



The impacts of gas impurities on the minimum miscibility pressure of injected CO₂-rich gas–crude oil systems and enhanced oil recovery potential

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Abstract

An effective parameter in the miscible-CO₂ enhanced oil recovery procedure is the minimum miscibility pressure (MMP) defined as the lowest pressure that the oil in place and the injected gas into reservoir achieve miscibility at a given temperature. Flue gases released from power plants can provide an available source of CO₂, which would otherwise be emitted to the atmosphere, for injection into a reservoir. However, the costs related to gas extraction from flue gases is potentially high. Hence, greater understanding the role of impurities in miscibility characteristics between CO₂ and reservoir fluids helps to establish which impurities are tolerable and which are not. In this study, we simulate the effects of the impurities nitrogen (N₂), methane (C₁), ethane (C₂) and propane (C₃) on CO₂ MMP. The simulation results reveal that, as an impurity, nitrogen increases CO₂-oil MMP more so than methane. On the other hand, increasing the propane (C₃) content can lead to a significant decrease in CO₂ MMP, whereas varying the concentrations of ethane (C₂) does not have a significant effect on the minimum miscibility pressure of reservoir crude oil and CO₂ gas. The novel relationships established are particularly valuable in circumstances where MMP experimental data are not available.

Keywords EOR exploiting impure flue gases · CO₂-crude oil minimum miscibility pressure (MMP) · Impact of gas impurities on MMP

1 Introduction

The life cycle of oil production from crude oil reservoirs involves three main stages: primary, secondary and tertiary. During primary recovery, reservoir-related forces, including gas cap expansion, aquifer expansion, dissolved gas expansion, rock and fluid expansion, gravity drainage, or a combination of these mechanisms provides sufficient energy to enable the oil to be produced through wellbores

drilled into the reservoir. The oil production rate decreases as the energy associated with these mechanisms decreases. As a result, some secondary injection methods, mainly gas or water injection, are widely applied to help boost recovery as the impact of primary drive mechanisms decline. Primary and secondary recovery is typically responsible for recovering about 35% of the in situ reservoir oil (Chatterjee 2006). Tertiary recovery methods, or enhanced oil recovery (EOR), when applied, are typically able to recover substantial additional oil from many reservoirs.

Chemical, thermal, microbial and hydrocarbon/non-hydrocarbon gas flooding methods are all successfully deployed as enhanced oil recovery methods (Green and Willhite 1998). In miscible flooding, various gases are injected into a crude oil reservoir leading to enhanced oil production rates caused by several mechanisms such as viscosity reduction and oil expansion (Ennin and Grigg 2016; Fathinasab et al. 2015; Lv et al. 2016). Among various gases, CO₂ is most commonly selected because it can establish miscibility by dissolving in reservoir oil at a

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relatively low minimum miscible pressure (Alomair and Garrouch 2016; Choubineh et al. 2016). Minimum miscibility pressure (MMP) plays an important role in the selection of which gas to use and in operational issues. MMP is defined as the minimum pressure at which reservoir oil and injected gas meet miscibility at a fixed temperature. Therefore, at this pressure, interfacial tension between the two fluids is approximately zero and a miscible condition is established between them. CO₂ injection into the subsurface also offers benefits for oil recovery from unconventional reservoirs (Wan and Liu 2018) and for CO₂ sequestration (Kalra et al. 2018).

There are two processes commonly used to develop miscibility during gas injection: (1) first contact miscibility (FCM) or (2) multi-contact miscibility (MCM). Where the reservoir oil and injected gas reach a single phase upon their initial contact at any arbitrary ratio, they are considered to be first contact miscible (Green and Willhite 1998). In most conditions, oil and gas cannot achieve first contact miscibility, yet they develop miscibility by means of continuous mass transfer between phases (Green and Willhite 1998). Multi-contact miscibility can occur in three different kinds of displacements: (1) a vaporizing gas drive; (2) a condensing gas drive and (3) a condensing/vaporizing drive (Zick 1986).

Minimum miscibility pressure can be determined by several different procedures, which can be grouped into two general divisions: (1) experimental and (2) non-experimental methods. The slim tube test (Yellig and Metcalfe 1998) is utilized as the standard experimental method to estimate MMP. Other experimental MMP techniques are the rising bubble/falling drop experiments (Zhang et al. 2018), mixing cell experiments (Stalkup 1983), interface or surface laser light scattering spectroscopy (ILLS or SLLS) (Dorshow 1995), the vanishing interfacial tension technique (Rao 1997) and the oil swelling/extraction test (Abedini et al. 2014; Sharbatian et al. 2018). Because the experimental measurement of MMP is time-consuming and costly, it is desirable to establish alternative (non-experimental) methods for determining and/or predicting MMP. Empirical correlations, ternary representation/limiting tie line approach (Whitson and Michelsen 1989), slim tube simulation (Elsharkawy et al. 1992), mixing cell models (Yuan 2003) and analytical methods (Orr 2007) are the five non-experimental approaches widely applied to establish MMP values (Ahmadi-Rahmatabadi 2011).

In recent decades, the effects of various parameters on the MMP of several gas–crude oil mixtures have been studied in some detail. In 1991, Huang and Dyer (1992) performed a laboratory study of the Weyburn reservoir in Canada. They concluded that the miscible displacement process using either pure CO₂ or a mixture of 90.1 mol%

CO₂ and 9.9 mol% methane would be an ideal EOR method for the studied reservoir.

Dong et al. (2000) investigated the impact of solution gas in oil on the MMP of CO₂–crude oil. Their results showed a decrease in the MMP of CO₂–crude oil as the amount of solution gas released from the oil increased and as the ratio of light-to-intermediate hydrocarbon components decreased. The latter, which typically occurs as solution gas, is progressively released from reservoir oil. Zhang et al. (2004) studied the effects of CO₂ impurities on the MMP of CO₂–crude oil for two light oils from Saskatchewan (Canada). Their results indicated that the MMP of CO₂–light crude oil increased as N₂ and/or C₁ concentrations increased in the CO₂ stream.

Sayegh and Huang (2007) studied the impacts of hydrogen sulfide (H₂S) and pressure depletion on the MMP of CO₂–crude oil, revealing an almost linear decrease in MMP with decreasing liberation pressure and increasing amounts of H₂S in the injection gas. This trend was accompanied by precipitation of solids. Vahidi and Zargar (2007) simulated the impacts of temperature, reservoir fluid and injected gas compositions on the MMP of N₂–light crude oil. Their results indicated that the most important factor in achieving miscibility is the ratio of light-to-intermediate hydrocarbon components in the crude oil. A relative increase in the intermediate components leads to a decrease in the MMP. The MMP of N₂–light crude oil was observed to increase slightly as temperature decreased.

Zolghadr et al. (2013) determined MMP between CO₂ gas and three distinct crude oil samples to explore the influence of temperature on CO₂–oil MMP. They concluded that for higher-pressure conditions (5.2 MPa), increasing temperature could lead to an increase in interfacial tension and MMP. On the other hand, this trend was indistinct for cases with pressures up to 5.2 MPa. In 2017, Guo et al. (2017) utilized a special kind of oil soluble surfactant, CAE, in order to decrease interfacial tension between injected gas and the fluid reservoir in efforts to diminish carbon dioxide–oil MMP. The optimum concentration of the CAE surfactant was shown to be 0.2% based on the experimental displacements. In this condition, minimum miscibility pressure was approximately 6.1 MPa lower than pure CO₂–oil MMP.

The effect of methane, changing from 1 to 4 mol% of the gas volume, as an impurity on CO₂ injection in the Bell Creek oil field (USA), was studied by Jin et al. (2017) using the vanishing interfacial method. Due to the restricted amount of gas impurities permitted in operational conditions, they determined an optimal composition range of the gas in order to obtain desired gas injection performance. Asmari and Sari (2018) simulated the influence of temperature and several kinds of impurities on the MMP of CO₂–crude oil. They detected that an increase in

temperature can lead to an increase in the MMP of CO₂–crude oil. Considering pure CO₂ as a base injection gas composition, they determined that the addition of 20% methane increased the MMP of CO₂–crude oil by 86%, whereas the addition of 20% ethane reduced the MMP of CO₂–crude oil by 13%, and the addition of 20% propane diminished the MMP of CO₂–crude oil by 33%.

Injecting the flue gases from power plants into oil reservoirs is an EOR and CO₂ sequestration method attracting significant attention. This article investigates the effects of compositional impurities of common injection gas on the minimum miscibility pressure employing the Eclipse reservoir simulator. The injection gas is pure carbon dioxide (CO₂) or various mixtures of CO₂ with nitrogen (N₂), methane (C₁), ethane (C₂) and propane (C₃). The PVTi Module of the Eclipse simulator (2010) was used for PVT simulation based on real data from an Iranian oil reservoir. The minimum miscibility pressure for different gas compositions (i.e., CO₂ plus various gas impurities) was estimated using customized codes written in Eclipse 300. The results obtained enable the impacts of the various gas impurities on the MMP of CO₂–crude oil to be compared and ranked. These results are relevant to projects planning to inject flue gases released from power plants into sub-surface reservoirs located in the vicinity of the power plants.

2 PVT simulation of experimentally determined measurements

In order to simulate the experimental tests carried out in the laboratory, the fluid PVTi Module, PVTi (2010 version), of the Eclipse Simulator (Schlumberger 2018) was employed. The PVTi equation of state (EOS)-based Eclipse simulator module was used to describe the various fluid samples. The experimental data generated comprised the relative volume, total formation volume factor, gas formation volume factor, gas gravity, gas oil ratio, liquid density and liquid viscosity.

2.1 Reservoir description

The oil sample was taken from an Iranian oil field at bottom hole sampling conditions. The oil gravity of the sample was approximately 30° API. The bubble point pressures at 135 (separator temperature) and 230 °F (reservoir temperature) were established as 4049 and 4449 psia, respectively. Table 1 displays the oil composition obtained from the laboratory analysis.

After splitting the plus fraction by the Whitson method, the hydrocarbon and non-hydrocarbon components were assembled into eight different groups (Table 2).

Table 1 Initial reservoir fluid composition of a 30° API crude oil from an Iranian oil field

Component	Mole percent, %
Nitrogen (N ₂)	0
Methane (C ₁)	49.665
Ethane (C ₂)	8.566
Propane (C ₃)	4.803
<i>i</i> -Butane (<i>i</i> -C ₄)	0.964
<i>n</i> -Butane (<i>n</i> -C ₄)	2.732
<i>i</i> -Pentane (<i>i</i> -C ₅)	1.109
<i>n</i> -Pentane (<i>n</i> -C ₅)	1.627
Hexanes (C ₆)	3.101
C ₇₊	27.433
C ₇₊ properties	
Specific gravity: 0.8873	
Molecular weight: 228.7	

Table 2 Reservoir fluid compositions expressed as compositional groups

Component	Mole percent, %
Nitrogen (N ₂)	0
Methane (C ₁)	49.665
Ethane (C ₂)	8.566
Propane (C ₃)	4.803
C ₄₋₆	9.533
C ₇₊	13.330
C ₁₄₊	9.061
C ₂₅₊	5.042

Although the crude oil sample did not contain any nitrogen, because our analysis involved checking the impact of nitrogen on MMP, it was necessary to define it as a separate group in the Eclipse PVTi module. The grouping applied for simulation depends upon the crude oil composition involved. Grouping is commonly applied in the PVTi module, because it leads to more efficient simulation than entering each of the molecular components individually. The liquid (C₄₊) molecular components of the groups assembled in Table 2 are further characterized as the pseudo-components listed in Table 3.

2.2 Simulation of the fluid sample

The PVT properties and fluid viscosity were predicted applying the three-parameter Soave–Redlich–Kwong (SRK) equation of state (EOS) and the Lorenz–Bray–Clark (LBC) correlation, respectively. It is necessary to tune the

Table 3 Properties of reservoir fluid pseudo-components

Component	Specific gravity	Molecular weight
C ₄₋₆	0.6313	72.26
C ₇₊	0.7893	136.64
C ₁₄₊	0.8791	248.07
C ₂₅₊	0.9927	498.85

EOS parameters to achieve an acceptable match between measured and simulated values. The PVTi module determines the optimum tuning parameters.

In the PVTi software (a compositional PVT equation-of-state software; 2010 version), there are four different methods for splitting C₇₊ hydrocarbon mixtures. These are constant mole fraction, Whitson, multi-feed and paraffinic–naphthenic–aromatic (PNA) distribution methods. Here, we apply the Whitson method in order to split the mixture into its components, applying the following assumptions:

First single carbon number (SCN) group in plus fraction:

7

Final single carbon number (SCN) group in plus fraction: 45

Number of pseudo-components after regrouping: 3

Critical properties correlation: Kesler–Lee

Acentric properties correlation: Kesler–Lee

Grouping technique: molecular weight

The main purpose of applying PVTi is to achieve an acceptable agreement between PVT experimental data and those obtained by simulation. The other three splitting methods (i.e., constant mole fraction, multi-feed and PNA distribution) did not provide results as consistent as the Whitson method and are therefore not considered further here.

Splitting the plus fraction (usually C₇₊) and subsequently grouping the components is a commonly adopted approach in PVTi. The main purpose of splitting is to better characterize the heavy components of the oil sample and to ensure that they are representative of real crude oil. On the other hand, grouping the previously split components reduces the run time of the ECLIPSE simulator compositional component E300 and leads to more efficient simulation than entering each of the molecular components individually. Here, the gas impurities considered are N₂, C₁, C₂ and C₃; therefore, these components are considered as separate groups. Furthermore, the quantity of these components is considerable. Since the mole fractions of *i*-C₄, *n*-C₄, *i*-C₅, *n*-C₅ and C₆ are quite low, they are combined into one group. The C₇₊ plus fraction selected are

C₇₊, C₁₄₊ and C₂₅₊ based on the outputs of the splitting process.

A reservoir fluid phase diagram before tuning is shown in Fig. 1 and highlights the impact of component grouping (Tables 2 and 3) on this analysis.

The EOS parameters were tuned by regression so as to obtain an appropriate agreement between predicted and observed values. The Ω_A , Ω_B and acentric factor of pseudo-components were selected as regression parameters, and their final values are given in Table 4. Depending on the parameters involved in the regression, several runs were performed to achieve the best matches. The results of the best matches are shown in Fig. 2, showing a match between experimental results and PVTi outputs for the oil sample at the two temperatures (135 °F and 230 °F). This figure is prepared on the basis of experiments of the relative volume, total formation volume factor, gas formation volume factor, gas gravity, gas oil ratio, liquid density and liquid viscosity.

The results listed in Table 5 indicate that the equation of state applied managed to achieve an excellent agreement between simulated and experimental data for the bubble point pressures at 135 and 230 °F.

Figure 3 shows reservoir fluid phase diagram after tuning.

2.3 Measurement of MMP by slim tube simulation (E300)

In the absence of experimental test data of minimum miscibility pressure for the studied reservoir, MMP was estimated using slim tube simulation (E300). That technique involves:

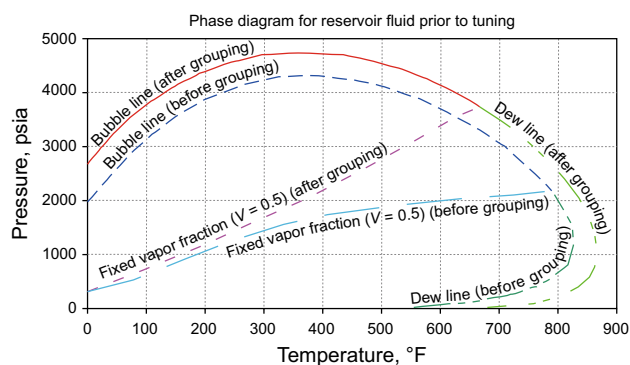


Fig. 1 Phase diagram for reservoir fluid before tuning generated by the PVTi module. The bubble point pressure at 230 °F is approximately 4000 psi before grouping and 4461 psi following the grouping procedure

Table 4 Values of Ω_A , Ω_B and acentric factor obtained from the ECLIPSE PVTi module

Component	Ω_A (Omega-A)	Ω_B (Omega-B)	Acentric factor
N ₂	0.427	0.0866	0.04
C ₁	0.427	0.0866	0.013
C ₂	0.427	0.0866	0.0986
C ₃	0.427	0.0866	0.1524
C ₄₋₆	0.427	0.0866	0.2494
C ₇₊	0.641	0.1087	0.5826
C ₁₄₊	0.427	0.0866	0.8667
C ₂₅₊	0.427	0.0866	1.231

1. Constructing a PVT new model on the basis of a wide variety of experimental tests.
2. Running sufficient slim tube simulations with different grid block sizes (i.e., 100, 500 and 1000 grid block size) and different pressures (i.e., 600, 1000, 1200, 1500, 1800, 2100, 2500, 2800, 3200, 3500, 3800, 4000, 4200, 4300, 4500, 4600, 4700 and 5000 psia).
3. Applying an extrapolation technique to determine a value for the oil recovery factor for an infinite grid block (from a plot of oil recovery factor versus $1/\sqrt{N}$, where N is grid block size).
4. Determining the break-over pressure in a graph of ultimate recovery (1.2 pore volume of injected gas) versus the displacement pressure and taking that value as the minimum miscibility pressure (Yellig and Metcalfe 1998).

3 Results and discussion

3.1 Pure CO₂ MMP

Slim tube simulations were performed for the reservoir fluid with pure CO₂ at twelve pressures ranging from 600 to 4300 psia. The results illustrated in Fig. 4 reveal the pure CO₂ MMP for this reservoir fluid to be 2885 psia at 230 °F. As there are no experimental data available for minimum miscibility pressure, it is appropriate to compare the MMP obtained by this simulation study with the results of Glaso (1985) and National Petroleum Council (NPC) (Klins 1984) based on the constraints of various correlations. MMP obtained by Glaso (1985) and Klins (1984) was 3070 and 3502 psia, respectively.

3.2 Impacts of N₂, C₁, C₂ and C₃ on the CO₂ MMP for the studied reservoir fluid

Figure 5 illustrates the impact of introducing various gas impurities into the CO₂ injection stream.

Increasing the nitrogen content increases the CO₂ MMP. In general, the presence of nitrogen in the injection stream, therefore, has a negative effect on oil recovery. Increasing the methane content also increases the CO₂ MMP. Compared to the reservoir oil–pure CO₂ system, addition of 5%, 10%, 15%, 20%, 25% and 30% C₁ as a contaminant to CO₂ increases the MMP of the system by 5%, 12%, 18%, 25%, 33% and 42%, respectively.

On the other hand, as illustrated in Fig. 5, increasing ethane (C₂) content does not have a significant effect on the CO₂ MMP, whereas increasing propane content by a small amount can significantly reduce CO₂ MMP.

3.3 Impact of combined gas impurity mixtures on CO₂ MMP

Figure 6 compares the impact of CO₂ MMP of various ternary mixtures of injection gases.

The MMP for a mixture of 10% N₂ and 10% C₁ is 3910 psia (Fig. 6). The results of slim tube simulation (Fig. 5) indicate that the addition of nitrogen increases CO₂ MMP more than the addition of similar quantities of methane to the injected gas stream. The ultimate recovery versus displacement pressure plot (Fig. 6) indicates that the MMP for reservoir fluid injected with a gas mixture of 10% N₂ and 10% C₂ is 3422 psia. In comparison with the reservoir oil–pure CO₂ system, the addition of a gas mixture with 10% N₂ and 10% C₃ as contaminants increases the system's MMP by 113 psia (Fig. 6).

For the impure CO₂ gas containing 10% C₁ and 10% C₂, the MMP was determined to be 3046 psia (Fig. 6). On the other hand, the MMP for the mixture of 10% C₁, 10% C₃ and 80% CO₂ was 2868 psia (Fig. 6) whereas the MMP for the mixture of 10% C₂, 10% C₃ and 80% CO₂ was 2504 psia. Note that the MMPs of these latter two mixtures are substantially lower than those involving nitrogen and/or methane as contaminants with CO₂.

4 Discussion

The principal gas injected into crude oil at sub-surface reservoir conditions for this research was CO₂. The novel relationships identified in this study for estimating CO₂ MMP with impurities related to four gases (constituting up to 30% of the injection gas stream) have particular

Match between experimental measurements and Eclipse Reservoir Simulator results for the crude oil sample from an Iranian oil field

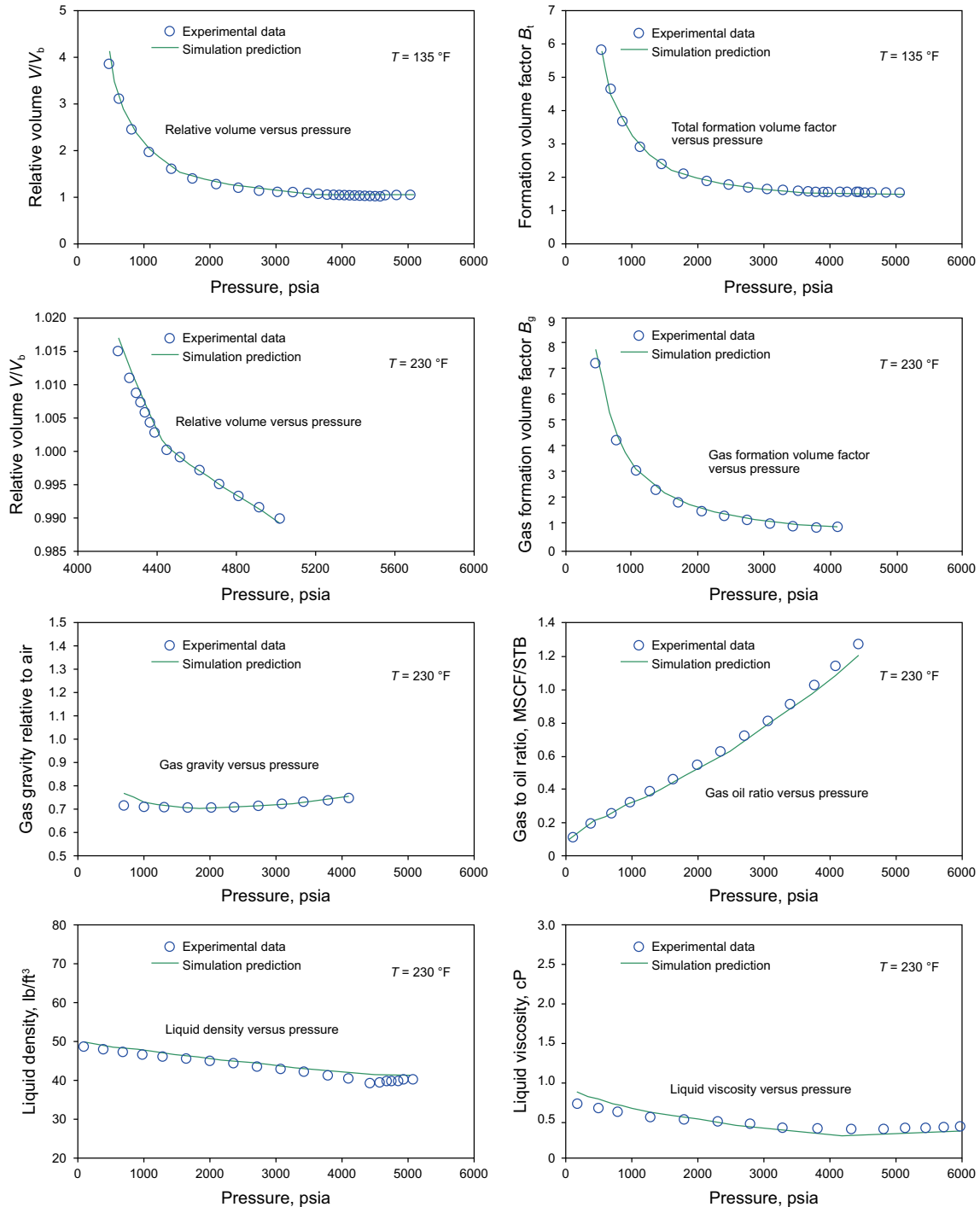


Fig. 2 Optimum matches achieved by regression analysis for the crude oil sample using the Eclipse reservoir simulator PVTi module

Table 5 Bubble point pressures at 135 and 230 °F

Temperature, °F	Experimental bubble point pressure, psia	Predicted bubble point pressure, psia	Error, %
135	4049	4061	0.29
230	4449	4461	0.27

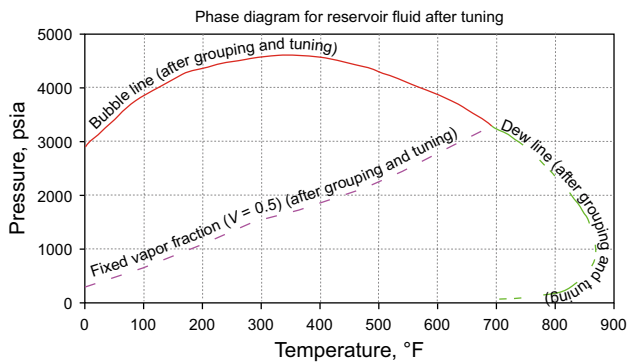


Fig. 3 Phase diagram for the 30° API crude oil reservoir fluid sample from an Iranian oil field after tuning

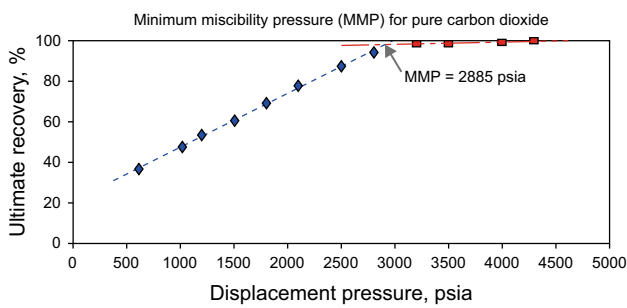


Fig. 4 Determination of MMP for pure CO₂ displacement

relevance to carbon capture and storage as well as enhanced oil recovery studies.

According to the simulation outputs, N₂ and C₁ as minor contaminants of that injected gas can have an adverse effect (i.e., an increase) on the minimum miscibility pressure (MMP) between the reservoir fluid and the injected

gas. This suggests that adding nitrogen or methane as contaminants into a CO₂-rich gas injection stream is likely to result in an increased MMP for the reservoir fluid system (i.e., a negative impact). On the other hand, C₂ and C₃ as minor contaminants in a CO₂-rich gas injection stream into the same oil sample were able to decrease the MMP between the reservoir fluid and the injected gas. The slope of the MMP versus CO₂ percent graph was steeper for C₃ compared to C₂. The addition of C₃ to a CO₂-rich gas injection stream is therefore likely to improve its performance as an EOR mechanism by reducing the MMP of the reservoir fluid-injected gas system.

These results also have significant implications for carbon capture and storage (sequestration) (CCS) into crude oil reservoirs, and specifically the CO₂ storage capacity of specific crude oil reservoirs. Since flue gases from power plants, refineries, steel works, etc. (i.e., major greenhouse gas emitting industrial plants) are not pure CO₂, but contain many impurities including N₂, C₁, C₂ and C₃, the results of this research provide insight into the likely impacts of these contaminants when injecting flue gas components with these impurities into underground reservoirs for CCS purposes. Achieving a low MMP for the reservoir fluid-injected gas system is likely to be advantageous from the perspective of being able to retain more of that injected flue gas in the reservoir fluid. Also, a lower MMP for the reservoir fluid-injected gas system will require lower injection pressures to achieve sub-surface miscibility and therefore incur lower costs. This suggests that the role of small quantities of C₃ in injected flue gases for CCS purposes is significant.

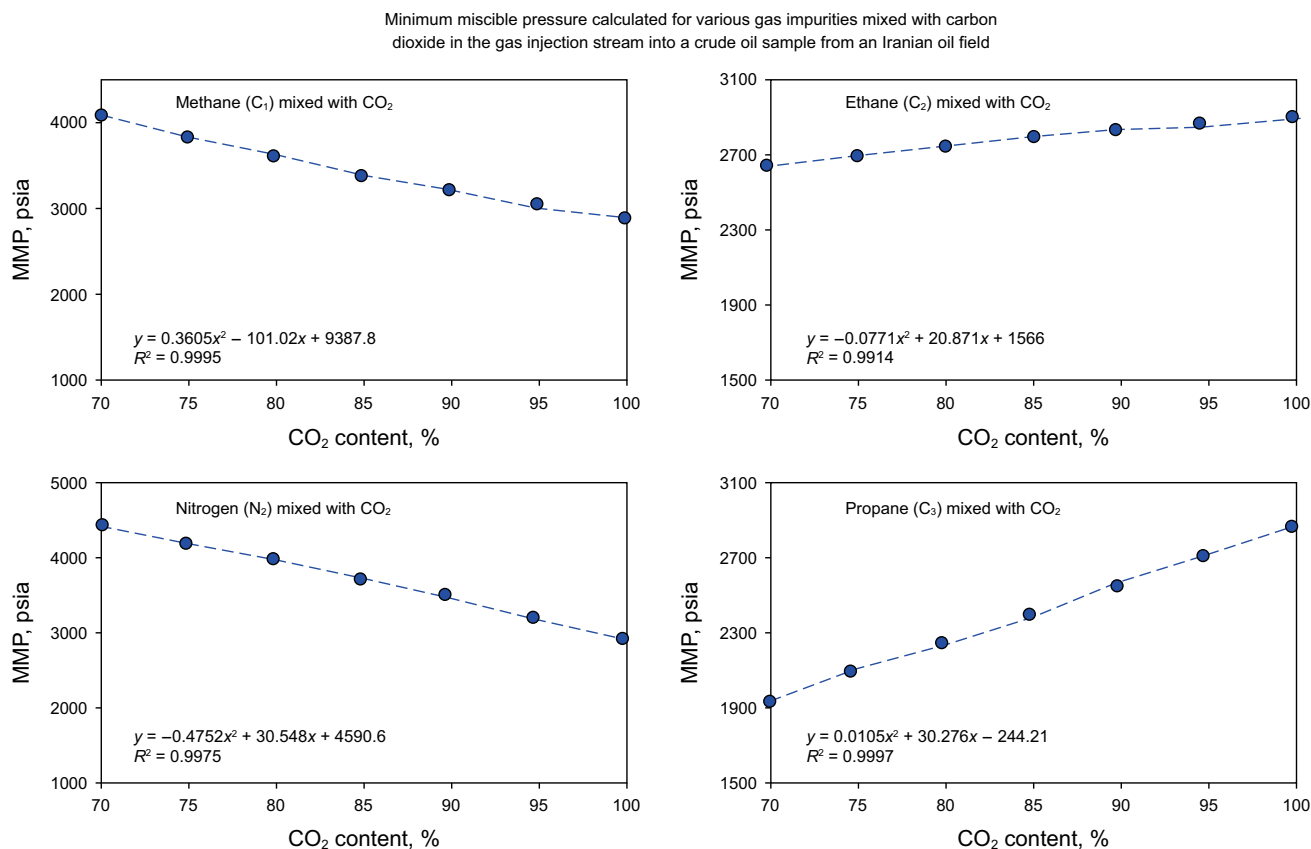


Fig. 5 MMP versus carbon dioxide (percent) for four binary mixtures of nitrogen, methane, ethane and propane with carbon dioxide CO₂

5 Conclusions

The MMP simulation results for a 30° API reservoir crude oil (Iran) lead to the following conclusions:

1. PVT properties of the oil sample could be successfully estimated with high precision by applying the three-parameter Soave–Redlich–Kwong (SRK) equation of state and Lorenz–Bray–Clark (LBC) correlation.
2. The minimum miscibility pressure (MMP) increases in an unfavorable manner when either C₁ and/or N₂ is involved as the dominant impurities in the CO₂ stream. The slim tube results obtained from Eclipse 300 indicate that nitrogen increases CO₂ MMP more than similar quantities of methane.
3. The presence of ethane (C₂) as an impurity in the CO₂ stream is able to slightly decrease CO₂ MMP, but not significantly. For instance, adding 30% ethane to CO₂ gas reduces the MMP of the pure CO₂–gas injection stream from 2885 to 2645 psia.
4. Addition of 5%, 10%, 15%, 20%, 25% and 30% propane (C₃) as an impurity to the CO₂ gas injection stream reduces the MMP of the system by 5%, 11%, 17%, 22%, 28% and 33%, respectively.
5. Collectively these results suggest that incorporating propane in contaminated CO₂-rich gas injection streams (e.g., extracted from flue gas) could have a beneficial impact by reducing the MMP of the system combining injection stream with crude oil at reservoir condition.

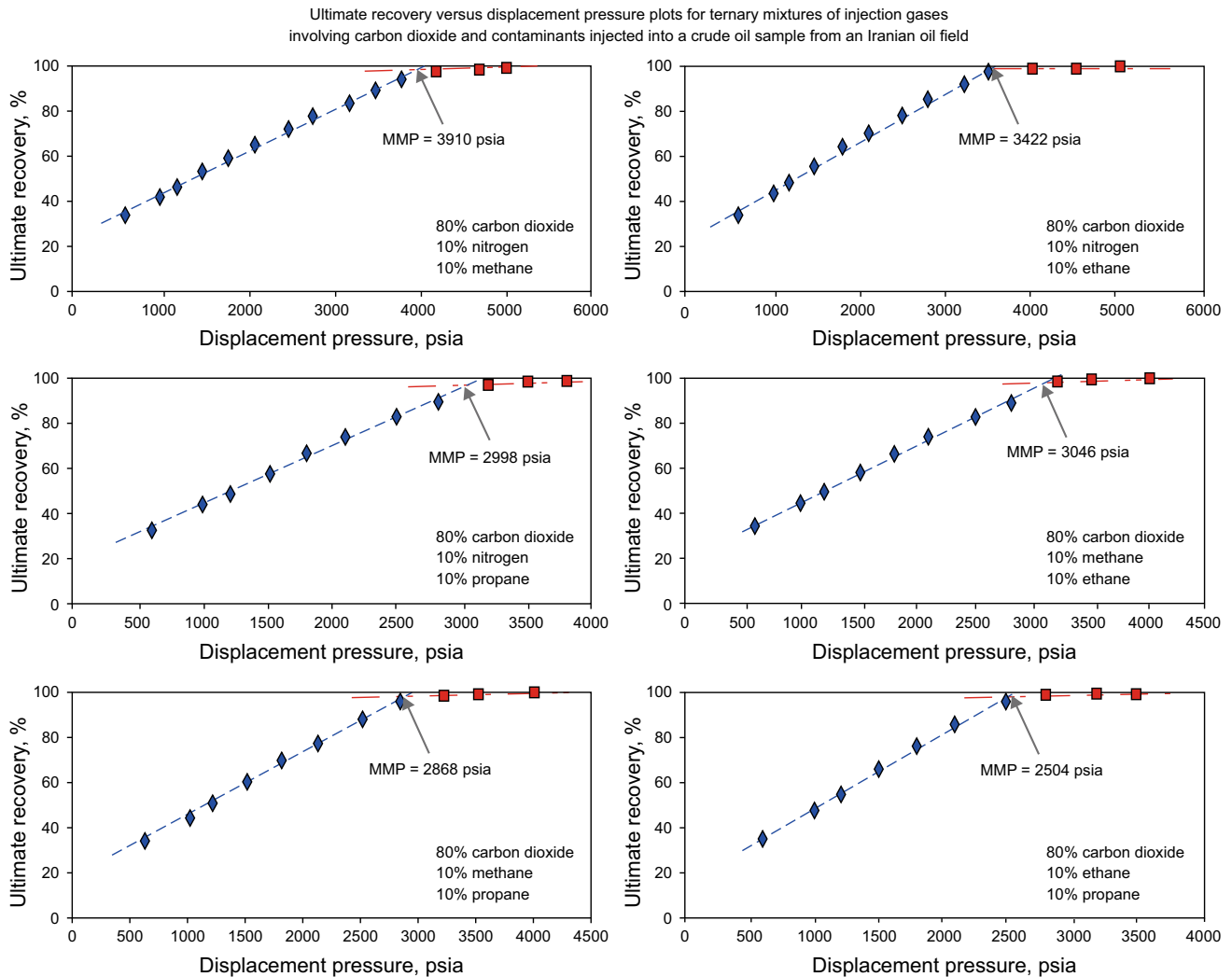


Fig. 6 MMP versus carbon dioxide (percent) for six ternary mixtures involving two impurity gases (nitrogen, methane, ethane and/or propane) mixed with carbon dioxide CO₂

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