Research Article

Investigation of CO₂ Enhanced Oil Recovery Using Dimensionless Groups in Wettability Modified Chalk and Sandstone Rocks

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The paper addresses enhanced oil recovery in chalk and sandstone rocks by CO₂ injection, with different wettability, porosity, and permeability as well as injection rate and flooding conditions. Results indicate that an increase in Bond number has a positive effect on oil recovery whereas for capillary number, there is a limit in which recovery is improving. This limit is estimated when the pressure drop by viscous force is approximately equal to the threshold balance between capillary and gravity forces. A dimensionless group is proposed that combines the effect of capillarity, injection rate, permeability, and CO₂ diffusion on the oil recovery. Recovery from all experiments in this study and reported data in the literature shows a satisfactory relationship with the proposed group.

1. Introduction

CO₂ flooding is the most widely used method for medium and light oil recovery in sandstone and carbonate reservoirs during the last decades [1, 2]. In the past five decades, there have been extensive laboratory studies, numerical simulations, and field applications of CO₂ EOR processes [1, 3–6]. The mechanisms affecting the displacement of oil by CO₂ injection include oil swelling, IFT (interfacial tension), and viscosity reduction as well as increasing the injectivity index due to solubility of CO₂ in water and subsequent reaction of carbonic acid with the minerals [7–9]. For immiscible CO₂ flow in porous media, displacement efficiency could depend on the wetting properties of the fluids, the rate of CO₂ injection and oil production, the difference of oil and CO₂ density, the viscosity ratio of fluids, and the oil and CO₂ relative permeabilities [7]. Miscible CO₂ EOR reduces viscosity and CO₂ can vaporize and extract intermediate hydrocarbons from the reservoir crude oil and/or condense to the oil for developing miscibility under certain pressures and temperatures [10]. Displacement efficiency for miscible

CO₂ flooding is also influenced by wetting properties of the rock, injection and production rates, density difference between oil and gas, viscosity ratio of fluids, and oil/CO₂ relative permeabilities [10].

Investigations in the laboratories and in the field [11–18] suggest the importance of the gas-oil gravity segregation in CO₂ EOR. Kulkarni and Rao [16] presented the effect of important dimensionless groups on the final oil recovery obtained from a number of miscible and immiscible CO₂ gravity drainage experiments. They stated that the main design parameters on a laboratory scale to evaluate the feasibility of gas injection depend on reservoir heterogeneity, rock type, fluid characteristics, injection gas, WAG ratio, and gravity considerations as well as miscibility development and compositions of oil and brine [16]. Wood et al. [19] introduced ten dimensionless groups to describe CO₂ flooding in a dipping water flooded reservoir and used them for experimental design purposes to develop screening criteria applicable to Gulf Coast reservoirs. Trivedi and Babadagli [20] proposed a new group incorporating the matrix-fracture

diffusion transfer for scaling of the miscible displacement in fractured porous media.

The objective of this paper is to study CO₂ enhanced oil recovery in chalk and sandstone rocks. The wettability of calcite and silicate minerals has been changed using fatty acid (stearic acid), amine (N,N dimethyldodecylamine), and asphaltene dissolved in the base solvents. A number of CO₂ flood experiments are performed with various ranges of permeability, porosity, wetting state, and CO₂ injection pressure and rate. These parameters are changed extensively to obtain a wide range of dimensionless groups. The results have been analyzed using capillary, Bond, and diffusion numbers at different wetting states. A new dimensionless group is proposed that combines the effect of capillarity, injection rate, permeability, and CO₂ diffusion during flooding processes.

2. Theory

To quantify the relative magnitudes of the prevailing forces in CO₂ flooding processes, the following set of dimensionless groups is usually suggested: (I) the Bond number (N_B) which is the ratio of the pore scale hydrostatic pressure drop to the capillary pressure and (II) the capillary number (N_c) which is the ratio of the pore scale viscous pressure drop to the capillary pressure [22, 24]:

$$N_B = \frac{\Delta P_{\text{grav}}}{\Delta P_{\text{cap}}} = \frac{\Delta \rho g (k/\phi)}{\sigma}, \quad (1)$$

$$N_c = \frac{\Delta P_{\text{vis}}}{\Delta P_{\text{cap}}} = \frac{\mu_o v_{\text{CO}_2}}{\sigma \phi}, \quad (2)$$

where μ_o is the wetting phase viscosity, v_{CO_2} is the Darcy velocity of the injected CO₂ through porous medium, which is equal to CO₂ volume injection rate divided by cross-sectional area of the porous medium, σ is the interfacial tension between CO₂ and oil, k and ϕ are the permeability and porosity of the porous medium, respectively, $\Delta \rho$ is the density difference between two fluids, and g is the gravity acceleration in the direction of flow.

The interfacial tension between CO₂ and oil can be calculated from the following equation using Parachor method [25]:

$$\sigma^{1/4} = (\rho_M^L \Sigma P_i X_i - \rho_M^V \Sigma P_i Y_i). \quad (3)$$

In this equation, ρ_M is density (mol/cc), P is Parachor number, and X_i and Y_i are liquid and vapor mole fractions, respectively.

It should be noted that the above-defined capillary number, N_c , does not take into account the length and pore size for a porous medium [24], which affect the viscous force through the porous medium. In the literature, the complete capillary number, N_{ca} , for a circular capillary, is defined as [24]

$$N_{ca} = \frac{\Delta P_{\text{vis}}}{\Delta P_{\text{cap}}} = \frac{4\mu_{\text{CO}_2} v_{\text{CO}_2} l}{\phi \sigma r}. \quad (4)$$

In (4), l and r are the length and radius of the circular capillary, respectively. In consideration of the actual pore size and

shape distributions in a reservoir rock, it is useful to relate the above-defined complete capillary number, N_{ca} , to some measurable reservoir characteristics, such as permeability and porosity of the oil reservoir, and Dullien [24] derived the following relation:

$$k = \frac{\phi r^2}{8}. \quad (5)$$

Here, k is the permeability, ϕ is the porosity, and r is the radius of the identical circular capillaries. Alternatively, the complete capillary number can be defined in terms of the permeability and porosity of the porous medium:

$$N_{ca} = \frac{\Delta P_{\text{vis}}}{\Delta P_{\text{cap}}} = \frac{\sqrt{2}\mu_{\text{CO}_2} v_{\text{CO}_2} l}{\sigma \sqrt{k\phi}}. \quad (6)$$

For drainage with high viscosity contrast between gas and oil, theoretical arguments based on percolation theory predict the following scaling law for the displacement under stabilized condition in three-dimensional (3D) pore structures:

$$l \approx (N_B^*)^{-1} \quad (7)$$

In this equation l is front width. The generalized Bond number, N_B^* , is also introduced by Løvoll et al. [26] as follows:

$$N_B^* = \frac{\Delta \rho g (k/\phi)}{\sigma} - \frac{\mu_o v_{\text{CO}_2}}{\sigma}. \quad (8)$$

They suggested that since the front width l in (7) diverges when $N_B^* \rightarrow 0$, the following criterion for interface stabilization is expected [26]:

$$N_B^* > 0. \quad (9)$$

After substitution and simplifications, the following criteria can be obtained:

$$\frac{\Delta \rho g k}{\mu_o} = v_c > v_{\text{CO}_2}. \quad (10)$$

This critical velocity also can be calculated during modeling of free gas gravity drainage as a rate of oil recovery before gas front reaches the capillary/gravity balance threshold. The appendix shows the analytical model describing free gravity drainage and the mathematical method to find the critical oil recovery velocity. The calculation of critical oil recovery velocity with another approach has been described also by Marle [27].

The diffusion number is defined as ratio of diffusive and convective dispersion (bulk flow or viscous force):

$$N_d = \frac{D\phi}{v_{\text{CO}_2} l}. \quad (11)$$

In (11), D is diffusivity coefficient in Fick's law between CO₂ and oil and ϕ is the porosity; l is the length of capillary tube and v_{CO_2} is the Darcy velocity of the injected CO₂ through porous medium, which is equal to CO₂ volume injection rate divided by cross-sectional area of the porous medium.

The number can scale the role of diffusive and convective forces in miscible processes.

Diffusivity coefficient (D) of CO_2 in oil can be estimated from the following equation [28]:

$$D_{\text{CO}_2, \text{oil}} = 10^{-9} \mu_{\text{oil}}^{-0.4562} M_{\text{CO}_2}^{-0.6898} \times V_{\text{CO}_2}^{-1.706} P^{-1.831} T^{4.524} \quad (12)$$

In this equation, $D_{\text{CO}_2, \text{oil}}$ is diffusivity coefficient of CO_2 gas in oil (m^2/sec), μ_{oil} is viscosity of oil (cp), M_{CO_2} is the molecular weight of CO_2 , V_{CO_2} is the molar volume of CO_2 (cm^3/mol), P (psia) is the pressure of CO_2 and oil system in equilibrium and T (Kelvin) is the temperature of CO_2 and oil system in equilibrium.

3. Experimental Section

3.1. Materials

3.1.1. Solid Phase. The core flood experiments were done in outcrop Stevns klint chalks near Copenhagen and outcrop Benthiermer sandstones. The dimension of cores varied between 5–7 cm in length and 3.8 cm in diameter. The outcrop chalks have approximate porosity of 44–48% and absolute permeability of 3–7 mD. Sandstone cores have the same porosity near 22–25%, while they are classified based on the permeability in the two groups with permeability range of 600–800 mD and with the permeability range of 1500–1700 mD. The capillary threshold of chalk cores for oil and gas system is measured near 2 bar by porous plate technique. The capillary threshold of Benthiermer sandstones was reported as 0.05 atm [29]. Table 1 is the cores detailed descriptions.

3.1.2. Oil Phase. The investigation is done for model oil containing asphaltene dissolved in toluene (95% purity), 0.01 M stearic acid (SA) dissolved in n-decane (95% purity) for chalk cores, and 0.01 M N,N-dimethyldodecylamine (NN-DMDA) dissolved in n-decane (95% purity) for sandstone cores. Synthetic seawater (SSW) has been used as irreducible water saturation. Tables 2 and 3 show the polar components used in this work and also the ionic composition of synthetic seawater.

3.1.3. Asphaltene Preparation Procedure. The model oil system is prepared from asphaltene precipitated from crude oil in excess of n-heptane (1:40). The mixture was shaken for at least twice a day and left for 48 hours to equilibrate. The mixture solution was then centrifuged and filtered through a 0.22 micrometer filter (Millipore) and dried for 1 day using a vacuum oven at room temperature. The dried asphaltene was then dissolved in toluene.

3.2. Experimental Setup. The detailed description of the experimental set up has been addressed elsewhere [30]. The major components of the experimental setup consist of a core holder, pressure regulators, two gas flow meters, pressure manometers, Gilson pump, CO_2 piston cell, graduated gas/oil

separator, and a lab view version 7.1 connected to a digital data acquisition system. Saturated core samples with synthetic seawater were inserted into a horizontally placed core holder that consists of a steel cylindrical body and rubber/nylon sleeve. A net overburden pressure of 20 bar is applied on the sleeve. The chalk core is flooded with oil sample at flow rate of 0.1 mL/min until oil breakthrough is occurred and then it is increased to 5 mL/min to overcome the capillary threshold using Gilson pump (806 manometric modules) to establish the irreducible water. The sandstone core is flooded with oil sample at flow rate of 3 mL/min until oil breakthrough is occurred and then it is increased to 5 mL/min to overcome the capillary threshold. Table 1 shows the irreducible water saturation, which is varying between 10 and 13% for chalks and 20 and 25% for sandstones. The cores were aged for at least two weeks to change the wettability of cores toward oil wet, when the saturating oil contains polar components. Then, CO_2 injection was carried out in two modes, namely, injection at constant rate and constant pressure. At constant rate, CO_2 is injected from piston connected to Gilson pump (806 manometric modules) with desired rate. At constant pressure modes, CO_2 is injected in immiscible and miscible processes. In the immiscible process, the pressure of CO_2 injection is lower than minimum miscibility pressure. Whereas, in the miscible mode, the oil saturated core was flooded with CO_2 at pressure equal to minimum miscibility pressure. CO_2 was injected into the core at constant pressures of 40 ± 0.2 bar and temperature of 25°C for immiscible process and 90, 70, and 140 ± 0.2 bar and at 50, 70, and 80°C for miscible processes. The constant pressure of CO_2 is provided by Gilson pump when the upper and lower pressure limits of pump are adjusted to the desired injection pressure plus/minus 0.5 bar. CO_2 is injected to the core passing mass flow meter (1) that records the inflow properties of CO_2 (mass flow rate, density, and total mass). A back pressure regulator is connected downstream the core to control the pressure during CO_2 flooding. The outflow properties (mass flow rate, density, and total mass) of the evolved gas were, also, recorded using flow meter (2) connected to the separator. The CO_2 injection continued for at least 3 pore volumes until there was no oil production. When the injection was terminated, the core was then removed from the core holder and dried using vacuum oven at a temperature of 120°C until a constant weight was obtained.

4. Results and Discussion

The experimental results with the relation to the selected dimensionless groups are discussed in this section.

4.1. Bond Number. The variations in Bond number were primarily performed by varying the permeability of the porous medium, where reduction in permeability leads to a reduction in Bond number. As it is seen in (2), permeability does not affect the capillary number, thus allowing investigation of the effect of Bond number on the oil recovery at a constant capillary number. Recovery factor after two pore volumes of injected CO_2 as a function of the Bound number is shown in

TABLE 1: Core descriptions of immiscible and miscible CO₂ flooding experiments.

EXP. number	S_{wi}	Phi	K (cm ²)	L (cm)	q (cc/min)	Type	Core
1a	0.22	0.23	$6.5E - 09$	6	0.2	Immiscible	Aged sandstone
2a	0.2	0.24	$7.5E - 09$	5.5	0.5	Immiscible	Aged sandstone
3a	0.25	0.25	$7.3E - 09$	5.8	2	Immiscible	Aged sandstone
4a	0.2	0.25	$8.3E - 09$	6.2	6	Immiscible	Aged sandstone
5a	0.25	0.23	$6.7E - 09$	6	8	Immiscible	Aged sandstone
5a*	0.21	0.22	$6E - 09$	5.5	8	Immiscible	Aged sandstone
5a**	0.22	0.24	$7.6E - 09$	6	8	Immiscible	Aged sandstone
1b	0.2	0.23	$1.5E - 08$	6	0.5	Immiscible	Aged sandstone
2b	0.25	0.24	$1.6E - 08$	5.5	2	Immiscible	Aged sandstone
3b	0.22	0.24	$1.7E - 08$	5.5	8	Immiscible	Aged sandstone
4b	0.25	0.25	$1.7E - 08$	5.8	10	Immiscible	Aged sandstone
1c	0.14	0.44	$5E - 11$	6	0.2	Immiscible	Aged chalk
2c	0.15	0.45	$3.5E - 11$	6.5	0.5	Immiscible	Aged chalk
3c	0.12	0.48	$6E - 11$	6.7	2	Immiscible	Aged chalk
4c	0.13	0.47	$5.5E - 11$	5.8	2.5	Immiscible	Aged chalk
5c	0.1	0.48	$6.5E - 11$	5.9	3.5	Immiscible	Aged chalk
5c*	0.13	0.45	$6.2E - 11$	6.5	3.5	Immiscible	Aged chalk
5c**	0.15	0.47	$6E - 11$	6	3.5	Immiscible	Aged chalk
1d	0.17	0.44	$5.5E - 11$	6	0.01	Immiscible	Aged chalk
2d	0.15	0.45	$4E - 11$	6.5	0.05	Immiscible	Aged chalk
3d	0.11	0.48	$2E - 11$	6.7	0.1	Immiscible	Aged chalk
1e [#]	0.16	0.45	$6.5E - 11$	6	0.05	Miscible	Aged chalk
2e [#]	0.14	0.46	$4E - 11$	6.5	0.05	Miscible	Aged chalk
3e [#]	0.12	0.48	$2.5E - 11$	6.8	0.05	Miscible	Aged chalk
1e	0.27	0.25	$6.5E - 09$	6	0.05	Miscible	Unaged sandstone
2e	0.3	0.23	$7.7E - 09$	6.5	0.05	Miscible	Unaged sandstone
3e	0.125	0.22	$8.2E - 09$	6.8	0.05	Miscible	Unaged sandstone
1g	0.22	0.23	$1.8E - 08$	6	0.5	Immiscible	Unaged sandstone
2g	0.23	0.26	$1.9E - 08$	5.5	2	Immiscible	Unaged sandstone
3g	0.24	0.3	$2.1E - 08$	5.5	8	Immiscible	Unaged sandstone
4g	0.24	0.25	$1.8E - 08$	5.8	10	Immiscible	Unaged sandstone
1h	0.14	0.44	$5.5E - 11$	6	0.05	Miscible	Unaged chalk
2h	0.15	0.45	$5.5E - 11$	6.5	0.05	Miscible	Unaged chalk
3h	0.17	0.47	$6E - 11$	6.8	0.05	Miscible	Unaged chalk

*Repetition of experiment (first time).

**Repetition of experiments (second times).

[#]Previously reported in Hamouda, A.A., Alipour Tabrizy, V., 2013, the effect of light gas on miscible CO₂ flooding to enhance oil recovery from sandstone and chalk reservoir, and journal of Petroleum Science and Engineering, volume 108, pages 259–266.

TABLE 2: Polar components.

Components	Supplier and purity	Structural formula
Stearic acid (SA)	Aldrich ≥99%	$CH_3(CH_2)_{16}COOH$
N,N-dimethyldodecylamine (NN-DMDA)	Fulka ≥99%	$CH_3(CH_2)_{11}N(CH_3)_2$

Figure 1. Results indicate that recovery at a constant capillary number increases with increasing the Bond number. This behavior is in line with the expectation about the effect of capillarity on oil drainage by gas injection. To examine

the relationship between oil recovery and Bond number, the recovery between all experiments is plotted versus Bond numbers in Figure 2. No satisfactory relation was found between the oil recovery and the balance between gravity and capillary forces.

The scaling of oil recovery using Bond number has three major limitations: (I) the effect of viscous force is not included. For example, by changing the gas injection rate, the oil recovery may be changed drastically, while the Bond number is remained unchanged; (II) the effect of viscosity ratio between displaced and displacing fluids is not included. For example, when the gas/oil viscosity ratio changes leading to a change in stability of the front and recovery, the Bond

TABLE 3: Synthetic seawater composition.

Ions	Concentration (mol/L)
NaCl	0.4
KCl	0.01
CaCl ₂ (2H ₂ O)	0.013
MgCl ₂ (6H ₂ O)	0.045
NaHCO ₃	0.002
Na ₂ (SO ₄)	0.024
TDS	0.494

FIGURE 1: CO₂ enhanced oil recovery at constant capillary number ($N_c \approx 3.50E-07$) as a function of Bond number.

number remains unchanged. The effect of CO₂ diffusion in oil and miscibility parameters that is, pressure, temperature, and oil composition, are not considered also in Bond number. The effects of viscous forces, viscosity ratio, and CO₂ diffusion are incorporated in the following sections.

4.2. Capillary Number. Large viscous forces in high-rate gas flooding, characterized by large capillary numbers, may result in unstable flooding fronts and the oil bypassing/trapping mechanism. From this point of view, an increase in the viscous force will result in reduction in the oil recovery because the microscopic displacement efficiency decreases. However, the low-rate gas flooding may lead to considerable residual oil since the pressure drop across the core may be lower than the capillary threshold. In the work reported by Rostami et al. [31], it has been observed that in unconsolidated bead pack porous media and high permeable water-wet outcrop sandstones, the oil recovery reduces as capillary number increases. This may not be applicable in the real reservoir cores, especially for tight oil-wet carbonate rocks with

FIGURE 2: CO₂ enhanced oil recovery from all experiments as a function of Bond number.

considerable capillary threshold. Experimental results are shown in Figure 4(a) to Figure 5(d), where oil recovery after two-pore volume CO₂ injection is plotted versus capillary number. Results in each figure are characterized by a Bond number that remains approximately constant. The interfacial tension between CO₂ and oil is estimated near to 20 mN/m for immiscible process and 0.1 mN/m for miscible process according to (3). Results indicate that oil recovery improves as capillary number increases up to the critical limit where afterward decreases. In other words, the recovery increases when the pressure drop due to viscous force increases near to the capillary threshold and then decreases due to flow instability. From Figure 3, it is clear that for experiments "a" and "c," the oil recovery is increasing up to a certain value and then it is decreasing by increasing the injection rate and viscous force. The maximum oil recovery may be linked to the pressure drop in which $\Delta P \approx P_{c_{th}}$. To observe the reliability of results, experiment "a5" and experiment "c5" are repeated and they are shown in Figure 4. The variations of oil recovery for similar conditions are near to 5%.

It can be concluded from Figures 3 and 4 that the oil recovery is not directly proportional to the capillary number; hence, the following modified capillary number is suggested:

$$N_c^* = N_c \left(1 - \text{abs} \left(1 - \frac{\Delta P}{P_{c_{th}}} \right) \right). \quad (13)$$

In this equation, abs is showing the absolute value of number. The other symbols are defined previously. Figure 5 shows the relation between modified capillary number and CO₂ enhanced oil recovery. Applying this number shows linear relationship of oil recovery to the number in semilog scale; hence, it might be more appropriate for scaling of oil recovery in gas flooding projects. The associated regression parameter for each figure is presented also in the graph.

The effect of injection rate on immiscible oil recovery can be also investigated by definition of displacement efficiency

FIGURE 3: CO₂ enhanced oil recovery at constant Bond number ($N_c \cong 3.50E-07$) as a function of capillary number.

number using the critical velocity derived in the appendix as follows:

$$N_{DE} = \frac{\nu}{\nu_c} \left(1 - \text{abs} \left(1 - \frac{\Delta P}{P_{c_{th}}} \right) \right). \quad (14)$$

In Figure 6, the oil recovery as function of displacement efficiency number is shown. This number takes into account the effect of injection rate, capillary threshold, and density difference between injected and displaced fluids and may be alternatively used instead of modified capillary number.

To explore an appropriate dimensionless number that describes the dynamics of the oil recovery, recoveries from all

experiments are plotted versus their modified capillary numbers (Figure 7). Again, no satisfactory relation is observed. This confirms that the capillary number alone is insufficient to predict the oil recovery behavior during the CO₂ flooding.

4.3. Diffusion Number. Diffusion number expresses the ratio of diffusivity to convective dispersion (bulk flow or viscous force). The number is important in miscible processes, when the diffusion of solvent in oil is considerable and the oil production is slow; hence, the viscous force and convective dispersion are small and the diffusion number should be taken into account according to (6). Experiments in porous media "e" and "f" are done in miscible mode in chalk and

FIGURE 4: Reproducibility of CO₂ enhanced oil recovery experiments.

sandstone cores, respectively. The minimum miscible pressure (MMP) for model oil composition at different temperatures is calculated according to SRK-Peneloux EOS, PVTsim version 17. The oil recovery for miscible CO₂ flooding experiments for porous media "e" and "f" is shown in Figure 8. The CO₂ flooding has been done at 50°C, 70°C, and 90°C and pressures 90 bar, 120 bar, and 140 bar for experiments "e1," "f1," "e2," "f2," "e3," and "f3," respectively. From the graph, it can be seen that as temperature and pressure are increasing, the diffusion number (N_d) increases and the recovery by CO₂ flooding is increasing. It is important to mention that the oil recovery rate before CO₂ breakthrough is kept near 0.05 cc/min for all experiments; accordingly the viscous force and capillary number of miscible experiments are small ($N_c \approx 10^{-7}$) and close to each other.

The dependency of all miscible and immiscible experiments to diffusion number is shown in Figure 9. As it can be seen, no relation could be drawn down for results, since the diffusion number is not playing a critical major role in all experiments.

4.4. Combined Dimensionless Group. Examination of oil recoveries as a function of dimensionless numbers (capillary number, Bond number, modified capillary number, and diffusion number) suggests that none of the above scaling groups individually can define the oil recovery relationship to affecting parameters. In the following section, a combined dimensionless group that could show the relationship of oil displacement over the whole range of variables is investigated. Figure 10 shows the results of all experiments, when ratio of the modified capillary number over Bond number is used. Results indicate that the oil recovery by CO₂ flooding is directly proportional to the ratio of modified capillary number over Bond number; however, no specific relation may explain this relationship.

In order to take into account the effect of diffusion number and appropriate ratio of the capillary number to Bond number, a combined dimensionless group, N_{Co} , is proposed as follows:

$$N_{Co} = \frac{N_d \times N_c^*}{N_B^a} = \frac{N_d \times N_c \times (1 - \text{abs}(1 - \Delta P/P_{cth}))}{N_B^a}, \quad (15)$$

where subscript Co. stands for the term "combined," and the parameter "a" is the scaling factor. It is found that in this research, the value of $a = 0.01$ provides a good relation. The small value of exponent suggests that the effect of the Bond number is much more than the importance of the capillary number. The results for the above dimensionless group are shown in Figure 11, where the recoveries of all experiments are plotted versus this number. This dimensionless group combines the effect of the three main forces that affect CO₂ enhanced oil recovery. The results suggest that within the range of experiments performed here, there is a logarithmic relationship between the oil recovery and proposed group.

In order to validate the applicability of the new group proposed for gas injection processes, the experimental results reported by Hudigins et al., (1990), Darvish [23], and Nobakht et al. [22] are used. Table 4 summarizes the properties of literature experimental data used in this study.

The first two experimental data are related to miscible N₂ injection in the slim tube and core, respectively; hence, the diffusion number and other relevant parameters are calculated for nitrogen. The third and fourth experiments were done for miscible CO₂ flooding in sandstone cores and the fifth experiment was reported for CO₂ flooding in saturated chalk core with live reservoir oil. Figure 12 compares the experimental data reported in this research and literature data reported previously.

FIGURE 5: CO₂ enhanced oil recovery as a function of modified capillary number.

TABLE 4: Literature experimental data used in this study.

Number	Type	S_{wi}	Φ	K (cm ²)	L (cm)	q (cc/min)	σ gas oil (mN/m)	Reference
11	Slim tube (N ₂ injection)	0	0.379	$1.2E-07$	2720	4.76	0.1	Hudgins et al. [21]
21	Core (N ₂ injection)	0.4	0.198	$4.6E-09$	60.96	0.8	0.1	Hudgins et al. [21]
31	Core (CO ₂ injection)	0.07	0.372	$1.8E-05$	28.8	0.5	4.22	Nobakht et al. [22]
41	Core (CO ₂ injection)	0.025	0.369	$1.8E-05$	28.8	0.5	1.13	Nobakht et al. [22]
51	Core (CO ₂ injection)	0	0.444	$4E-11$	60	0.02	0.05	Darvish (2007) [23]

FIGURE 6: CO₂ enhanced oil recovery as a function of displacement efficiency number.

4.5. Effect of Wettability. To understand the role of wettability in miscible and immiscible CO₂ flooding, some experiments are done for unaged cores and the results of oil recovery at CO₂ breakthrough and 2 PV of injected CO₂ are compared in Figures 13 and 14. Figure 13 shows that the oil recovery after 2 PV of injected CO₂ for unaged samples is higher than the aged samples. In fact, in aged sample (oil-wet porous media), oil is the wetting phase and occupies the corner of pores; water is the nonwetting phase which occupies the center of pores. During the flowing of oil in an oil-wet media, oil flows through wetting films, gas flows principally as continuous nonwetting filaments, and water is in the form of isolated ganglia; hence, the existing residual water plays a blocking

role during the displacement and the recovery of aged cores after 2 PV of injected CO₂ is significantly lower than unaged core. In contrast, for unaged core (water-wet porous media), water is not mobilized but wets the pore walls and the continuity of oil is maintained along the porous media because the spreading of oil within water and gas phases is positive. The spreading coefficient of gas, oil water system is calculated based on the equation proposed by Blunt et al. [32] as follows:

$$S = \sigma_{gw} - (\sigma_{go} + \sigma_{wo}). \quad (16)$$

Assuming $\sigma_{gw} = 70$ mN/m [32], $\sigma_{ow} = 19$ mN/m (measured by pendent drop technique) and $\sigma_{go} = 20$ mN/m (calculated

FIGURE 7: CO₂ enhanced oil recovery from all experiments as a function of modified capillary number.

FIGURE 8: CO₂ enhanced oil recovery as a function of diffusion number in miscible experiments.

FIGURE 9: CO₂ enhanced oil recovery as a function of diffusion number in miscible and immiscible experiments.

FIGURE 10: CO₂ enhanced oil recovery in all experiments as a function of modified capillary number to Bond number.

FIGURE 11: CO₂ enhanced oil recovery in all experiments as a function of combined dimensionless number.

FIGURE 12: CO₂ enhanced oil recovery versus combined dimensionless number in all experiments and the experimental data reported in the literature.

FIGURE 13: Immiscible CO₂ enhanced oil recovery versus combined dimensionless number in aged and unaged experiments.

FIGURE 14: Miscible CO₂ enhanced oil recovery versus combined dimensionless number in aged and unaged experiments.

by (3)) the value of spreading coefficient is +31 mN/m. According to the classification reported by Blunt et al. [32] when $S > 0$, the oil can flow as continuous phase among initial water and injected gas and the film flow contributes significantly to the recovery but with slow rate. From Figure 13, it is also clear that the oil recovery is decreasing as N_{Co} is decreasing both for aged and unaged cores at 2 PV and breakthrough of injected CO₂. However, the slope of this relation for aged core (oil wet) at breakthrough time is larger than unaged cores, which indicates that the recovery of aged cores is strongly dependent on the flooding parameters. This has been observed previously by Shahidzadeh-Bonn et al. [33] for forced and free gravity drainage of gas in glass bead pack media, where the wettability of glass beads was changed by chemical treatment with TCMS (Three-Chloro-Methylsilane).

Figure 14 shows that the oil recoveries after 2 PV of injected CO₂ for unaged samples are higher than the aged samples. Similar trend to immiscible CO₂ flooding can be seen for miscible CO₂ flooding.

In unaged core (water-wet porous media), water is not mobilized and wets the pore walls and oil is maintained in the center of pores; hence, the CO₂ diffuses to oil and the recoveries at CO₂ breakthrough and 2 PV injected CO₂ are higher than aged core. In aged core (oil wet), the water droplet occupies the center of pore and the injected CO₂ will dissolve in water and then the mass transfer between oil and CO₂ may occur; hence, the water will act as a barrier. Figure 15 shows the schematic of CO₂ enhanced oil recovery in hydrophobic (aged) and hydrophilic (unaged) porous media. In this figure, oil is shown by brown; water is shown by blue and CO₂ is represented by white color.

The diffusivity of CO₂ in seawater can be found based on the following equations [34]:

$$\log(D_{CO_2,w}) = -4.1764 + \frac{712.52}{T} - \left(\frac{2.5907 \times 10^{-5}}{T} \right), \quad (17)$$

$$\log\left(\frac{D_{CO_2,sw}}{D_{CO_2,w}}\right) = 0.87 \log\left(\frac{\mu_{sw}}{\mu_w}\right), \quad (18)$$

where T is temperature in Kelvin, μ_{sw} is viscosity of seawater (cp), μ_w is viscosity of distilled water, and $D_{CO_2,sw}$ is diffusivity of CO₂ in seawater (cm²/sec). In Figure 16 the diffusivity of CO₂ in seawater based on (17) and diffusivity of CO₂ in model oil are calculated based on the (7) and are compared at different temperatures and corresponding minimum miscibility of CO₂ within oil. The figure clearly indicates that the diffusion of CO₂ in seawater is higher than oil; hence, most of CO₂ will diffuse in water rather than oil.

5. Conclusion

Analysis of oil recovery with Bond number, at constant capillary number, shows a direct relation, but when the capillary number is changing, dependency of oil recovery with the

FIGURE 15: Schematic of rock pore in miscible CO₂ enhanced oil recovery for hydrophobic (aged) and hydrophilic (unaged) porous media [30].

FIGURE 16: Comparison of CO₂ diffusion in seawater and model oil at different temperatures and corresponding minimum miscibility pressures [30].

Bond number is varying also. In addition, at constant Bond number, oil recovery increases as capillary number increases up to a certain limit and then decreases drastically as a result of higher viscous forces and fluid flow instability. Results presented in this work suggest that the Bond, capillary, and diffusion numbers could not scale the oil recovery, individually. A dimensionless group was defined to relate the competition between three forces affecting oil recovery over a wide range of petrophysical and experimental conditions. A logarithmic relationship was observed between the proposed group and oil recovery. Applicability of the proposed relation beyond the range of experiments reported in this work needs to be further examined. It is observed also that the recovery by CO₂ injection for unaged rocks (water wet) is higher than the aged (oil wet) rocks due to higher diffusivity of CO₂ in water (occupying the center of pores in oil-wet cores) compared to that in oil.

Appendix

By using the continuity equation and Darcy's law, and the concepts of relative permeability and capillary pressure, one-dimensional displacement of multiphase flow can be

described mathematically. The one-dimensional Darcy's law for two-phase flow in vertical direction is

$$u_o = \frac{KK_{ro}}{\mu_o} \left(\frac{\partial P_o}{\partial z} - \gamma_o \right), \quad (\text{A.1})$$

$$u_g = \frac{KK_{rg}}{\mu_g} \left(\frac{\partial P_g}{\partial z} - \gamma_g \right), \quad (\text{A.2})$$

where the z axis is vertically downward, u_o and u_g are the oil and gas velocity, and g is the acceleration due to gravity. The gas-oil capillary pressure (P_c) is defined by

$$P_c = P_g - P_o. \quad (\text{A.3})$$

By substituting the flow equation of oil in the continuity equation and combining it with gas-oil capillary pressure, the equation of 1D displacement of oil by gas in vertical direction is gained:

$$\frac{\partial}{\partial z} \left(\frac{KK_{ro}}{\mu_o} \left(\frac{\partial (P_g - P_c)}{\partial z} - \gamma_o \right) \right) = -\frac{\partial}{\partial t} (\phi S_o). \quad (\text{A.4})$$

Gas has a very low viscosity with respect to oil; therefore, the assumption of infinite gas mobility is viewed as a reasonable assumption. Consider the Darcy law for gas phase in (A.2), when u_g is a real number and mobility of gas (i.e., (K_{rg}/μ_g)) approaches infinity, the value of driving force, that is, $(dP_g/dz - \rho_g g)$, is relatively small. This implies that hydrostatic gas pressure gradient is a good estimation in free gravity drainage modeling. By substituting the gas pressure in (A.4) and assuming constant porosity the following equation is obtained:

$$\frac{\partial}{\partial z} \left(\frac{KK_{ro}}{\mu_o} \left(\frac{\partial P_c}{\partial z} + (\rho_o - \rho_g) g \right) \right) = -\phi \frac{\partial}{\partial t} (S_o). \quad (\text{A.5})$$

Since $P_c = P_c(S_o)$, the above equation can be written as

$$\frac{\partial}{\partial z} \left(\frac{KK_{ro}}{\mu_o} \left(\frac{\partial P_c}{\partial S_o} \frac{\partial S_o}{\partial z} + (\rho_o - \rho_g) g \right) \right) = -\phi \frac{\partial}{\partial t} (S_o). \quad (\text{A.6})$$

Equation (A.6) is written in diffusion-convection form for simplifying:

$$\frac{\partial}{\partial z} \left[D(S_o) \frac{\partial S_o}{\partial z} \right] + U(S_o) \frac{\partial S_o}{\partial z} = \frac{\partial S_o}{\partial t}. \quad (\text{A.7})$$

In this equation $U(S_o)$ is the coefficient of first derivative term and is defined in (A.9). This is a convective term and is affected by oil-gas density difference and derivative of oil relative permeability. $D(S_o)$ is a coefficient of second derivative term and is defined in (A.8). This diffusive term is a function of oil relative permeability and first derivative of capillary pressure with respect to the oil saturation:

$$D(S_o) = -\frac{KK_{ro}}{\mu_o \phi} \frac{dP_c}{dS_o}, \quad (\text{A.8})$$

$$U(S_o) = -\frac{K}{\mu_o \phi} (\rho_o - \rho_g) g \frac{dK_{ro}}{dS_o}. \quad (\text{A.9})$$

The boundary and initial conditions for (A.7) are as follows.

Initial Condition: $S(z, 0) = 1.0 - S_{wi}$.

Inlet Boundary Condition:

$$u_o = \frac{KK_{ro}}{\mu_o \phi} \left(\left(\frac{dP_c}{dS_o} \right) \left(\frac{\partial S_o}{\partial z} \right) + (\rho_{oil} - \rho_{gas}) g \right) = 0 \quad (\text{A.10})$$

$$z = 0, \quad t > 0.$$

The inlet boundary condition occasionally can be rewritten as $S_o = S_{org}$ @ $z = 0, t > 0$. This assumption only is valid when viscosity of displaced phase (i.e., oil phase) is relatively small.

Outlet Boundary Condition: $S(z, t) = 1$ @ $l < z < l$. L is the length of rock column and H is the height of capillary/gravity equilibrium. Since the governing partial differential equation of free gravity drainage is nonlinear it is not possible to find an exact solution analytically. Instead, if the capillary pressure is neglected, (A.7) is converted to (A.11) which is linear and can be solved by separation of variable method:

$$-\frac{K}{\mu_o \phi} (\rho_o - \rho_g) g \left(\frac{dK_{ro}}{dS_o} \right) \left(\frac{\partial S_o}{\partial z} \right) = \frac{\partial S_o}{\partial t}. \quad (\text{A.11})$$

Equation (A.12) shows the Corey-type function for oil relative permeability; using this correlation, Equation (A.11) is written as (A.13):

$$K_{ro} = K_{ro \max} \left(\frac{S_o - S_{org}}{1 - S_{org} - S_{wi}} \right)^{no}. \quad (\text{A.12})$$

In this correlation $K_{ro \max}$ is the end point oil relative permeability, no is Corey exponent and affects the degree of curvature in relative permeability curve, and S_{org} is the residual gas-oil saturation:

$$-\left[\frac{KK_{ro \max}^{no} (\rho_o - \rho_g) g}{\mu_o \phi (1 - S_{org} - S_{wi})} \right] \left(\frac{S_o - S_{org}}{1 - S_{org} - S_{wi}} \right)^{no-1} \times \left(\frac{\partial S_o}{\partial z} \right) = \frac{\partial S_o}{\partial t}. \quad (\text{A.13})$$

Assuming the above equation and following dimension less parameter this equation is simplified as (A.15):

$$S_{OD} = \frac{S_o - S_{org}}{1 - S_{org} - S_{wi}}, \quad (\text{A.14})$$

$$-\left(\frac{KK_{ro \max}^{no} (\rho_o - \rho_g) g}{\mu_o \phi (1 - S_{org} - S_{wi})} \right) (S_{OD})^{no-1} \left(\frac{\partial S_{OD}}{\partial z} \right) = \frac{\partial S_{OD}}{\partial t}. \quad (\text{A.15})$$

Assuming $s_{OD} = F(z) * G(t)$ and using (A.15) the following differential equation for $F(z)$ and $G(t)$ can be deduced:

$$\left(\frac{KK_{ro \max}^{no} (\rho_o - \rho_g) g}{\mu_o \phi (1 - S_{org} - S_{wi})} \right) F(z)^{no-2} \left(\frac{\partial F(z)}{\partial z} \right) = -\frac{\partial G(t)}{\partial t} G(t)^{-no}. \quad (\text{A.16})$$

Equation (A.16) can be solved by calculation of $F(z)$ and $G(t)$ using separation of variable method. The saturation function can be found by multiplication of these two functions and can be written as

$$S_{OD} = \left[\frac{\mu_o \phi (1 - S_{org} - S_{wi}) z}{KK_{ro \max}^{no} (\rho_o - \rho_g) g t} \right]^{1/(no-1)}. \quad (\text{A.17})$$

Occasionally this equation can be simplified as (A.19) using the following definition of dimensionless time:

$$t_{Dg} = \frac{\Delta \rho g K t}{\mu \phi (1 - S_{wir} - S_{org}) l}, \quad (\text{A.18})$$

$$S_{OD} = \left[\frac{z}{no t_{Dg} K_{ro \max} l} \right]^{1/(no-1)}. \quad (\text{A.19})$$

It should be noted that the above saturation function is valid for z values which are less than the gas penetration depth from top of the block at specific time. For larger z values the oil column is not affected by gas invasion and oil saturation is equal to $1 - S_{wi}$. Consequently, the solution of (A.16) should be written as

$$S_{OD} = \left[\frac{z}{no t_{Dg} K_{ro \max} l} \right]^{1/(no-1)} \quad @ 0 \leq z \leq l(t), \quad (\text{A.20})$$

$$S_{OD} = 1 \quad @ l(t) \leq z \leq l. \quad (\text{A.21})$$

In this solution $l(t)$ represents the penetration depth of gas from top of the block and can be found when the dimensionless saturation (s_{OD}) in (A.21) is equal to 1:

$$h(t) = no K_{ro \max} t_{Dg} l. \quad (\text{A.22})$$

At early stage, gas invades in the column and the invasion depth can be calculated by (A.22). The gas moves down

the oil in the column until it approaches capillary/gravity equilibrium. For this transition time, l ($t = t_{\text{transition}}$) becomes equal to l . From (A.22) one may conclude that the gas front moves linearly with time. For later stage, the gas cannot displace the oil beyond the capillary/gravity equilibrium height due to capillary end effect. The value of threshold height can be calculated by (A.23). In this equation $r_{\min}$ represents the smallest pore throat of porous media:

$$Z_{\text{threshold}} = \frac{P_{c\text{Threshold}}}{\Delta\rho g} = \frac{2\sigma_{og} \cos\theta_{og}}{r_{\min}\Delta\rho g}. \quad (\text{A.23})$$

The oil recovery factor can be found using saturation function as it is defined by following equation:

$$R = 1 - \frac{\overline{S_o}}{1 - S_{wi}} = 1 - \frac{(1/l) \int_0^l S_o dz}{1 - S_{wi}}. \quad (\text{A.24})$$

In early stage when the gas does not reach the gravity/capillary equilibrium the average residual oil saturation should be calculated as follows:

$$\overline{S_o} = \left(\frac{1}{l}\right) \int_0^l S_o dz = \left(\frac{1}{l}\right) \left[\int_0^{l(t)} S_o dz + \int_{l(t)}^l (1 - S_{wi}) dz \right]. \quad (\text{A.25})$$

Using (A.25), (A.26), and (A.21) the value of average residual oil saturation and recovery factor are

$$\overline{S_o} = 1 - S_{wi} - \left[(1 - S_{wi} - S_{\text{org}}) (K_{ro\text{max}} t_{Dg}) \right], \quad (\text{A.26})$$

$$R = \left(\frac{1 - S_{wi} - S_{\text{org}}}{1 - S_{wi}} \right) (t_{Dg} K_{ro\text{max}}). \quad (\text{A.27})$$

The value of cumulative oil production is also

$$Q_p = (Al\phi) (1 - S_{wi} - S_{\text{org}}) (t_{Dg} K_{ro\text{max}}). \quad (\text{A.28})$$

This equation shows that the cumulative oil production increased linearly as a function of time until the gas front reaches capillary/gravity equilibrium height. The velocity of oil in this stage is constant and is equal to

$$v_c = \frac{dQ_p}{Adt} = \frac{KK_{ro\text{max}}\Delta\rho g}{\mu_o}. \quad (\text{A.29})$$

Nomenclature

A :	Cross-sectional area
S_o :	Oil saturation
S_g :	Gas saturation
S :	Saturation
S_i :	Initial water saturation
S_{wi} :	Irreducible water saturation
S_{OD} :	Dimensionless saturation
ϕ :	Porosity
μ :	Viscosity
ρ :	Density
q :	Injection rate, volumetric flux
q_c :	Vertical flow rate
P :	Capillary pressure/parachor confident
P_c :	Pressure
$P_{c_{th}}$:	Threshold pressure
t_{Dg} :	Dimensionless time
N_c :	Capillary number
N_g :	Gravity number
N_B :	Bond number
N_d :	Diffusion number
$N_{Co.}$:	Combined number
N_{DE} :	Displacement efficiency number
$K_{ro\text{max}}$:	End point oil permeability
$K_{rg\text{max}}$:	End point gas permeability
K_r :	Relative permeability
K_{ro} :	Relative oil permeability
K_{rg} :	Relative gas permeability
ng :	Corey exponent in gas relative permeability function
no :	Corey exponent in oil relative permeability function
σ :	Interfacial tension
S :	Spreading coefficient
R :	Recovery factor
M :	Mobility ratio
PV :	Pore volume
l :	Length or height.

Subscripts

g:	Gas
w:	Water
o:	Oil
abs:	Absolute
wg:	Water-gas system
wo:	Water-oil system
og:	Oil-gas system.

Conflict of Interests

The author declares that there is no conflict of interests regarding the publishing of this paper. The paper will not reflect the viewpoints of the affiliated institutions by any means.

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