.5 × 10⁶, 6 × 10⁶, 1.2 × 10⁷, 1.8 × 10⁷ and 2.4 × 10⁷ cells/ml, respectively.

In the case of Ca²⁺, the apparent $D^*$ recorded a trend of increase with increase in B. coagulans suspension. Values of $-1.3 \times 10^8$, $-9.27 \times 10^{-10}$, $1.16 \times 10^{-9}$, $1.54 \times 10^{-9}$, $-3.18 \times 10^{-9}$ and $5.08 \times 10^{-9}$ m²/s were recorded for B. coagulans suspension density of 0, 1.5 × 10⁶, 6 × 10⁶, 1.2 × 10⁷, 1.8 × 10⁷ and 2.4 × 10⁷ cells/ml, respectively. The $\tau_0$ values between -16.43 and 6.41generally increased for B. coagulans suspension density from 0 cell/ml up to 2.4 × 10⁷ cells/ml.

Table 1: Effect of pore solution concentrations on electrical conductivity

| Depth in soil (mm) | Electrical conductivity (dS/m) |
|-------------------|-------------------------------|
| 0 /ml            | 0.2                           |
| 1.5E+8/ml        | 0.4                           |
| 6.0E+8/ml        | 0.6                           |
| 12.0E+8/ml       | 0.8                           |
| 18.0E+8/ml       | 1.0                           |
| 24.0E+8/ml       | 1.2                           |

3.1.3 Effect on pore solution electrical conductivity

The effect of electrical conductivity (EC) on soil slices after diffusion with depth is shown in Fig. 3. The electrical conductivity generally decreased with depth of slices. In the case of natural soil, marginal changes were observed in EC values with depth of soil slices. Lateritic soil-B. coagulans mixture significantly influenced the EC of the soil. Ghosh et al. (2013) reported that EC is related to the increase in the degree of salinity of the solution formed from the reaction of leachate with soil. The recorded changes may be associated with the interaction between ions in the clay structure and the leachate. Ion exchange may be responsible for such behaviour.
organic chemical pollutants (Raymond, 2001). Geochemical processes, formation of calcite precipitate, and other microbial interaction with the soil could be responsible for the lower attenuation capacity of the treated soil (Osimubi et al., 2020b).

The effects of varied contaminant solutes on diffusion coefficients was also reported by Robinson and Stokes (1959), Jost (1960) and Lerman (1979). The implication of the recorded negative values indicate that B. coagulans treated lateritic soil has lower capacity to attenuate the identified cations and anions with negative $D^*$ and $\tau_a$ values. The higher the values of $D^*$ and $\tau_a$, the higher the contaminants migration. However, not all cases recorded negative values of $D^*$ and $\tau_a$ for the treated soil. It is presumed that pH variation, environmental factors and high leachate concentration may be responsible for the reduced potency of lateritic soil-B. coagulans mixture to minimize the contaminant flow.

In general, the sorption capacity of cation and anions can be explained in terms of their respective apparent $D^*$ and $\tau_a$. All the cations considered recorded a general trend of increase for apparent $D^*$ and $\tau_a$ with increase in B. coagulans suspension density with the exception of Mg$^{2+}$ that initially decreased from $5.98 \times 10^{-9}$ m$^2$/s at 0 cells/ml to a minimum value of $4.30 \times 10^{-8}$ m$^2$/s at 1.5 $\times$ 10$^8$ cells/ml and thereafter increased to $1.64 \times 10^{-9}$ m$^2$/s at B. coagulans suspension density of 2.4 $\times$ 10$^9$ cells/ml. In the case of anions, apparent $D^*$ and $\tau_a$ initially decreased and thereafter increased.

Fig. 4. Variation of pore solution concentration with depth for potassium (K$^+$).

Fig. 5. Variation of pore solution concentration with depth for sodium (Na$^+$).

Fig. 6. Variation of pore solution concentration with depth for calcium (Ca$^{2+}$).

Fig. 7. Variation of pore solution concentration with depth for magnesium (Mg$^{2+}$).

Fig. 8. Relationship of pore solution concentration with depth for sulphate (SO$_4^{2-}$).

Fig. 9. Variation of pore solution concentration with depth for chloride (Cl$^-$).
Fig. 10. Variation of apparent diffusion coefficient of lateritic soil with B. coagulans suspension density.

Fig. 11. Variation of Tortuosity factor of lateritic soil with B. coagulans suspension density.

4 CONCLUSIONS

Results obtained show that diffusion testing water content, pH and electrical conductivity within the soil column decreased with depth. Generally, all the cations considered had $D^+$ and $\tau_v$ values that increased with increase in B. coagulans suspension density. However, the value for Mg had value that initially decreased from $5.98 \times 10^{-3}$ m$^2$/s at 0 cells/ml to a minimum value of $-4.30 \times 10^{-3}$ m$^2$/s at 1.5 x 10$^9$ cells/ml and thereafter increased to $1.64 \times 10^{-3}$ m$^2$/s at B. coagulans of 2.4 x 10$^9$ cells/ml. The apparent $D$ and $\tau_v$ values for the anions considered initially decreased to a minimum and thereafter increased. The pore fluid concentration profile for the numerous chemical classes tested showed that the compacted lateritic soil-B. coagulans can attenuate $K^+$ and $Cl^-$ ions in an MSW containment application.

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