

Solubility of CO₂ in Brine and Reservoir Oil for Depleted Reservoirs in China

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Abstract: Many countries play an important role to carbon capture, utilization, and Storage (CCUS) to reduce emission. Geological storage is an important approach to cope with climate change. Live oil combined with saline aquifers and gas reservoirs are among the candidates for carbon dioxide (CO₂) sequestration, owing to their proven sealing capabilities and characterization. CO₂ mainly from power industrial was captured to improve oil recovery in some oilfields in China, and solubility is a key factor to evaluate storage ability of reservoir and saline aquifer. In this paper, CO₂ sequestration mechanisms in deplete reservoir and saline aquifer were explained. An experimental method to test solubility of CO₂ in emulsion system of liver oil and brine was developed. The experimental results showed that with the salinity increased, the solubility of CO₂ in reservoir oil increased slightly, and the solubility of CO₂ in brine decreased. Salinity has little impact on solubility of CO₂ in reservoir oil. With the pressure increased, the solubility of CO₂ in reservoir oil and brine both increased. Molecular diameter of reservoir oil and white oil have inverse proportion relationship with the solubility. Research on solubility of CO₂ in brine and reservoir oil for depleted reservoir in China enabled us to better predict and deal with the challenges for rapid development of carbon storage.

Keywords: CO₂ storage, solubility, depleted reservoir, saline aquifers.

1. INTRODUCTION

With the rapid development of economics, greenhouse gas (GHG) was continuously increasing in the past three decades. More than 70% of GHG emissions are composed of carbon dioxide (CO₂). CO₂ is one of the most important factors causing global warming [1]. Recent studies indicate that the atmospheric concentration of CO₂ increased from preindustrial level of 280 ppm to 380 ppm in 1994, and it is expected to 550 ppm by 2050 [2, 3]. Geological storage of CO₂ has been recognized as an efficient way to reduce greenhouse gas emissions and mitigate global warming. Besides, CO₂ capture, utilization, and storage (CCUS) is a technological approach to mitigate the emission of CO₂ from anthropogenic sources. Geological sequestration in saline aquifers, depleted reservoir, and gas reservoirs is an attractive option to reduce CO₂ emissions into the atmosphere.

Hydrocarbon reservoirs, either depleted or active, provide a large potential storage volume for CO₂ sequestration. The captured CO₂ was injected into oil fields to enhance oil recovery, usually called CO₂ enhanced oil recovery (CO₂-EOR). It is believed that CO₂ sequestration is not only an efficient method to increase the oil recovery, but also mitigate the global warming by permanently storing CO₂ into underground

[4, 5]. Mechanisms to increase oil recovery in CO₂ injection process include oil viscosity reduction, oil swelling due to dissolution of gas in crude oil, solution gas drive by gravity drainage, vaporization of lighter components of oil, interfacial tension reduction, and relative permeability effects. There was a good case that CO₂ immiscible and miscible flooding was carried out in Jilin Oil Field located in Songliao Basin of China, which was one of the most CO₂-CCS field pilot tests [6]. From 2008 to 2010, valid geological storage reached to 0.17 ton/bbl. CO₂ was also injected in Caoshe oil field in Jiangsu of China and accumulation injection reached to 0.17 million ton until last year [7]. The Weyburn CO₂ enhanced oil recovery project is forecasted to add another 130 million barrels and extend the life of the oilfield by 25 years, located in the province of Saskatchewan in Western Canada [8]. The CO₂CRC Otway Project is the first demonstration of geological CO₂ storage in Australia [9].

CO₂ capture in reservoirs during EOR is one of the most important techniques for CO₂ sequestration, and the injection of CO₂ into deep saline aquifers is the second importance method. According to an assessment report of the International Energy Agency (IEA) and the special committees of the Intergovernmental Panel on Climate Change (IPCC), the CO₂ storage capacity of global saline aquifers is estimated at 400 ~ 10,000 Gt [10]. The theoretical storage capacity of China's deep saline aquifers is approximately 3.02×10^{15} t; but the actual available capacity is only about 1.19×10^{11} t, which will still play an important role from a century-long perspective,

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considering security, economics and other factors [11]. In addition, depleted reservoirs have a higher storage capacity compared with aquifers. Current pilot CO₂ storage projects around the world are focusing on saline aquifers, such as the Sleipner site in the North Sea, the In Salah site in Algeria, the Snøhvit site in the Barents Sea [12], the pilot Gorgon project in northwest Australia [13], the Frio Brine Project in Texas US [14], and the Shenhua Carbon Capture and Storage (CCS) project in China [15]. A million-ton-scale CO₂ geological storage project in Western Taiwan was planned by the Taiwan Power Company in late 2010 [16].

Additionally, five oil reservoirs selected from Shengli and Jiangsu oilfields in China were carried out and the results show that CO₂ storage capacity can be greatly increased if including the adjacent aquifers. Oil and water displacement and CO₂ dissolution in remaining oil are the main forms of CO₂ trapping in oil reservoirs, trapping and dissolution in saline water are the main mechanisms for CO₂ storage within aquifers, which is similar to the gas injection design of the in Salah CO₂ storage project. CO₂ injection into combined reservoirs is a favorable option for the oil industry in the next years, and the potentials of CO₂ EOR and storage deserve further attention.

CO₂ solubility is a key factor for evaluating the CO₂ storage capacity in depleted reservoirs and aquifers. There were many experimental and analytical methods to obtain the CO₂ solubility [17]. However, the CO₂ solubility in reservoir oil combined with brine was rarely reported. Whether reservoir oil could influence the solubility of CO₂ in brine is very important to evaluate the storage capability of depleted reservoir combined with brine, and this will provide more accurate evaluation on storage capacity.

This paper is organized as follows. Firstly, emulsification phenomenon between reservoir oil and brine was observed by high speed microscope. Secondly, an experimental method to measure the CO₂ solubility in emulsion system was established. Thirdly, calculation method on solubility of CO₂ in brine and

reservoir oil was developed. Finally, the CO₂ solubility in reservoir oil or brine was determined respectively.

2. EXPERIMENTS

2.1. Materials

Carbon dioxide with a purity of 99.7% was supplied by Beijing Zhaoge Gases Industry Company. White oil was supplied by CNPC Corp and carbon number is from C₁₂ to C₂₈. Reservoir oil is supplied by the east of Hebei oil field Co. Ltd. in China. The oil density at 20 °C is 0.8376g/cm³. Component of the oil consisted of 0.59 mol% CO₂, 47.50 mol% N₂+C₁, 9.45 mol% C₂-C₆ and 42.46 mol% C₇₊. Distilled water was supplied by Beijing Chemical Industry Company. Table 1 shows the ion composition of brine.

2.2. Experimental Setup

A schematic diagram of the PVT apparatus (Figure 1) was manufactured by Sanchez Technology (ST) Co. Ltd. (France). Photo of PVT apparatus was shown in Figure 2 and internal profile of the PVT apparatus was shown in Figure 3. It was composed of four parts, which are an optical PVT cell, a pressure generator system, a sampling system, and data collecting system. The high-pressure visualization cell was mainly made of stainless steel, with total sample volume of 240 mL rated to 150 MPa and 473.15 K, equipped with a magnetic stirrer and a trunk piston was utilized for the experiment. To observe miscibility phenomenon, a sapphire window with a camera was fixed in PVT cell. The temperature of the experiment was adjusted by electrical heating around the steel. The temperature of cell was measured with three thermocouples calibrated inside the cell, and can be controlled within ±0.2 K. The pressure of the PVT cell was measured by a pressure sensor and the pressure was controlled by ±0.1 MPa. Computer software can record and take photos which were installed in the computer in advance.

High speed microscope was manufactured by Vision Research of America. It has a 12 bit and 1280 × 800 resolution sensor, the full picture of the rate of

Table 1: Ion Composition of Water (mg/L)

No.	NaHCO ₃	Na ₂ SO ₄	Na ₂ CO ₃	CaCl ₂	NaCl	MgCl ₂ ·6H ₂ O	Total
1	13680	4870	4035	880	11270	5270	8000.69
2	5.507	1.937	1.890	0.346	5.125	0.209	15014.07
3	39925	9685	9450	1730	38165	1045	20000.83

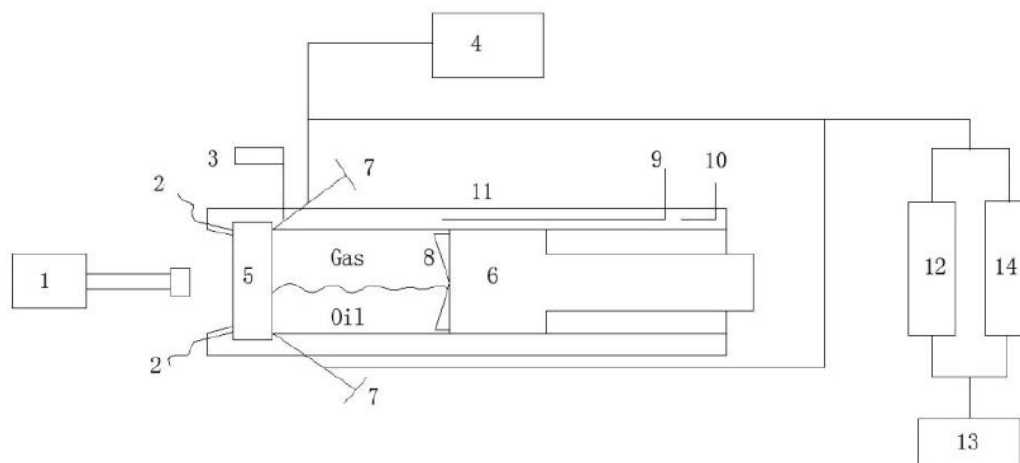


Figure 1: Schematic diagram of the PVT apparatus used in this work (1. Camera; 2. Fiber; 3. Pressure transducer; 4. GC; 5. Sapphire window; 6. Piston cylinder; 7. Valve; 8. Stirring Paddle; 9. Heating tube; 10. Temperature transducer; 11. PVT cell; 12. CO₂ gas bottle; 13. Pump; 14. Reservoir oil bottle).

3140 frames / second, and the highest rate of 500,000 frames / second shot. High speed centrifugal machine was manufactured by Beckman of America. Its max rotate speed was 12,000 r/min.



Figure 2: The photo of PVT apparatus.

2.3. Method

The operation procedures are similar to that of conventional reservoir oil and condensate gas PVT test described by Ali Danesh [18]. The whole system including PVT cell, the tubes, and sample containers which were cleansed, dried, and evacuated. High pressure CO₂, brine and reservoir oil stored in three different transfer cylinders, were then loaded sequentially to the PVT cell through the valve V7. The volume of CO₂ and brine were not crucial for the equilibrium as long as there were enough CO₂ and the system of brine and reservoir oil phases at equilibrium. PVT setup was then set to the experimental

temperature at 323.15 K. After the temperature was stabilized, the pressure was increased to the experimental pressure. The mixture was then stirred for 2 h, after that it was kept overnight under the experimental pressure in the 90 degree position as shown in Figure 3.

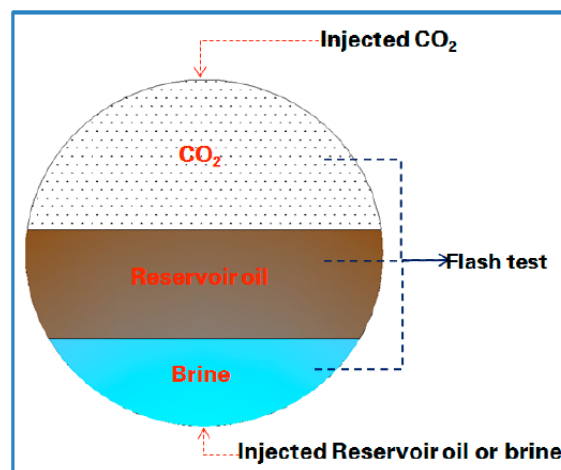


Figure 3: Internal profile of the PVT apparatus.

The experimental pressure was kept constant during the whole thermodynamic equilibration period, because the PVT setup was actually a plunger pump. Before sampling, it is necessary to purge the connection tubing at least 20 ml equilibrium phase to ensure representative sampling. The sampling cylinder was calibrated using distilled water before the experiment was evacuated, weighed and then attached to the PVT setup. The weight of the evacuated sampling cylinder was denoted by W_0 . Another evacuation from the top of the cylinder was conducted to remove the air in the short connection tubing between the cylinder and the valve V7. The mixture of reservoir oil and brine sample could be taken by

discharging the liquid phase slowly to the sampling cylinder. The whole sampling procedure was controlled by using the constant pressure mode of the PVT pump. Precaution must be taken to make sure that only the equilibrium mixture phase was sampled. The single phase high pressure brine was flashed into two phases in the sampling cylinder. Once the sampling cylinder was cooled down to room temperature T_{room} , its weight was measured again and denoted by W_1 . To determine how much CO₂ was dissolved in the sample, the dissolved gas was released slowly from the top of the sampling cylinder until no more gas was released and the weight of the cylinder did not change. The final weight of the cylinder, which contained CO₂ and mixture of reservoir oil and brine solution at atmospheric pressure, was denoted by W_2 . Then the mixture in the sampling cylinder was charged into a test tube and mixture volume was denoted by V_{co2} . All the weights were measured using an analytical balance with an accuracy of 0.001 g. Each sample was charged more than 10 ml. Following the procedures mention above, the 10MPa, 20MPa and 30MPa CO₂ solubility in reservoir oil and brine were measured respectively.

The total volume of CO₂ dissolved in the mixture, V_{co2} , consists of two parts:

$$V_{co2} = V_{co2}^w + V_{co2}^o \quad (1)$$

where V_{co2} , V_{co2}^w , and V_{co2}^o stand for the total volume of CO₂ released from the cylinder, the CO₂ dissolved in the brine, and the CO₂ dissolved in the oil in the cylinder at room temperature and atmospheric pressure, respectively. All the volume terms in Eq. (1) are at normal conditions of 273.15 K and 0.1013 MPa. In the following equations for calculating CO₂, we have used g, ml, and g/ml for all the weight, volume, and density quantities, respectively.

$$M_{o+w} = W_2 - W_1 \quad (2)$$

$$m_w + m_o = M_{o+w} \quad (3)$$

$$v_w + v_o = V_{co2} \quad (4)$$

Where M_{o+w} is the mixture weight of oil and brine. m_w and m_o are the weight of brine and oil in the test tube, respectively. v_w and v_o are the volume of the brine and oil at atmospheric pressure and room temperature, respectively. Substituting Eq. (4) into Eq. (3), Eq. (5) was got which was shown as follows.

$$\rho_w v_w + \rho_o v_o = M_{o+w} \quad (5)$$

In Eq. (5), ρ_w and ρ_o stands for the density of brine and dead oil at atmospheric pressure and room

temperature, respectively. See from Eq. (5), there were two unknown variations (v_w and v_o) and another equation was necessary. So, flash experiment was done twice. Another equation like Eq. (5) was got "" stands for the second flash experiment.

$$\rho'_w v'_w + \rho'_o v'_o = M'_{o+w} \quad (6)$$

Eq. (5) and Eq. (6) were both solved. v'_w , v'_o , v_w , and v_o were obtained.

$$GOR_o = \frac{V_{co2}^o}{v_o} \quad (7)$$

$$GOR_w = \frac{V_{co2}^w}{v_w} \quad (8)$$

$$GOR_o = \frac{V_{co2}^{o'}}{v'_o} \quad (9)$$

$$GOR_w = \frac{V_{co2}^{w'}}{v'_w} \quad (10)$$

Where GOR_o and GOR_w were gas liquid ratio. Substituting Eq. (7), Eq. (8) Eq. (9), and Eq. (10) into Eq. (1).

$$GOR_o v_o + GOR_w v_w = V_{co2} \quad (11)$$

$$GOR_o v'_o + GOR_w v'_w = V_{co2} \quad (12)$$

$$S_o = \frac{\frac{PV_{co2}^o}{ZRT}}{\frac{PV_{co2}^o}{ZRT} + \frac{m_o}{\bar{M}_0}} \quad (13)$$

$$S_w = \frac{\frac{PV_{co2}^w}{ZRT}}{\frac{PV_{co2}^w}{ZRT} + \frac{m_w}{18}} \quad (14)$$

Where S_o and S_w are the CO₂ solubility in oil and brine both in mol /mol, respectively. P , Z , R , and T are the atmospheric pressure, deviation factor, gas equilibrium constant, and room temperature, respectively. m_o , m_w and \bar{M}_0 are the weights of oil, the weights of water, and average molecular weight of oil, respectively.

3. CO₂ SEQUESTRATION MECHANISM IN DEPLETE RESERVOIR AND SALINE AQUIFER

The CO₂ geological sequestration technology can be classified into three major types: ocean

sequestration, geological sequestration, and vegetation sequestration. Geological sequestration is the technology that deposits the CO_2 under the ground, in the pores. For petroleum industry, geological sequestration is the most economic and practical one among the 3 types. Aiming at geological sequestration, there are four dominating storage mechanisms for geological sequestration, which are geological structure sequestration, dissolved gas sequestration, mineral reaction sequestration, and residual gas sequestration.

Firstly, geological structure sequestration is the main form of CO_2 storage after CO_2 injection through injection wells, in which mobile CO_2 gas is retained under buoyancy forces by impermeable cap rock, similar to condensate gas. Secondly, dissolved gas sequestration is formed due to CO_2 dissolve in formation water (including injected water and natural saline) and reservoir oil, which can be the main and the safest CO_2 storage way. The variation of dissolved quantity depends on the scale of the storage medium, temperature, pressure, and salinity. When the water salinity is in 3%, dissolving capacity is about the $47\text{--}51\text{kg/m}^3$; CO_2 will be filled with the corresponding pore volume $6.7\%\text{--}7.3\%$. If dissolved CO_2 reaches saturation in formation water, it will be a very important storage mechanism. Thirdly, mineral reaction sequestration may occur when CO_2 reacts with minerals in formation rocks and form solid carbonates and aqueous complexes, but the reaction is slow and its contribution to storage capacity is usually small. Because of new mineral in the course of mineralization process, mineral sequestration considered is the most durable and stable storage method. CO_2 and dissolution of rock minerals in chemical reaction can

form an ionic substance. Over millions of years, the CO_2 will be converted to carbonate mineral solid (clastic rock) or HCO_3^- ion (carbonate rock) to storage the CO_2 permanently. At last, residual gas sequestration is the CO_2 gas remained in small pores due to capillary forces after CO_2 displacement of water.

All in all, most of injected CO_2 will be trapped by structures and as residual gas in early stages, and then slowly transferred to dissolution and mineral reaction sequestration for long term storage. The processes of CO_2 injection into oil reservoirs and saline aquifers are different. When CO_2 is injected into oil reservoirs, it will displace oil and water, and some CO_2 will be dissolved in formation water and remaining oil and reacts with minerals in the reservoir. When CO_2 breaks through in production well, it will be recycled or re-injected to the reservoir. By contrast, CO_2 injection into saline aquifers is simpler than that of reservoir since the only formation fluid there is brine. The injection rate could be very high if the bottom hole pressure does not exceed the fracture pressure of the formation.

For the oil reservoirs associated with large scale saline aquifers, the aquifers can be a lateral or underlying extension of the upper oil formations. CO_2 will migrate upward or along the up dip direction due to buoyancy forces if it is injected at a lower part of the aquifer. CO_2 invasion into oil reservoir from associated aquifers will take an impact on EOR performance. The potential of CO_2 -enhanced oil recovery and storage can be optimized *via* the selection of injection location and process design based on the geological features of the sites.



(a)

(b)

Figure 4: The emulsion picture of reservoir oil and brine.

4. DISCUSSION AND RESULTS

4.1. The Emulsification of Reservoir Oil and Brine

Atypical picture of reservoir oil and water emulsion by microscopy is shown in Figure 4. It can be seen from the picture (a) in Figure 4 that the mixture from cylinder was centrifuged at 10,000 r/min, but was not separated into the water phase and oil phase, and the stable oil/water emulsion was observed using microscope. If water and oil was separated in two phases, water and oil could be transferred to cylinder and measured respectively, and dissolved gas from water or gas was also measured respectively. Aiming at mixture which cannot be separated before flash experiment, an experiment method to test the solubility of CO₂ in mixture was developed in this paper (in section 2.3).

4.2. Influence of Salinity on the Solubility

Solubility of CO₂ in reservoir oil and brine at different salinities was shown in Figure 5, respectively. It can be seen that with the salinity increased, the solubility of CO₂ in brine decreased and reservoir oil increased slightly. From 0 mg/L to 15000 mg/L, the slope changed from 10⁻⁶ to 10⁻⁷. When salinity reached 20000 mg/L, the slope was near zero and the solubility did not decrease as the salinity increased. If salinity increased, the space in the water molecular was occupied by more brine ion and the solubility of CO₂ in brine decreased. That was the reason that the solubility of CO₂ in deionized water was the most than that of brine. Contrary to brine, the solubility of CO₂ in reservoir oil increased lightly as the salinity increased. The salinity had little influence on the solubility of CO₂ in reservoir oil because of adequate injected CO₂.

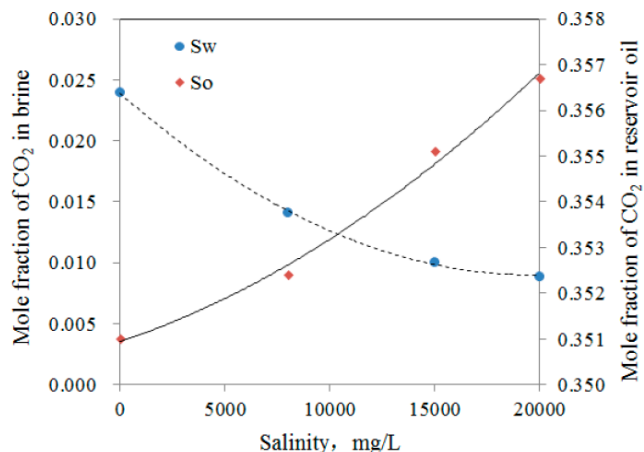


Figure 5: Solubility of CO₂ in reservoir oil and brine at different salinity.

There exists competition between reservoir oil and brine, so with the salinity increased, the solubility of CO₂ in reservoir oil increased and the solubility of brine decreased. Salinity took a weak role in solubility of CO₂ in reservoir oil.

4.3. Influence of Pressure on the Solubility

Solubility versus pressure of CO₂ in reservoir oil and brine was shown in Figure 6. It can be seen from Figure 6 that with the pressure increased, solubility of CO₂ in brine and reservoir oil both increased. The salinity of brine was constantly 20000 mg/L. Mole fraction of CO₂ in brine changed from 0.0089 to 0.0278, and pressure from 10 MPa to 30 MPa. However, Mole fraction of CO₂ in reservoir oil changed from 0.3567 to 0.7648, and pressure from 10 MPa to 30 MPa. Pressure has larger impact on the solubility of CO₂ in reservoir oil than brine. According to similarity principle, CO₂ which was non-polar molecular was easier to dissolve in non-polar reservoir oil than polar brine. Besides, coefficient of compressibility of reservoir oil was bigger than that of brine [19] indicates that when pressure was less than 10MPa, increment extent of CO₂ solubility in reservoir oil or brine was very big. When pressure was more than 10 MPa, increment extent of CO₂ solubility in reservoir oil or brine was very slower. CO₂ density phase diagram could explain that phenomenon.

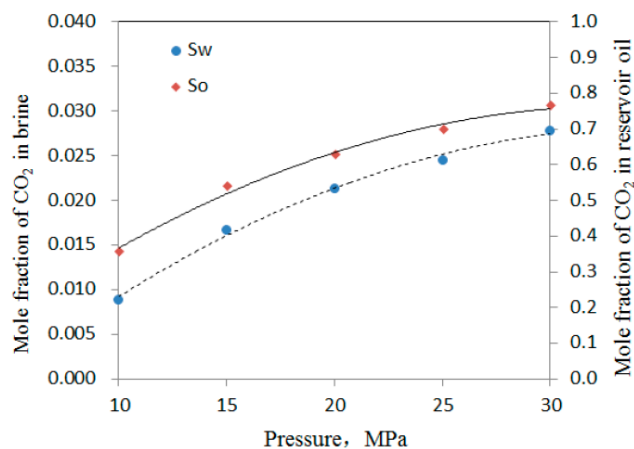


Figure 6: Solubility versus pressure of CO₂ in reservoir oil and brine, respectively.

4.4. Influence of Oil Type on the Solubility

Solubility of CO₂ in reservoir oil and white oil was shown in Figure 7. With pressure increases, the solubility of CO₂ in white oil and reservoir oil increases, and the solubility of CO₂ in white oil were higher than that of reservoir oil. Firstly, the intermolecular force

operating within white oil is smaller than that of reservoir oil because it is easier for CO₂ molecule to be squeezed in white oil under the same pressure, so that the solubility of CO₂ in white oil is stronger than that in reservoir oil. Another reason is that the smaller molecule of the oil, the smaller intermolecular force within CO₂ and the oil molecules is, and also the distance between CO₂ molecules and the oil molecules is large under the same pressure. Hence, white oil had more CO₂ than that of reservoir oil under the same pressure.

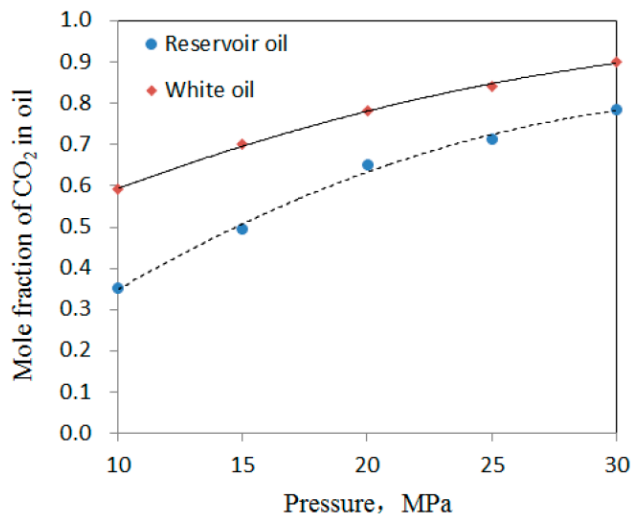


Figure 7: Solubility of CO₂ in reservoir oil and white oil.

5. CONCLUSIONS

Based on the results of the study, the conclusions can be made as follows:

1. CO₂ sequestration mechanisms in deplete reservoir and saline aquifer were illustrated in details.
2. Aiming at mixture of reservoir oil combined with brine in depleted reservoir cannot be separated after flash experiment, an experiment method to test the solubility of CO₂ in mixture was developed in this paper.
3. As the salinity increases, the solubility of CO₂ in reservoir oil increased lightly, and the solubility of CO₂ in brine decreased. Salinity plays only a small part in solubility of CO₂ in reservoir oil.
4. As the pressure increases, solubility of CO₂ in both brine and reservoir oil increases. The smaller molecular diameter scale was the bigger solubility of CO₂ in oil was.

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