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Modeling the Diffusion Rate of Biogenic Gases Produced During Microbial Enhanced Oil Recovery

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Abstract*—Different transport mechanisms occur within the subsurface region. To have a vast comprehension of these transport modes, researchers, scientists, scholars etc. are charged with the responsibility of providing concrete answers to lingering subsurface mass transfer questions. In this study, the prediction of diffusion rates of biogenic gases for a MEOR subjected core is investigated using a developed predictive model, which when compared to results from experiments conducted showed a great level of precision. The 50 -50 mixtures of both microbes for the MEOR investigation were also found to reduce the formation permeability and heavy crude viscosity, all of which constitute mobility improvement techniques.

***Index Terms*—Biogenic gases, Diffusion, Microbes.**

I. INTRODUCTION

The concept of microbial introduction for oil enhancement dates back to more than 60yrs years ago, with a varying degree of success in field trials [1]. The introduction of a technology offering a non-expensive and environmental friendly method for oil enhancement by the activation of microbes both in-situ and ex-situ for metabolite production has now been a pedestal of investigation in recent times [2]. Donaldson et al reported that the range of metabolic products from microbial activity on crude oil is very broad, depending on the prevailing conditions, the presence of nutrients available for cell metabolism and the choice of microbe selected for the investigation of its interaction with the crude oil [3]. In general terms, metabolites could be gases (methane, hydrogen, Carbon dioxide, Hydrogen sulphide), Carboxylic acids (formic, acetic, valeric), solvents (alcohols, ketone, aldehydes), polymers (proteins, polysaccharides), surface-active compounds (poly anionic lipids) and many other compounds ranging from simple to very complex macromolecules [4]. Biogases are products of the biological breakdown of organic matter in the absence of oxygen. These biogases when produced subsurface have the capacity to re-pressurize the reservoir as well as reducing heavy crude viscosity. Some of these biogenic gas producers include Bacillus , pseudomonas and methanogens that produce about 60% methane and 40% carbon dioxide[5]. Foglar discussed the fundamentals of gas diffusion, molar flux and presented the gas mole balance in terms of molar fluxes for a rectangular and cylindrical coordinate [6]. Using Fick's law, the presentation of a comprehensive equation describing the flow, reaction and the gas diffusion was established.

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The kinetic theory was then used to explain the diffusion rate by introducing Boltzmann constant and assuming that mean kinetic energies of the gases are of the same Kelvin temperature. For a system of diffusion through cell membranes, the rate of diffusion was affected by a number of cell properties, the diffusing molecule and surrounding solution [7].

II. MATERIALS AND METHODS

A. Core sample and Crude oil

A synthetic core with dimension (14cm ×14cm×9cm) was used as a representation of a reservoir for a Niger-Delta formation, with a cement-sand ratio of about 1:25, withdrawn from a depth of 900ft. the physical properties of the oil is shown in Table 1 below. The crude sample was gotten from a well located at Ebocha field of Nigerian Agip Oil Company.

Table 1. Oil properties

P_R (psi)	3717
P_b (psi)	1048
(cp)	19
API gravity (^{o}API)	19.4
B_w (bbl/stb)	1.062
Z	0.915
S.G	0.567
T_r	136
R_s	116

B. Flooding Agent and Nutrient

Distilled water was used as a flooding agent, autoclaved at a temperature of about 12oC, without altering then chemical properties of the water to avoid interference with oil or nutrient composition. The nutrient was prepared by dispersing 13g of "E" broth powder in 1litre deionized water. The mixture was heated to dissolve the powder properly and then sterilized by autoclaving at 121oC for 15mins. Table 2 shows the nutrient composition. Nutrient pH was found to be 7.4 ± 0.2.

Table 2. Nutrient composition

Composition	Concentration (g/cm^3)
Beef extract	1.0
Yeast extract	2.0
Peptones	5.0
Sodium chloride	5.0

C. Microbe selection

Two variants of microbes were selected for this study, Bacillus Substilis and Pseudomonas Aerogenosa.

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The Persian type culture method was adopted for the culture of these microbes (PTC). Both were cultured on a liquid growth medium, presented in table 3. These cultures were centrifuged at 2000rpm for 30mins and collected at a stationary state, suspended in autoclaved in distilled water. The microbial suspension was placed on a magnetic stirrer at room temperature for 8mins with solution centrifuged and washed again with water.

Table 3 composition of liquid growth media for A and B

Composition	A	B
NH_4Cl	2.0	2.0
Glucose	5.0	3.0
Peptone	10	10
Meat infusion	5.0	5.0
Na_2H	2.0	2.0
Sodium Chloride	0.3	0.25

D. Description of Apparatus

Hand lens, Pumps and flowline

A magnifying hand lens was use to magnify the bioreactor for a better capture of the process scanner. The hand lens was spherical in shape with a diameter of 6cm and glass convex lens. An injection syringe was used for bacteria injection. The total volume of the Syringe was 45ml and the rate of its injection was manually controlled. All system tubing of the flowline was 1/7 in OD PTFE Teflon. The choice of this small sized flow line was to attain approximate flow line pore volume proportions. Image analyzer Glass micro models have been used regularly as tools for qualitative study, some researchers have employed image analyzing technique for the saturation measurement in micro models. A computerized image processing system was used to monitor the effects of bacteria on the water- oil system as well as the core. The basic system consists of a Pentium IV PC, scanning machine which allows us to capture higher quality images at time intervals and a 14'' color monitor.

The procedure adopted for the experiment is outlined as follows:

1. A bioreactor sterilized with xylene and moist dried with self-indicating silica gel.
2. Core was then saturated with distilled water until the connate water saturation was reached.
3. It was the saturated with crude oil to the point of initial water saturation.
4. The bioreactor was placed in an incubator in a steady room temp. air was then injected at a P of 28psi until no more fluid was produced in the effluent flow line. Water was pumped at 28psi with until no more fluid was produced in the effluent flow line.
5. 1.7% porev9olume of the mixture of bacterial- nutrient (50-50) was injected into the model.
6. The system was incubated for a period of 48hrs (shut-in period) at a steady ambient temperature of 25°C.
7. Following the shut in period, the system was then flooded until no oil was produced at the effluent.
8. The same step was followed using pseudomonas and a mixture (50-50) of *Bacillus Substilis* and *Pseudomonas Aerogenosa*.

Model development

Assumptions

- No reaction between the nutrients.
- Mass transfer occurs only by diffusion.
- Negligible gas influx.
- Gas composition is homogenous.
- Diffusion occurs only at the incubation stage.
- Diffusion occurs predominantly on the x axis

Bulk volume for a rectangular element $V_b=\Delta x\Delta y\Delta z$

Law of mass conservation stated as;

[Rate In] – [Rate Out] + [Rate Of Generation] = [Rate Of Accumulation]

The gas mass flow rate is given as; $F_g = A_c W_g$

In a 3D representation,

$$F_{gx} = A_x W_g \quad (1)$$

$$F_{gy} = A_y W_g \quad (2)$$

$$F_{gz} = A_z W_g \quad (3)$$

Mass balance through a differential element Δx

$$\left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_x - \left[\begin{matrix}\text{rate}\\ \text{out}\end{matrix}\right]_{X+\Delta x} + \left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_Y - \left[\begin{matrix}\text{Rate}\\ \text{out}\end{matrix}\right]_{Y+\Delta y} + \left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_z + \left[\begin{matrix}\text{Rate}\\ \text{out}\end{matrix}\right]_{Z+\Delta z} + \left[\begin{matrix}\text{Rate}\\ \text{of}\\ \text{generation}\end{matrix}\right] = \left[\begin{matrix}\text{Rate}\\ \text{of}\\ \text{accumulation}\end{matrix}\right] \quad (4)$$

The gas flow rate is given as; $F_g = A_c W_g$ (5)

Rate of gas generation = $V_b r_g$ (6)

Rate of gas accumulation = $\frac{V_b\,\partial C_g}{\partial t}$ (7)

$$[A_x W_{gx}]_x - [A_y W_{gx}]_{x+\Delta x} + [A_y W_{gy}]_y - [A_y W_{gy}]_{y+\Delta y} + [A_z W_{gz}]_z - [A_z W_{gz}]_{z+\Delta z} + V_b r_g = V_b \frac{\partial C_g}{\partial t} \quad (8)$$

Considering unit volume,

$$\left[\frac{W_{gx}}{\Delta x}\right]_x - \left[\frac{W_{gx}}{\Delta x}\right]_{x+\Delta x} + \left[\frac{W_{gy}}{\Delta y}\right]_y - \left[\frac{W_{gy}}{\Delta y}\right]_{y+\Delta y} + \left[\frac{W_{gz}}{\Delta z}\right]_z - \left[\frac{W_{gy}}{\Delta z}\right]_{z+\Delta z} + r_g = \frac{\partial C_g}{\partial t} \quad (9)$$

Taking limits as $\Delta x, \Delta y$ and Δz tends to zero

$$-\frac{\partial W_{gx}}{\partial x} - \frac{\partial W_{gy}}{\partial y} - \frac{\partial W_{gz}}{\partial z} + r_g = \frac{\partial C_g}{\partial t} \quad (10)$$

The corresponding balance on a cylindrical coordinate is;

$$-\frac{1}{r}\frac{\partial (r W_{gr})}{\partial r} - \frac{\partial W_{gz}}{\partial z} + r_g = \frac{\partial C_g}{\partial t} \quad (11)$$

To study the diffusivity of the biogas with time, the nutrient consumption rate must first be determined and thus a mole balance of produced biogenic gas is equated to the rate of nutrient consumption. The mass balance for nutrient consumption rate is given below assuming a steady state consumption of nutrient by microbes.

Recall,

$$\left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_x - \left[\begin{matrix}\text{rate}\\ \text{out}\end{matrix}\right]_{X+\Delta x} + \left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_Y - \left[\begin{matrix}\text{Rate}\\ \text{out}\end{matrix}\right]_{Y+\Delta y} + \left[\begin{matrix}\text{Rate}\\ \text{in}\end{matrix}\right]_z + \left[\begin{matrix}\text{Rate}\\ \text{out}\end{matrix}\right]_{Z+\Delta z} + \left[\begin{matrix}\text{Rate}\\ \text{of}\\ \text{generation}\end{matrix}\right] = \left[\begin{matrix}\text{Rate}\\ \text{Of}\\ \text{Accumulation}\end{matrix}\right] \quad (12)$$

Assuming diffusion occurs predominantly on the y axis and also assuming no nutrient accumulation

$$[A_c W_N]_y - [A_c W_N]_{y+\Delta y} + 0 = 0 \quad (13)$$

Considering unit volume,

$$\left[\frac{W_N}{\Delta y}\right]_y - \left[\frac{W_N}{\Delta y}\right]_{y+\Delta y} = 0 \quad (14)$$

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Taking limits as $\Delta y = 0$

$$\left[\frac{W_N}{\Delta y}\right]_y = 0 \quad (15)$$

But

$$W_A = -D_A \frac{\partial C}{\partial y} \quad (16)$$

And

$$W_N = -D_e \frac{\partial C_A}{\partial y} \quad (17)$$

Differentiating the above with respect to y, we have

$$\frac{\partial W_N}{\partial y} = -D_e \frac{\partial^2 C_A}{\partial y^2} \quad (18)$$

$$D_e \frac{\partial^2 C_A}{\partial y^2} = 0 \quad (19)$$

Boundary conditions

Y=0, C_A=C_{AO}, t=0

Y=L, C_A=0, t > 0

Y=δ, t > 0, C_A =C_{Ai}

Integrating Eq. (19) we have

$$\frac{\partial C_A}{\partial y} = k_1 \quad (20)$$

$$C_A = k_1 y + k_2 \quad (21)$$

Using boundary conditions to eliminate k_1 and k_2 we obtain

At y=0, $C_A = C_{Ao}$; hence

$$k_2 = C_{A0} \quad (22)$$

$$C_A = k_1 y + C_{Ao} \quad (23)$$

$$C_{Ai} = k_1 \delta + C_{Ao} \quad (24)$$

Dividing (23) by (24)

$$\frac{C_A - C_{Ao}}{C_{Ai} - C_{Ao}} = \frac{y}{\delta} \quad (25)$$

Solving for C_A in the above

$$C_A = C_{Ao} + (C_{Ai} - C_{Ao})\frac{y}{\delta} \quad (26)$$

Differentiating the above with respect to y, we have

$$\left[\frac{\partial C_A}{\partial y}\right] = \frac{C_{Ai} - C_{Ao}}{\delta} \quad (27)$$

From (17), W_N can be written as

$$W_N = -D_e \frac{\partial C_A}{\partial y} = -D_e \frac{C_{Ai} - C_{Ao}}{\delta} \quad (28)$$

Applying mass balance for the generation of biogas with no influx and production

$$\left[\text{Rate in}\right]_x - \left[\text{Rate out}\right]_{X+\Delta x} + \left[\text{Rate in}\right]_Y - \left[\text{Rate out}\right]_{Y+\Delta y} + \left[\text{Rate in}\right]_Z + \left[\text{Rate out}\right]_{Z+\Delta z} + \left[\begin{matrix}\text{Rate}\\ \text{of}\\ \text{generation}\end{matrix}\right] = \left[\begin{matrix}\text{Rate}\\ \text{of}\\ \text{accumulation}\end{matrix}\right]$$

$$[0 - 0 + r' V_b] = V_b \left[\frac{\partial C_A}{\partial t}\right] \quad (29)$$

Considering unit volume

$$[r'] = \left[\frac{\partial C_A}{\partial t}\right] \quad (30)$$

Rate of nutrient diminished = rate of generation of biogas, hence:

$$[r'] = -[W_N] = \frac{\partial C}{\partial t} \quad (31)$$

Solving (29) yields

$$\left[\frac{\partial C_g}{\partial t}\right] = -D_e \frac{y_{Ai} - y_{Ao}}{\delta} \quad (32)$$

Expressing diffusion rate in terms of mole fraction,

$$\left[\frac{\partial C_A}{\partial t}\right] = -D_e C_t \frac{y_{Ai} - y_{Ao}}{\delta} \quad (33)$$

Where C_t is the total molar concentration

The above is the model for biogas diffusion rate

Solving for gas concentration profile by integrating (32) we obtain

$$C_g = D_e [C_{Ai} - C_{Ao}] \frac{1}{\delta} \quad (34)$$

Effective diffusivity determination, D_e

$$D_e = \frac{D_{AB} \emptyset_p \sigma_c}{\tau} \quad (35)$$

D_{AB} is the Knudsen diffusivity constant =10^{-2}cm^2/sec

τ is the actual distance between A and B/ the shortest distance between them

$$\tau = \frac{2L}{\sqrt{2}l} = \sqrt{2} = 1.414$$

$$\emptyset = \frac{\text{Pore volume}}{\text{Bulk volume}} = \frac{264.6}{1764} = 0.15$$

$$\sigma_c = \frac{A_2}{A_1} = 1.0$$

Substituting the above values we obtain

$$D_e = 1.0608 \times 10^{-3} \text{cm}^2/\text{sec}$$

III. RESULTS AND DISCUSSION

Three experiments were conducted in order to study the transport process of biogas produced in-situ and a significant reduction in viscosity was recorded. This phenomenon could not only have been due to reduction in average molecular weight of the heavy crude component, but traceable to biogas production. Gases, when dissolved in crude oil also have the capacity to reduce viscosity of oil. The gases produced were suspected to be carbon dioxide and hydrogen based on the chemistry of the two microbes used of the oil. The initial gas concentration was 0g/cm^3. From the experiments conducted, gas production was strained to amount of nutrient injected as 0.1cm^3 of gas produced was subjected to 1lb-mass of nutrient injected. The microbe diffusion coefficient was estimated to be 5.1cm^2/min. Table 4 shows the experimental results of diffusion parameters. A 50 -50 mixture of *B. substilis* and *P. aeroginosa* produced the greater amount of biogases, thus reducing the oil viscosity at a higher percentage when compared to a single bacteria solution.

Table 4 show data from mathematical model developed for the prediction of biogas diffusion rate.

Parameters	Core 1(*B.substilis*)	Core 2 (*P.aeroginosa*)	Core 3 (50-50 mixture of both)
Nutrient concentration (g/cm^3)	0.131	0.138	0.195
Injected bacteria concentration (cells/cm^3)	0.8×10^{-9}	0.8×10^{-9}	0.8×10^{-9}
Cumulative gas produced (cm^3)	0.81	0.91	1.18
Injected nutrient (cm^3)	30	30	30

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From analysis, the diffusion rate value ranges from 0 – 1.63 × 10^{-5}g/sec-cm^3 when using the developed mathematical model while that from experimental results ranges from 0 - 1.59 × 10^{-5}g/sec-cm^3. Table 5 shows the diffusion data using the developed model while that from experimental result is as shown in the table 6.

Table 5 Model results for biogas diffusion

Y (cm)	t × 10^2 (sec)	y/L	C_{Ai} (g/cm^3)	C_{Ai}/C_{Ao}	C_{Ai} - C_{A0} (g/cm^3)	C_g × 10^{-3} (g/cm^3)	N_g × 10^{-5} (g/cm^3)
1	0	0.11	1.195	1	0	0	0
2	0.5	0.22	0.145	0.74	0.05	1.33	2.65
3	1	0.33	0.116	0.59	0.079	2.79	2.80
4	1.5	0.44	0.097	0.5	0.098	3.9	2.60
5	2	0.56	0.087	0.45	0.108	4.58	2.29
6	2.5	0.67	0.077	0.4	0.118	5.22	2.09
7	3	0.78	0.067	0.34	0.128	5.82	1.94
8	3.5	0.89	0.057	0.29	0.138	6.40	1.83
9	4	1	0.057	0.29	0.138	6.50	1.63

Table 6 Experimental data for biogas diffusion

Y (cm)	t × 10^2 (sec)	y/L	C_{Ai} (g/cm^3)	C_{Ai}/C_{Ao} (g/cm^3)	C_{Ai} - C_{A0} (g/cm^3)	C_g × 10^{-3} (g/cm^3)	N_g × 10^{-5} (g/cm^3)
1	0	1	1.195	1	0	0	0
2	0.5	0.76	0.148	0.76	-0.047	1.25	2.49
3	1	0.61	0.119	0.61	-0.076	2.69	2.68
4	1.5	0.51	0.100	0.51	-0.095	3.78	2.52
5	2	0.46	0.090	0.46	-0.105	4.46	2.23
6	2.5	0.41	0.080	0.41	-0.115	5.08	2.03
7	3	0.36	0.070	0.36	-0.125	5.68	1.89
8	3.5	0.30	0.060	0.30	-0.135	6.27	1.79
9	4	0.30	0.060	0.30	-0.135	6.30	1.59

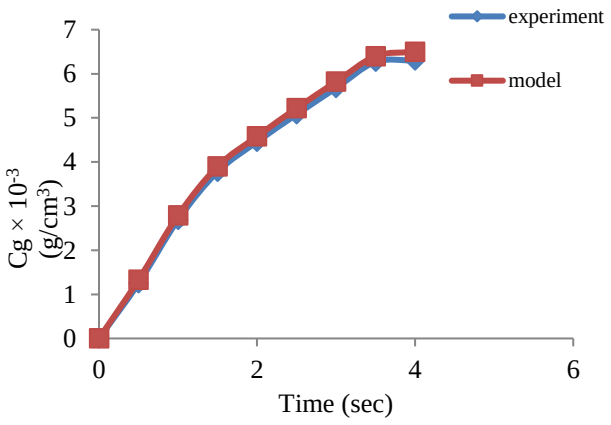

Fig 1. Plot of biogas concentration against time

The above figure shows the model and experimental trend of biogas concentration against time with biogas concentration increasing considerably with time. As nutrient injection rate increased, gas production increased. Diffusion rate peaked at layer 3 and gradually decreases as the gas concentration increased. A graph of diffusion rate vs nutrient concentration ratio shows a linear relationship with a negative slope to a C_A/C_{Ao} value of 0.74. This value represents the K-value as reported by Tiem [7]. At K-value of 0.74 and above, the diffusion rate vs C_A/C_{Ai} followed a parabolic pattern having a negative slope depicting a decreasing order of nutrient concentration. This is represented in fig 4. A dimensionless parameter that could be used to estimate diffusion rate is shown in fig 5.

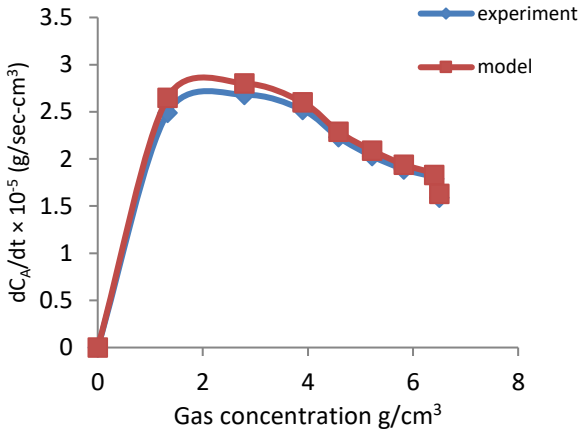

Fig 2. Biogas diffusion rate against biogas concentration

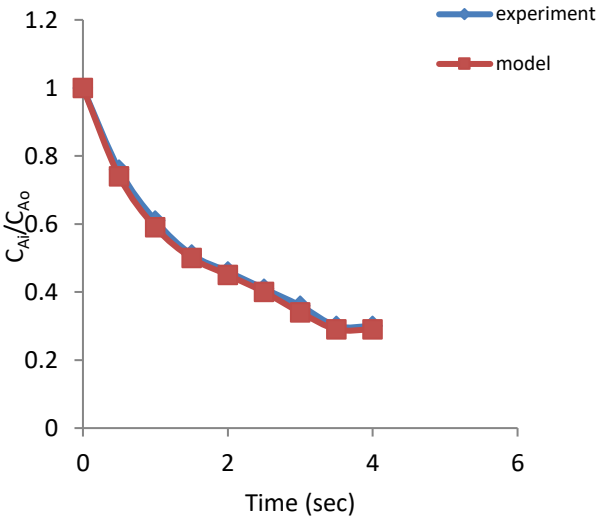

Fig 3 Plot of C_A/C_{Ai} against time

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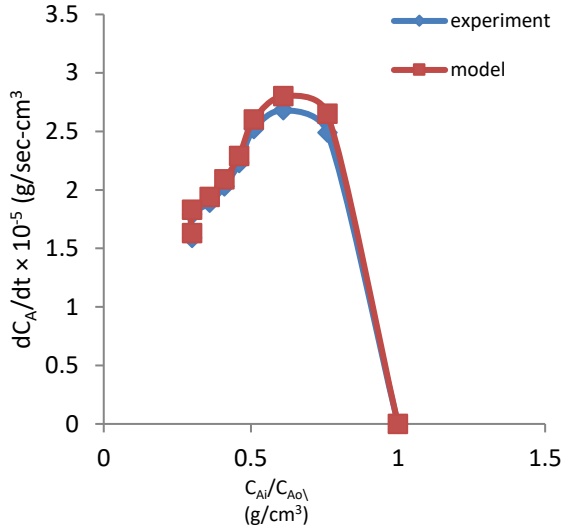

Fig 4 Plot of diffusion Rate against C_A/C_{Ai}

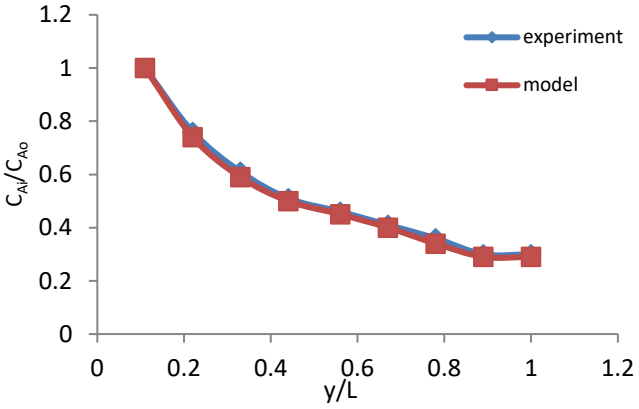

Fig 5 Dimensionless diffusion plot

A. *Viscosity reduction*

Crude oil recovered from the MEOR process was tested for viscosity with a remarkable reduction achieved by all three systems (A, B and A+B). The mixture bacteria solution was observed to record the highest reduction in viscosity. Oil viscosity was reduced from an initial value of 19cp to 12.1cp, 11cp and 9.86cp for B. subtilis, P. aeroginosa and the mixture bacteria solution respectively. This high reduction in viscosity confirms a trace level of biogas production in a bacteria mixture of B. sublitis and P. aeroginosa.

Table 7 Percentage reduction of viscosity by each bacteria system.

sample	Bacteria solution	μ_o (cp)	μ_f (cp)	% reduction
Core A	B. substilis	19	12.2	35.8
Core B	P. aeroginosa	19	11	42.1
Core C	Mixture of both (50 - 50)	19	9.86	48.1

B. *Permeability reduction*

Bio clogging could be achieved by using polymer producing bacteria. Bacteria choice and crude oil composition determines the type of by-products produced. A small reduction was noticed on the permeability values after the microbial activity on the cores originally saturated with oil and water. Percentage reduction in formation permeability is shown Table 8. With the mechanism of viscosity reduction and biogas production, 32% of oil was recovered.

Table 8. The percentage reduction in permeability values

Sample	Bacteria solution	K_i (mD)	K_f (mD)	% reduction
Core A	B. substilis	340	313	7.9
Core B	P. aeroginosa	340	310	8.8
Core C	Mixture of both (50 - 50)	340	306	9.8

IV. CONCLUSION

Gas production by microbes consist one of the essential mechanisms through which oil production can be enhanced. Results from the above figures have shown the reliability of the diffusion model when correlated with diffusion values deuced from experimental results. The 50 – 50 microbial mixtures posed a higher reduction in heavy crude viscosity. Conclusively, for every biogas producing microbe, gas concentration increases with increasing nutrient consumption and thus increased diffusion rate is achieved.

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