

KING SAUD UNIVERSITY  
College of Engineering  
Chemical & Petroleum Engineering Departments

Preprints

Symposium on Production and Processing  
of Natural Gas

26 - 28 Shaban 1412 H  
29 Feb. - 2 March 1992 G

## NATURAL GASES FROM BACTERIA AND THEIR USE IN TERTIARY OIL RECOVERY

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### Abstract

Bacteria are the only microorganisms that have been proposed for tertiary oil recovery processes. They are microscopic in size, exponentially grow and produce natural gases such as methane ( $\text{CH}_4$ ) and some other gases such as nitrogen ( $\text{N}_2$ ), hydrogen ( $\text{H}_2$ ) that could improve oil recovery by increasing pressure and by reducing the crude oil viscosity which lead to improvement in mobility ratio.

Many species of bacteria produce carbon dioxide ( $\text{CO}_2$ ). In recent years, carbon dioxide is used successfully to achieve a miscible displacement which is an important method of tertiary oil recovery processes. Some of these miscible systems are more difficult to achieve and maintain than the others due to limitations on temperatures and pressures.

This paper is an insight look into the classifications of the natural gases and carbon dioxide that are produced by bacteria and their possible applications in tertiary oil recovery.

### Introduction

The interest in applications of bacteria in petroleum industry and technology results from the low cost and versatility of bacterial formulations. Furthermore, bacteria are self-replicating and therefore inexpensive to produce using inexpensive nutrient sources. With recent declines in

oil prices, enhanced oil recovery by bacteria, being a cost-effective EOR method, might be the answer to increasing future world proved reserves(1).

Bacteria are able to produce a wide spectrum of chemical products and gases (mainly CO<sub>2</sub>) under variety of conditions. The mechanisms of oil recovery by bacteria are still not well understood because the biological bacterial systems are complicated living cells. It was concluded that bacterially enhanced oil recovery should be considered for reservoirs with a permeability of about 100-150 mD(2-4).

The reduction of heavy oil viscosity through carbon dioxide dissolution is a well known phenomenon. Under 1000 psi pressure, CO<sub>2</sub> dissolution can swell heavy crude by 10% and cut its viscosity by an order of magnitude(5).

It is extremely important to prevent any adverse effects on the environment when applying any oil recovery method. The bacteria used in EOR are not pathogenic. One possible adverse environmental effect is the stimulation of indigenous sulfate reducing bacteria which can produce toxic hydrogen sulfide causing corrosion(6).

The basic mechanisms of Microbial Enhanced Oil Recovery (MEOR) are: (i) reduction of the interfacial tension of the oil/water rock system, (ii) improvement in the mobility ratio of the fluids involved, and (iii) increase in the permeability of the reservoir rock.

It is well known that microorganisms such as alga, yeasts and fungi can plug formation of high permeability. The degree of plugging varies widely, depending upon the numbers and kinds of bacteria present in the water and upon the permeability, porosity, and pore size distribution of the reservoir rock(7). Bacterial spreading in a highly permeable formation is affected

by the mineralogical composition of the porous media, physicochemical and microbiological properties and the injection rate.

### Classification of Gases That Can be Produced from Bacteria

Growth of bacteria can create metabolic products that fall into five general classes<sup>(8)</sup> : organic acids, inorganic acids, gases, surfactants and biopolymers. This growth of bacteria may have a number of potentially important interactions with the inorganic matrix of the rock and the oil present in the reservoir. The classification of the gases produced by bacteria was reported by Jack et al<sup>(8)</sup> as follows :

- Gases produced as metabolic by-products from organic substrate utilization include hydrogen (H<sub>2</sub>), methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>).
- Gases produced as metabolic by-products from inorganic anions include nitrogen (N<sub>2</sub>) from NO<sub>3</sub> reducing bacteria and hydrogen sulfide (H<sub>2</sub>S) from SO<sub>4</sub> reducing bacteria which can dissolve sulfate-containing minerals to increase permeability.
- Gases from the dissolution of organic acids produced by bacteria and carbonates, which can increase the permeability of the reservoir and release carbon dioxide (CO<sub>2</sub>).

Some of the microbial culture currently employed in MEOR field tests are<sup>(9)</sup> :

Anaerobes and facultative anaerobic cultures such as Clostridium, Bacillus, Pseudomonas, Leuconostoc.

Salt tolerant bacteria of the genus clostridium have been isolated and shown to produce large amounts of carbon dioxide and solvents in 5-6 percent salt solutions. Spores of this bacteria do not adsorb on sandstone rock samples. A field test in the United States with clostridium acetobutylicum resulted in a two and half increase in production from a two-spot injected test and production of 35,000 Kg of acids and 5700 m<sup>3</sup> of carbon dioxide

during the nine month of test period<sup>(10)</sup>.

Acetobutylicum belongs to the group of clostridium comprising the butyric acid bacteria<sup>(11)</sup>. They are characterized by the formation of acetic and butyric acids, carbon dioxide, and hydrogen. The natural gases products of the fermentation of glucose by some species of butyric acid bacteria are given in Table 1. This type of bacteria is of interest for enhanced oil recovery because it will cause partial pressurization of the reservoir due to the release of carbon dioxide and hydrogen. The acids produced will also react with reservoir carbonates to form additional carbon dioxide. Figure 1 shows the overall pathway of glucose fermentation by acetobutylicum.

The alteration in gases that are produced by organisms using different porous media was presented by Russell and Gurla<sup>(12)</sup>. Figure 2 depicts total gas production as a function of time of clostridium culture grown in the presence of different core samples (0.5 gm/tube) as obtained by Russell and Gurla.

Microscopic and photographic observations on bacteriological laboratory displacement tests were reported<sup>(13)</sup> Figure 3 shows the gas production, in some of these tests, due to the in-situ growth of organisms. It is seen that more gases are accumulated at the face of the sandpack. The pressure was increased by about 0.0014 psi (in some other experiments it was about 0.0075 psi) due to biogenic gas production.

#### Miscibility Conditions in the Oil Reservoir Due to the Production of Natural Gases from Bacteria

In order to mobilize the residual oil droplets, miscibility and/or a high pressure gradient is required. The residual oil is an essential target for

enhanced oil recovery. This recognize that, the efficiency of a given process to recover this amount of oil depends upon capillary and interfacial forces. The miscible process consists of displacing oil by injecting microorganisms that are producing a solvent which is completely soluble in oil.

### Natural Gas

Figure 4 shows that phase diagram of solvent/hydrocarbon system assuming the solvent (produced gas) consists entirely of the light component (G). Point O represents the oil composition which must be rich in intermediate components. The displacement process is not first-contact miscible since the dilution path  $OC_1$  passes through the two-phase region.

The crude oil (O) and the produced gas (G) are not in thermodynamic equilibrium. Phase exchange takes place and as a result the overall composition  $M_1$ . The mixture will split into two phases, a gas  $G_1$  and a liquid  $L_1$ , determined by the tie lines. The gas  $G_1$  will have a much higher mobility than  $L_1$  and comes into contact with newly formed residual oil of composition O. Phase exchange takes place to form mixture  $M_2$  which splits into gas  $G_2$  and liquid  $L_2$ ,  $G_2$  flows to form mixture  $M_3$ , and so forth.

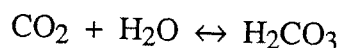
This process continues until the composition of the gas in contact with oil becomes  $G_t$  (i.e. the gas phase will no longer form two phases on mixing with the crude). The process, at this stage, has developed miscibility. The formation of the miscible bank is schematically shown in Figure 5. The miscibility bank is extremely stable. This process requires crude oil rich in intermediates. Design of a miscible process by injecting bacteria needs extensive laboratory studies and pilot tests. The parameters involved in the design counter both physical properties of crude oil viscosity and reservoir

characteristics such as depth, temperature, pressure and permeability.

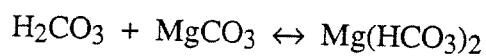
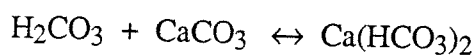
### Carbon Dioxide

There is increasing interest in using carbon dioxide in oil displacement from porous media. When carbon dioxide (gas) is produced from microorganisms injected into a reservoir, it dissolves with crude oil and water at reservoir conditions and reduces its viscosity causing it to swell. In miscible carbon dioxide flooding, liquid CO<sub>2</sub> mixes with oil and through a mechanism of multicontact phase equilibrium, eventually achieve miscibility. This will increase recovery efficiency. Miscibility conditions are required in order to mobilize the trapped oil droplets. An example of CO<sub>2</sub> - crude oil equilibria is shown in Figure 6. It is seen that the phase equilibria is strongly dependent on reservoir pressure and temperature.

Another principal effect of carbon dioxide is the action of carbonic acid, formed in solution with water<sup>(14)</sup>.



Its effect on calcareous rocks is to increase injectivity by partially dissolving the rock according to the following equations<sup>(14)</sup>:



The biocarbonates formed are soluble in water. This may increase absolute permeability of the carbonate rock. It was found by Bubela<sup>(15)</sup> that the porosity and permeability of calcite were increased in the presence of bacteria from 17% to 45% and from 2 darcy to 47 darcy, respectively, while those of the dolomite were altered from 24% to 27% and from 2 darcy to 14 darcy, respectively. No differences were observed in the absence of bacteria. This increase is due to the biological production of fatty

acids and CO<sub>2</sub>. Cementation resulting from diagenesis of carbonates, due to alteration of CO<sub>2</sub> level in the interstitial fluids, decreased permeability of the simulated sediments quite considerably.

The residual oil saturation obtained by producing CO<sub>2</sub> from bacteria is lower than that obtained by producing natural gases. This will provide an improvement in the oil recovery factor.

### Mechanisms of Natural Gases Bacterial Flooding

The bacterial recovery approach has been to ferment molasses anaerobically in the reservoir to generate agents of oil release in-situ. These include gases, acids and solvents as shown in Figure 7. The major agent for stimulated oil release will be gas formation.

Gas products fermentation (produced from molasses) such as CO<sub>2</sub>, H<sub>2</sub> and CH<sub>4</sub> can give sufficient reservoir pressure to drive more oil. Acid products will modify reservoir rocks (especially limestone) to increase rock porosity and permeability.

Generation of more carbon dioxide (CO<sub>2</sub>) as a result of reaction of organic acids produced by bacteria and calcareous material in a reservoir will serve as a swelling agent for oil(16).

Gas products, especially carbon dioxide, dissolve in oil reducing its viscosity. Microorganisms can grow on heavy crude oil which reduce its viscosity. This mainly due to(17): (i) reduction of the average molecular weight of the heavy oil, and (ii) the production of specific metabolic products essentially gases and surfactants. This will improve the mobility ratio of the displacing and displaced fluids.

In-situ miscibility between gases and crude oil will lead to the formation of a miscible bank which will mobilize more of the residual



oil.

Bacteria plugging results from bacterial growth especially in the vicinity of the well bore. The following equation shows the relative injection rate before and after plugging by bacterial treatment:

$$\frac{Q_p}{Q} = \frac{\frac{K_p}{K} \log \frac{R_e}{R_w}}{\log \frac{R_p}{R_w} + \frac{K_p}{K} \log \frac{R_e}{R_p}}$$

where,  $Q_p$  = flow rate after bacterial plugging

$Q$  = flow rate before bacterial plugging

$K_p$  and  $K$  = rock permeability in plugged area and original area, respectively.

$R_w$ ,  $R_e$  = well radius and drainage radius, respectively.

$R_p$  = plugged radius.

This equation is useful for relating the fluid injection rate after bacterial treatment to the fluid rate before treatment. The flooding rate has a great effect on relative flooding time and therefore the economics of the oil recovery program will be affected<sup>(18)</sup>.

Plugging mechanism is of great importance in the success of both waterflooding and bacterial flooding. Thus, bacterial plugging represents an important limiting factor for MEOR. Some types of bacteria could provide a feasible system for plugging the high permeable zones which may improve oil recovery through modifying the homogeneity of the formation.

### Economics of MEOR

At the end of 1990, the world has a total of 1015 billion barrels

of crude oil proved reserves. This figure is available to us through conventional production technology. The remaining oil in place, however, can reach up to three times as much. This indicates that at least 3000 billion barrels of oil are targeted for enhanced oil recovery technology. In Saudi Arabia alone, there will be more than 500 billion barrels of oil left in place after depleting its proved reserves (amounts to 260 billion at the of 1990) by conventional recovery. The ever increasing world demand for oil calls for new recovery technology to economically produce oil left in place. One of the emerging EOR technology which holds promise as a cost-effective method for producing more oil is microbial Enhanced Oil Recovery (MEOR).

#### Advantages of MEOR from an Economical Point of View

Several literature publications have shown that MEOR is potentially cost-effective technology for increasing oil production<sup>(2)</sup>. Many advantages of using MEOR can be sited from which some of the most important are<sup>(2),(19)</sup>:

1. The injected microorganisms and nutrients are relatively inexpensive and easy to obtain and handle in the field.
2. MEOR is economically attractive for marginal producing fields.
3. Injected fluid cost is not dependent on oil prices.
4. Implementation of the process needs only minor modifications to existing field facilities which reduces cost.
5. Easily applied with typical surface equipment for waterflooding.
6. MEOR is less expensive to install and more easily applied than any other EOR technique.
7. MEOR products are all biodegradable and will not accumulate in the environment which make it very attractive nowadays.

### Reported Results of Recovery Improvement Using MEOR

The following is an attempt to summarize some reported results of recovery improvement using MEOR.

The Journal of Petroleum Technology reported in its Technology Digest (September 1991) that the results from tests at the Alton field in Queensland, Australia, showed that Biological Oil Stimulation (BOS) increased oil output by 70%(20). The same study has estimated that MEOR technology could unlock as much as 300 billion bbl of Australian oil left in place after conventional technology.

Injection of microorganisms and molasses has improved the rate of oil production at the Mink Unit project by about 13%(21). WOR in the same application was reduced in producing wells by as much as 35%. Bryant et al.(22) showed that microorganisms and molasses nutrient can recover an average of 32% more of a light crude oil (Delaware-Childers) from Berea sandstone cores than after laboratory simulated waterflood. Kianipey et al.(13) reported that the in situ growth metabolism of injected bacteria decreased residual oil saturation in the unconsolidated thin reservoir flow cells by 9-24%. Another study(23) showed that a field core produced 28% more residual Mink crude oil than the waterflood process. Richard et al.(24) have shown that between 10 and 39% of the oil left in place behind in the cores after waterflood was recovered.

### Field Application of Producing gases Bacteria

Most commonly, a consortium of bacteria believed to produce gas is introduced into the oil reservoir with nutrients and water. As a result, oil production is increased due to the increase in reservoir pressure from

fermentation of gases helping in displacing oil trapped in the rock pore space.

A review of the available literature which concerns field tests performed using microorganisms that are producing gases is shown in Table 2.

The average changes in free carbon dioxide observed during the Romanian Field test period (1975-1983) is shown in Figure 8. A significant amount of increased oil production was observed during this test and it was considered to be dependent on maintaining in the nutrient addition<sup>(9)</sup>. This increase may be due to the increase in carbon dioxide from bacteria. The average values of the responding two reservoirs (Baicoi and Vata) in the Romanian field show that the oil viscosity and the amount of CO<sub>2</sub> increased as shown in Table 3. Oil viscosity increased indicates slight oxidation and loss of dissolved CO<sub>2</sub>.

The test conducted in Lisbon unit in Arkansas showed that the oil production rate had increased significantly. Table 4 shows the gases products observed during this test. The variation of CO<sub>2</sub> production is shown in Figure 9. The carbon dioxide produced have exerted beneficial effects in the reservoir and total oil recovery. In the field test carried out in Okmulgee in Oklahoma, produced water showed presence of carbon dioxide (CO<sub>2</sub>). The test was considered a success. Carbon dioxide (CO<sub>2</sub>) observed was considered to be biogenic and played an important role in the oil recovery<sup>(9)</sup>. The composition of the gas during the test is shown in Table 2.

The test conducted on the Royal Loco Unit, Oklahoma indicated that oil production increased after the initial shut-in. Clostridiums which are capable of fermenting carbon dioxide and neutral solvents were injected.

The viscosity reduction from in-situ carbon dioxide production was the most important factor affecting oil production in this test<sup>(19)</sup>. More favorable mobility ratio was obtained due to the improvement in oil viscosity. The bacterium used in the Loco test produced some surfactant. The surfactant/CO<sub>2</sub> reaction creates foam and oil/water emulsion which play an important role in improving oil recovery<sup>(25)</sup>.

### Conclusions

Field application indicated that injection of natural gases producing bacteria was a feasible procedure of enhanced oil recovery to increase oil production. The mechanisms of oil mobilization in the reservoir rock pore space by bacterial flooding process are still not well understood due to the complexity of the process. Laboratory and field tests results indicate that much more work will be required to evaluate the mechanisms of oil recovery by bacterial flooding. Some field results indicated that the mechanisms of oil recovery by bacteria that are producing carbon dioxide (CO<sub>2</sub>) suggested the improvement in vertical and areal sweep efficiencies.

An extensive laboratory work on microbial formulation and flooding using reservoir fluids and cores should be carried out prior to any MEOR field trial. The success of an in-situ MEOR process depends on the selection of useful organisms that can function in the environment of the reservoir. Nutrients are essential for bacteria growth in the porous media. Bacteria can alter the viscosity of heavy oil successfully through degradation of high molecular weight asphaltenes or production of metabolic products such as gases and surfactants.

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Table 1 - Gases products from three butyric acid clostridia(11)

<u>Product</u>	<u>C. Butyricum</u> (moles/100 moles glucose)	<u>C-acetobutylicum</u> (moles/100 moles glucose)	<u>C-butylicum</u> (moles/100 moles glucose)
CO <sub>2</sub>	188	221	204
H <sub>2</sub>	235	135	78





Table 2 : Some Field Applications on MEOR - CONTINUED

Year	Location	Geology and lithology	Injected Bacteria	Response	Depth	Pressure	Temperature	Permeability	Salinity	Oil Viscosity	Oil Density or API	Oil Saturation
1985-86	Minejuz-Rockdale Field. Micro-Bac Gulf Coast Inc.	Fine silty sandy shale - highly laminated from sand shale to shaley sand	-----	• Oil production increased from 1.5 to 4 BOPD	908-1022 ft	90°F	90°F	---	---	---	---	---
1987	Delaware, Childers, Ok	Sandstone	<ul style="list-style-type: none"> <li>• Bacillus</li> <li>• Clostridium</li> <li>• Nutrient = molases 22 gal / injection well</li> </ul>	<ul style="list-style-type: none"> <li>• Bacteria produces : gases (mainly CO<sub>2</sub>) acids and surfactants.</li> <li>Produced gas composition : Methane 60% CO<sub>2</sub> 25.8% Butane 2.8% Pentane 2.6% Octane 3%</li> </ul>	600 ft (porosity = 20%)	535 psi	80°F	52 mD	<0.02%	7 cp	35°	31% (Reservoir gases are : Methane = 77.6% CO <sub>2</sub> = 15.8%)
---	West Germany	Limestone	---	• About 19% increase in recovery	4068 ft	---	150°F	10-50 mD	23% TDS	---	30.6°	---
1986	USSR	Sandstone	<ul style="list-style-type: none"> <li>• Bacteria in 4% molases (from 6 wells).</li> </ul>	<ul style="list-style-type: none"> <li>• 3% CO<sub>2</sub> produced</li> <li>• Oil viscosity increased</li> <li>• Oil recovery = 37-40 m ton/d for 4 months.</li> </ul>	(1000-1200 m) 5577 ft φ=20-23%	---	90°F	500-1000 mD	0.02% TDS	---	Heavy oil (asphalt)	---
1990	BWN Oil Co. Australia	---	---	---	---	---	---	260 mD	---	---	---	---
1961-70	Hungary	Sandstone and Limestone	<ul style="list-style-type: none"> <li>• Anaerobic thermophiles in molases (from 10 wells)</li> </ul>	<ul style="list-style-type: none"> <li>• Recovery = 126% yield from 12 wells.</li> </ul>	656-8061 ft	---	207°F	10-700 mD	---	---	---	---
961-69	Poland	φ = 13-25%	<ul style="list-style-type: none"> <li>• Clostridium in molases (from 17 wells)</li> </ul>	<ul style="list-style-type: none"> <li>• CO<sub>2</sub> produced increased.</li> <li>• Recovery = 20 - 200%</li> </ul>	1325-3753 ft	---	---	---	---	---	---	---
954-58	Czechoslovakia	φ = 22-36%	<ul style="list-style-type: none"> <li>• Desulfouibric Pseudomonas in molases.</li> </ul>	<ul style="list-style-type: none"> <li>• Oil viscosity decreases</li> <li>Recovery = 50% (from 7 wells)</li> </ul>	164-230 ft	---	---	3000-8100 mD	---	---	---	---

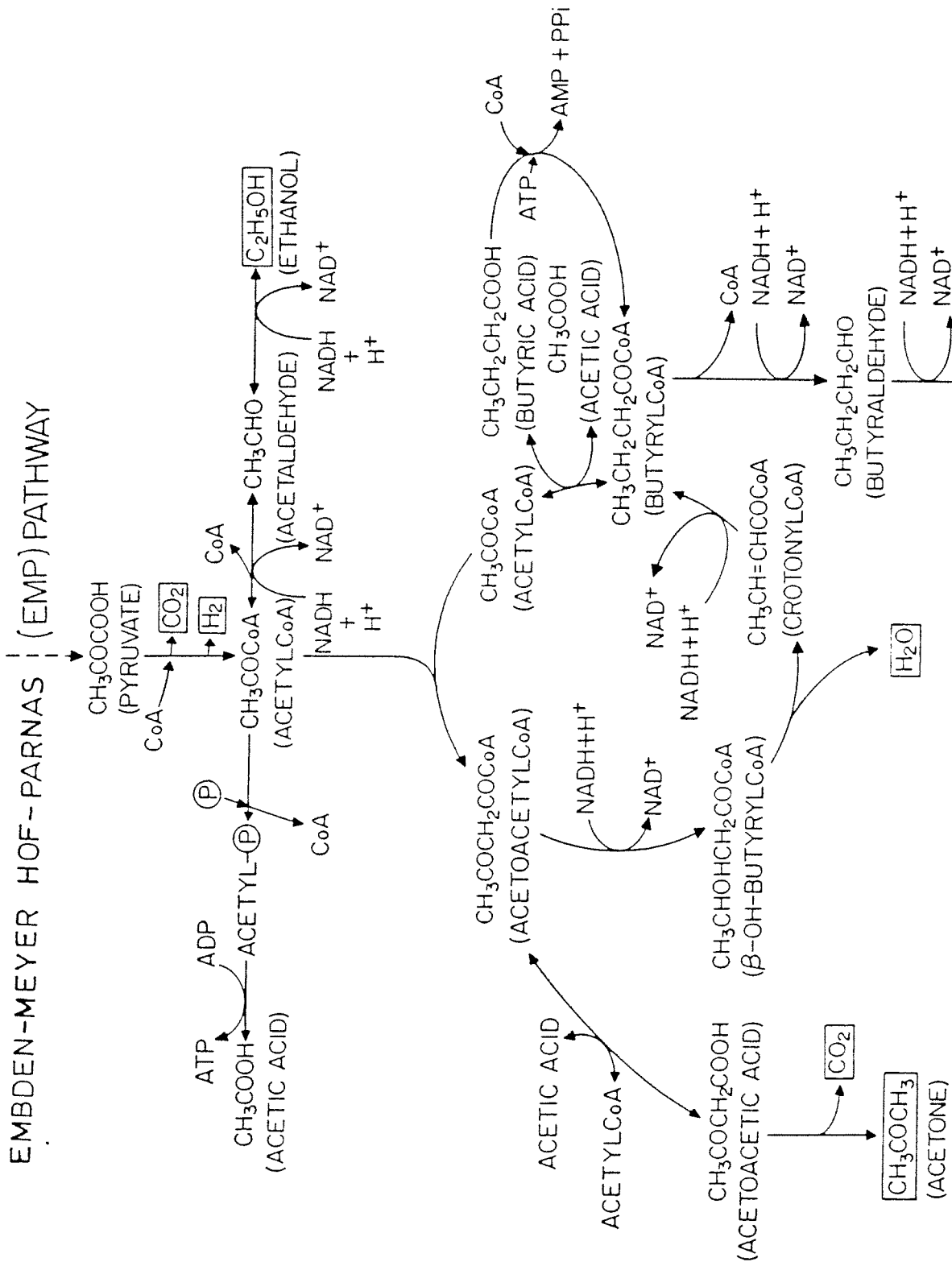


FIGURE 1 : Pathway of glucose fermentation by *C. acetobutylicum* (1,1)

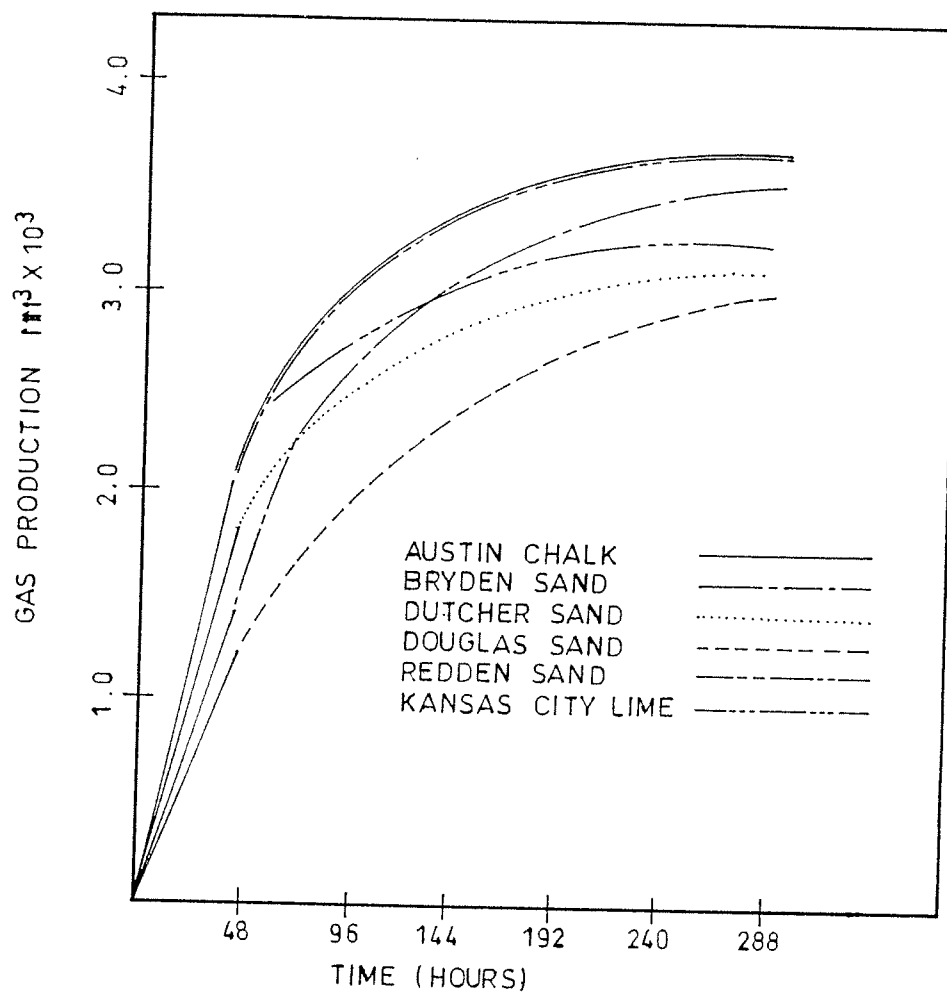
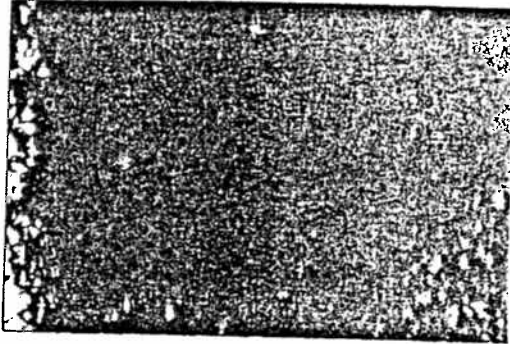


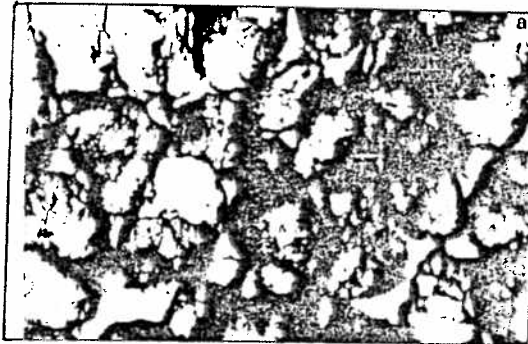
FIGURE 2 TOTAL GAS PRODUCTION IN THE PRESENCE OF VARIOUS CORES AS A FUNCTION OF TIME. (12)

Before Injection of Microorganism

After Injection of B. Lichenitorum



A.) At the Inlet



B.) In matrix sand

Figure 3 : Laboratory Biogenic Gas Production (13)

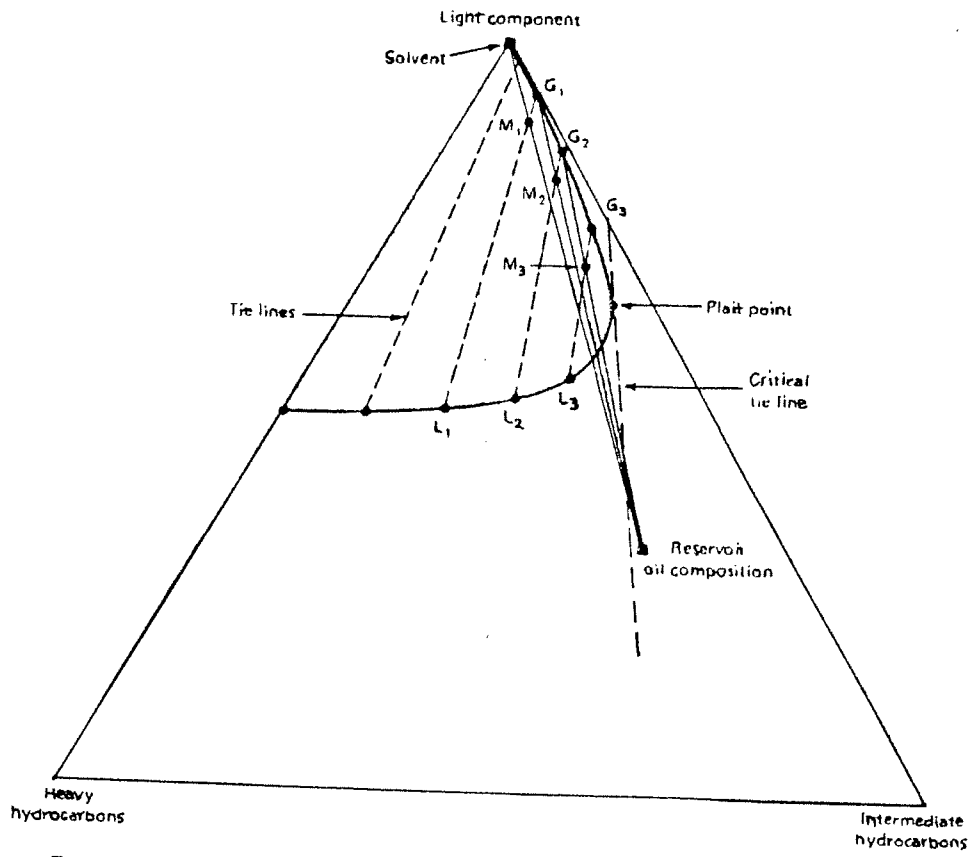


Figure 4 : Schematic of the vaporizing gas drive process

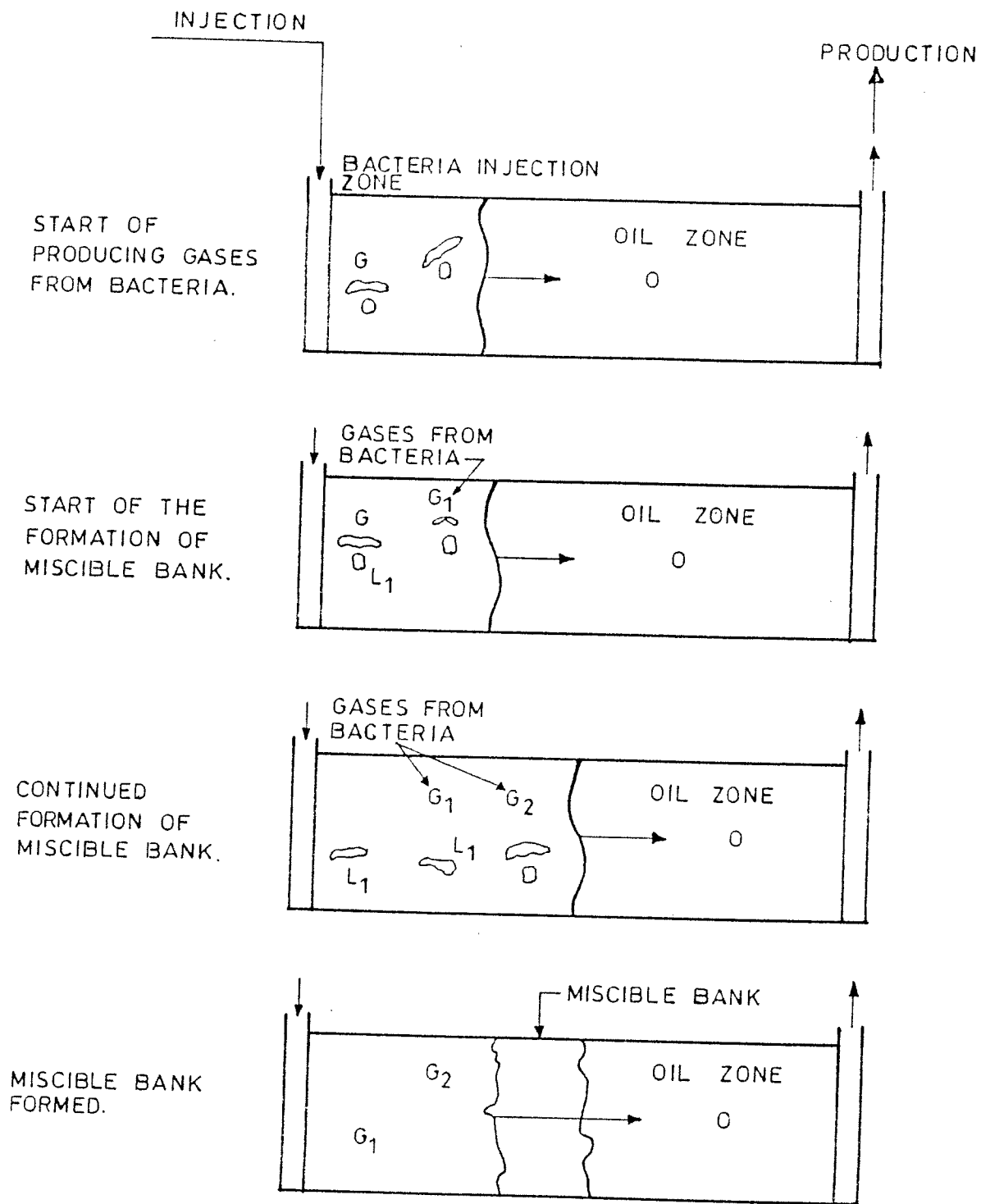


Figure 5 : FORMATION OF MISCIBLE BANK BY GASES FROM BACTERIA

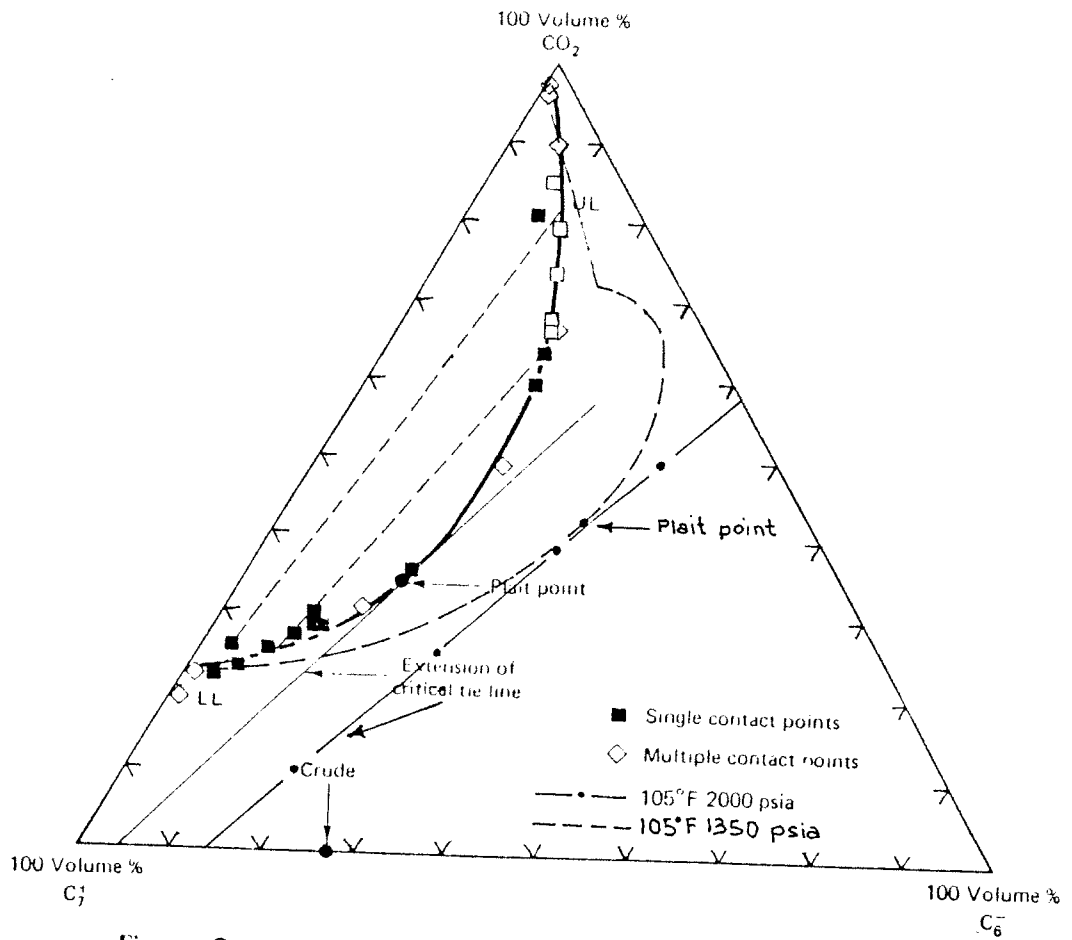
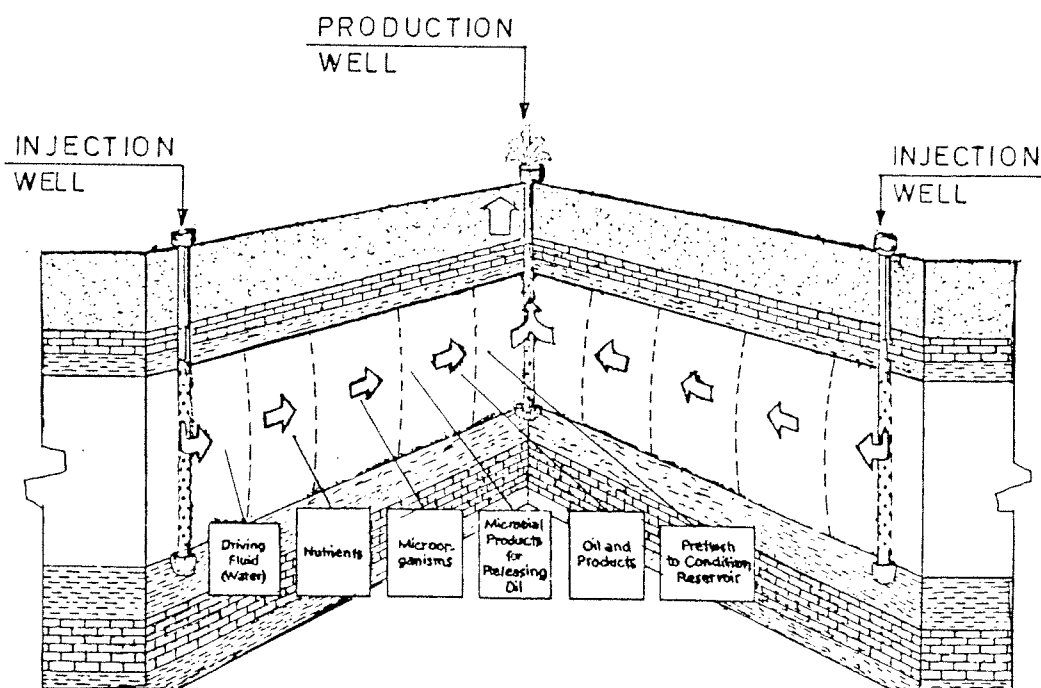
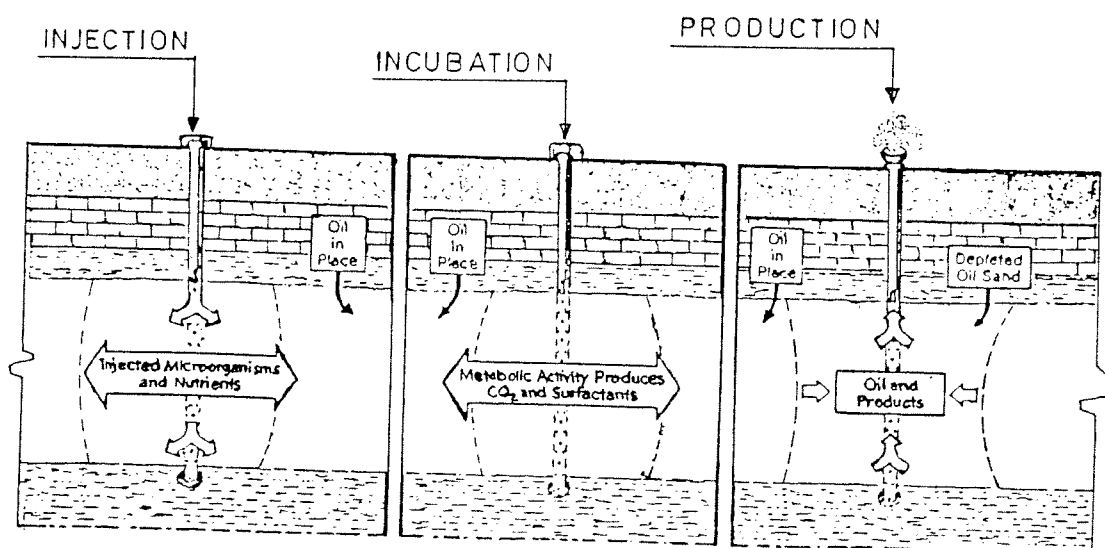


Figure 6 Ternary equilibria for  $CO_2$ -recombined Wasson crude mixture (25)





a.) MICROBIAL - ENHANCED WATERFLOODING



b.) CYCLIC MICROBIAL STIMULATION

FIGURE 7: SCHEMATIC DIAGRAM OF MEOR PROCESS

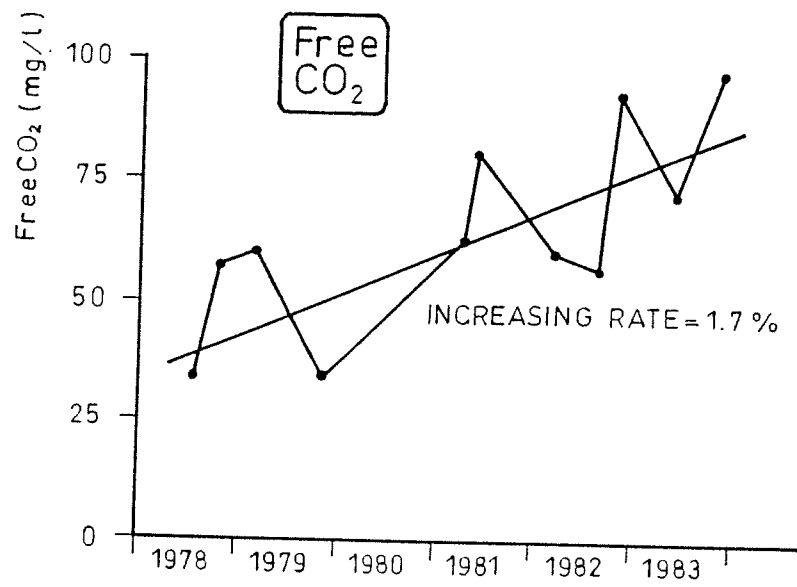


FIGURE 8 : CO<sub>2</sub> CHANGES OBSERVED IN THE RESERVOIR WATERS (9)

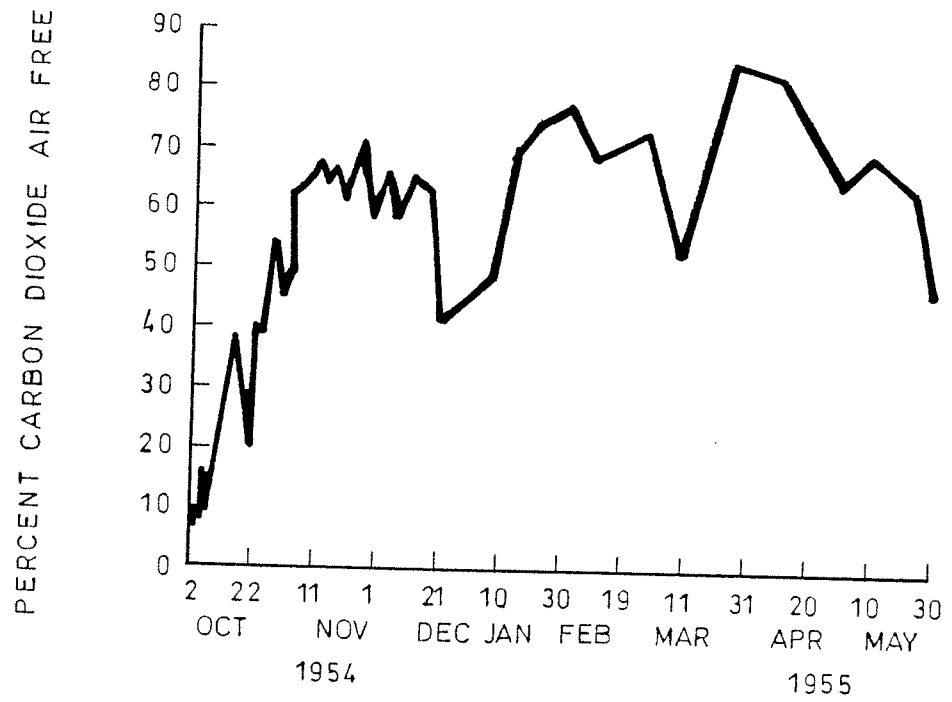


FIGURE 9 : CARBON DIOXIDE PRODUCTION (WELL NO. 31) (9)

الغازات الطبيعية من البكتيريا واستخداماتها  
فى الاستخلاص الثلاثى للزيت

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خلاصة

تعتبر البكتيريا من الكائنات الدقيقة التى اقترح حديثا استخدامها فى الاستخلاص الثلاثى للزيت. وهى ميكروسكوبية الحجم وتنمو بدالة أسية كما أنها مصدرا من مصادر انتاج الغازات الطبيعية مثل الميثان وبعض الغازات الأخرى مثل النيتروجين والهيدروجين التى تحسن عائد الزيت بزيادة الضغط ويخفف لزوجة الزيت الخام التى تؤدى الى تحسين النسبة الحركية .

ويمكن انتاج ثانى أكسيد الكربون من بعض أنواع البكتيريا. وفى السنوات الحديثة أمكن التوصل الى ازاحة امتزاجية باستخدام ثانى أكسيد الكربون حيث تعتبر من طرق تحسين عائد الزيت الثلاثية .

ويتناول هذا البحث تصنيف الغازات الطبيعية وثانى أكسيد الكربون التى يمكن انتاجها بواسطة البكتيريا وامكانية استخدامها فى الاستخلاص الثلاثى للزيت.