

## Research Article

# Effect of Contact Time and Gas Component on Interfacial Tension of CO<sub>2</sub>/Crude Oil System by Pendant Drop Method

Xin Wang,<sup>1</sup> Lifeng Liu,<sup>2</sup> Zengmin Lun,<sup>1</sup> Chengyuan Lv,<sup>1</sup> Rui Wang,<sup>1</sup>  
Haitao Wang,<sup>1</sup> and Dong Zhang<sup>1</sup>

<sup>1</sup>EOR Technology Division, Petroleum Exploration & Production Research Institute, Sinopec, Beijing 100083, China

<sup>2</sup>Research Institute of Petroleum Exploration & Development, PetroChina, Beijing 100083, China

Correspondence should be addressed to Rui Wang; [wrzhdsg@126.com](mailto:wrzhdsg@126.com)

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Pendant drop method has been used to measure the equilibrium interfacial tension and dynamic interfacial tension of CO<sub>2</sub>/crude oil system under the simulated-formation condition, in which the temperature is 355.65 K and pressure ranges from 0 MPa to 30 MPa. The test results indicated that the equilibrium interfacial tension of CO<sub>2</sub>/crude oil systems decreased with the increase of the systematic pressure. The dynamic interfacial tension of CO<sub>2</sub>/original oil, CO<sub>2</sub>/remaining oil, and CO<sub>2</sub>/produced oil systems is large at the initial contact and decreases gradually after that, and then finally it reaches dynamic balance. In addition, the higher the pressure is, the larger the magnitude of changing of CO<sub>2</sub>/crude oil interfacial tension with time will reduce. Moreover, by PVT phase experiment, gas-oil ratio, gas composition, and well fluid composition have been got, and different contents of light components in three oil samples under reservoir conditions have also been calculated. The relationship between equilibrium interfacial tensions and pressures of three different components of crude oil and CO<sub>2</sub> system was studied, and the higher C<sub>1</sub> is, the lower C<sub>2</sub>-C<sub>10</sub> will be, and the equilibrium interfacial tension will get higher. Therefore, the effect of light weight fractions on interfacial tension under formation conditions was studied.

## 1. Introduction

The EOR technology of gas injection is theoretically better than water injection in the enhancement of oil recovery. Gas injection has developed rapidly in recent years overseas, and it has become the most important EOR method besides thermal recovery [1–3]. There are a variety of gases that can be injected, and CO<sub>2</sub>, for its wide sources and good flooding effect, has been widely put into practical application in oil fields [4–7]. When CO<sub>2</sub> is in a supercritical state, at the temperature above the critical temperature of 31.26°C and the pressure higher than the critical pressure of 7.2 MPa, its property will change. The density of CO<sub>2</sub> is close to liquid, and the viscosity of it is close to gas at this condition; moreover, the diffusion coefficient of the CO<sub>2</sub> is 100 times higher than liquid. Therefore, CO<sub>2</sub> has a great ability to dissolve in liquid [8–15]. This is helpful in improving the overall mass transfer rate when supercritical solubility increases and the reservoir

structure is conducive to increase inner diffusion and external diffusion, in order to increase the opportunity of the contact of CO<sub>2</sub> with oil and make them easier to mix. Therefore, CO<sub>2</sub> miscible flooding can meet the requirements for injected solvent in many oil fields [16–22].

The data of gas and crude oil interfacial tension under conditions of formation has very important theoretical and practical value, and the key of miscible flooding technology is to determine the minimum miscibility pressure of injection agent and crude oil [23–27]. The phenomenon that the interfacial tension changes with pressure significantly affects the reservoir fluid transfer behavior [28–31]. Miscible flooding technology is considered as one of the most cost-effective methods of EOR [32–35].

Pendant drop has been used to measure the relationship between the equilibrium interfacial tension and the pressure of CO<sub>2</sub>/crude oil system under the condition of the stratum temperature. Changes of interfacial tension with time

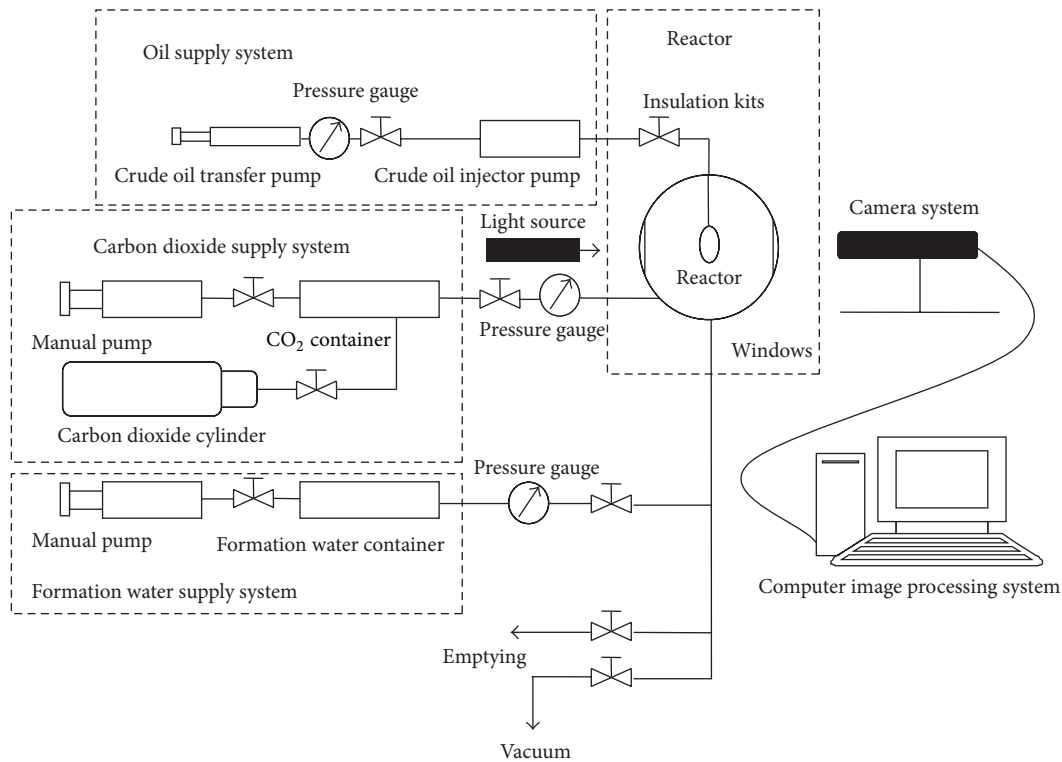


FIGURE 1: The schematic for the determination of interfacial tension by pendant drop method.

and pressure were measured by this method. By PVT phase experiment, the gas-oil ratio, gas composition, and well fluid composition have been measured. And the effect of light weight fractions on the interfacial tension under formation conditions was also calculated.

## 2. Experiment

**2.1. Experimental Apparatus.** High temperature and pressure interfacial tension meter has been used in the experiments, made by a French production company ST. The core of the device is a reactor with a window, which has an operating temperature of 0~200°C and a maximum working pressure of 70 MPa. Diameter of the needle used in the experiment is 0.81 mm, which is used for hanging droplets and using PVT analyzer for routine analysis.

Pendant drop method technology is the most accurate method to measure the interfacial tension under high temperature and high pressure conditions. First, the pump is used to form droplets on stainless steel needle department, and then the droplets shape photos are shot by amplifying camera system; after that the computer image processing systems are used to get the outer contour of the oil droplets. Ultimately calculated the interfacial tension, by using corrected image magnification, the density of light phase and heavy phase. The apparatus is shown in Figure 1.

**2.2. Experimental Samples.** Crude oil was provided by Zhongyuan oilfield, and formation temperature was 355.65 K. Experiment with CO<sub>2</sub> gas was produced by Beijing Hua

Yuan Co. with a purity of 99.995% and petroleum ether was produced by Sinopharm Chemical Reagent Company.

## 3. Experimental Phenomena and Results Analysis

### 3.1. CO<sub>2</sub>/Oil Equilibrium Interfacial Tension

#### 3.1.1. Experimental Phenomena

(1) *The Dissolution and Extraction Effect.* The experimental temperature is 82.5°C, and the pressure ranged from 0 MPa to 30 MPa. During the experiment, there is a medium exchange as the oil drop from the tip of the needle interacts with the CO<sub>2</sub> from the reactor on the condition that the experiment pressure is higher than the bubble point pressure [16–18]. CO<sub>2</sub> has been dissolved into the oil droplets constantly, and the light group of oil droplets also spread to CO<sub>2</sub> [19–22]. At the beginning of the contact of oil and CO<sub>2</sub>, the reaction is much stronger, and the light component of crude oil has been dissolved by supercritical CO<sub>2</sub> constantly [23–25], as shown in Figure 2. After some dissolution and extraction, the heavy component of crude oil will be left behind, and the crude oil and CO<sub>2</sub> will eventually reach equilibrium state as shown in Figure 3. The interfacial tension of this time can be seen as the equilibrium interfacial tension.

(2) *The Effect of Pressure on the Dissolution and Extraction.* It can be seen from the picture that, with the increase of the pressure, the extraction of crude oil increases, and the



FIGURE 2: The initial phase.



FIGURE 3: The equilibrium phase.

interface between  $\text{CO}_2$  and crude oil becomes unstable. A small amount of light components can be extracted out at the pressure of 16 MPa, but the extraction effect becomes more significant when the pressure reaches 30 MPa, as shown in Figure 4, taken at the initial state. At this time, the oil droplets gravity is equal to the  $\text{CO}_2$ /oil interfacial tension, so that the oil droplets could suspend on the needle without dripping.

**3.1.2. The Experimental Curve.** The relationship between interfacial tension and pressure is shown in Figure 5. The equilibrium interfacial tension between crude oil and  $\text{CO}_2$  decreases with the increase of pressure. The minimum miscibility pressure of the system calculated by extrapolation method is 18.97 MPa. When the pressure is lower than 18.97 MPa, the interfacial tension decreased rapidly; but while the pressure reaches 18.97 MPa, the reduction of interfacial tension will get slower.

**3.2. The Dynamic Interfacial Tension between Crude Oil and  $\text{CO}_2$ .** The experiment showed that the interaction between  $\text{CO}_2$  and crude oil is strong at the early stage, but with the extraction of light weight fractions of crude oil by  $\text{CO}_2$  and the dissolution of  $\text{CO}_2$  into the oil, the interfacial tension between them changes. In order to study the effect of this process on the interfacial tension, the contact time's effect on the interfacial tension between crude oil and  $\text{CO}_2$  should also be tested.

TABLE 1: The equilibrium interfacial tension of three  $\text{CO}_2$ /crude oil systems.

Oil sample	Equilibrium interfacial tension/MPa
Original oil	18.9744
Remaining oil	19.4748
Produced oil	20.5144

Figures 6, 7, and 8 show how the three oil samples' interfacial tension changes with time. The oil samples include original oil, remaining oil, and produced oil. It can be seen from the figure that the interfacial tension of  $\text{CO}_2$  and oil is large at the initial contact; but as the contact time is getting longer, the interfacial tension decreases gradually and eventually reaches dynamic balance.

Figures 9 and 10 compared the curves of  $\text{CO}_2$ /crude oil interfacial tension change with time in two different pressures. It can be seen from the figure that the higher the pressure is, the larger the magnitude of  $\text{CO}_2$ /crude oil interfacial tension changing with time will reduce. The value of equilibrium interfacial tension under 12 MPa is more than 90% of the initial interfacial tension, but it turns to 80% at the pressure of 21 MPa. Obviously, the interaction between  $\text{CO}_2$  and oil is stronger, and the change of interfacial tension is bigger at a higher pressure. This phenomenon is due to the fact that more lightweight components are extracted out and the dynamic mass transfer is getting active at high pressure. The actual reservoir  $\text{CO}_2$  flooding belongs to multicontact miscible flooding, it's a process that after the contact of  $\text{CO}_2$  and oil, and many times extraction and dissolution, the oil and  $\text{CO}_2$  eventually get miscible. This leads to the interfacial tension between crude oil and  $\text{CO}_2$  which becomes an inevitable result of dynamic change.

**3.3. Effects of Gas Composition on  $\text{CO}_2$ /Oil Interfacial Tension.** Using PVT phase experiment, gas-oil ratio, gas composition, and well fluid composition have been got as shown in Figures 11, 12, and 13. And calculate different contents of light weight fractions in the three oil samples under reservoir condition. Contents of  $C_1$ ,  $C_2-C_{10}$ , and  $C_{11+}$  of three oil samples are shown in Figure 14.

As can be seen from Figure 13, the higher the  $C_1$  is, the lower its corresponding  $C_2-C_{10}$  will be, and the  $C_{11+}$  content of the three oil samples is relatively at a low level.

Using extrapolation method, calculate the equilibrium interfacial tension of  $\text{CO}_2$ /original oil system,  $\text{CO}_2$ /remaining oil system, and  $\text{CO}_2$ /produced oil system. The equilibrium interfacial tension of three different systems is shown in Table 1.

It can be seen from Figure 13 and Table 1 that the content of  $C_1-C_{10}$  component is continually increasing. The higher  $C_1$  is, the lower  $C_2-C_{10}$  will be, and the equilibrium interfacial tension is getting higher. On the contrary, the lower  $C_1$  is, the higher  $C_2-C_{10}$  will be, and the equilibrium interfacial tension becomes lower. In other words,  $C_1$  is negative for  $\text{CO}_2$  and oil miscibility, and  $C_2-C_{10}$  promotes the miscibility of  $\text{CO}_2$  and oil system. Therefore, factors that affect the  $\text{CO}_2$  and crude oil system interfacial tension are pressure, gas composition,

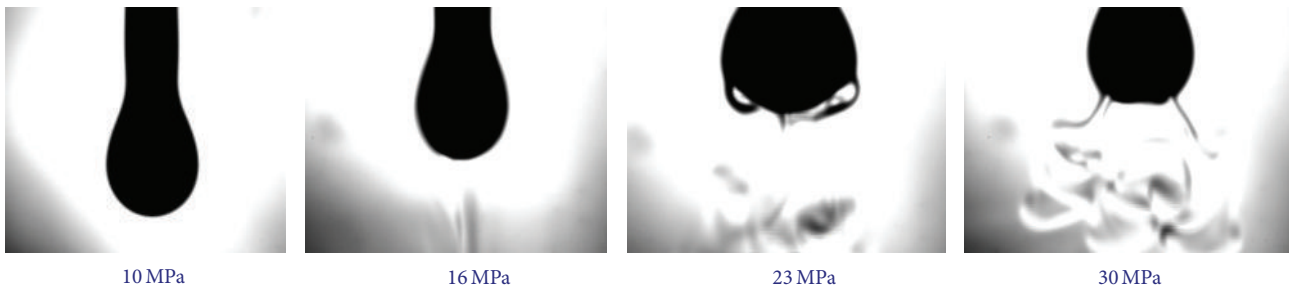


FIGURE 4: Images of the pendant oil drop in CO<sub>2</sub>/crude oil system under different pressure.

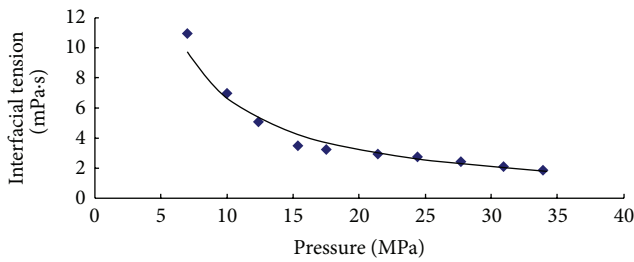


FIGURE 5: Interfacial tension of CO<sub>2</sub>/crude oil system under different pressure.

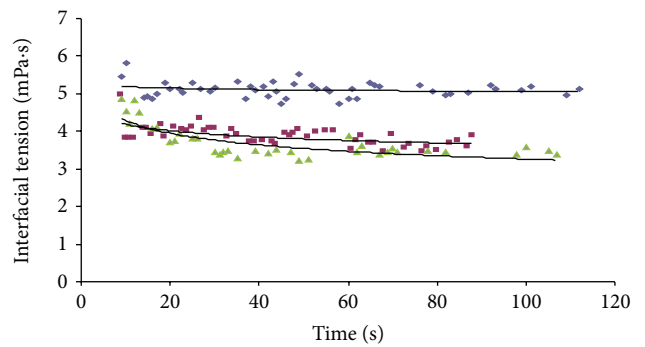


FIGURE 7: The relationship between interfacial tension and time of CO<sub>2</sub>/remaining oil system.

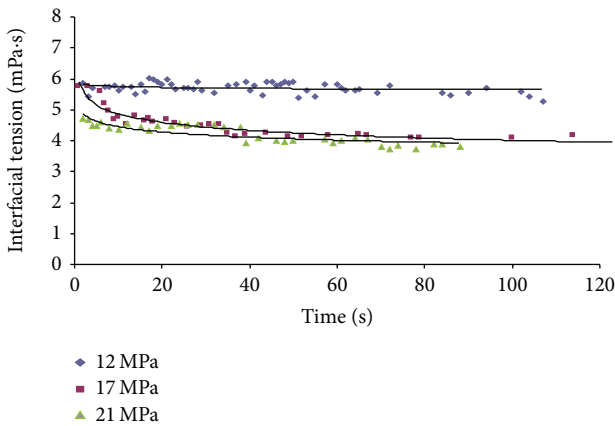


FIGURE 6: The relationship between interfacial tension and time of CO<sub>2</sub>/original oil system.

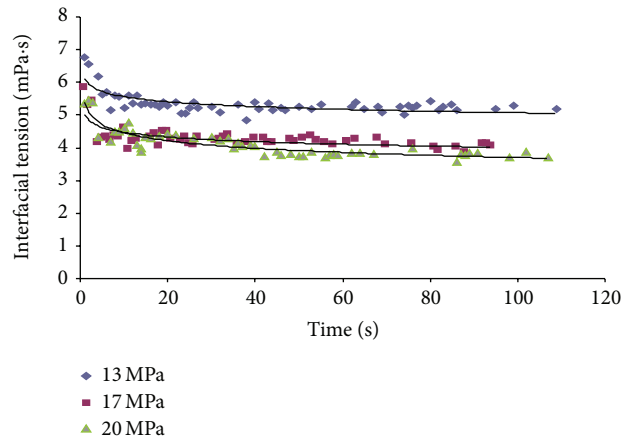


FIGURE 8: The relationship between interfacial tension and time of CO<sub>2</sub>/produced oil system.

and time. In hydrocarbons and CO<sub>2</sub> compound flooding process, the selection of C<sub>2</sub>-C<sub>10</sub> and CO<sub>2</sub> as a composite system will achieve better results in flooding.

Recent studies mostly focused on the research and application of equilibrium interfacial tension, and the study of dynamic interfacial tension is rare. But the actual reservoir CO<sub>2</sub> flooding belongs to multicontact miscible flooding, and this leads to the interfacial tension between crude oil and CO<sub>2</sub> becoming an inevitable result of dynamic change. So the dynamic interfacial tension can better simulate the situation under reservoir conditions and is more valuable. Historical studies do not mention hydrocarbons and CO<sub>2</sub> compound flooding, but the data from the experimental measurements are conducive to the development of hydrocarbons and

CO<sub>2</sub> compound flooding technology. The findings not only provide experimental evidence for indoor hydrocarbons and CO<sub>2</sub> compound flooding but also make it not just stick to a single CO<sub>2</sub> flooding. The experimental results provide a broad prospect for CO<sub>2</sub> flooding and opened up new areas for hydrocarbons and CO<sub>2</sub> compound flooding technology.

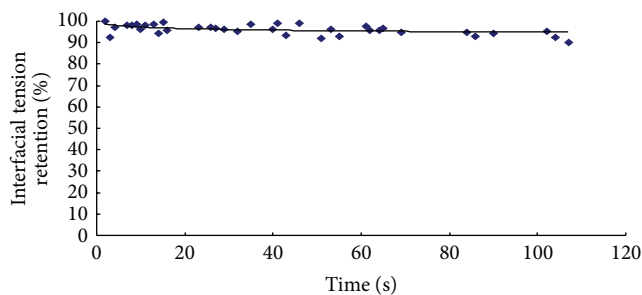


FIGURE 9: The curve of CO<sub>2</sub>/crude oil system interfacial tension change with time under 12 MPa.

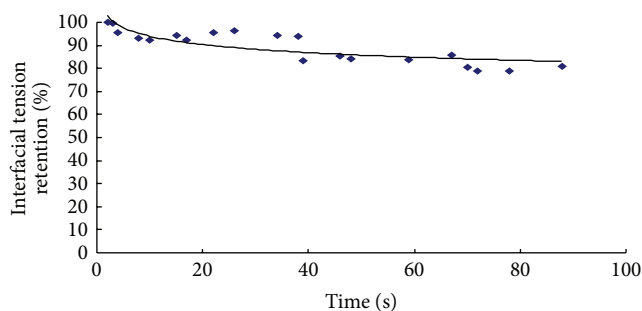


FIGURE 10: The curve of CO<sub>2</sub>/crude oil system interfacial tension change with time under 21 MPa.

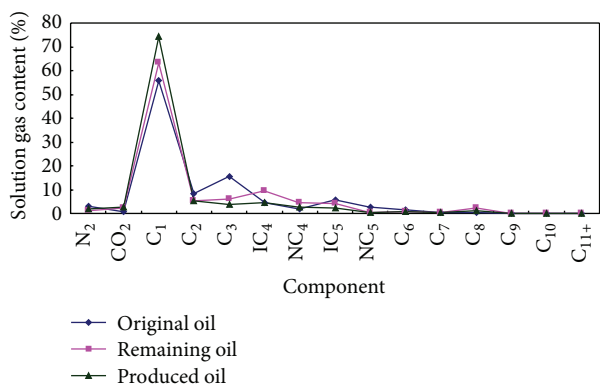


FIGURE 11: Gas phase composition of crude oil.

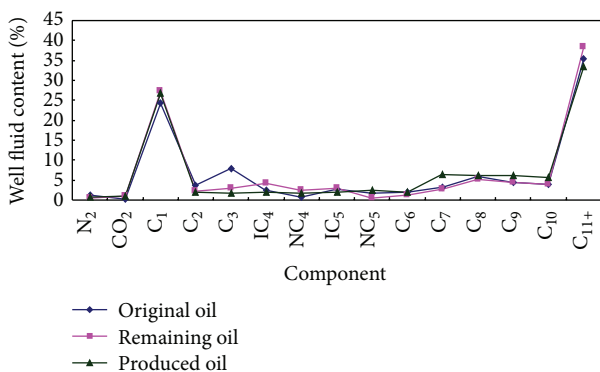


FIGURE 12: Well fluid composition of crude oil.

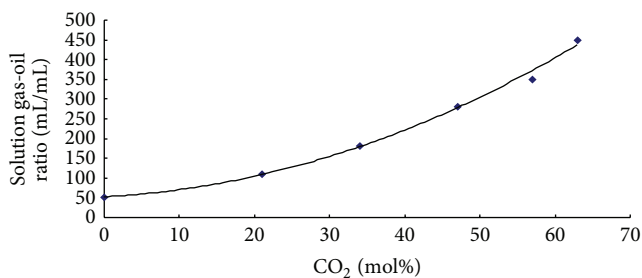


FIGURE 13: Gas-oil ratio of crude oil.

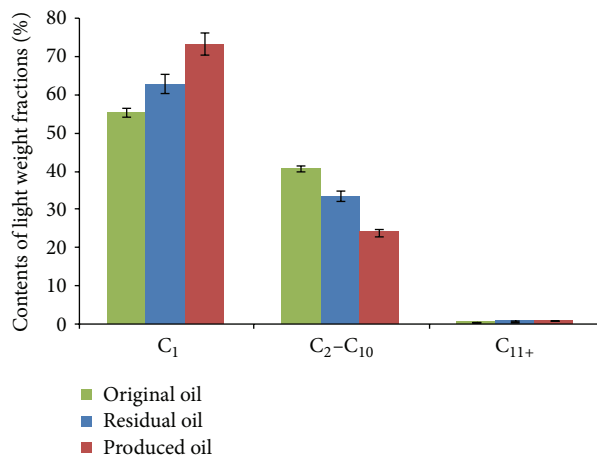


FIGURE 14: Content of different light weight fractions of original oil, remaining oil, and produced oil.

### 4. Conclusions

(1) Using the pendant drop method, the interaction between CO<sub>2</sub> and crude oil can be seen through the reactor under simulated-formation conditions of temperature and pressure. There is a strong mutual diffusion at the beginning of the contact of CO<sub>2</sub> and crude oil, and as the pressure goes higher, the dissolution and extraction become easier.

(2) Data of CO<sub>2</sub> and crude oil interfacial tension under conditions of the temperature of 355.65 K and the pressure ranging from 0 MPa to 30 MPa were measured by experiment. Experimental results show that CO<sub>2</sub>/crude oil equilibrium interfacial tension decreases with the increasing of pressure.

(3) The interfacial tension of CO<sub>2</sub> and oil is large at the initial contact, and as time is getting longer, the interfacial tension decreases gradually and eventually reaches dynamic balance. Moreover, the higher the pressure is, the more the magnitude of changing of CO<sub>2</sub>/crude oil interfacial tension with time will reduce.

(4) The higher the content of C<sub>1</sub> in crude oil is, the lower the content of C<sub>2</sub>-C<sub>10</sub> will be, and the equilibrium interfacial tension will get higher. On the contrary, the lower the content of C<sub>1</sub> is, the higher the content of C<sub>2</sub>-C<sub>10</sub> will be, and the equilibrium interfacial tension will become lower. In other words, C<sub>1</sub> is negative for the miscibility of CO<sub>2</sub> and oil, and C<sub>2</sub>-C<sub>10</sub> can promote the miscibility of CO<sub>2</sub> and crude oil system.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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