OIL RECOVERY PERFORMANCE IN THE CO-INJECTION FLOODING OF CO_2 WITH NON-POLAR CHEMICAL MODIFIERS USING MICROMODEL SYSTEM

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DECLARATION

I hereby declare that the materials in this thesis are original except for quotations, excerpts, summaries and references, which have been duly acknowledged

13th May 2019

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ABSTRACT

This research work investigated the possibility for the significant influence of the addition of non-polar chemical modifiers with CO_2 on improving the extraction capability of CO_2 with crude oil, with regards to the solvents rule of 'like dissolves like' by using a micro-model system. Micromodel system allows oil recovery to be studied at the pore scale which determines the large scale flow patterns of oil reservoirs. Thus, by optimizing oil recovery at the pore scale, ultimate oil recovery can be achieved. Other than that, not all oil reservoirs in Malaysian waters are operating at miscible conditions. Thus, the research also investigated the performance of the co-injection of CO_2 and non-polar chemical modifiers at both immiscible conditions and miscible conditions. An economic analysis had also been conducted from the oil recovery data obtained from the experiments to evaluate the potential of the technology to be implemented in the Malaysian reservoirs. From the experiments conducted using micro-model system at immiscible conditions, it was found that extraction of crude oil improved with the addition of non-polar chemical modifiers with the highest oil recovery achieved of 52.54% by co-injection of gaseous CO_2 and toluene, as compared to oil recovery of 27.73% by gaseous CO_2 alone after 20 Pore Volume (PV) of fluid injected. At miscible conditions, the extraction of crude oil also improved with the addition of non-polar chemical modifiers with the highest oil recovery, 71.87%, achieved by co-injection if CO_2 and toluene, as compared to oil recovery of 44.77% by injection of supercritical CO_2 alone after 20 Pore Volume (PV) of fluid injected. From the economic analysis conducted, it can be concluded that miscible process is more economically attractive with injection of supercritical CO_2 at miscible conditions producing profits of RM582.36, whereas its counterpart, gaseous CO_2 at immiscible conditions only produces profit of RM394.47, with the same amount of fluids injected, 16000 litres (L).

ABSTRAK

PRESTASI PEROLEHAN MINYAK MENTAH DALAM SUNTIKAN BERSAMA KARBON DIOKSIDA DENGAN PENGUBAH-SUAI KIMIA BUKAN 'POLAR' *DENGAN MENGGUNAKAN SISTEM MIKRO-MODEL*

Kerja penyelidikan ini menyiasat kebarangkalian pengaruh ketara penambahan pengubah-suai kimia bukan 'polar' dengan karbon dioksida dalam meningkatkan kebolehan pengekstrakan karbon dioksida dengan minyak mentah, dengan menggunakan peraturan pelarut 'sama melarutkan sama' dengan menggunakan sistem mikromodel. Sistem mikromodel membolehkan peningkatan perolehan minyak mentah dipelajari dalam skala liang yang menentukan corak aliran skala besar dalam telaga minyak. Dengan mengoptimakan peningkatan perolehan minyak mentah pada skala liang, peningkatan perolehan minyak mentah muktamad dapat dicapai. Selain itu, ia juga dapat diperhatikan bahawa tidak semua telaga minyak mentah dia perairan Malaysia beroperasi dalam keadaan terlarut campur. Oleh itu, penyelidikan ini juga menyiasat prestasi suntikan bersama karbon dioksida dengan pengubah-suai kimia bukan 'polar' dalam keadaan terlarut campur dan keadaan tidak terlarut campur. Daripada eksperimen yang dijalankan dengan sistem mikromodel, dalam keadaan tidak terlarut campur, ia dijumpai pengekstrakan minyak mentah meningkat dengan penambahan pengubah-suai kimia bukan 'polar' *dengan perolehan minyak 52.54% dengan suntikan bersama karbon dioksida dan toluena, berbanding perolehan minyak 27.4% dengan suntikan karbon dioksida sahaja dalam fasa gas selepas 20 isi padu liang cecair disuntik. Dalam keadaan terlarut campur, pengekstrakan minyak mentah juga meningkat dengan penambahan pengubah-suai kimia bukan* 'polar' *dengan perolehan minyak tertinggi, 71.87%, dengan suntikan bersama karbon dioksida dan toluena, berbanding perolehan minyak 44.77% dengan suntikan karbon dioksida sahaja dalam fasa superkritikal selepas 20 isi padu liang cecair disuntik. Oleh itu, dapat disimpulkan bahawa suntikan bersama karbon dioksida dengan pengubah-suai kimia bukan 'polar' dapat meningkatkan perolehan minyak mentah dalam keadaan terlarut campur dan keadaan tidak terlarut campur dan sesuai untuk dilaksanakan di telaga minyak di Malaysia. Daripada analisa ekonomi yang dilakukan, proses dalam keadaan terlarut ialah paling menarik dari segi ekonomi dengan, ialah suntikan karbon dioksida superkritikal dalam keadaan terlarut dengan keuntungan RM582.36, manakala rakan proses yang sama, gas karbon dioksida dalam keadaan tidak terlarut hanya membuat keuntungan sebanyak RM394.47 dengan menggunakan jumlah bahan mentah yang sama, 16000 liter.*

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LIST OF ABBREVIATIONS

AP	Alkaline-Polymer
AOS	alpha olefin sulfonates
API	American Petroleum Institute
ASP	Alkaline-Surfactant-Polymer
BHFP	Bottom Hole Flowing Pressure
BOPD	Barrels of oil per day
CCS	Carbon Capture and Storage
cP	centrePoise
CTAB	cetytrimethylammoniumbromide
DEC	diethylcarbonate
DMC	dimethylcarbonate
EOR	Enhanced Oil Recovery
GC-FID	Gas Chromatography-Flame Ionisation Detector
HP	horsepower
HPAM	Hydrolyzed Poly-Acrylamide
IOIP	Initial Oil In Place
IOR	Improved Oil Recovery
ISCGHP	In-Situ-CO_2-Generation-Huff-and-Puff
LAPB	Lauramidopropyl Betaine
MMP	Minimum Miscible Pressure
MPa	mega Pascal
MRI	Magnetic Resonance Imaging (MRI)
NA	Not Available
NP	nano-particles
NYMEX	New York Mercantile Exchange
OOIP	Original Oil In Place
PEF	Polymer Enhanced Foam
PETRONAS	Petroliam Nasional Berhad
PV	Pore Volume
PVI	Pore Volume Injected
RF	Recovery Factor
ROIP	Residual Oil in Place
SAG	Solution Alternating Gas
SC	supercritical
SDS	Sodium Dodecyl Sulfate
SI	Solubilization Enhancing Indicator
SP	Surfactant Polymer
STB	stock tank barrel
TSCF	trillions of standard cubic feet
VI	Vaporization Enhancing Indicator
WAG	Water Alternating Gas

LIST OF SYMBOLS

A_o	area of oil
A_c	area of carbon dioxide or chemicals
Al_2O_3	aluminium oxide
°C	degree Celcius
$CaCl_2$	calcium chloride
CH_4	methane
C_2H_6	ethane
C_3H_8	propane
C_4H_{10}	butane
C_5H_{12}	pentane
C_6H_{14}	hexane
CO_2	carbon dioxide
cm^3	cubic centimetre
CuO	copper oxide
D	Darcy
°F	degree Fahrenheit
H_2S	hydrogen sulfide
km^2	square kilometres
KOH	potassium hydroxide
m^3	cubic metre
mD	mili Darcy
$MgCl_2$	magnesium chloride
N_2	nitrogen
NaCl	sodium chloride
Na_2CO_3	sodium carbonate
Na-DDBS	sodium dodecylbenzenesulfonate
NaOH	sodium hydroxide
ppm	parts per million
Psi	pound force per square inch
S_{gr}	residual gas saturation
S_o	saturation of oil
S_{of}	final oil saturation
S_{oi}	initial oil saturation
S_{or}	residual oil saturation
S_{wi}	initial water saturation
S_{wr}	residual water saturation
SiO_2	silicon dioxide
T_{inj}	injection temperature
TiO_2	titanium dioxide
wt%	weight percent

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Currently, due to the declining production of crude oil in mature reservoirs after extraction by primary and secondary oil recovery, which results in increase of number of idle reservoirs, more focus are directed at the tertiary and final oil recovery which is known as enhanced oil recovery (EOR) in the attempt of recovering more oil from the idle reservoirs. Among the types of EOR technologies used in the industry is chemical injection, gas injection, and thermal injection.

Currently, the potential for EOR technologies in Malaysia is promising. EOR efforts have been kick-started in 2012 by the signing of Production Sharing Contract between PETRONAS and Shell Malaysia for the use of EOR chemical injection technologies with an investment of $USD12 billion for over 30 years to extend the production lives of Malaysia's oldest oilfields (Shell, 2012).

Although, so far, Malaysia has not ventured into EOR projects involving carbon dioxide flooding, its potential for oil production in Malaysia is promising as analysed by Sugiatmo and Idris (1997) with potential additional recovery of 500 million stock tank barrel (STB) of oil. The potential was supported by the fact that Malaysian oil fields are surrounded by large CO_2 reserves with the reserves of CO_2 from K5 gas field located offshore Sarawak and the surrounding area from Natuna gas field (Indonesia) which is located in Greater Sarawak basin.

Initial study by Hui (1995) indicated that the estimated minimum miscible pressure (MMP) of Malaysian reservoirs are in the range of 2300 and 4380 psig, which is higher than Malaysian reservoir pressures. However, the miscibility can be improved and MMP of crude oil and CO_2 can be reduced by blending the CO_2 with solvents such as ethane, propane and butane (Ikhsan *et. al.*, 1997). Another method proposed to reduce the MMP of high temperature reservoir (more than 38°C) is by using cold CO_2 injection which is injected at temperature lower than critical temperature of CO_2 of 31°C which may lead to better efficiency and thus significantly reduce the MMP (Hamdi and Awang, 2014; 2013).

1.2 Problem Statement

In order to improve extraction capability of carbon dioxide (CO_2), in the chemistry industry, joint application of supercritical CO_2 with some chemical modifiers such as alcohols is usually employed. Polar chemical modifiers such as methanol, was well known for its capability in extracting polar components of crude oil such as asphaltic. However, most of the components in crude oils are non-polar, so there is a possibility for the significant influence of the addition of non-polar chemical modifiers with CO_2 on improving the extraction capability of CO_2 with crude oil, with regards to the solvents rule of 'like dissolves like', where polar solvents would dissolve polar solutes, and vice versa (Dobbs *et. al.*, 1986). In the previous studies, it was found that CO_2 extraction accompanied with chemical modifiers can yield crude oil extracts almost 3 times over the CO_2 extraction only (Hwang and Ortiz, 2000).

Therefore, it is worthy of investigating the influence of nonpolar chemical modifiers on the CO_2 injection displacement at the pore scale by using micro-model system for enhanced oil recovery. Through the work in this thesis, enhanced oil recovery through co-injection of carbon dioxide and non-polar chemical modifiers are studied in great detail by using a micro-model system. This is important because the flow on the pore scale decides the large scale flow patterns in the oil reservoirs, and thus by optimizing oil recovery at the pore scale in this research, the ultimate oil recovery can be achieved.

1.3 Research Objectives

1) To investigate the increase in oil recovery during CO_2 flooding, flooding of non-polar chemical modifiers, and co-injection of CO_2 non-polar chemical modifiers with CO_2 at immiscible conditions by using micro-model system.
2) To investigate the increase in oil recovery during CO_2 flooding, flooding of non-polar chemical modifiers, and co-injection of CO_2 non-polar chemical modifiers with CO_2 at miscible conditions by using micro-model system.
3) To conduct an economic analysis of the process of CO_2 flooding, non-polar chemical modifiers flooding, and co-injection flooding of CO_2 and non-polar chemical modifiers in enhancing oil recovery at immiscible and miscible conditions by using micro-model system.

1.4 Novelty of Research

Research works on the utilization of non-polar chemical modifiers in carbon dioxide injection for enhanced oil recovery has not been done yet by any other researchers by using a micro-model system, and by conducting this research, the utilization of nonpolar chemical modifiers in carbon dioxide injection for enhanced oil recovery can be studied in great detail at the pore scale.

Other than that, this research will also investigate the effect of injection of non-polar chemical modifiers on oil recovery which has not been done yet by any other researchers. The findings from this research will also indicate of its potential for application in low and high pressure (20 bars to 85 bars) and low temperature reservoirs (25 to 32°C) in Malaysia as Malaysian crude oil was used in this research.

The improvement of crude oil recovery which is the main objective this research would also help to supply the increasing petroleum and energy demands in Malaysia, and thus can contribute to the society, economy, and nation. Furthermore, the utilization of carbon dioxide would also help to lessen the greenhouse gas emission in the atmosphere by using the carbon dioxide for enhancing oil recovery.

1.4 Scope of Work

The scope of work of this research was to conduct a series of experiments by using micro-model system to investigate the performance of the addition of non-polar chemical modifiers in CO_2 injection in enhancing oil recovery and the effect of miscibility on its performance. Experiments conducted compared the performance of co-injection of CO_2 with injection of CO_2 alone and non-polar chemical modifiers alone as well as oil recovery of the different phases of CO_2 which were gaseous or supercritical and was calculated by image analysis method using Adobe CC 2017 Photoshop software. The second part of the research was to conduct an economic analysis on the technology and process in this research to indicate of its potential for implementation in low temperature and low and high pressure Malaysian reservoirs as Malaysian crude was used in this research and the experiments were conducted at the temperature of 25 to 32°C and pressure of 20 to 85 bar.

The limitation of this research was that the experiments were conducted by using a micro-model glass instead of real core samples from the oil reservoir. Other than that, the glass was fabricated with symmetrical two dimensional imprint instead of being constructed from two dimensional thin-sections of real porous rocks due to the in-availability of the data. However, the glass is still reliable and capable of giving basic accurate data on the oil recovery in a real reservoir and is suitable for this research as the investigation of oil recovery performance at the pore scale with controlled flow conditions of the real reservoir is the main objective of this research.

Another limitation for this research was the temperature and pressure used for the experiments due to the limitation of the micro-model system used in this research. This is because the maximum pressure for the injection by the micro-model system is 120 bar and the maximum temperature that the system can withhold is 70°C. Due to this reason, the experimental temperature for the miscible experiment was chosen to be 32°C with the experimental pressure of 85 bar, to ensure that miscible displacement would occur during the experiments and that the CO_2 would be in supercritical phase at the lowest temperature possible while at the same time ensuring the safety while conducting the experiment by making sure the system would not be over-pressurized.

CHAPTER 2

LITERATURE REVIEW

2.1 Crude Oil

Crude oil is a naturally occurring hydrocarbon that is found accumulated in various porous rock formations within the Earth's crust that is extracted for fuel purposes and processed into chemical products. The colour of extracted crude oil varies from light golden yellow to red, green or most common dark brown to deep dark black. In its natural unrefined state, the extracted crude oil can vary considerably in its density and consistency from a very volatile and thin liquid to extremely thick, semi-solid heavy weight oil. In the United States, it is common practice by the petroleum industry to measure capacity of crude oil by volume and to utilize the English system of measurement. Due to this, in the United States, crude oil is measured in barrels, with each barrel containing 42 gallons of oil. In most other parts of the world, capacity of crude oil is measured by the weight of materials processed and measurements are recorded in metric units. Therefore, outside of the United States, crude oil is measured in metric tons.

2.1.1 Types of Crude Oil

According to Plainsman Manufacturing Inc. (2017), the four main types of crude oil are: very light oils, light oil, medium oil, and heavy fuel oils. It is discussed in **Table 2.1** below.

Table 2.1: Types of crude oil.

Types of Crude Oil	Descriptions
Very light oil	It has a tendency to be very volatile. It evaporates within a few days and as a consequence, evaporates its toxicity levels. Examples of this type of oil are: jet fuels, gasoline, kerosene, petroleum ether, petroleum naphtha, and petroleum spirit.
Light oil	It is moderately volatile and toxic. Examples of this type of oil are Grade 1 and Grade 2 fuel oils, diesel fuel oils and most domestic fuel oils.
Medium oil	It is the most common type of crude oil. It generally has low volatility and higher viscosity than the light oils which in turn leads to higher toxicity and causes greater environmental impact during clean-up.
Heavy fuel oil	It is the most viscous and least volatile crude oil and is the most toxic. Examples of this type of oil are heavy marine fuels and the heaviest Grade 3, 4, 5, and 6 fuel oils.

Source: Plainsman Manufacturing Inc. (2017)

American Petroleum Institute (API) is a major United States trade association for oil and gas industry. One of the most important standards that was set by API is the API gravity which is a method used to measure the density of petroleum ("Petroleum - Classification - API", 2015). API gravity is calculated using the specific gravity of oil, which is the ratio of density of the oil relative to that of water. Specific gravity values for API measurements are always measured at 60 degrees Fahrenheit (^{0}F). **Equation 2.1** below shows the formula for API gravity.

$$\text{API Gravity} = \frac{141.5}{\text{Specific Gravity}} - 131.5 \qquad \textbf{Equation 2.1.}$$

The API gravity is used to determine the categories of crude oil such as light, medium, heavy and extra heavy. API gravity is exceptionally important as the 'weight' of the oil is the biggest determinant of its market value. The API values for each category of crude oil are shown in **Table 2.2.** In the industry, less dense or 'light' oil is preferable as compared to more dense oil as it contains more quantity of hydrocarbons that can be converted to gasoline and diesel fuel.

Table 2.2: API Gravity values of different types of crude oil.

Type of Crude Oil	API Gravity
Light	API more than 31.1^0
Medium	API between 22.3^0 and 31.1^0
Heavy	API less than 22.3^0
Extra Heavy	API less than 10^0

Source: "Petroleum - Classification - API" (2015)

2.1.2 Crude Oil Composition

Composition of crude oil varies depending on the location of the oil reservoir and the process performed to extract the oil. According to The Editors of Encyclopedia Britannica (2017), crude oil is a mixture of relatively volatile liquid hydrocarbon, and contains some oxygen, nitrogen and sulphur. Regardless of variations, almost all crude oil ranges from 82 to 87 percent carbon by weight and 12 to 15 percent hydrogen by weight.

Table 2.3 shows the hydrocarbon composition in mass percentage (% mass) of different types of crude oil from various parts of the world. As can be seen in the table, hydrocarbon composition varies according to the type of crude oil and the location of crude oil retrieved, with light crude oil having C12+ components within the range of 41-81 (%mass), while medium crude oil within the range of 74-84 (%mass) and one sample of heavy crude oil having 74.14 (%mass) of C12+ components.

The composition of the crude is also used to categorize the crude oil. This is because crude oil is sometimes categorized as sweet or sour depending on the amount of sulphur in its composition, which occurs as elemental sulphur or in compounds such as hydrogen sulphide. Sweet crudes contain 0.5 percent or less of sulphur, while sour crudes contain more than 1 percent of sulphur. During refining, excess sulphur is usually removed from the crude as sulphur oxides released into the atmosphere during combustion of oil are major pollutants.

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