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Research Article
Fatty Acid Methyl Ester from Rubber Seed Oil as Additives in Reducing the Minimum Miscibility Pressure of CO_2 and Crude Oil

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Abstract

Background and Objective: Malaysian reservoir is one of the best candidate to perform CO_2 flooding with abundant CO_2 gas presence in the reservoir and the light crude oil. The high temperature in Malaysian reservoir leads to high miscibility pressure; higher than the fracturing pressure of most reservoirs, which hindering the CO_2 miscible displacement to be achieved. Injection at Minimum Miscibility Pressure (MMP) is crucial in maximizing oil recovery for miscible gas flooding. Chemicals as co-solvent with crude oil and CO_2 such as alcohol has been proved successfulin reducing the MMP. Regardless, the cost and residue of this chemical in crude oil prevent the mass application of this chemical in the reservoir. In addition to the depletion of non-renewal fossil feedstock as chemical sources, renewable biomass source is seen as a promising alternative of a chemical commodity due to abundantly available and biodegradable. The main aim of this study is to study the potential of using chemicals extracted from biomass in EOR. Fatty Acid Methyl Ester (FAME) is one of the main chemicals derived from the biomass by the fatty acid transesterification process. **Materials and Methods:** The ability of the fatty acid methyl ester inenhancing the miscibility between CO_2 and Malaysian crude oil was tested with the slim tube test conducted at pressure ranging from 18 MPa up to 31.03 MPa and ata constant temperature of 90°C. Carbon dioxide gas with 99.99% purity was used to eliminate any impurities that might affect the result. **Results:** Injection of 10% of slug ofthe renewable FAME as additive shows an MMP reduction of 15% lower than the MMP of the crude oil without the additive. **Conclusion:** With this finding, alternative green chemical sources have been found, which indirectly utilizing the use of biomass and which is suitable for mass application in the reservoir.

Key words: Enhanced oil recovery, CO_2 miscible flooding, chemical enhanced oil recovery, minimum miscibility pressure, slim tube test, biomass, rubber seed, FAME, tapis crude oil, methyl linoleate

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

One of the widely used Enhanced Oil Recovery (EOR) methods is miscible gas displacement and CO_2 gas is often used as the injected gas due to its inert characteristics, cost and abundant. Gas injection processes are most effective when the gas injected is at near or completely miscible in the oil reservoir[1]. The most important parameter in designing a gas miscible injection is the Minimum Miscibility Pressure (MMP). There are various definitions on the MMP have been mentioned in the literature previously. The definition of the MMP was the pressure at which the ultimate recovery approached 100%[2-4], the MMP also determined at the break over point on the plot of recovery versus pressure[5]. While others define MMP where the oil recovery of 80-90% on the plot[6].

The MMP is dependent on many factors; reservoir conditions such as the temperature and pressure of the reservoir and the composition of crude oil and injected gas[7,8]. Most of the Malaysian reservoirs have high temperature and since MMP is a direct function of temperature and its increases linearly corresponding to the temperature[9]. High MMP in these type of reservoirs and most of the time achieving miscible flooding is impossible. Estimated MMP for Malaysian crude is in the range of 2300-4380 psi[10] which is higher than reservoir fracturing pressure. Injection above fracture formation pressure resulting to fracture which leads to CO_2 loss resulting to the failure of displacement.

Carbon dioxide miscibility with crude oil isusually utilized with the addition of chemicals. In this method, chemicals that have solubility with crude oil and CO_2 gas were selected in enhancing the miscibility thus lowering the MMP in reservoir condition. Alcohol has been a popular example of a chemical additive[9,11-16] with CO_2. The polar structure owns by alcohol responsible for dipolar bonding between molecules[14]. By analogy, CO_2 is not a good solvent at high temperatures where dispersion interactions are dominant[17]. Due to the structural symmetry, CO_2 does not have a dipole moment, but it does have a substantial quadrupole moment due to the carbon and oxygen electron affinities differences[18].

Currently, with the depletion of the non-renewable chemical's resources, biobased chemicals, which produced from the renewable resources appear to be a great alternative[19,20]. The fatty acid is one of the potential feedstocks for fuels and chemical manufacture in the future[21-23]. About 15-20% of biomass are made of oil and fatty acid, which comprising of carbon, hydrogen and oxygen[24]. The FAME is considered as "green solvent" because it is biodegradable, environmentally safe and non-toxic[25-27]. It is well known that polar chemicals such as alcohol can improve the miscibility between CO_2 and crude oil[28], however, considering that crude oil mostly made of non-polar component[29,30], the presence of a long carbon chain in FAME could significantly influence the equilibrium of CO_2-crude oil system.

Malaysia is one of the largest rubber producer in the world with abundant rubber trees plantations in Malaysia, which cover more than 1.2 million hectares all over Malaysia[31,32]. The oil rubber seeds yield a good amount of oil between 40-50% weight[33]. Rubber seed has a potential as a good feedstock with high free Fatty Acid Content (FFA) of 45% for FAME production[27].

In this study, an experimental approach is adopted to study the application of FAME extracted from rubber seed oil on enhancing miscibility between CO_2 and crude oil. Despite of there are many chemicals have proposed to be used as additives in reducing the MMP, this is the first time to the knowledge where the FAME from biomass has been used for the purpose mention. With the findings, a cheaper chemicals and from renewable sources have been found which can be considered for mass reservoir application. These findings also will help in researching for more chemicals from different biomass which are cheaper and safe that can be used in chemical EOR generally.

MATERIALS AND METHODS

Materials: The dead crude oil sample with API 42.90 from Tapis field that was used in this study was obtained from Petronas Refinery at Melaka, Malaysia. The properties and composition of the crude oil used in are provided in Table 1 and 2. The bubble point pressure of the reservoir is 15.81 MPa. Industrial-grade CO_2 with 99.9% purity is used to eliminate external factors that may affect the experimental outcomes.

Table 1: Properties of crude oil at 25°C

Properties	Value
Density (g cm^{-3})	0.8107
Viscosity (cP)	3.9234
API gravity	42.90

Table 2: Composition of crude oil

Component	Mole friction (%)
Pentane	2.6
Hexane	4.38
Heptanes	1.15
Octanes	2.27
Nonanes	5.05
Decanes	6.68
Undecanes plus	77.87
Total	100
Molecular weight (kg mol^{-1})	224.46

Table 3: FAMEs composition analysis for rubber seed oil methyl ester

Formula	IUPAC name	Composition (%)
$C_{17}H_{32}O_2$	Methyl palmitoleate	0.50
$C_{17}H_{34}O_2$	Methyl palmitate	12.56
$C_{19}H_{34}O_2$	Methyl linoleate	32.73
$C_{19}H_{36}O_2$	Methyl oleate	8.68
$C_{19}H_{38}O_2$	Methyl octadecanoate	9.56

Table 4: Properties of methyl linoleate[34]

Chemical	Carbon No.	Formula	Density ($g\ cm^{-3}$)	Molecular weight ($g\ mol^{-1}$)
Methyl linoleate	19	$C_{19}H_{36}O_2$	0.87	296.49

Molecular structure

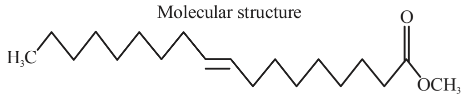

Table 5: Slim tube specifications

Outer diameter (inches)	0.25
Wall thickness (inches)	0.083
Length (ft)	20
Porosity	30%

Slim tube is filled with Ottawa sand 120-270 US Mesh (53-125 µ)

The rubber seed methyl ester was obtained from the Biomass Processing Laboratory, Center of Biofuel Biochemical, Green Technology, Department of Chemical Engineering of Universiti Teknologi Petronas. The FAME compositions of rubber seed methyl ester are provided in Table 3.

Based on Table 3, methyl linoleate made up the most FAME compositions in rubber seed oil methyl ester. The properties of rubber seed methyl ester will be influenced by the properties of methyl linoleate. Thus the properties of methyl linoleate are listed in Table 4.

Apparatus description

CO_2-crude Oil MMP measurement: The MMP measurements of CO_2 with mixtures of crude oil and FAME were carried out with MMP -100 slim tube provided by the core laboratories as shown in Fig. 1. The specifications of the slim tube used are listed in Table 5.

Experimental conditions

Samples preparation: The FAME was dissolved in the crude oil, based on the volumetric ratio method:

- Crude oil (95% by volume)+methyl ester (5%)
- Crude oil (90% by volume)+methyl ester (10%)

The mixture is then stirred at 25°C or 15 min to ensure both fluids totally mix with each other.

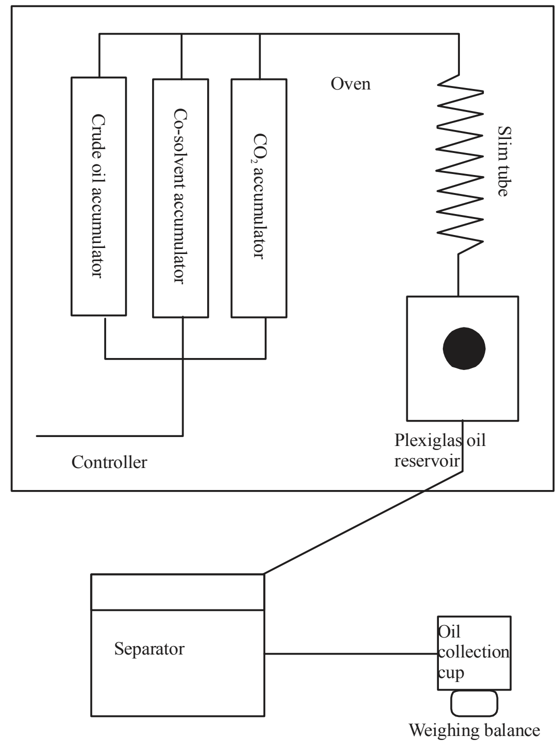

Fig. 1: Schematic diagram of slim tube

Slim tube test: Before the experiment, the slim tube is saturated with crude oil at the tested pressures and temperature. The chemicals then injected until the tested volume required are achieved. When the system achieved the experimental condition, CO_2 was injected at a flow rate of 0.10 $cm^3\ min^{-1}$ until the injected CO_2 volume reached 1.2-1.4 Pore Volume (PV). The crude oil which recovered by CO_2 was collected and weighed. In this study, pressure ranges between 12.41-31.03 MPa and the temperature of 90°C were applied to the samples to simulate the reservoir conditions. The initial measurement was conducted with CO_2 and crude oil as the base case.

MMP measurement: The recoveries are calculated using Eq. 1. In this experiment, the Minimum Miscibility Pressure (MMP) is defined as the lowest pressure at which we have a distinct point of maximum curvature when recovery of oil at 1.2 PV gas injected is plotted versus pressure[10,35]. The recovery at 80 and 90% were also calculated:

$$\text{Oil recovery (\%)} = \frac{V_{\text{Oil recovered}}}{V_{1.2\times\text{pore volume}}} \times 100 \quad (1)$$

RESULTS AND DISCUSSION

MMP reduction with addition of 5% volume of methyl linoleate: Oil recovery for each pressure after the injection of 1.2 PV of CO_2 is plotted and the slope changes correspond to the minimum miscibility pressure. The results of the slim tube test shown in Fig. 2-4.

The MMP value for each test then summarized in Table 6. Generally, the MMP reduction can be observed with the addition of 5% vol., the concentration of FAME. A 4% lower MMP was observed with the addition of methyl linoleate compared to the base case with 16% higher recovery were observed at the same pressure. The addition of methyl linoleate did enhance the property of crude oil inducing the dipole moment with the presence of carbonyl compound in the FAME tail[36,37].

The specific interactions between CO_2 and carbonyl compounds have been extensively studied before[38-40]. Carbon dioxide has many properties that make it an attractive solvent, regardless, CO_2 is a poor solvent for many polar and non-polar compounds although it is miscible with small non-polar molecules at moderate pressure[41]. Thus the term CO_2-philic has been coined to describe the molecules that exhibit high solubility[42] with CO_2.

The most important ability of a compound to interact with CO_2 is to have an ability to be an electron donating, which CO_2 will be the electron acceptor. Kazarian *et al.*[43] in their studies found that, in a temperature sensitive specific interactions, present of certain compound that possess electron donating functional groups such as carbonyls, exhibits specific interactions with CO_2, where the oxygen atoms in oxygen acts as acceptor while carbonyl oxygen acts

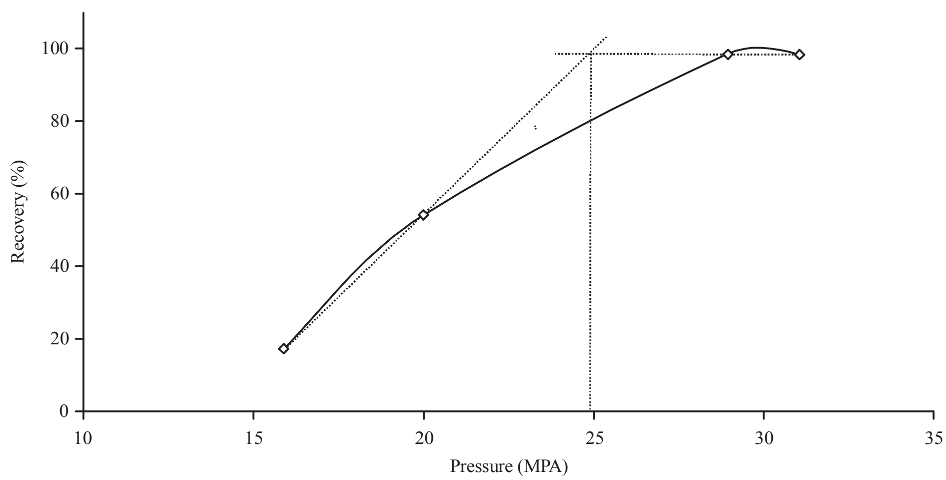

Fig. 2: Oil recovery versus test pressure for base case

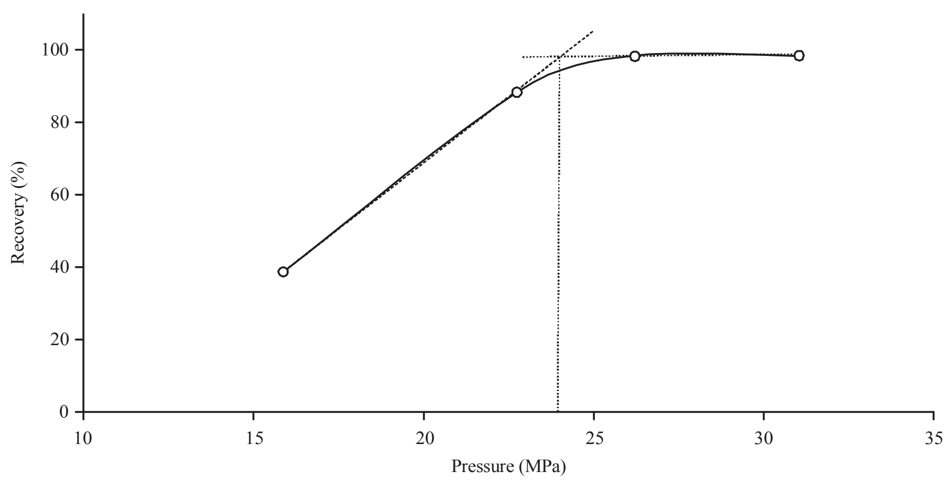

Fig. 3: Oil recovery versus test pressure of crude oil with 5% volume of methyl linoleate

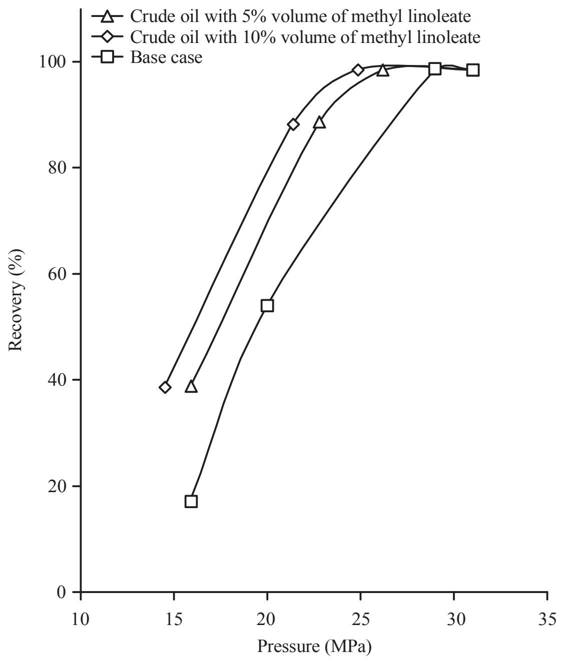

Fig. 4: Oil recovery versus test pressures

Table 6: Slim tube test result for crude oil with FAME

Additives	Concentration (Vol. %)	MMP (MPa)	MMP reduced (MPa)	Reduction (%)
Base case		25.0		-
Methyl linoleate	5	24.0	1.0	4.0

Table 7: MMP summary at 80 and 90% recovery

Additives	Concentration (Vol. %)	MMP (MPa) at 80% recovery	Reduction (%)	MMP (MPa) at 90% recovery	Reduction (%)
Base case		25.0		28.0	
Methyl linoleate	5	21.2	15.2	23.0	17.86
Methyl linoleate	10	20.0	20.0	21.5	23.21

as an electron acceptor. The addition of co-solvent such as of carboxylic acid esters with CO_2, where the carboxylic acid portion has from 2-4 carbon atoms and the ester portion with 1-10 carbon atoms has successfully formed a single phase in CO_2 flooding[44].

The MMP value comparison at 80 and 90% recovery and the effect of concentration. Summaries of MMP reduction for each test are listed in Table 7.

In the literature, the MMP was defined as the pressure at 80 and 90% recovery[2,3,6] while the other resources determined the MMP at the break-even point of the oil recovery versus pressure[45]. Regardless, both definitions were fairly accepted in the industry in determining the MMP. Following the definitions, the value obtained for the MMP of methyl linoleate at 5% vol., concentration was much lower compared to the first part with differences up to 12%. On the other hand, the recovery for the MMP as the first part was much higher at 97%, which higher than recovery according to the definitions. Thus, the MMP values obtain were directly determined by the MMP definition used.

With the results obtain, it's clearly shown that 80% recovery occurred at pressure 20% lower than the MMP of the base case with the addition of 10% of methyl linoleate and an impressive 23.21% lower MMP can be observed at the 90% recovery. Noted that, higher recovery were observed with concentration increases. Regardless of the pressure increases the trend of recovery obtains remained constant, while recovery only shows an increment of 5-6% with the increase in the concentration. Thus, a lower concentration can be used which much more economical for a huge reservoir applications.

CONCLUSION

From the results obtained, the following conclusions can be drawn:

- The addition of rubber seed oil methyl ester has proven to reduce the CO_2-crude oil MMP with methyl linoleate as the main component, shows an MMP reduction of 4% with the addition of 5% vol., concentration with crude oil
- The MMP value was directly determined by the definition used, the MMP difference varied according to the definition applied
- Higher recovery was obtained with the increase in the concentration of methyl linoleate at low pressure, meanwhile, as the pressure increase, a plateau trend can be seen even with added concentration. Thus, a low concentration methyl linoleate can be applied at the reservoir with high temperature and pressure which more economics for a mass application

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