

Microbial selective plugging and enhanced oil recovery

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SUMMARY

The ability of indigenous populations of microorganisms in Berea sandstone to improve the volumetric sweep efficiency and increase oil recovery by in situ growth and metabolism following the injection of nutrients was studied. Cores of differing permeabilities connected in parallel without crossflow and slabs of sandstone with differing permeabilities in capillary contact to allow crossflow were used. The addition of a sucrose-nitrate mineral salts medium stimulated the growth and metabolism of microorganisms in the sandstone systems. This resulted in a preferential decrease in permeability in the core or slab with the higher initial permeability, diverted flow into the lower-permeability core or slab and improved the volumetric sweep efficiency. Injectivity into the slab with the lower initial permeability in the crossflow system increased during subsequent nutrient injections. Thus, microbial selective plugging does occur in laboratory systems that have the complex flow patterns observed in petroleum reservoirs without losing the ability to inject fluids into the formation. In situ microbial growth and metabolism increased oil recovery 10 to 38% of the original oil in place. Biogenic gas production accompanied oil production, and much of the gas was entrained within the produced oil suggesting that gas production was an important factor leading to increased oil recovery. Quantitation of the amount of phospholipid in the core confirmed that microbial growth preferentially occurred throughout the core with the higher initial permeability. These data showed that in situ microbial growth in the high-permeability regions improved not only the volumetric sweep efficiency but also the microscopic oil displacement efficiency.

INTRODUCTION

Permeability variation and crossflow limit oil recovery from petroleum reservoirs [8,23,25,29,30].

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When water is injected into a reservoir, flow will be preferentially conducted into the high-permeability layer, and flow into lower-permeability layers will be diverted into high-permeability layers whenever possible. Increased permeability variation decreases volumetric sweep efficiency of enhanced oil recovery processes, and crossflow complicates this problem by allowing flow between contrasting layers [33].

Several methods to selectively plug petroleum reservoirs have been proposed, including polymers, gels, clays, paraffins, and resins [2,7,13]. These have not been widely used in the field since these agents do not selectively plug larger pores, do not penetrate deeply into the reservoir, migrate when injection pressure is decreased, and are physically or chemically incompatible with the reservoir in question.

Bacterial selective plugging has been proposed to correct microscopic and volumetric sweep efficiencies within geologic formations [3]. This selective plugging could correct stratification problems within a reservoir by penetrating the high-permeability layers of a reservoir. Since bacteria multiply their mass exponentially, plugging could be enhanced under growth conditions. This increase in cell mass could divert flow to less-permeable zones, and increase oil recovery in those zones.

Much laboratory work has been directed to the study of microbial selective plugging. Jenneman et al. [17,18] have shown that nutrients necessary for growth and metabolism can be transported through sandstone cores. Jang et al. [15,16] and Jenneman et al. [17,18] have shown that bacteria can penetrate through sandstone cores. Jenneman et al. [19] showed that microbial penetration is reduced in cores below 100 mD due to increasing probability of cells encountering pores too small to penetrate. Torbati et al. [37] suggested that live cells preferentially plug larger pore throats. Shaw et al. [32] found that bacteria produce an exopolysaccharide film, which contributes to pore plugging by suspending clay particles, cells, and other suspended solids. This polymer was not removed by fluid pass-

ing through the porous media or by bacteriocidal agents. Raiders et al. [27] have reported that semi-continuous nutrient injection under anaerobic conditions will limit facial plugging in cores and that the microbial plugging process selectively reduces permeability and improves the volumetric sweep efficiency.

Jack and DiBlasio [14] and Jang et al. [16] have reported that injecting pure cultures of a specific bacterial species will increase oil recovery beyond residual oil saturation. We have previously shown that the in situ growth and metabolism of microorganisms significantly reduces the permeability of Berea sandstone core [17,18]. However, it is not known whether the use of indigenous bacteria will enhance oil recovery and improve volumetric sweep efficiency.

Mathematical and physical model systems have been developed to study the factors which affect fluid flow in stratified petroleum reservoirs [4,9,25,31,33]. These studies showed that reducing the permeability only in the vicinity of the wellbore does not result in a significant improvement in volumetric sweep efficiency [28,33]. Once the fluid has moved past the plugged region it will reenter the high-permeability regions due to crossflow between the regions. Thus, the plug must penetrate deeply into the formation to alter flow patterns enough to recover additional oil [28,33]. Mathematical models assuming no flow between the stratified layers did not simulate experimental systems where crossflow was evident. However, when crossflow was accounted for in the mathematical models, they adequately simulated experimentally obtained data.

In this paper, we determine whether the in situ growth of microorganisms enhances oil recovery and improves sweep efficiency in Berea sandstone model systems. We have constructed a crossflow system using two slabs of Berea sandstone with different permeabilities. Thus, we have studied the effects of bacterial growth and metabolism on fluid flow patterns using an experimental system that more accurately represents the flow conditions of a petroleum reservoir.

MATERIALS AND METHODS

Sandstone systems

Blocks of Berea sandstone (Cleveland Quarries, Amherst, OH) were cut into 6-inch-long, 2-inch-diameter cores for parallel core experiments or 12-inch \times 12 inch \times 2 inch slabs for crossflow experiments. Cores were steam-cleaned for 2 weeks, dried for 24 h at 125°C, and stored in a vacuum desiccator until used [27]. Two-inch sections were removed from two sides of the crossflow slabs and these sections were cored in the three principal directions to determine initial rock conditions in each direction (Fig. 1). All cores were coated with Conap Easy-poxy. Plexiglass endplates were epoxied to the parallel cores, and pressure taps were drilled into the crossflow cores (Fig. 2). The crossflow system was assembled with a sheet of Whatman chromatography paper between the slabs of sandstone. All cores and crossflow systems were then wrapped with fiberglass cloth and coated with fiberglass resin. An additional layer of epoxy was then applied to the cores.

Each sandstone system was flushed with CO₂ for 15 min and vacuum-saturated with 5% NaCl + 0.1 M CaCl₂ brine. The calcium was added to stabilize clays in the rock [35]. All injected fluids were filtered through 0.22 μ m filters and saturated with N₂ to limit the amount of dissolved O₂ in the brines. After saturation, 100 pore volumes (PV) of brine were flowed through each parallel core (10 PV through

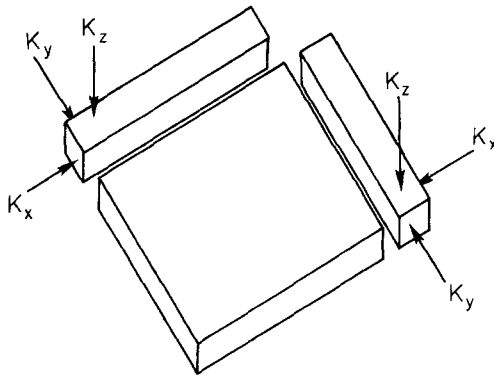


Fig. 1. Location of the cores taken from the crossflow system to determine the initial permeability (K) in all three dimensions.

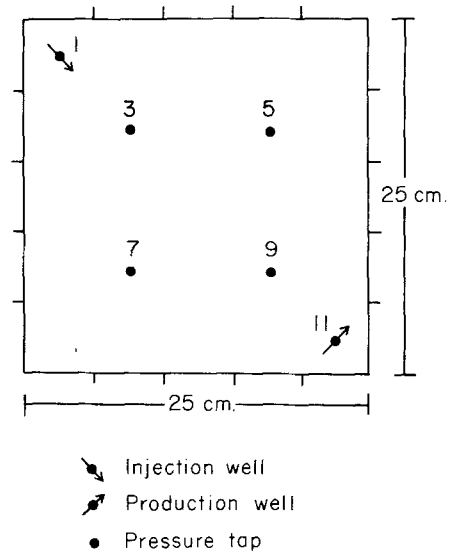


Fig. 2. Top view of the crossflow system indicating the location of the injection port (I) and production port (II) and location of the pressure taps.

the crossflow systems) at constant flow rate (0.02 ml/s for parallel cores, 0.05 ml/s for crossflow), using a Gilson HP4 Minipuls peristaltic pump. Fifteen PV of 5% NaCl brine were then injected, to reduce calcium concentrations in the cores, since high calcium concentrations may interfere with microbial growth.

Parallel cores were connected together at the injection source at this time [27]. The injection pumps used for the crossflow system were calibrated to inject fluids into each slab at the same potential. All parallel cores and one crossflow system were saturated by flooding with *n*-hexadecane. All oil-saturated systems were waterflooded to residual oil saturation.

A series of treatments was then performed on each system. A treatment consisted of the injection of 1 PV of medium E [17] with 0.1% NaNO₃ into the high-permeability zone or until pressure response was observed, incubation of the system at room temperature for 24 h, and waterflood with 5% NaCl brine until a stable pressure was reached. The treatment process was repeated until a stable permeability reduction factor was reached or the pressure limits of the system were reached.

Petrophysical measurements

Porosity and PV of the sandstone systems were determined as previously described [19]. The pressure drop across the cores was measured using a pressure transducer [27]. Pressure of the injection line and at each pressure tap of the crossflow system was measured using pressure gauges. Flow rates were measured volumetrically.

The permeability of the sandstone system or core was calculated from Darcy's law using the physical dimensions of the system, the volumetric flow rate, the differential pressure across the core or a section of the crossflow system, and the dynamic viscosity of the fluid [19,27]. The dynamic viscosity of the fluid was measured as previously described [26]. The initial permeability of the systems was measured with 5% NaCl brine after the calcium treatment. The permeability was measured after each nutrient treatment during the waterflood with 5% NaCl brine. The permeability reduction factor (PRF) is the percent of the initial permeability remaining after each treatment [27].

The effective permeability to oil was calculated when the sandstone system was saturated with *n*-hexadecane. *n*-Hexadecane was flushed into the sandstone system until no more brine was detected in the effluent. The flow rate of *n*-hexadecane and differential pressure of the system were measured and the effective permeability to oil was calculated using Darcy's law. The effective permeability to brine was calculated by flooding the *n*-hexadecane-saturated sandstone system with 5% NaCl until no more *n*-hexadecane was observed in the effluent, i.e., until residual oil saturation was reached. The flow rate of brine and differential pressure of the system at this time were used to calculate the effective permeability to brine using Darcy's law.

The percent oil saturation was calculated by determining the percent of the PV that was *n*-hexadecane at residual oil saturation. The amount of *n*-hexadecane in the sandstone system was determined volumetrically.

Effluent analyses

A sample of the effluent was collected within the first pore volume of each waterflood and the pH

was measured using a pH meter. The sample was centrifuged at $10\,000 \times g$ for 10 min at 4°C and the supernate was frozen (-20°C) until further analyzed. The oil-effluent interfacial tension was measured using a Dunouy ring tensiometer [21]. Gas was collected in an inverted graduated cylinder filled with 5% NaCl adjusted to a pH of 3 with citric acid.

Gas analysis was performed with a gas chromatograph equipped with a Porapak Q column and a thermoconductivity detector [1]. Volatile fatty acid concentrations in the effluent samples were determined with a gas chromatograph equipped with a flame ionization detector and a fused silica column [20]. Carbohydrate and nitrate concentrations of effluent samples were determined colorimetrically [11]. The amount of polymeric carbohydrate was determined after dialyzing samples in dialysis tubing with a 500 dalton exclusion size against 2 liters of distilled water. Sections of some cores were crushed and the amount of biomass present was determined by extraction and quantitation of the phospholipids as described by White et al. [38].

Isolation and characterization of bacteria in core effluents

Samples of core effluents were serially 10-fold diluted in sterile 5% NaCl and the highest dilutions were inoculated onto plates of plate count agar (Difco) with 5% NaCl or medium E [17] with 0.1% NaNO₃ and 0.05% yeast extract. Plates of each medium were incubated at room temperature aerobically or anaerobically using a Gas Pac (BBL) jar. Five colonies from plates of each medium inoculated with the highest dilution incubated under either condition were restreaked until pure cultures were obtained. The morphology, biochemical characteristics and ability to grow at different salt concentrations were determined. Routine biochemical tests and staining procedures were performed according to published procedures [34]. Ability to grow in different salt concentrations was determined by changing the amount of NaCl added to medium E from 0 to 15%.

Mathematical model of crossflow system

A mathematical model was developed to simulate the crossflow core system. A modification of Poisson's equation was used to model single-phase incompressible flow in an incompressible heterogeneous anisotropic porous medium [6,36]. The differential equation represents steady-state flow behavior controlled by sources and sinks to represent injection and production locations within the porous medium. The equation is second-order with respect to each of the flow dimensions. The solution gives the flow potential field for the porous medium. Because the porous medium can be heterogeneous, numerical methods are required for solution. The second-order derivatives of flow potential in the three space dimensions were represented using second order correct, centered finite difference approximations. The resulting system of linear algebraic equations were solved using the point successive overrelaxation (SOR) method. The validity of the method was tested by simulating several three-dimensional experiments in steady-state, non-growth systems. The model was utilized to predict whether permeability reduction alone could explain changes in flow patterns within the crossflow systems.

RESULTS

Parallel core experiments

Physical parameters of the cores are shown in Table 1. Oil recovery and permeability reduction data are shown in Table 2. For the first three experiments, nutrients were injected into the cores until pressure response was observed. In the fourth experiment, only 1 PV of nutrients were injected into the high permeability core after the initial treatment, which was continued until pressure response was observed. Experiment 3 was not waterflooded completely to determine whether mobile oil saturation affected the microbial plugging process. The first 20 ml produced from each core during each waterflood were analyzed. Samples from the first three experiments were analyzed for pH changes and interfacial tension changes. Samples from experiment four were analyzed for material balance determinations.

In all experiments, the level of permeability reduction was similar, 6–8% for high-permeability cores, 14–18% for low-permeability cores. Permeability reduction occurred more quickly in high-permeability cores than in low-permeability cores

Table 1
Physical parameters of cores before treatment

	Core number ^a							
	H1	L1	H2	L2	H3	L3	H4	L4
Length of core (cm)	14.2	14.2	13.5	13.5	14.2	14.2	11.4	11.4
Porosity (%)	21	15	20	17	19	17	23	20
Pore volume (ml)	65	48	59	51	59	55	51	44
Initial brine permeability (mD)	866	103	292	109	1210	112	460	110
Initial oil saturation (%)	62	52	45	57	54	50	68	55
Effective oil permeability (mD)	250	55	179	37	435	65	120	52
Effective brine permeability (mD)	90	5.7	67	7.5	79	5.8	67	11.3
Residual oil saturation (%)	41	41	30	39	39	50	41	34

^a H and L represent the high- and low-permeability cores, respectively, from each experiment, followed by the experiment number in which they were used.

Table 2

Oil recoveries and permeability reductions obtained in core experiments

	Core number							
	H1	L1	H2	L2	H3	L3	H4	L4
Number of treatments	13	13	12	12	17	17	13	13
Waterflood brine injected (PV)	2.9	0.3	7.4	0.5	0.3	0	0.5	0.1
Total brine injected (PV)	12.8	3.3	35.4	4.3	23.1	10.1	25.1	5.3
Average brine injected per treatment (PV)	1	0.3	3	0.4	1.4	0.6	1.9	0.4
Total nutrients injected (PV)	13	2.7	26.3	2.5	28.5	6.6	18.4	10
Average nutrient injected per treatment (PV)	1	0.2	2	0.2	1.7	0.4	1.4	0.8
Waterflood oil recovery (% OIP)	33.7	21.1	34	31.7	27.9	0	39.4	36
Total oil recovery (% OIP)	48.4	35.9	56.3	42	55.1	38.9	56.5	54
Average oil recovery per treatment (% OIP)	1.1	1.1	1.8	0.9	1.6	2.3	1.3	2
Final PRF (%)	8.3	14	6.6	15	7	18	7.7	14.2
Total gas produced (ml)	82	15	55	13	78	25	82	26

^a % OIP = incremental oil recovery.

(Fig. 3). Oil recovery and gas production did not occur in significant quantities until PRF dropped below 50%. Most of the oil recovered by the treatment process in high-permeability cores was produced within three or four treatments after PRF had fallen below 50%. Gas bubbles were entrained in the oil as it was produced from the cores.

Correcting for the initial waterflood front from the third experiment, the oil recovery from that low-permeability core correlates with similar cores. Between 10 and 39% of the oil left behind in the cores after waterflood was recovered by the process. Generally more oil was recovered from the high-permeability cores than from low-permeability cores.

Fig. 4 shows the volumetric sweep efficiency improvements in the core system as each experiment progressed. The best sweep efficiency improvements were observed in the third dual core experiment, which had the greatest difference in permeability contrast between cores. The smallest improvement in sweep efficiency occurred in experiment 2, where there was the least permeability contrast.

Material balance calculations for carbon were performed for each treatment in the fourth experiment. The carbon balance data are shown in Table 3. These results are summarized in Fig. 5. Initially,

some nutrients were found in the initial effluent sample, L0, taken from the first 20 ml of the first nutrient injection. This suggests that the pore structure of core L4 was very heterogeneous, and that most of the flow was through a few large pores. The initial peak in both curves showed that little metabolism of the nutrients occurred at the start of the experiment. Some of the nutrients injected into the cores during the first treatments may have been adsorbed onto the clays. After this initial adjustment period, the microorganisms utilized the carbon source and produced acids, gases, and polymers. At that time, significant oil was released from the cores, and the PRF values decreased below 50%.

Fig. 6 shows the cell concentration calculated from the phospholipid content found in cores H4 and L4 after the treatment process. While the cell concentration in core H4 was fairly constant from inlet to outlet, the cell concentration in core L4 decreased by two orders of magnitude from inlet to outlet. Growth was more concentrated at the inlet sections of core L4.

The effluent from the eleventh waterflood of core H4 was analyzed to determine the numerically dominant bacteria. Viable cell counts using medium E with 0.1% NaNO₃ and 0.05% yeast extract (Dif-

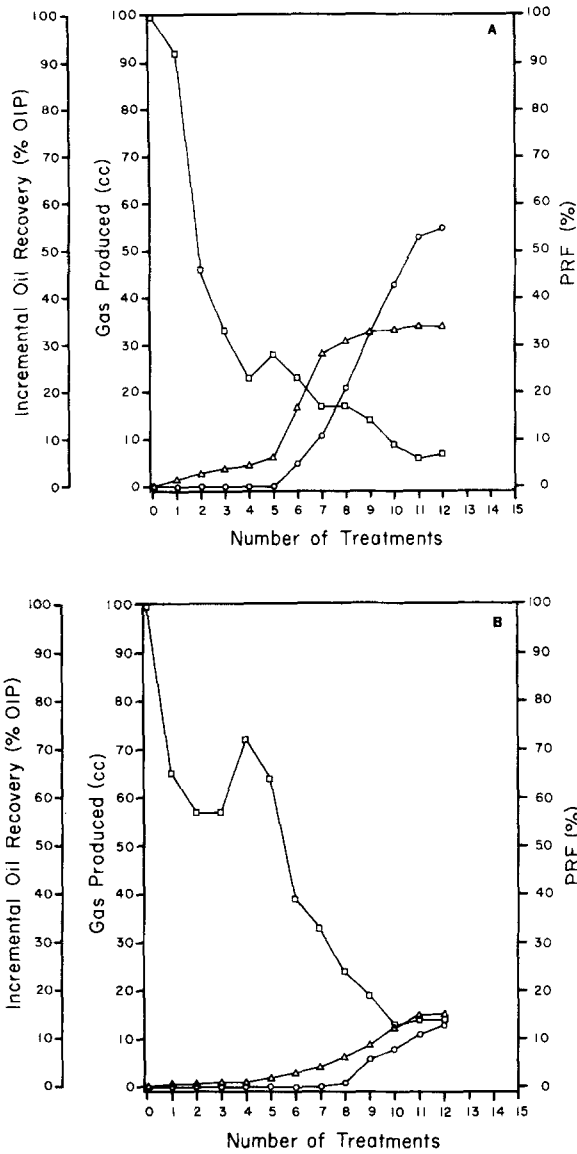


Fig. 3. Permeability reduction factor, incremental oil recovery, and gas production from cores H2 (A) and L2 (B) per treatment.

co, Inc.) incubated under aerobic and anaerobic conditions were 5.7×10^7 cells/ml and 2.4×10^7 cells/ml, respectively. Viable cell counts using plate count agar with 5% NaCl under aerobic and anaerobic conditions were 4.6×10^7 cells/ml and 2.0×10^7 cells/ml, respectively.

Five colonies from plates of each medium inoculated with the highest dilutions under aerobic and

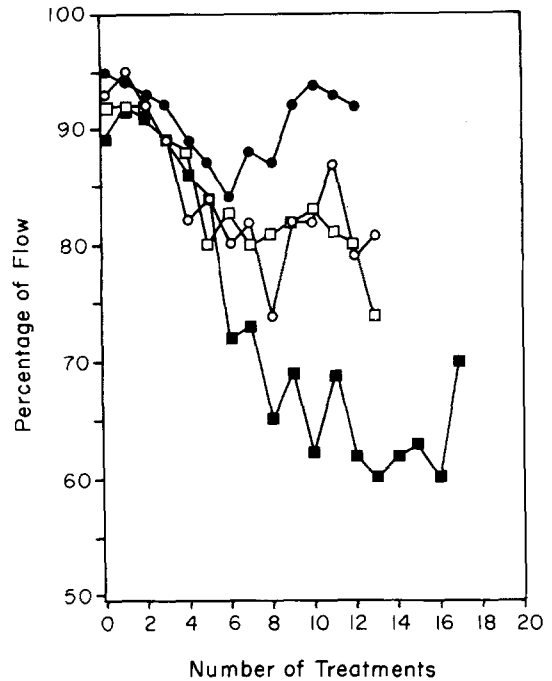


Fig. 4. Percentage of flow entering the high-permeability core after each treatment for each parallel core experiment. ○, ●, ■, □: experiments 1 through 4 respectively.

anaerobic conditions were restreaked until pure cultures were obtained. All isolates grew aerobically as well as anaerobically in thioglycollate medium, plate count agar, and medium E with and without nitrate. All were gram-negative rods. Further biochemical and morphological characterizations showed that the isolates could be separated into two groups. One group was composed of facultative coccobacilli with some diplococoid and long filamentous forms detected especially in medium with 5% NaCl. These isolates were non-motile, oxidase-negative, ornithine decarboxylase-negative, reduced nitrate, produced indole, hydrolyzed urea, decarboxylated lysine, fermented malonate, produced acid and gas from glucose, lactose, maltose, sucrose but not from insulin, did not produce hydrogen sulfide, formed acetyl methyl carbinol, and hydrolyzed esculin. These isolates grew in medium E with up to 10% NaCl added. The above characteristics indicate that these isolates are indole-positive variants of *Klebsiella pneumoniae*. The other group of isolates did not grow in medium E without

Table 3

Carbon balance for effluent samples from cores H4 and L4

Sample ^a	Acetic acid (mmol)	Butyric acid (mmol) ^b	CO ₂ (mmol)	Sucrose (mmol) ^d	Polymer (mmol) ^d	Total (mmol)	Carbon recovery ^e (%)
L0	1.7	0	0.9	106.8	0.0	109.4	31.2
L1	2.7	0	1.4	429.3 ^f		433.4	123.7
L2	3.1	0	1.6	372.7 ^f		377.4	107.7
L3	6.9	0	3.5	123.3 ^f		133.7	38.1
L4	13.1	0	6.6	25.1	7.2	52.0	14.8
L5	16.3	0	8.2	21.5	52.5	98.5	28.1
L6	14.3	0	7.1	0.0	77.8	99.2	28.3
L7	13.3	0	6.7	52.6	103.7	176.3	50.3
L8	13.5	0	6.8	48.2	24.8	93.3	26.6
L9	14.3	0	7.1	167.8	14.2	203.4	58.0
L10	13.9	0	7.0	34.0	80.1	135.0	38.5
L11	12.7	0	6.4	53.7	19.3	92.1	26.3
L12	15.9	0	8.0	46.1	23.9	93.2	26.8
L13	18.7	0	9.4	90.0	39.7	157.8	45.0
H0	0.0	0	0.0	0.0	0.0	0.0	0.0
H1	2.3	0	1.2	252.2	22.5	278.2	79.4
H2	2.6	0	1.3	19.1	23.7	46.7	13.3
H3	7.3	0	3.6	333.8	16.5	361.2	103.1
H4	20.4	0	10.7	340.7 ^f		371.8	106.1
H5	27.5	1.1	15.9	122.7	21.3	188.5	53.8
H6	30.2	1.2	17.5	86.9	46.4	182.2	52.0
H7	25.4	1.0	14.7	47.3	24.5	112.9	35.0
H8	38.2	1.5	22.1	0.0	97.3	159.1	45.5
H9	31.8	1.6	19.1	130.9	17.0	200.4	57.2
H10	27.6	1.2	16.1	53.0	115.0	212.9	60.8
H11	31.2	1.5	18.7	0.0	148.2	199.6	57.0
H12	38.0	1.8	22.7	73.4	102.1	238.4	68.0
H13	36.2	1.6	21.4	103.3	92.2	254.7	73.0

^a H and L represent core H4 and L4, respectively, followed by the treatment number in which the sample was collected. Samples from treatment 0 were collected during the first 20 ml of the first nutrient injection. All other samples were collected during the first 20 ml of waterfloods.

^b Butyrate was detected in measurable quantities only in samples H5 through H13.

^c CO₂ was determined by calculating carbon balance by known metabolic pathways [32]. One millimole CO₂ would be made from each millimole acetic acid, and 2 mmol CO₂ would be made from each millimole butyric acid.

^d Sucrose and polymer concentrations are given in mM hexose equivalent.

^e Sucrose was injected into the cores at 350.4 mM during nutrient injection.

^f This represents the sum of the polymer and sucrose concentrations.

5% NaCl. These isolates were facultative, motile, oxidase-positive, catalase-positive, rods that reduced nitrate and contained poly- β -hydroxybutyrate. These characteristics are consistent with being members of the genus *Photomicrobium*.

A decrease in pH of the effluent from 7.0 to 6.0 occurred and a decrease in the oil-effluent interfacial tension of 20% was found throughout the study. Some CO₂ was observed in the gas samples (0.1–1% of the gas phase). The remainder of the gas

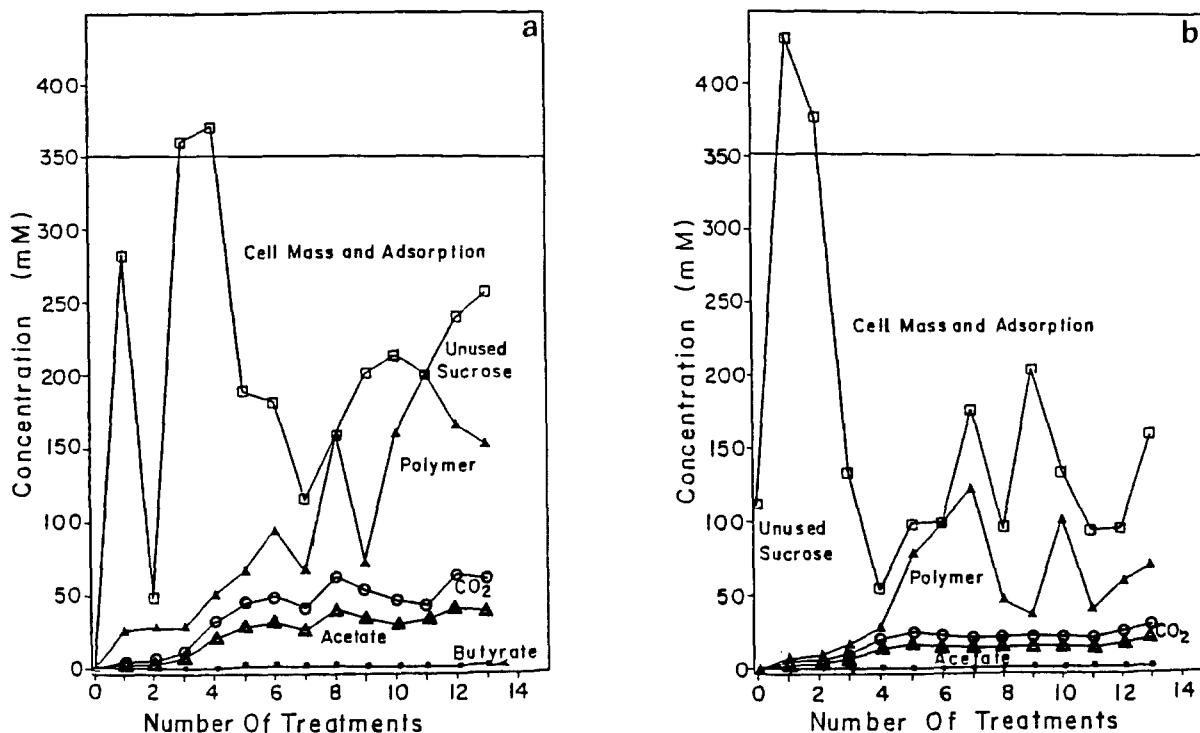


Fig. 5. Carbon balance for each treatment for cores H4 (a) and L4 (b). Data are taken from Table 3. The carbon that was not detected as sucrose or metabolic products was assumed to be the result of the absorption of organic components by the sandstone and the production of cellular mass inside the core.

was presumed to be N_2 , since other common gaseous products (H_2 , NO , N_2O , and CH_4) were not detected in any gas samples. Nitrite was detected in early effluent samples of core L4 (less than 0.2 mM).

Nitrate was not detected in any effluent samples. Since the concentration of nitrate in the influent was 17 mM, these data suggest that denitrification or dissimilatory nitrate reduction was the dominant metabolism in the cores.

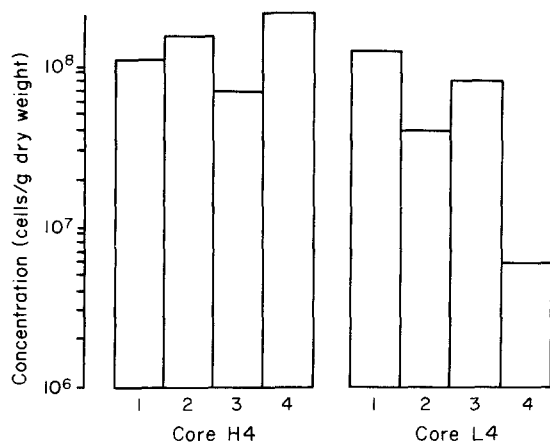


Fig. 6. Concentration of cells calculated from the phospholipid content in consecutive core segments from inlet (1) to outlet (4) for cores H4 and L4 after the treatment process.

Crossflow experiments

The simulation model results agreed with previously published experimental data and other published crossflow simulation results. The literature, and our model, suggests that selective plugging is effective in changing crossflow patterns within plugged regions by modifying flow profiles within porous media. Wellbore plugging alone will not correct the flow profile if layers are in capillary contact, since flow can be diverted around the plugged higher-permeability region past the wellbore [22,28,33].

Flow diversion and selective plugging by indigenous bacteria in crossflow core systems were first examined in a brine-saturated system. The porosity

of the high- and low-permeability layers was 20% and 17%, respectively. Average permeabilities for the high-permeability layer were 423 mD in the x-direction, 415 mD in the y-direction, and 342 mD in the z-direction. For the low-permeability core, average permeabilities were 107 mD in the x-direction, 105 mD in the y-direction, and 54 mD in the z-direction.

Five treatments were conducted in this experiment. In the first two treatments, very little change was observed in the potential distributions from the baseline data (Table 4). This was due to the small number of bacteria residing in the core, or found in the effluent, at the beginning of the treatment process. During later treatments, large numbers of bacteria were found in the effluent. As the bacterial populations became large enough to modify pore structure, the potential distribution was modified.

Fig. 7 shows the percentage of flow into and out of the high-permeability layer from this experiment. After microbial plugging occurred, the amount of flow conducted by the low-permeability layer increased up to 60% of the total flow from the system.

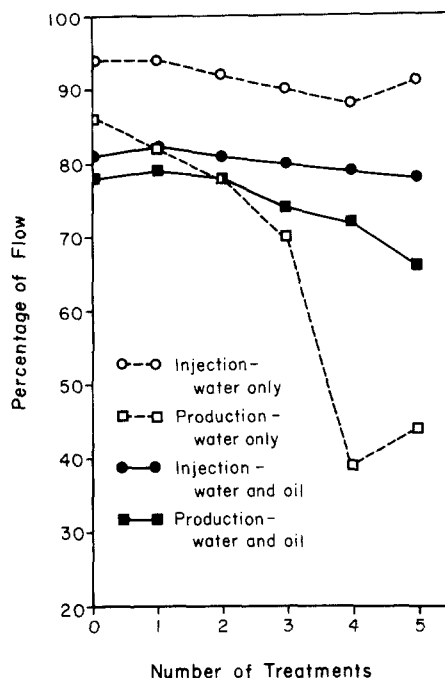


Fig. 7. Percentage of flow entering (circles) and exiting (squares) high-permeability layer of crossflow apparatus after each treatment for brine-saturated (open symbols) and oil brine-saturated (closed symbols) crossflow systems.

Table 4

Potential distributions in atmospheres at discrete points in the brine-saturated crossflow system

Location ^a	Treatment					
	0	1	2	3	4	5
1	1.095	1.109	1.143	1.211	1.449	2.374
3	1.054	1.054	1.061	1.095	1.229	2.143
5	1.027	1.027	1.048	1.088	1.286	2.143
7	1.034	1.027	1.034	1.082	1.279	2.129
9	1.024	1.02	1.02	1.082	1.279	2.143
2	1.095	1.102	1.143	1.224	1.449	2.401
4	1.054	1.061	1.075	1.122	1.327	2.129
6	1.408	1.034	1.048	1.088	1.313	2.075
8	1.054	1.048	1.054	1.088	1.299	2.102
10	1.054	1.054	1.068	1.102	1.286	2.061

^a Odd numbers represent the high-permeability layer, which was placed on top of the system. The potential at points 11 and 12 is given as atmospheric.

Relative injectivity into the low-permeability layer increased by as much as threefold.

Wellbore plugging could not have caused the uniform plugging and flow diversion found in the crossflow system, according to our simulation data (not shown). Since the plug, and sustained increases in potential, occurred throughout the system, and not just in the injection and production ports, we concluded that flow diversion occurred throughout the entire high-permeability layer. The mathematical simulator accurately predicted the actual waterflood data and data from treatments before the microbial plugging began (data not shown). After the increase in potential, the simulator no longer uniquely solved for the permeability distribution, injectivity indexes, productivity indexes, or the amount of crossflow. None of the solutions adequately explained the amount of crossflow from Fig. 6 that occurred in the brine-saturated system. All solutions showed diversion into the low-permeability layer, but the attempted solutions showed an

order of magnitude less flow diversion than the experimental data.

After five treatments, the pressure on the system exceeded the operating limits of the equipment. One PV (atmospheric) of biogenic gas was collected from the system. No significant changes in cross-flow patterns were observed after the gas was drained from the system. Potential decreased slightly during this waterflood.

When an oil phase was introduced into the system, similar results were obtained, except that the magnitude of the observed changes was reduced (Table 5). This was expected since the volume of active pore space available for bacteria was reduced by introducing oil into the system [27]. The porosities of this system were 17% and 14% for the high- and low-permeability cores, respectively. Average permeabilities for the high-permeability layer were 225 mD for the x-direction, 235 mD in the y-direction, and 80 mD in the z-direction. For the low-permeability layer, permeabilities were 95 mD in the x-direction, 88 mD in the y-direction, and 27 mD in the z-direction. Initial oil saturations were 52% and 44% for high- and low-permeability layers, respectively. Residual oil saturations were 34%

and 25%. Relative permeabilities to brine were 40% and 10%.

Significant flow diversion was observed, and the potential distribution was changed as bacterial growth occurred. Relative injectivity into the low-permeability slab increased by 50%. In the five treatments, 10% of the residual oil in the high-permeability layer was recovered through the high-permeability port, and 2% of the residual oil in the low-permeability layer was recovered through the corresponding port.

The simulator was able to duplicate the initialization data and results for treatments before the plug was established. Simply reducing permeability in the high-permeability layer did not explain the observed crossflow in the later treatments. The mathematical model simulated single-phase (brine) flow and did not model the presence of additional phases (oil and gas). It is likely that multiphase flow effects not accounted for in the simulator, such as relative permeabilities, affected interlayer flow. Unfortunately, it was not possible to measure the necessary data to validate a more complex model in the experimental system.

Table 5

Potential distributions in atmospheres at discrete points in the oil brine-saturated crossflow system

Location ^a	Treatment					
	0	1	2	3	4	5
1	1.102	1.095	1.109	1.177	1.367	1.728
3	1.082	1.061	1.082	1.163	1.265	1.422
5	1.068	1.054	1.068	1.150	1.259	1.408
7	1.054	1.054	1.054	1.136	1.245	1.395
9	1.041	1.020	1.048	1.109	1.231	1.388
2	1.102	1.095	1.116	1.177	1.395	1.952
4	1.082	1.068	1.082	1.163	1.259	1.414
6	1.082	1.054	1.075	1.136	1.244	1.401
8	1.054	1.034	1.054	1.122	1.224	1.381
10	1.082	1.054	1.068	1.129	1.204	1.361

^a Odd numbers represent the high-permeability layer, which was placed on top of the system. The potential at points 11 and 12 is given as atmospheric.

DISCUSSION

The decrease in permeability observed in these experiments is believed to be the result of the in situ growth and metabolism of microorganisms, rather than abiotic factors, because of the following observations. The time lag between the initiation of nutrient injection and the pressure increase in the cores, and the exponential pressure increase observed throughout each experiment are indicative of microbial growth. Since clay swelling was controlled by CaCl₂ injection, and NaCl concentration was not changed during experiments, clay swelling would not cause the pressure increase [24,35]. The fluid velocity was kept below the critical fines migration velocity, so fines migration would not be responsible [10].

Cells and biologically produced substances were observed in the effluent from each core, and the amount of these substances increased as the PRF

decreased. After four to six treatments, gas, acid, and polymer production began from each core. The production of CO_2 and presumably N_2 suggests that the denitrification pathway was selected for by the choice of nutrients. Since gas was not initially produced from cores, and the quantity of gas produced increased as PRF decreased, the gas produced from the cores resulted from microbial action.

While all experiments exhibited some volumetric sweep efficiency improvement, the best improvement was shown in the third parallel core experiment, where the permeability contrast was greatest. The sweep efficiency improvement decreased with decreased contrast. Mobile oil saturation in a core seems to decrease microbial activity in that core. PRF declined more slowly and erratically in core L3 than in comparable cores. Once the mobile oil was produced from the core, PRF decline and oil and gas production correlated with other low-permeability cores.

While PRF data for crossflow experiments are not directly available, the plugging process is comparable to the parallel cores through other experiments. The plugging process occurs more slowly in oil-saturated cores than in brine-only cores without crossflow [27]. When crossflow models were tested, the plugging process was again slower for oil-saturated systems than for brine-only systems. The relative injectivity increases observed in the crossflow experiments were far more dramatic than those observed in parallel cores. We attribute the increased injectivity changes to the influence of interlayer flow.

Microscopic sweep efficiency may have also contributed to the oil recovery process. Very little oil was recovered from any of the swept cores until a substantial reduction in PRF occurred. Larger pores are preferentially plugged during in situ microbial growth in porous media [14,37]. The lack of oil recovery until the PRF declined suggests that plugging the larger pores diverted flow into smaller pores containing higher oil saturations, which had been previously unswept.

Biogenic gas production was an important factor in the oil recovery process. At the beginning of each

experiment, little oil and no gas were produced from each core. During the first four to six treatments in which gas was produced, most of the incremental oil was produced. Since in the crossflow experiments only two treatments were run after the beginning of gas production, little of the oil recovery activity was observed before apparatus pressure limits were reached. Much of the gas produced at this time was entrained in the oil. The gas acts to increase pressure, increase mobility, and change flow patterns by introducing another mobile phase. As oil production decreased, gas was produced as a free phase, not entrained in the oil. However, in an actual petroleum reservoir, the in situ pressure would be great enough to prevent the formation of a free gas phase. Thus, it is uncertain whether biogenic gas production would be an important mechanism in oil recovery.

Potentially, microbial enhanced oil recovery processes may affect the quality of oil that is produced. The two major ways to detrimentally affect the composition of the oil would be by the degradation of the lighter molecular weight fractions of the crude oil and by the production of hydrogen sulfide. Since our process is an anaerobic one, little or no modification of the hydrocarbons present in the crude oil will occur in the time-course of the treatment process. Also, we use a nutrient solution containing high levels of nitrate to inhibit sulfide production [20]. Thus, we do not expect that the quality of the recovered oil would be changed.

The largest increases in oil recovery correlated with the largest increases in the amount of gas produced. These increases occurred after most of the PRF decline and the initiation of acid and polymer production. The relatively small pH and interfacial tension decreases suggest that fermentation acids or surface-active agents were not of major importance in the recovery process, although they may act with the gas, with the selective plugging, or separately to increase oil production.

The large number of PV of nutrients injected into the cores through most of this study was a result of experimental design. Since nutrients were injected into cores until pressure increases were observed, large amounts of nutrients were injected into cores

at the beginning of each experiment. Since initial cell populations are low and the residence time of nutrients is short compared to the time initially required to metabolize the nutrients, much of the nutrients initially injected into the cores were not metabolized. Thus, in later experiments, the nutrient injections were restricted to 1 PV into the high-permeability core or layer, and no significant changes in the process were observed. As the experiments progressed and the cell density increased from *in situ* growth, the amounts of nutrients needed to observe a pressure increase were much lower than initially required.

Throughout the fourth parallel core experiment, not all of the sucrose injected was metabolized in the core, and significant quantities of exopolymer were found in the effluent. This polymer may serve as a carbon source for bacteria downstream from the point of sucrose utilization. When the cell density in the core was increased by injecting bacteria, PRF declined more rapidly and less nutrients were required [27]. Also, treatments were continued until PRF stabilized or no more oil was produced from the cores. Thus, a greater number of treatments were used in this study than would be necessary in a field trial, with far more nutrients per treatment than would be needed.

Significant reductions in permeability were observed in low-permeability cores in dual-core experiments, even during the first several treatments. This was particularly true when the permeability contrast was small, and little sweep efficiency correction occurred. Much of this plugging was likely due to facial plugging. The amounts of nutrients injected into low-permeability cores during early stages of experiments often were less than 1 PV. This would force most of the growth near the entry. Also, the penetration rate of bacteria through Berea sandstone of permeability < 100 mD is slow and dependent upon the length of the core [19]. Since the average pore-throat size of these cores is less than the size of the average bacterial cell (1–5 μm), the probability that a pore will end or be constricted increases with increasing length. This suggests that bacteria will not deeply penetrate low-permeability regions. This is verified by decreased lipid concen-

tration observed in the outlet end of low-permeability cores, relative to that of high-permeability cores. This effect is enhanced in the crossflow experiments, since facial plugging in the low-permeability layers was minimal relative to the growth in the high-permeability layers. The plugging in the low-permeability layers was never enough to divert significant flow back into the high-permeability layers.

The observation of such high numbers of bacteria in the effluent and the isolation of cells compatible with the applied experimental conditions suggests that injecting cells into cores is not necessary to increase oil recovery, and the process could then be applied on a larger scale without cell injection.

A major problem that must be addressed is the development of the microbial biomass over a large portion of the petroleum reservoir. The volume of even the smallest oil reservoir is in the millions of barrels, which would require the injection of very large amounts of nutrients. One must be able to inject these large amounts of nutrients into the reservoir without plugging the wellbore. Biofilms tend to develop at the source of substrates and nutrients, which would prevent the injection of nutrients into the deeper portions of the reservoir. We propose to solve this problem in a variety of ways. In our method, we do not inject bacteria into the formation. Thus, the concentration of bacteria near the well bore will be small and metabolically inactive. If the well bore has higher populations as a result of prior activities or if a biofilm develops during nutrient injection, it can be removed by treatment with sodium hypochlorite [5]. Nutrients will be injected in high concentrations (10–20% molasses slugs); the high osmolarity will inhibit microbial growth. Once injected the nutrient slugs will be diluted deeper in the reservoir by areal dispersion. Also, one may alternate the injection of the substrate followed by a needed nitrogen or phosphorus source and allow for mixing to occur in the reservoir by diffusion/dispersion processes. The use of more refined sugar products would be beneficial in this regard. Finally, the analysis of previous microbial field trials have shown that large amounts of nutrients can be injected into petroleum reservoirs without detrimentally affecting the injectivity [12]. Thus, results ob-

tained in small-scale laboratory systems do provide some measure of predictability when large-scale field operations are planned.

Growth of *in situ* bacteria selectively plugged high-permeability cores in parallel flow systems with oil present, and in crossflow core systems, with or without oil present. The selective plugging increased microscopic and volumetric sweep efficiency of waterflood brine in the cores and resulted in an increase in oil recovery beyond residual oil saturation. This selective plugging increased the relative injectivity of fluids into less-permeable sandstone, in systems with or without capillary contact between the layers. The selective plugging, along with biogas and acid production, is responsible for increased oil recovery beyond residual oil saturation in core systems. These results show that microbial selective plugging is technically feasible in laboratory experimental systems which accurately simulate the fluid flow regimes of petroleum reservoirs. Our process depends only on the stimulation of the production of microbial biomass in the high-permeability zone, not on the growth of a particular microorganism or the production of a particular metabolite. Thus, it should be a feasible process to control under actual field conditions.

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