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# Role of molecular diffusion in the recovery of water flood residual oil

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A B S T R A C T

Traditionally, carbon dioxide ($CO_2$) injection has been considered an inefficient method for enhancing oil recovery from naturally fractured reservoirs. Obviously, it would be useful to experimentally investigate the efficiency of waterflooding naturally fractured reservoirs followed by carbon dioxide ($CO_2$) injection. This issue was investigated by performing water imbibition followed by $CO_2$ gravity drainage experiments on artificially fractured cores at reservoir conditions. The experiments were designed to illustrate the actual process of waterflooding and $CO_2$ gravity drainage in a naturally fractured reservoir in the Brass Area, Bayelsa. The results demonstrate that $CO_2$ gravity drainage could significantly increase oil recovery after a waterflood. During the experiments, the effects of different parameters such as permeability, initial water saturation and injection scheme was also examined. It was found that the efficiency of the $CO_2$ gravity drainage decrease as the rock permeability decreases and the initial water saturation increases. Cyclic $CO_2$ injection helped to improve oil recovery during the $CO_2$ gravity drainage process which alters the water imbibition. Oil samples produced in the experiment were analyzed using gas chromatography to determine the mechanism of $CO_2$-improved oil production from tight matrix blocks. The results show that lighter components are extracted and produced early in the test. The results of these experiments validate the premises that $CO_2$ could be used to recover oil from a tight and unconfined matrix efficiently.



**Capsule Summary:** The efficiency of water flooding naturally fractured reservoirs followed by carbon dioxide ($CO_2$) injection, for the recovery of oil residues was investigated and it was found that $CO_2$ gravity drainage could significantly increase oil recovery and has may play a considerable role in oil residue recovery.

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## INTRODUCTION

The displacement of oil by $CO_2$ has been the subjects of numerous studies, and a considerable volume of experimental data now exists for the process. In these studies, $CO_2$ usually displaces oil from a fully saturated slim tube or test core in a secondary displacement under conditions where phase behavior favors the development of miscibility. Foe these conditions, recoveries are observed high and rate-insensitive, provided that the effects of gravity segregation, viscous fingering, and bypassing are minimized. In direct contrast, for tertiary recovery experiments, $CO_2$ is injected into a previously watered-out test core, recoveries of residual oil which observed to be considerably lower, dependent on both flood rate and core length, and different for water-wet and oil-wet systems. A $CO_2$ pilot test is now being conducted in various fields in different parts of the world for naturally fractured oil reservoirs. Traditionally, in 1982, $CO_2$ injection was considered an inefficient method for enhancing oil recovery from naturally fractured reservoir. Horle (2004) noted that it would be useful to experimentally to investigate the efficiency of waterflooding naturally fractured reservoirs. To understand the mechanism of waterflooding residual oil by $CO_2$ injection, a series of investigation were performed in by George (1998). In order to optimize the $CO_2$ pilot test design in the world, (Brass community) trend areas, previous experimental work were scaled up to field scale using the mathematical model proposed (Schechter and Guo 1996; Campbell, 1983; Chartizis et al., 1983; Conway et al., 19996; Davis et al., 1967). The $CO_2$ injection will naturally and significantly enhance oil recovery after waterflooding in the naturally fractured reservoir. Oil produced during $CO_2$ injection process yields an increased concentration of components such as undecane to icosane ($C_{11}$-$C_{20}$). This indicates significant extraction by $CO_2$ during the $CO_2$ injection process. During the investigation of the role of molecular diffusion, it was examined that the effect of different parameters such as injection schemes, the efficiencies of the $CO_2$ injection decreases as the injection scheme decreased and the initial water saturation also enhanced. Cyclic $CO_2$ injection helped improve oil recovery, during the $CO_2$ injection process after water imbibitions. Investigation of the role of molecular diffusion in the recovery of waterflood residual oil by $CO_2$ injection is a huge topic for researchers. The subject topic is the overall goal of assessing the economic feasibility of $CO_2$ flooding the naturally fractured reservoir (John, 1986; Luan, 1994; Orr and Tabber; 1983; Schecker et al., 1981, Schecker et al., 1998; Scheckter and Guo 1996; Suffridge et al., 1994; Taylor; 1975; Turek and Metcalfe, 1984; Wang, 1982). Effects of other critical parameters (temperature, pressure, injection rate, injection pattern and initial water saturation) on $CO_2$ injection which was also the goal of investigation, is very difficult to analyze. The objective of this work will be focused on the following areas; extensive characterization of the reservoirs, experimental studies of crude oil/brine frock interaction in the reservoirs. Experimental investigation of $CO_2$ injectionand analytical and numerical simulation of molecular diffusion in the recovery of waterflood residual oil was also aim of the work (Elkins, 1996; Grogan and Pinczewski, 1987; Holm, 1985; Holm and Josendal, 1974; Huang and Tracht, 1974). The principle objective of the project was investigation of the role of molecular diffusion in the recovery of waterflood residual oil by $CO_2$ injection, $CO_2$ gravity drainage for naturally fractured reservoirs and the rock permeability to brine as a measure of various constant pressure drops and flow rates at ambient temperature.

## MATERIAL AND METHODS

### Samples collection and apparatus

An experimental investigation is designed to model the actual field experiment of waterflooding of residual oil by $CO_2$ injection in the naturally trend areas of Brass Community. The objective of the experiment is to investigate the effect of water imbibitions followed by $CO_2$ injection on oil recovery under reservoir conditions. Two experiments were conducted on Field A cores.

The apparatus used in the experiment are as follows; Cylinder or core holder, Visual cell for collecting produced fluids, Back Pressure Regulator (BPR), Pump, $CO_2$ and brine accumulators, Oil-gas separator and Graduated or calibrated glassware used to measure oil, gas and brine produced. The materials used in the experiment are Brine and Dead oil and samples.

Synthetic brine made using a measured composition was used in the experiments. The density and the viscosity of the brine are 1.08g/cm$^3$ and 1.2lcP, respectively at ambient temperature (26°C) and pressure (12.6 psia).

Dead oil was used in the experiment. The average molecular weight of the dead oil is 230g, the oil density was 0.85g/cm$^3$ and the viscosity was 2.96CP, measured at ambient temperature and pressure. The composition of the dead oil was measured by gas chromatography and was used a as base for comparing produced oil samples in the experiment.

Two core samples from field A were used in the experiments. Core X and Core Y. Core X was 24.45cm long, 10.16cm diameter, 197 1cm$^3$ bulk volume, 348cm$^3$ pore volume (PV), and 17.7% porosity. Rock permeability to brine was measured seven times at ambient temperature, using three different constant pressure drops and four different flow rates. The permeability ranged from 196 to 215 md with an average of 210 md. Core Y was 48.74cm long, 10.16cm diameter, 3949cm$^3$ bulk volume, 781 cm$^3$ pore volume, and 9.8% porosity.

The rock permeability to brine was also measured at various constant pressure drops and flow rates at ambient temperature. The permeabilities ranged between 394 and 413 md with an average of 404 md. The physical properties of those cores are presented in Table 2, Figure 1 below, schematically illustrates the setup used in the experiments), and were used to measure oil, gas and brine produced.

### Experimental procedure

The whole system was placed in an oven at reservoir temperature of 58.9°C. The cores were first put into a core holder and cleaned, After cleaning, it was taken from the core holder and put into an oven for drying at reservoir temperature (58.9°C), The core and core holder were oriented vertically and the system evacuated to about 0.6 psia. The bottom valve of the core holder was then opened to synthetic brine. After the core was saturated with brine, additional brine was flushed through the core. Permeability was then measured at various constant pressure drops and flow rates at ambient temperature.

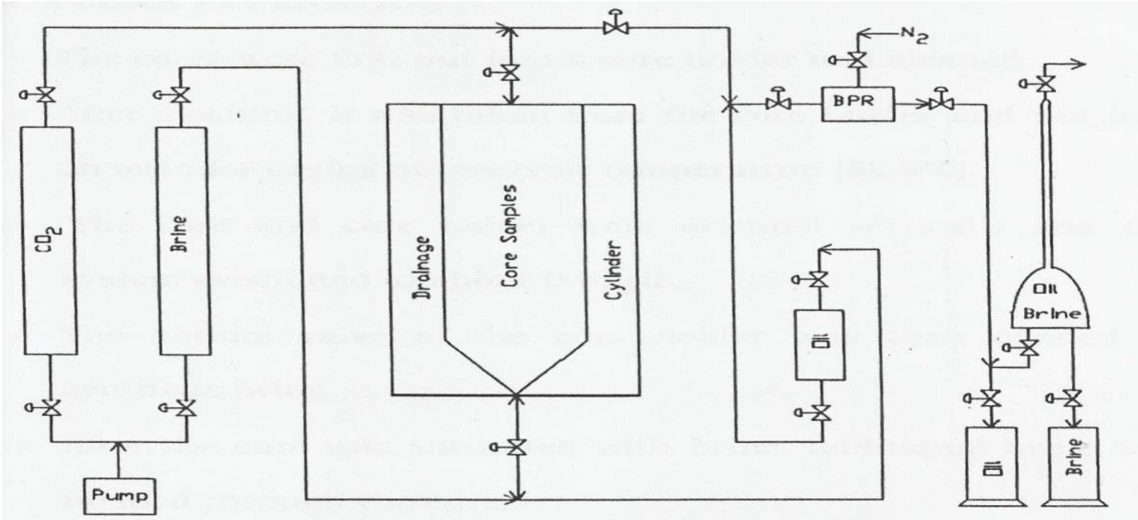


**Fig .1:** Schematic setup for $CO_2$ injection

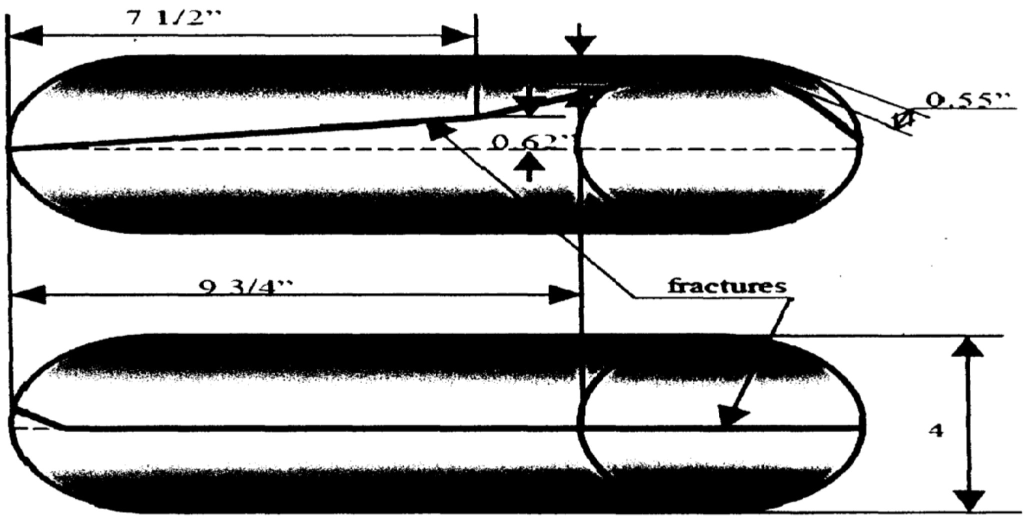


**Fig. 2:** Sketch of the fracture split on core X

Dead oil was then injected into Core X from the top of the core at 50 $cm^3$/hr. Oil broke through after 215 $cm^3$ of brine had been produced.

The total dead oil injected into the core was 1003 $cm^3$ (2.88 pore volume, PV) and the total volume of brine produced from the core was 264 $cm^3$. Thus, the initial water saturation, $S_{wi}$, and the initial oil saturation, SM, were 24.1% and 75.9%, respectively, (Table 2, Figure 2). The core was aged five days at reservoir temperature. It was artificially fractured into two pieces as shown in Fig. 2.

The fracture went through the middle on one side, and deviated from the centre by about one-fourth of a radius to the other side: The two ends of the core were then sealed to leave only the fracture open for fluid flow.

Core Y was prepared in a similar way. The core was cleaned and dried., it was put into a core holder in an oven at 58.9°C, dead oil was injected into the top of the core at 50$cm^3$/hr and then flow was reversed by injecting oil into the bottom of the core holder at 20$cm^3$/hr., the total volume of brine produced from the core was 518$cm^3$. The initial water saturation ($S_{wi}$) and the initial oil saturation ($S_{oi}$) were 33.7% and 66.3%, respectively, the core was aged for 55 days at the reservoir temperature and it was artificially fractured into six pieces with two horizontal and three vertical fractures as shown in Figure 3.

## Water imbibition

Water injection was performed for Core X at reservoir temperature for 28 days, the core holder was vertically placed in the oven, brine was injected into the bottom of the core at lOcm$^3$/hr for 15 days and then at 50cm$^3$/hr for the

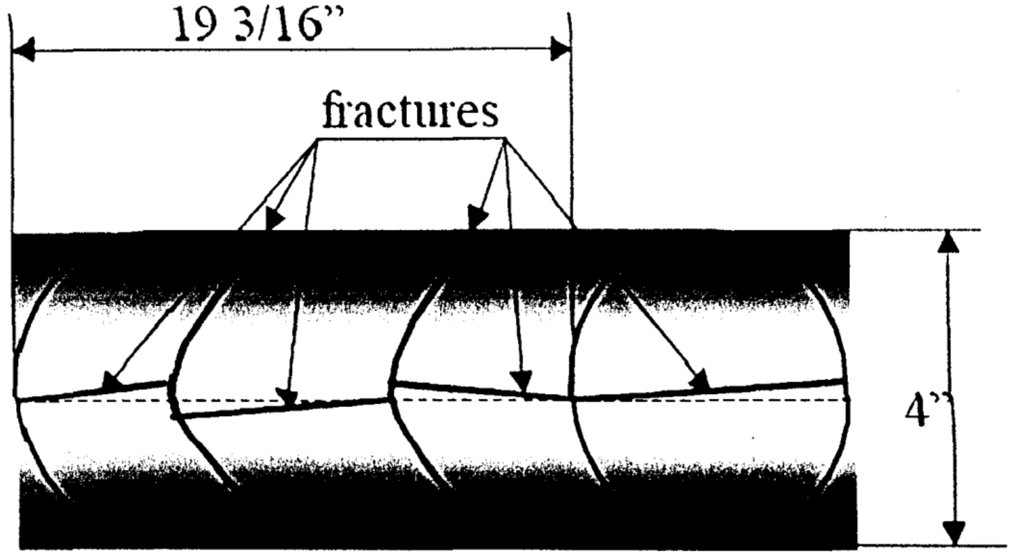


**Fig 3:** Sketch of the fracture split on core V

last 13 days, the system backpressure was set above 1750 1750 psi. A total of 43.7 cm$^3$ or 16.6% initial oil in the core (IOIC) was produced.

For water imbibitions, initial oil in the core (IOIC) equals the original oil in place (OOIP). The oil saturation decreases from 75.9% to 63.3%, and the water saturation increases to 36.7%, water imbibitions was similarly conducted for Core Y. at reservoir temperature for 31 days, brine was injected into the bottom of the core at 1.0cm$^3$/hr for three days and then at 0.5cm$^3$/hr for 28 days, the system backpressure was again set above 1750 psig and 242 cm3 or 46.9% initial oil in core (IOIC) was produced. The oil saturation decreased from 66.3% to 35.3% and the water saturation increased to 64.8% and during water imbibitions, 427cm$^3$ (0.55pore volume PV) of water was injected with 199cm$^3$ of water produced.

**RESULTS AND DISCUSSION**

A series of experimental tests were designed to aid in optimizing the $CO_2$ pilot design in the Brass Trend Area in delta state. Six experiments were performed, three on Field A and three on Field B reservoir cores. The experiments were performed to investigate the effects of initial water saturation, $S_{wi}$, and rock permeability on the efficiency of $CO_2$ gravity drainage. The properties of core samples used in the experiments and the experimental results are summarized in Table 1 as shown below.

These studies show that $CO_2$ gravity drainage could significantly enhance oil recovery in the naturally fractured Brass Area. The efficiency of $CO_2$ gravity drainage decreases as rocks become tighter and water saturation increases. The effect of water saturation on oil recovery seems to be more significant than rock permeability. Core discontinuity and impermeable layers at the top and bottom of the pay zone could affect the efficiency of $CO_2$ gravity drainage. Changes of $CO_2$ injection rate could help in enhancing oil recovery during $CO_2$ gravity drainage process.

The experiments presented in this work were designed and performed to investigate $CO_2$ gravity drainage after water injection in an artificially fractured core. These were done because reservoirs in the Brass Trend area are naturally fractured with very fight matrices. Initial investigations used Field A cores. The properties of Cores 6, X, and Y are given in Table 2. For comparison, Core #6 has no fractures, but was placed in the gravity drainage cell during water imbibition and carbon dioxide ($CO_2$) drainage where all the surfaces were exposed to either water or $CO_2$. The results obtained in the experiments are presented in Table 3a and 3b. The total oil recovery after water imbibtion and $CO_2$ for reservoir sample six (6) and cores X and Y are given in Table 4. The following analyses are based on these results.

**Carbon dioxide ($CO_2$) injection**

For core X:

1. $CO_2$ gravity drainage was conducted at reservoir temperature for 38 days.
2. The core holder was placed vertically in the oven.
3. The system backpressure was initially set at 1750 psig and later increased to 1950 psig.
4. $CO_2$ was injected vertically into the Core with flow from top to bottom.
5. The initial flow rate was 10 cm$^3$/hr, which was decreased to 5 cm$^3$/hr after six hours.
6. $CO_2$ gravity drainage continued for 806 hours, the backpressure in the system was decreased gradually

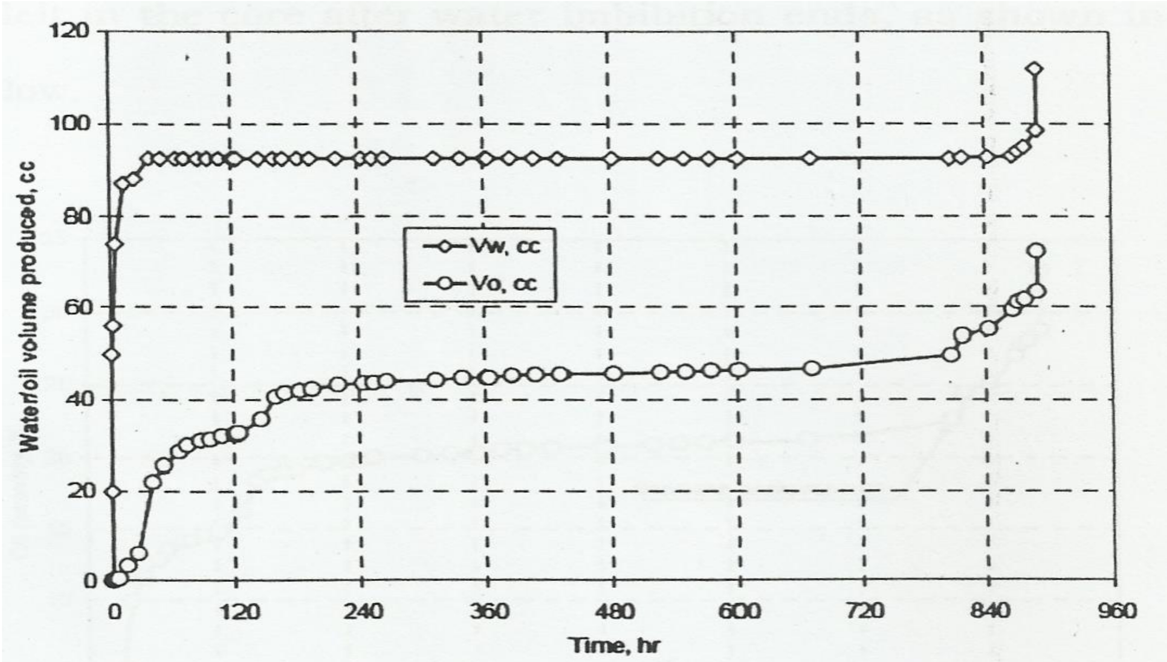


**Fig 4:** Water and oil produced during $CO_2$ gravity drainage for core X

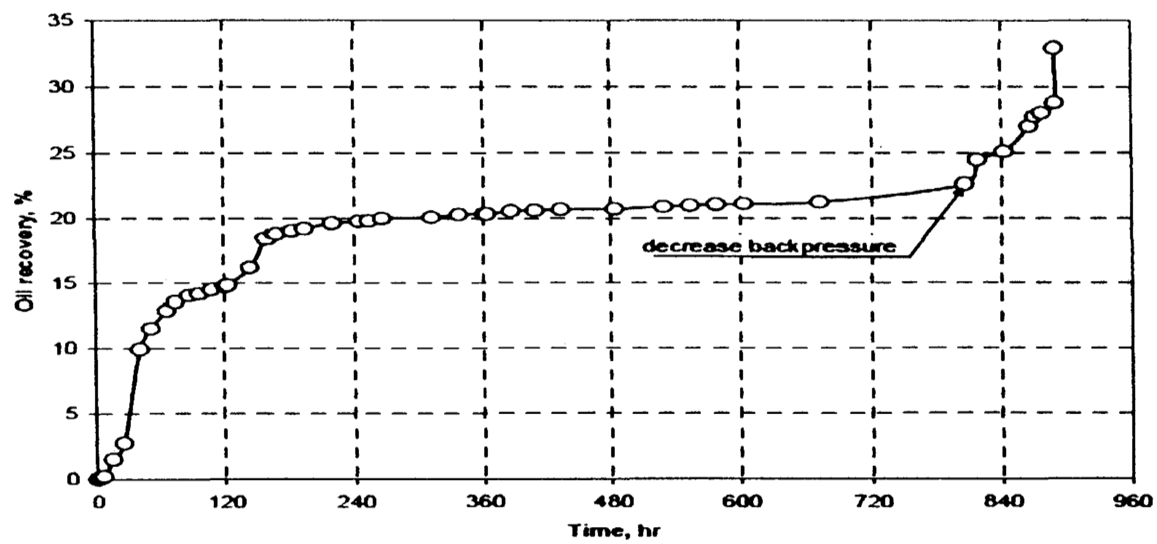


**Fig. 5:** Oil produce during $CO_2$ gravity drainage for core X

from 1950 psig to 0 psig. Figure 4.1 below shows water and oil produced during $CO_2$ drainage for core K.

The total volume of oil produced from the core was 90.5cm$^3$ (including captured liquids and estimated gas produced with $CO_2$.

Oil recovery with $CO_2$ gravity drainage was 41.1% initial oil in core (IOIC). For $CO_2$ gravity drainage, initial oil in core (IOIC) is the amount of oil within the core before $CO_2$ gravity drainage began or oil left in the core after water imbibition ends, as shown in Fig. 4.2 below. The residual oil saturation was 34.0%. The total oil recovery was 57.7% during water imbibition and $CO_2$ gravity drainage.

For core Y:

1. $CO_2$ gravity drainage was conducted at reservoir temperature for 18 days.
2. The core holder was also placed vertically in the oven at a backpressure of 1700 psig.
3. $CO_2$ was injected into the core from the top at flow rates between 0.5cm$^3$/hr and 1.0cm$^3$/hr. The total volume of oil produced from the core was 65.6 cm$^3$ and the total volume of water produced was 137cm$^3$. Oil recovery by

**Table 1:** Physical properties of the core sample used in the experiments

| Core sample number | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| Core type | A | A | B | B | B | A |
| Configuration | Continuous | Continuous | Continuous | Continuous | Stacked | Continuous |
| $CO_2$ injection time | Initial | Initial | Initial | Initial | Initial | After water injection |
| Length, Cm | 55.52 | 55.52 | 55.0 | 55.52 | 24.77 & 25.08 | 55.25 |
| Diameter, Cm | 10.16 | 10.16 | 10.16 | 8.89 | 6.53 & 6.58 | 10.16 |
| Porosity% | 18.7 | 13.0 | 10.0 | 11.1 | 10.7 | 22.43 |
| Brine permeability, md | 500 | .50 | 0.01 | 0.38 | 0.057 | 610.00 |
| Initial water saturation $Sw_i$, % | 35.0 | 29.3 | 38.6 | 45.0 | 37.6 | 66.5 |
| Residual oil saturation $S_{or}$ % | 37.5 | 32.5 | 42.5 | 41.8 | 50.5 | 28.0 |
| Initial oil in core (IOIC) $cm^3$ | 544.5 | 411.1 | 273.8 | 209.3 | 111.0 | 336.3 |
| Initial water in core (IWIC) $cm^3$ | 293.2 | 171.2 | 172.1 | 171.3 | 67.0 | 667.7 |
| total oil recovery % | 40.0 | 54.0 | 30.8 | 24.0 | 19.1 | 18.5 |
| Time, day | 6 | 220 | 190 | 167 | 331 | 36 |

$CO_2$ gravity drainage was 23.8 % initial oil in core (IOIC), as shown in Figure 3.

The residual oil saturation was 22.8 %. The total oil recovery was 70.7 % during water imbibition and $CO_2$ gravity drainage. The results obtained from $CO_2$ gravity drainage are summarized in Table 3a and 3b.

**Efficiency of $CO_2$ gravity drainage**

The experimental results show the efficiency of $CO_2$ gravity drainage after water injection is significant, which is consistent with previous results. The water saturations of the cores are high after water injection with initial water in core (IOIC) reduced well below original oil in place (OOIP). The oil recovery, however, is improved by more than 20% original oil in place (OOIP), by $CO_2$ gravity drainage. This is explained by the low interfacial tension between oil and $CO_2$ in the core. Much of the higher oil recovery for Core X was from production during system pressure blow down just before the experiment was terminated. This action produced 12% initial oil in core (IOIC). In addition, brine was displaced during $CO_2$ drainage. This indicates that some water would be produced during $CO_2$ gravity drainage in the Brass Trend when $CO_2$ injection is proceeded by water flooding.

Six oil samples were taken during $CO_2$ gravity drainage and analyzed using gas chromatography (GC). The results are shown in Figure 4.

The injected oil was dead oil, with which the core was saturated. Figure 4 shows that most of the oil produced during $CO_2$ gravity drainage consists of the component group undecane to icosane ($C_{11}$ - $C_{20}$) (~80 weight %). This figure shows that there were essentially no decane (Cio-) components in the produced oil samples, as found in the dead oil injected into the core. The decane ($C_{10}$-) components were produced with the high volume of $CO_2$ separated at the

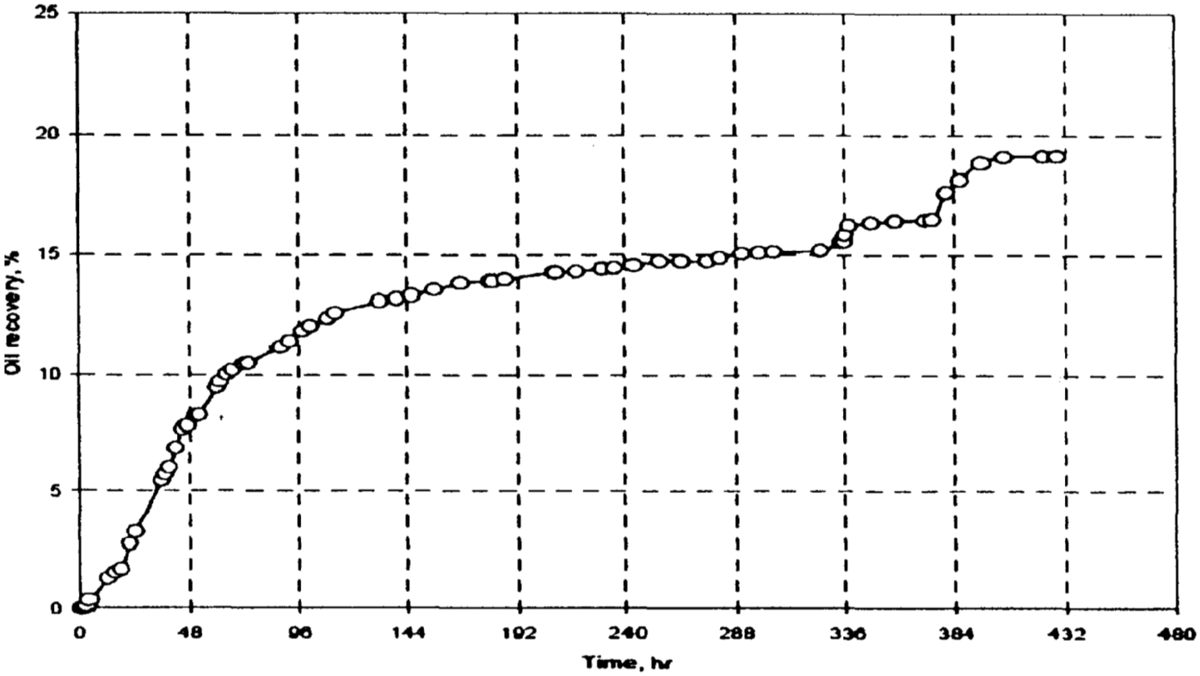


**Fig 6:** Oil recovery by $CO_2$ gravity drainage for core Y

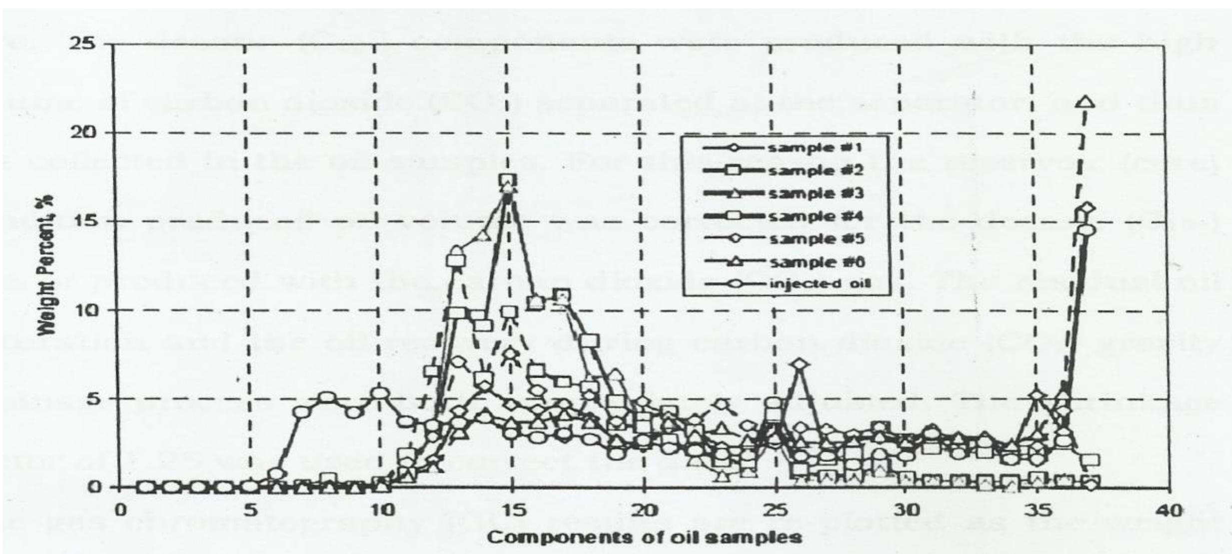


**Fig. 7:** Weight composition of oil samples produced during carbon dioxide ($CO_2$) drainage

separator, and thus not collected in the oil samples. For this reason the reservoir (core) condition produced oil volume was corrected for the decane ($C_{10}$-) loss or produced with the $CO_2$ gas. The residual oil saturation and the oil recovery during $CO_2$ gravity drainage process were both appropriately adjusted. The shrinkage factor of 1.25 was used to correct the data.

The gas chromatography (GC) results are re-plotted as the weight percent of component groups with time as shown in Figure 5.

The figure shows that the undecane to icosane ($C_{11-20}$) components were being disproportionally extracted from the oil by the second sample that was taken during the fourth day after the start of $CO_2$ injection. The undecane to icosane ($C_{11-20}$) component group was roughly 80 wt% of the undecane ($C_{11+}$) oil production until near the end of the test. The last two samples were taken during blow down and had compositions closer to that of the original oil, but with higher heavier ends. These last two samples were probably a combination of oil that had the lighter components stripped by $CO_2$ and oil that had not been contacted or at least

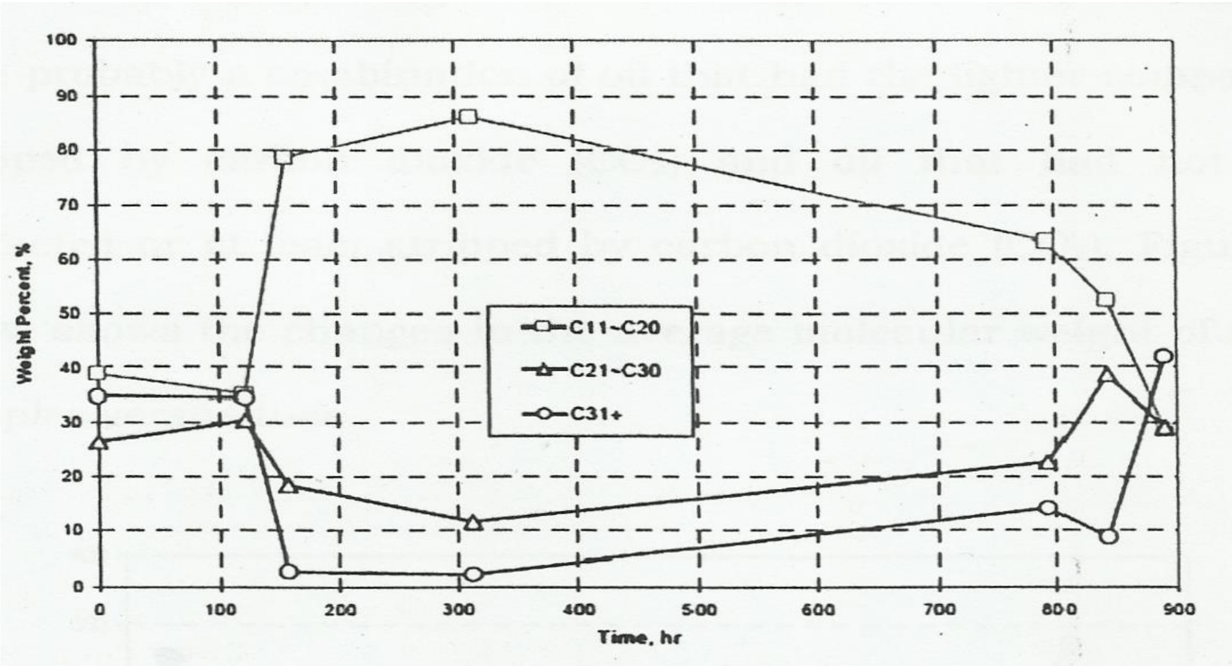


**Fig. 8:** Changes of undecane ($C_{11+}$) component distribution in weight % versus $CO_2$ injection time

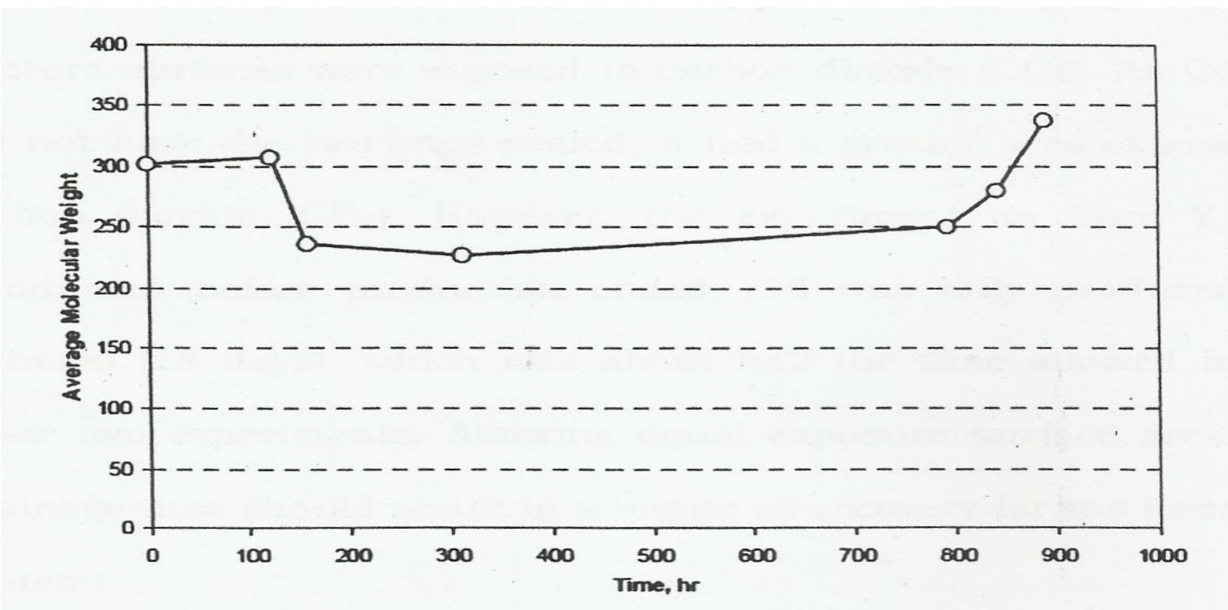


**Fig. 9:** Changes of average molecular weights of undecane ($C_{11+}$) components with time

stripped by carbon dioxide (CU2). Figure 4.6 below shows the changes in the average molecular weight of the oil samples versus time.

**Effects of fractures**

Core # 6 was placed in a gravity drainage cell where all the outside surface area was exposed to $CO_2$ during $CO_2$ drainage, thus, exposing greater surface area. Because of the fractures in Cores X and Y, they were not placed in the gravity drainage cell, but separately in a core sleeve in a core holder. The two ends on Core X were sealed so that only the fracture surfaces were exposed to $CO_2$. As Core Y did not have the two ends scaled, it had a greater area exposed to $CO_2$. However, the experiment on Core Y was terminated before production ended. Oil was only produced for eighteen (18 days), which was about half the time allowed in the other two experiments. Allowing equal exposure surface area and drainage time should result in a higher oil recovery for the fractured systems.

**Table 2:** Properties of the artificially fractured and non-fractured field A Core samples

| Core sample number | 6 | 7 | 8 |
|---|---|---|---|
| Core type | A | X | Y |
| Fractures | 0 | 1 | 5 |
| Sealed end section orientation | No | Yes | No |
| System orientation | Vertical | Vertical | Vertical |
| Length, Cm | 55.25 | 24.45 | 48.74 |
| Diameter, Cm | 10.16 | 10.16 | 10.16 |
| Bulk volume Cm3 | 4476.6 | 1971.3 | 3949.2 |
| Pore volume Cm3 | 1004.0 | 348.0 | 781.0 |
| Porosity% | 22.43 | 17.65 | 19.80 |
| Brine permeability, md | 610.0 | 210.0 | 404.0 |
| Initial water saturation, $S_{wi}$ % | 42.1 | 24.1 | 33.7 |
| Initial oil saturation, $S_{oi}$,1 % | 57.9 | 75.9 | 66.3 |
| Initial oil in core (IOIC) $Cm^3$ | 581.3 | 264.0 | 518.0 |
| Initial water in core (IWIC) $Cm^3$ | 422.7 | 84.0 | 263.0 |
| Aging Time, day | 10 | 5 | 55 |

**Effect of initial water saturstion ($S_{wi}$)**

Comparing the initial water saturations before $CO_2$ injection and the oil recoveries for the three cores, it appears that lower water saturation at the start of $CO_2$ injection results in higher oil recovery. It should be noted that Core X had a lower water saturation when water injection was terminated. The probable reason is that water flow occurred only in the fracture. If allowed to go to termination, Core Y could well have achieved a higher oil recovery by $CO_2$ drainage. Oil recovery, however, may not be as high as Core X, because of the higher water saturation at the start of $CO_2$ drainage, see Table 3a and 3b for details.

**Effect of permeability**

There appears to be a simple relationship between oil recovery and rock permeability (See Tables 4.2, 4.3a and 4.3b). Fractures in a core introduce more complexity into the analysis; however, initial water saturation when carbon dioxide ($CO_2$) drainage starts affects oil recovery more than does permeabity.

**CONCLUSIONS**

From the series of experiment performed, molecular diffusion plays a dominant role in the recovery of waterflood residual oil on the micro or pore scale. In laboratory-scale corefloods used to determine unit displacement efficiency, sufficient contact time must be allowed for diffusion of $CO_2$ to swell the residual oil effectively if high displacement efficiencies are to be realized. This necessitates the use of flooding rates as low as 0.76cm/h [0.6ft/D] for 244-cm [8-ft] cores. Shorter cores requires the use of correspondingly lower flooding rates. It is unlikely that molecular diffusion plays a significant role in reducing diverse effects of large-scale bypassing resulting from gravity segregation, reservoir stratification, and unfavorable mobility ratio (viscous fingering) in tertiary field floods. Although unit or local displacement efficiencies are high as a result the large contact times, overall recoveries may be low because of the presence of large-scale bypassing. $CO_2$ injection could significantly enhance oil recovery after waterflooding in the naturally fractured oil reservoir. The results from these experimental investigations support our subject topic. This study therefore supports previous indications that the efficiency of $CO_2$ injection is significantly affected by the water saturation of the start of $CO_2$ injection. Oil produced during much of $CO_2$ injection process, yields an increased

**Table 3a:** Comparison of results from the artificially fractured and non- fractured field A core samples during water imbibition

| Core Sample Number | 6 | X | Y |
|---|---|---|---|
| Initial oil saturation $S_{wi}$, % | 42.1 | 24.1 | 33.7 |
| Initial oil saturation, $S_{oi}$,% | 57.9 | 75.9 | 66.3 |
| Initial oil in core (IOIC) $Cm^3$ | 581.3 | 264.0 | 518.0 |
| Initial water in core (IWIC) $Cm^3$ | 422.7 | 84 | 263.0 |
| Water saturation, $S_{we}$,% | 61.5 | 36.7 | 64.8 |
| Oil saturation, $S_{oe}$, % | 38.5 | 63.3 | 35.2 |
| Percentage oil volume, $V_{op}$, $Cm^3$ | 194.4 | 43.7 | 242.7 |
| Oil recovery by water imbibition, % | 33.4 | 16.6 | 46.9 |
| Time, day | 20 | 28 | 31 |

**Table 3b:** Comparison of results from the artificially fractured and non-fractured field A core samples during carbon dioxide ($CO_2$) gravity drainage

| Core Sample Number | 6 | X | Y |
|---|---|---|---|
| Initial oil saturation $S_{wi}$, % | 61.5 | 36.7 | 64.8 |
| Initial oil saturation, $S_{oi}$,% | 38.5 | 63.3 | 35.2 |
| Initial oil in core (IOIC) $Cm^3$ | 386.5 | 220.3 | 275.3 |
| Initial water in core (IWIC) $Cm^3$ | 617.5 | 127.7 | 505.7 |
| Residual water saturation, $S_{we}$,% | 38.4 | 8.4 | 47.2 |
| Residual oil saturation, $S_{oe}$, % | 23.4 | 34.1 | 22.8 |
| Percentage Oil Volume, $V_{op}$, $Cm^3$ | 116.3 | 90.5 | 65.6 |
| Oil recovery by $CO_2$ gravity drainage | 30.1 | 41.1 | 23.8 |
| Time, day | 36 | 38 | 18 |

**Table 4:** Total oil recovery during water inibibition and carbon dioxide ($CO_2$) gravity drainage

| Core sample number | 6 | X | Y |
|---|---|---|---|
| Total oil recovery, % | 63.5 | 57.7 | 70.7 |

concentration of components. This indicates significant extraction by $CO_2$ during the $CO_2$ injection. During $CO_2$ injection after waterfooding, water in the sample is immoveable. This is because high water saturation indicates that water could be produced during $CO_2$ injection in oil reservoirs if waterflooding was performed before $CO_2$ injection.

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