

# Carbon-dioxide for EOR in Upper Assam Basin

Subrata Borgohain Gogoi

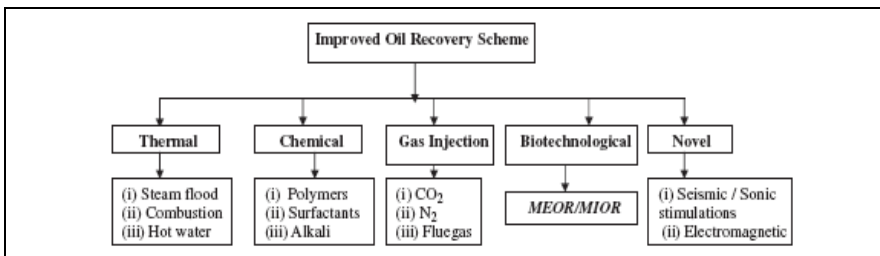
Department of Petroleum Technology, Dibrugarh University, Assam, India

**Abstract.** This paper will deal with the injection of carbon-dioxide (CO<sub>2</sub>) for the purpose of enhanced oil recovery (EOR) in mature and depleted oil and gas reservoirs of Upper Assam basin, India. CO<sub>2</sub> injection system is a procedure used to extract maximum oil from the reservoir. This system is performed through injecting natural gases like CO<sub>2</sub> into the oil wells. The main objective of the CO<sub>2</sub> injection is to stimulate the oil droplets that are inside the oil reservoir rock. Minimum Miscibility Pressure (MMP) is achieved by lowering the viscosity of the oil to make it flow easily to the surface. This paper will calculate the MMP at which miscible recovery takes place. This minimum dynamic miscibility pressure depends upon several factors, such as the composition of injected gas, reservoir temperature and pressure and the characteristics of the oil in place fluids.

**Keywords:** Carbon-dioxide, Upper Assam basin, Minimum Miscibility Pressure, factors.

## 1 Introduction

Petroleum is present in complex capillary networks in underground oil reservoirs. Production in most of the oil fields includes three stages namely, primary, secondary and tertiary or enhance oil recovery (EOR). EOR is defined as oil recovery by injection of fluids not normally present in reservoir which changes the intrinsic properties of the oil by using chemical thermal, Gas etc and excludes pressure maintenance or water flooding. The flow diagram in Figure 1 shows the type of EOR processes that are currently employed in the oil industry [1].



**Fig. 1** Flow sheet diagram showing the process steps in enhanced oil recovery (EOR)

Oil displacement by CO<sub>2</sub> injection depends on the phase behavior of the CO<sub>2</sub> and crude oil mixtures that are strongly dependent on reservoir temperature, pressure and crude oil composition. This method also plays an important role in the reduction of the green house effect representing how utilization of CO<sub>2</sub> can be combined with the rational use of natural resources and the saving of energy sources. These mechanisms range from oil swelling and viscosity reduction for the injection of immiscible fluids (at low pressures) to complete miscible displacement in high pressure applications.

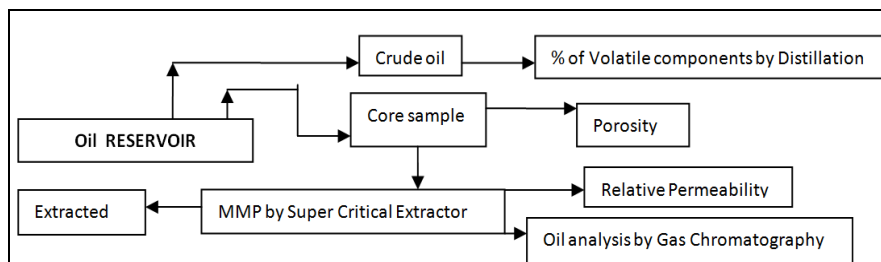
The CO<sub>2</sub> miscible processes are first-contact miscible process or multiple-contact miscibility process. A volume of relatively pure CO<sub>2</sub> is injected to mobilize and displace residual oil. Through multiple contacts between the CO<sub>2</sub> and the oil phase, intermediate and higher-molecular-weight hydrocarbons are extracted into the CO<sub>2</sub> rich phase. Under proper conditions this CO<sub>2</sub>-rich phase will reach a composition that is miscible with the original reservoir oil. From that point, miscible or near-miscible conditions exist at the displacing front interface [2, 3]. To achieve high sweep efficiency CO<sub>2</sub> and water are injected in alternate cycles.

In these applications, half to two-thirds of the injected CO<sub>2</sub> returns to the production well and is usually re-injected into the reservoir. CO<sub>2</sub> vaporizes or extracts hydrocarbon from the crude oil as heavy as the gasoline and the gas-oil fractions. Vaporization occurs at temperatures where the fluid at the displacement front is CO<sub>2</sub>-rich gas, and extraction occurs at temperatures where the fluid at the displacement front is CO<sub>2</sub>-rich liquid.

If reservoir pressure is considerably below MMP, large volumes of CO<sub>2</sub> will be needed to obtain the MMP. A semi-analytical method for predicting the MMP was later presented by Wang and Orr [4] who played an important role in the development and application of the analytical theory of gas injection processes [5]. The density of oil and water are similar in many reservoir conditions, but in reservoirs which are water flooded prior to CO<sub>2</sub> flooding, a density contrast may occur between water and CO<sub>2</sub>, which may cause segregation.

## 2 Experiment

Figure 2 shows the experiments which will be carried out in this work.



**Fig. 2** Layout of the Experiment

### 3 Materials

The core samples were collected from a depth 3929.8 – 3981.5 m, taken from the Oligocene sandstone reservoirs (Barail Formation) of Nahorkatiya in the Upper Assam basin, India. The porosities of the core samples were found to be 21.6%, 23.2% and 24.8% by the Helium Porosimeter, model no. TPI-219 made by Coretest systems. The average air permeability of the core sample was 2.7 mD measured by Temco's Gas Permeameter model no. RCHR-1, TEMCO INC. Tulsa, USA, using Darcy's equation. The core samples have an oil saturation of 33-40% after water flooding.

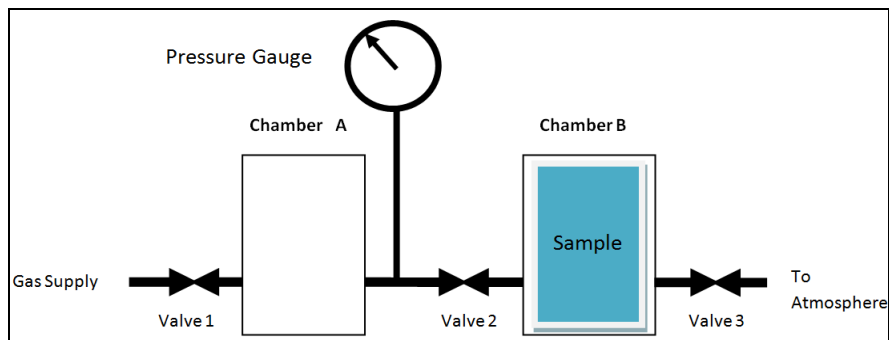
The crude oils obtained from Nahorkatiya oil fields have API gravities in the range of 15-58°API with an average of 30°API with significant wax content of 0.11-22%, low sulphur content and are generally moderately mature. Based on bulk geochemical characteristics the Assam oils are characterized to be from two major groups, (i) naphthenic aromatic and (ii) paraffinic. The former have 15-32° API gravity owing to their coal source while the latter group have 30-58° API. The crude oil for this study has an API gravity of 30° (sp. gr. = 0.88) and a viscosity of 10 cp. A deoiler by the trade name Catflo-T (Cationic Polyelectrolyte) was supplied by Thermax, Puna, India for the separation of the oil from the solvent.

If the temperature and pressure are both increased from standard temperature and pressure (STP) to be at or above the critical point for CO<sub>2</sub> it can adopt properties midway between a gas and a liquid. More specifically, it behaves as a supercritical fluid above its critical temperature (31.1 °C) and critical pressure (73.9 bar), expanding to fill its container like a gas but with a density like that of a liquid.

### 4 Methods

#### 4.1 Porosity Calculation

This method relies on the expansion of helium gas, and also measures the effective porosity of the rock. However, since helium is a slippery little molecule, it can penetrate pores which are much smaller. Hence, this method provides slightly higher porosity measurements on any given rock sample. This method uses the apparatus shown in Figure 3.



**Fig. 3** Porosity Experiment

Plugs from core samples were cleaned and the bulk volumes ( $V_B$ ) were obtained. In each case, valve 2 was closed and the helium gas was introduced into the chamber. A using valve 1 until a pressure  $P_i$  was reached. This pressure was approximately 6.89 bar (100 psi). The plug was inserted in chamber B, which was at atmospheric pressure and valve B was closed. Slowly valve 2 was opened to let the gas equilibrate through both the chambers. The gas penetrates into the pores of the rock sample. During this process the pressure was decreased to a new stable level,  $P_f$ . The drop of pressure depends on how much space there was in Chamber B, and that depends on how much of chamber B was occupied by solid rock particles. The measured gas pressures  $P_i$ ,  $P_{atm}$  and  $P_f$  alone were not sufficient to obtain the volume of the sample rock matrix. However, the system was calibrated by putting a range of metal cylinders of accurately known volume into chamber B and the experiment was repeated. When this was done, the calibration constant, and the pressures allowed the volume of the rock matrix ( $V_M$ ) to be obtained. The porosity was then calculated using the following formula.

Helium porosity,  $\Phi$  calculated as:

$$\Phi = \frac{V_B - V_M}{V_B}$$

Where Bulk Volume =  $V_B$

$$V_B = \frac{\pi}{4} d^2 l$$

$$V_B = \frac{\pi}{4} (2.4)^2 \times 3.4 = 61.56 \text{cc}$$

**Table 1** Calculation of Porosity

Serial no.	$V_B$ (cc)	$V_M$ (cc)	$\Phi = \frac{V_B - V_M}{V_B}$ (%)
1	61.56	48.25	21.6
2	61.56	47.27	23.2
3	61.56	46.29	24.8

#### 4.2 Distillation of Crude Oil (IP/55)

100 ml of the crude oil sample was centrifuged in a closed container to remove the water. The sample was then filtered at atmospheric pressure and heated in a closed steel container fitted with a thermometer and a pressure gauge. The container was filled upto 70% capacity with the sample and heated till the temperature was 200°C. The container was allowed to cool and the water separated by decantation. 100 ml of the sample was distilled in a specified glass apparatus under standard conditions. The volume of the distillate obtained at each multiple of 25°C was recorded, up to a maximum of 300°C when the distillation was stopped. The volumes of distillates collected at 75°C, 100°C, 125°C etc were noted.

### 5 Minimum Miscibility Analysis by Super Critical Extractor

#### 5.1 Preparation of the Sample

Experimental analyses were performed on cylindrical core plug samples of 3.4 cm length and 2.4 cm diameter from depth of 3981.5 m, 3946.5 and 3929.8m. The samples were first placed in an oven for 24 hours in order to remove moisture. The dry and clean samples were weighed and placed in the saturation apparatus. Oil was introduced from one side of the flask, and the vacuum was created by a vacuum pump. The samples were kept for 48 hours in the same air tight flask to achieve proper saturation. After the saturation the excess liquid was removed and the sample was weighed. The weight of the saturation oil was calculated.

#### 5.2 Test Procedure

The core plug samples were transferred one by one into the extraction vessel. First supercritical CO<sub>2</sub> at 31°C and pressure of 74 bar was injected into the sample in the vertical position. The samples were 100% saturated with reservoir oil at 70°C, 60°C and 50°C respectively in the pressure difference range between the inlet and exit of the extraction vessel of 100 – 430 bar and a constant flow rate of 4L/min

was maintained. This is an unsteady state operation at a constant flow rate. The extracted oil samples were collected at different pressures in the sampling vessels. The oil of the sampling vessels was separated by using a deoiler by the trade name Catflo-T (Cationic Polyelectrolyte). The separated oil samples were assessed both quantitatively and qualitatively. Quantitative assessment was done volumetrically, while qualitative assessment was done in the Gas Chromatograph (GC).

### 5.3 Equipment Operation

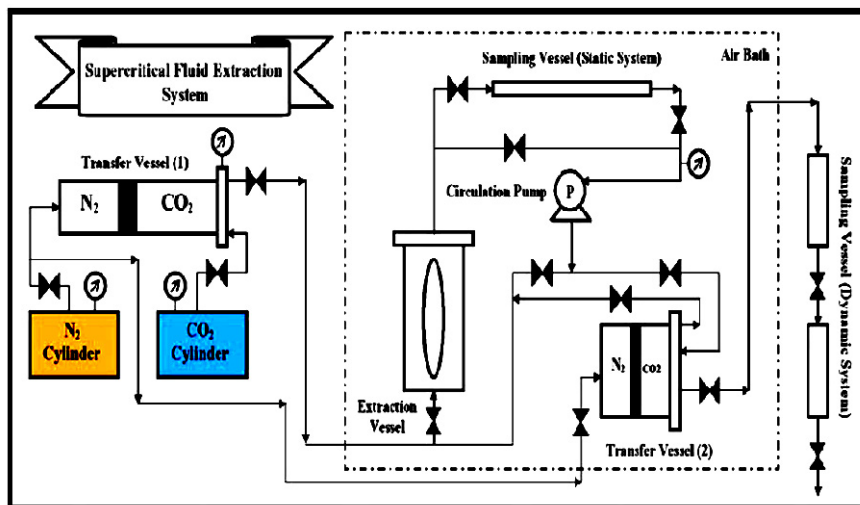
To remove the oil extracted from the core sample by supercritical fluid CO<sub>2</sub>, a thermodynamic machine was required to extract the oil at high pressure. The design method of the Supercritical fluid extractor (SCFE) has been presented by different researchers [6-8]. In this research, SCFE can be used for separation and extraction of oil from plugs using SCFE. As it can be seen in Figure 4, this system can function in static and dynamic conditions. In this system two specially designed Transfer Vessels were used to provide system pressure using Nitrogen (N<sub>2</sub>) gas. The possibility of establishing the flow of CO<sub>2</sub> gas in the machine in two different directions by fixing the existing valve in the machine, is a characteristic of the system. The extraction vessels were made of stainless steel like other parts of the system which can resist up to 689.5 bar (10,000 psi) pressure. The container has a side glass made of silicon material, and can withstand high pressures. The ability of the designed pump for rotating the supercritical fluid within the system is another unique feature of this machine in comparison to other devices. The mechanical part of this pump was designed and made manually. It has the ability to pump a two-phase fluid in a thermal range up to 120°C and with a flow rate of 4 L/min, regardless of the creation of Cavitations in the system, can be tuned by operator. This device uses an air bath system to provide temperature. The designed air bath was able to provide a uniform temperature of 120°C.

Extracted oil samples were collected at 100, 130, 160, 180, 200.....430 bar pressures in the sampling vessels as in Figure 4. The results were interpreted graphically and the MMP was obtained. The composition of the oils extracted was determined by GC analysis. The extraction yield (Y) in supercritical carbon dioxide extraction (SC-CO<sub>2</sub>) was calculated by equation [9]:

$$Y = \frac{m_{ext}}{m_{mat}} \times 100\%$$

where:  $m_{ext}$  = mass of extract,  
 $m_{mat}$  = mass of raw material.

This review provides a detailed and updated discussion of applications of SC-CO<sub>2</sub> extraction in the EOR of crude oil from the underground porous medium.



**Fig. 4** Schematic diagram of the Supercritical Fluid Extraction System (SCFE) for Extraction of oil from core samples with supercritical carbon dioxide

#### 5.4 Gas Chromatography (GC)

The GC analysis of oil samples was carried out in a *Thermo Fisher TRACE GC* coupled to a *DSQ, 2005*. In order to carry out GC analysis 0.05g of methylheptadecane was taken and diluted with 100ml of pentane reagent (grade 98%). Oil samples by SC- $CO_2$  collected from the SCE at a fixed temperature (50°C, 60°C, 70°C) and at 180 bar were diluted in test tubes so that the tubes were half filled with samples. The samples were injected into the heater zone, vaporized and transported by carrier gas helium into a packed column, which contained partitions for separating different samples. The column partitioned the components according to their boiling points, the eluted compounds were then carried by carrier gas into a detector, where their concentration was related to the area under the detector response-time curve. Individual peaks can be identified by comparing their retention times inside the column with those of known compounds previously analyzed under the same GC condition [10].

## 6 Results and Discussions

### 6.1 Distillation of Crude Oil (IP/55)

The results predict the crude oil sample contains 44.6% distillates up to 300°C and are in Table 2.

**Table 2** Showing the distillates in crude oil upto 300°C

Serial no.	Temperature range (°C)	Cumulative Distillate obtained (% vol)	Carbon no.
1.	0-75	3	C <sub>1</sub> -C <sub>4</sub>
2.	75-100	8	C <sub>5</sub> -C <sub>12</sub>
3.	100-125	13	C <sub>5</sub> -C <sub>12</sub>
4.	125-150	20	C <sub>5</sub> -C <sub>12</sub>
5.	150-175	24	C <sub>5</sub> -C <sub>12</sub>
6.	175-200	28.5	C <sub>5</sub> -C <sub>12</sub>
7.	200-225	33	C <sub>12</sub> -C <sub>16</sub>
8.	225-250	36.5	C <sub>12</sub> -C <sub>16</sub>
9.	250-275	43.5	C <sub>16</sub> -C <sub>19</sub>
10.	275-300	44.6	C <sub>16</sub> -C <sub>19</sub>

It was important to know the amount of volatiles because when CO<sub>2</sub> is injected into an oil reservoir, volatiles or light hydrocarbons from the oil dissolve into CO<sub>2</sub>. This occurs most readily when the CO<sub>2</sub> density is high i.e., when it is compressed. Below some minimum pressure, CO<sub>2</sub> and oil will no longer be miscible. As the reservoir temperature increases the density of CO<sub>2</sub> decreases or as the oil density increases due to the decrease of the light hydrocarbon fraction the minimum pressure needed to attain oil/CO<sub>2</sub> miscibility increases. For this reason, oil field operators must consider the pressure of a depleted oil reservoir when evaluating its suitability for CO<sub>2</sub> EOR. Low pressured reservoirs may need to be re-pressurized by injecting water.

## 6.2 Minimum Miscibility Analysis by Super Critical Extractor

The results of the tests are shown in Figure 5. The graphs have irregular shapes, the peaks matching the highest rates of recovery were observed at 180 bar and at temperatures of 70°C, 60°C and 50°C. This MMP was far from the value indicated by three slim-tube results for oil that give an MMP of about 74 bar at 31°C [11-14]. The oil recovery rates in the pressure range from 100-430 bar were close at all three temperatures. The graphs for 60°C and 50°C were more similar. The graph at 70°C has more deviations, which do not coincide with the deviations in the curves at the other two temperatures. The oil displaced at all the three temperatures was highest at 180 bar. The increase in oil recovery rates in the range of 100-430 bar was temperature dependent. It was more gradual at 70°C and 50°C, while there was a sharp step at 160 bar for the 60°C curve. The visual observation of the oil samples shows that the viscosity decreases and flows easily as the pressure increases from 100 bar to 180 bar, but after crossing the 180 bar pressure the oil becomes viscous and slowly ceases to flow. During the whole process of extraction 37.39% of oil was extracted at 70°C, 32.27% at 60°C and 25% at 50°C as shown in Table 3.



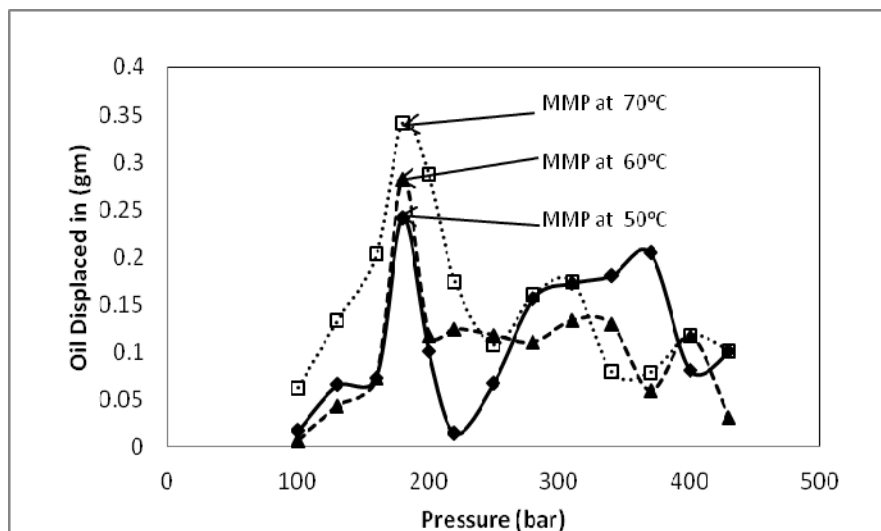


Fig. 5 Amount of Oil displaced Vs saturation pressure at different temperatures

Table 3 Showing the crude oil recovery from the SCE

Temp (°C)	MMP (bar)	Total wt. of oil recovered (gm)	Density of oil (gm/cc)	API gravity of oil (°API)	Vol. of oil recovered (cc)	Saturat ed wt. of core sample (gm)	Dry wt. of sample (gm)	Wt. of oil satura ted in core (gm)	% recovery
70	180	1.92	0.84	37.09	2.29	30.68	25.54	5.13	37.39
60	180	1.48	0.84	37.09	1.76	31.25	26.67	4.57	32.27
50	180	1.34	0.84	37.09	1.60	32.46	27.09	5.37	25.01

It was observed that the miscibility between CO<sub>2</sub> and crude oil requires restricted conditions of temperature and pressure rather than simply dissolving the CO<sub>2</sub> in the oil. Miscibility is attained when the IFT between CO<sub>2</sub> and oil is eliminated. At temperatures of 70°C, 60°C and 50°C the MMP was observed to be 180 bar, below which the interface will remain and it will be an immiscible flooding process. In addition CO<sub>2</sub> and crude oil, because of the difference in their properties and composition, will not become miscible at the first contact regardless of pressure. These materials will have what is called multiple contact miscibility i.e., CO<sub>2</sub> must repeatedly contact the oil. The concentration gradient from oil to CO<sub>2</sub> enables many hydrocarbon molecules specially C<sub>5</sub>-C<sub>30</sub> to leave the oil and enter the CO<sub>2</sub>. After several contacts many of these hydrocarbons will

join the vapour ( $\text{CO}_2$ ) phase, so that the vapour phase becomes miscible with crude oil. At this point the interface will disappear, capillary force will become zero and theoretically about 100% oil can be displaced from the part of the reservoir contacted by  $\text{CO}_2$ .

There is a difference between  $\text{CO}_2$  dissolving in crude oil and  $\text{CO}_2$  being miscible with crude oil. As pressure is applied to  $\text{CO}_2$  – crude oil system, the crude oil will readily dissolve until the oil is saturated with  $\text{CO}_2$  at the existing pressure and temperature. At that time both free  $\text{CO}_2$  and  $\text{CO}_2$  saturated oil will be present with an interface between the two phases. Dissolving the  $\text{CO}_2$  in this manner will result in an expansion of the liquid phase and a reduction of the liquid viscosity. A  $\text{CO}_2$  solution will take place regardless of the composition or API gravity of crude oil. It is obvious that the swelling of the oil will increase the oil saturation and therefore enhance the relative permeability of the rock to oil ( $K_{ro}$ ). In addition the residual oil saturation that will remain after  $\text{CO}_2$  flooding will be swollen with a relatively large volume of  $\text{CO}_2$ . Both reduction of  $\mu_o$  and  $K_{ro}$  will facilitate flow of the swollen oil to the production well.

### 6.3 Gas Chromatography

To determine the hydrocarbon peak distribution in the oil samples obtained from SCE at the value of MMP (180 bar) and at temperatures of  $70^\circ\text{C}$ ,  $60^\circ\text{C}$  and  $50^\circ\text{C}$ , a “standard” chromatography from the selected crude oil sample was run, and peaks from the sample were compared to the known peaks on the standard for identification. Figure 6 to Figure 8 shows the majority of n-alkane distributions for whole-oil gas chromatograms. A broad spectrum of n-alkanes ranging from n-C10 through n-C35 is observed. Modest odd-carbon preference in the n-C15 through to n-C19 range was also observed. The presence of isoprenoids pristane ( $\text{C}_{19}\text{H}_{40}$ ) and phytane ( $\text{C}_{20}\text{H}_{42}$ ) was also detected. In Figures 6 to 8 the heavy

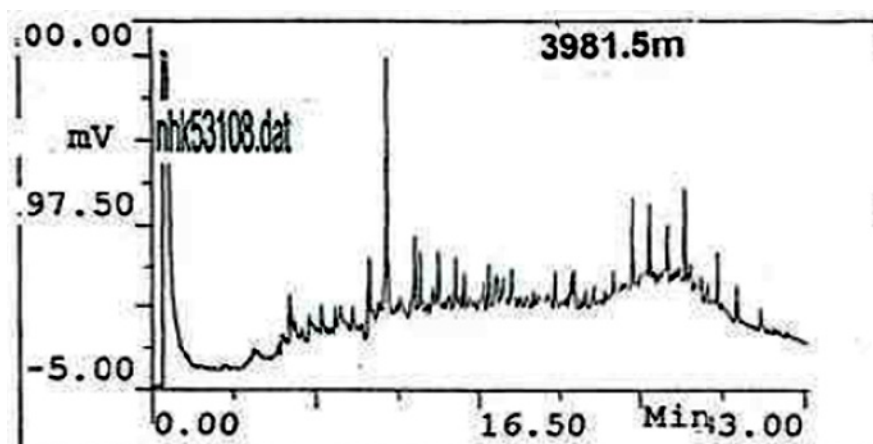


Fig. 6 Shows the GC analysis of oil collected at 180 bar and temperature of  $70^\circ\text{C}$  from the core sample

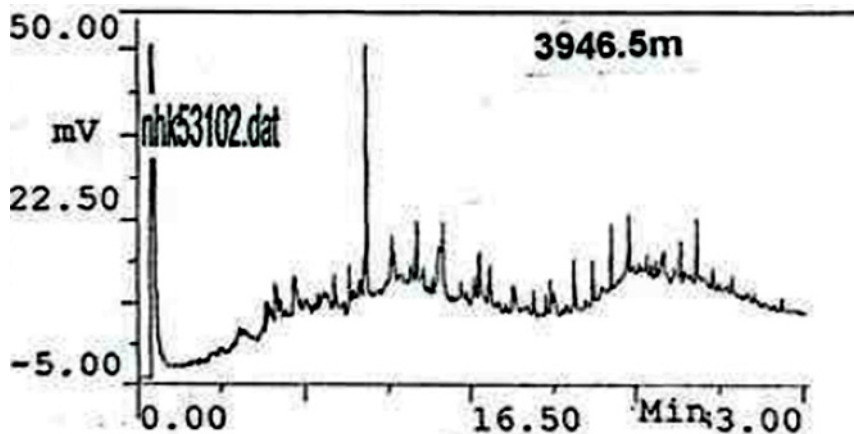


Fig. 7 Shows the GC analysis of oil collected at 180 bar and temperature of 60°C from the core sample

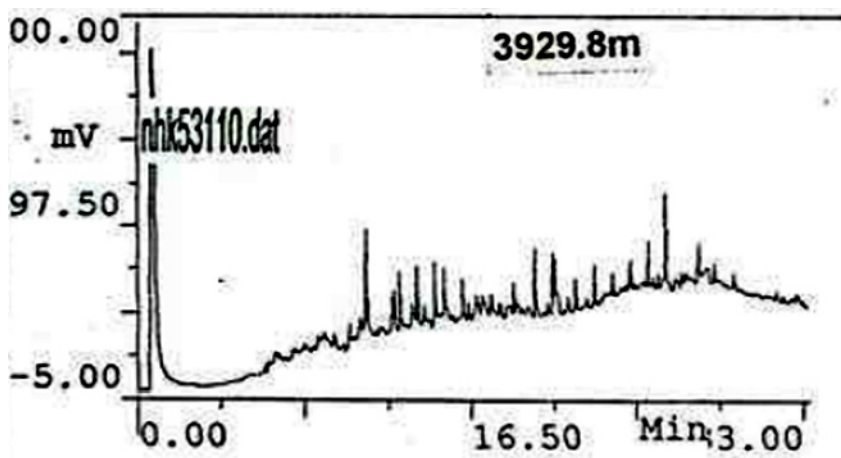


Fig. 8 Shows the GC analysis of oil collected at 180 bar and temperature of 50°C from the core sample

hydrocarbons from C18 to C20 can be seen which is a confirmation that CO<sub>2</sub> can displace heavy hydrocarbons by reducing the viscosity and improving the mobility ratio.

## 7 Conclusion

The percentage of lighter hydrocarbons from C<sub>1</sub>-C<sub>12</sub> in the reservoir crude oil, as obtained from Table 2, was 8% and 44.6% of the distillates (C<sub>1</sub>-C<sub>19</sub>) up to 300°C.

When compressed CO<sub>2</sub> was injected into the plugs of the core at high density samples saturated with crude oil, became miscible with 44.6% distillates of the oil.

From visual observations it was clear that the viscosity of the extracted oil had reduced which indicates that CO<sub>2</sub> displaced the oil from the pores of the rock. As CO<sub>2</sub> dissolves in the oil the oil swells and reduces its viscosity which improves the efficiency of the displacement process.

From the GC analysis (Figures 6, 7 & 8) of the extracted oil samples, it was observed that a broad spectrum of n-alkanes ranging from n-C<sub>10</sub> through n-C<sub>35</sub> was present. Modest odd-carbon preference in the n-C<sub>15</sub> through n-C<sub>19</sub> range was observed and both isoprenoids pristane (C<sub>19</sub>H<sub>40</sub>) and phytane (C<sub>20</sub>H<sub>42</sub>) were also detected. The presence of heavy hydrocarbons from C<sub>18</sub> to C<sub>20</sub> predicts that CO<sub>2</sub> had displaced heavy hydrocarbons by reducing the viscosity and improving the mobility ratio.

The Nahorkatiya crude oil used for the study had an average API gravity of 30°. However, the extracted crude oil from the SCE had an average API gravity of 37° as shown in Table 3, which predicts that the oil became lean due to it swelling from the addition of lighter components

Results of Figure 5 predicts that during the whole process of extraction in the SCE 37.39% of oil was extracted at 70°C, 32.27% at 60°C and 25% at 50°C at MMP of 180 bar in all the cases. This means that the higher the temperature of the plugs the better the oil recovery at MMP will be.

**Acknowledgements.** I would like to express my deepest gratitude to my supervisor, Professor Narendra Nath Dutta, who contributed immensely in carrying out the work. It was my privilege to be his scholar and to complete the work under his supervision.

## References

1. Sen, R.: Biotechnology in petroleum recovery: The microbial EOR. *J. Progress in Energy and Combustion Science* 34, 714–724 (2008)
2. Green, W., Willhite, G.: *Enhanced Oil Recovery*, Richardson, TX, USA. SPE Textbook Series, vol. 6 (1998)
3. Nedjad, M., Reza, A., Vafaie, S.M.: Determination of Minimum Miscibility Pressure by Analytical Method, *Iran. J. Chem. Eng.* 26, 11–17 (2007)
4. Wang, Y., Orr Jr., F.M.: Calculation of Minimum Miscibility Pressure. *J. Pet. Sci. Eng.* 27, 151 (2000)
5. Jessen, K., Standord, U., Stenby, E.H.: Fluid Characterization for Miscible EOR projects and CO<sub>2</sub> sequestration. In: SPE Paper 97192, SPE Annual Technical Conference and Exhibition, Dallas, Texas, October 9-12 (2005)
6. Rudyk, S., Spirov, P., Sogaard, E.: Extraction of Heavy Oil by Supercritical Carbon Dioxide. In: CHISA, Symposium on Supercritical Fluid Applications, C6.4, Serial number 0310 (2010)
7. Rudyk, S.N., Sogaard, E.G., Abbasi, W.A., Jorgensen, L.W.: Determination of minimum miscibility pressure in supercritical reactor using oil saturated sample. *Chemical Engineering Transactions* 17, 1531–1535 (2009a)

8. Rudyk, S.N., Sogaard, E.G., Abbasi, W.A., Jorgensen, L.W.: Determination of minimum miscibility pressure in supercritical reactor using oil-saturated sample. AIDIC Conference Series 9, pp. 253–260 (2009b)
9. Sovilj, M.N., Nikolovski, B.G., Spasojevic, M.D.: Critical Review of Supercritical fluid extraction of selected spice plant materials 30, 197–220 (2010)
10. Curvers, J., Van den Engel, P.: Gas Chromatographic method for simulated distillation up to a boiling point of 750<sup>0</sup>C using temperature-programmed injection and high temperature fused silica wide-bore columns. J. High Resolution Chromatography 12, 16–22 (1989)
11. Rahmatabadi, K.A.: Advances in Calculation of Minimum Miscibility Pressure. Dissertation for Doctor of Philosophy, The University of Texas at Austin (2011)
12. Creek, J.L., Sheffield, J.M.: Phase Behavior, Fluid Properties and Displacement Characteristics of Permian Basin Reservoir Fluid/CO<sub>2</sub> Systems. J. SPE Reservoir Engineering 8(1), 34–42 (1993)
13. Fong, W.S., Sheffield, J.M., Ehrlich, R., Emanuel, A.S.: Phase Behavior Modeling Techniques for Low-Temperature CO<sub>2</sub> Applied to McElroy and North Ward Estes Projects. In: Proceedings of SPE/DOE Enhanced Oil Recovery Symposium (1992)
14. Winzinger, R.: Design of a Major CO<sub>2</sub> Flood, North Ward Estes Field, Ward County, Texas. SPE Reservoir Engineering 6(1), 11–16 (1991)