Enhanced solvent dissolution into in-situ upgraded heavy oil under different pressures

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Abstract

During a solvent-based heavy oil recovery process, asphaltene precipitation may occur when a solvent is dissolved into heavy oil under certain reservoir conditions. Thus, such an in-situ upgraded heavy oil with a lower asphaltene content is less viscous and easier to be recovered. In this paper, enhanced solvent dissolution into the in-situ upgraded heavy oil is studied. First, three heavy oil samples with different asphaltene contents are tested to model the upgraded heavy oils to rather different extents. Then the propane solubilities in these three heavy oil samples and the viscosities of the three heavy oil–propane systems are measured at five equilibrium pressures ranging from 200 to 800 kPa. Also, the propane diffusivities and oil-swelling factors of the three heavy oil–propane systems are measured by applying the newly developed dynamic pendant drop volume analysis (DPDVA) method in the same pressure range. The detailed experimental results show that the asphaltene content in heavy oil strongly affects propane dissolution into the heavy oil. The propane solubility in the maltenes is found to be the highest, which results in the largest oil-swelling factor among the three heavy oil samples tested. Expectedly, the viscosity of the heavy oil–maltene system is the lowest and accordingly the propane molecular diffusivity in the maltenes is the largest. The in-situ upgrading of heavy oil during a solvent-based recovery process greatly enhances further solvent dissolution into heavy oil and reduces its viscosity.
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Keywords: Solvent-based heavy oil recovery; In-situ upgrading; Asphaltene precipitation; Heavy oil–solvent system

1. Introduction

Depletion of conventional petroleum reserves and increase of hydrocarbon fuel demands require effective and economical recovery of unconventional heavy oil and bitumen resources. The Western Canada contains tremendous heavy oil and bitumen with estimated original-oil-in-place (OOIP) of 2.5 trillion barrels [1]. Nevertheless, heavy oil and bitumen cannot be effectively recovered by using some conventional oil recovery methods due to their extremely high viscosities. The cold heavy oil production (CHOP) is a major recovery process, in which heavy oil and bitumen are produced with sands under solution–gas drive [2]. After the cold production, there is still 85–95% of the OOIP unrecovered at the economic limit [3]. In practice, thermal-based tertiary oil recovery methods, such as steam-assisted gravity drainage (SAGD) process [4], are often applied because they can dramatically reduce the heavy oil viscosity. However, large heating requirement makes the thermal-based methods ineffective and uneconomical for many Canadian heavy oil reservoirs with thin pay zones, underlying bottom water, overlying gas caps, low rock conductivities, large vertical fractures, and high water saturations [5].

In the past, a number of studies have been conducted to explore the potential of non-thermal enhanced oil recovery (EOR) methods for the heavy oil reservoirs. Solvent-based recovery processes, such as vapor extraction (VAPEX) [6–9] and cyclic solvent injection [3,10], are among the most promising heavy oil recovery methods under investigation. In the VAPEX process, for example, gaseous condensable solvents [11], in conjunction with carrier gases [12], are used to extract heavy oil from the reservoir formations. The major oil recovery mechanism in this process is significant viscosity reduction of heavy oil through sufficient solvent dissolution and possible asphaltene precipitation. The solvent-based heavy oil recovery

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processes can eliminate several major technical problems associated with the thermal-based EOR methods, such as source water supply, formation heat losses, and produced water treatment [7].

In the solvent-based heavy oil recovery processes, solvent dissolution into heavy oil plays a dominant role in determining the subsequent heavy oil recovery rate. The molecular diffusivity and solubility of a solvent in heavy oil are the two most important parameters to quantify the solvent diffusion rate and dissolution amount respectively, both of which strongly depend on the physicochemical properties of the heavy oil–solvent system and the operating pressure and temperature as well. On the other hand, it has long been found that the heavy oil properties are largely determined by its heaviest and most polar components, i.e., asphaltenes [13,14]. When certain solvent is made in contact with heavy oil at a sufficiently high reservoir pressure, asphaltene precipitation occurs so that the heavy oil in the reservoir is in-situ upgraded. Such an in-situ upgraded heavy oil has a lower viscosity and a higher API gravity [7,15]. In the literature, several experimental studies report that asphaltene precipitation has been observed to occur in different VAPEX physical models [15–17]. However, it is unknown whether such an in-situ upgrading process enhances further solvent dissolution into heavy oil under the practical reservoir conditions.

In this paper, three heavy oil samples with different asphaltene contents of 0.0 wt.% (i.e., the maltenes), 7.0 wt.% (the reconstituted oil), and 14.5 wt.% (the original oil) are tested to model the heavy oils that are in-situ upgraded to rather different extents. These heavy oil samples are saturated with propane at five different equilibrium pressures in the range of $P = 200$–800 kPa and $T = 23.9\,°C$. It is found that the following properties of three heavy oil–propane systems with different asphaltene contents are rather different even at the same equilibrium pressure. In general, the propane solubility is higher and the oil-swelling factor is larger in the heavy oil sample with a lower asphaltene content. Consequently, the viscosity of the heavy oil–propane system is lower, whereas its molecular diffusivity is larger if the heavy oil contains a lower asphaltene content. In addition, a commercial phase behavior simulation module with the Peng–Robinson equation of state (P–R EOS) is used to predict the propane solubilities in different heavy oils at different equilibrium pressures. Finally, some existing empirical correlations are applied to analyze the overall relationships among the propane concentration, oil-swelling factor, viscosity, and molecular diffusivity.

2. Experimental

2.1. Materials

The original heavy oil is collected from the Lloydminster area, Canada. The density and viscosity of the cleaned field heavy oil sample are equal to $\rho_{oil} = 988\,kg/m^3$ and $\mu_{oil} = 24{,}137\,mPa\,s$ at the atmospheric pressure and reservoir temperature of $T = 23.9\,°C$, respectively. The compositional analysis result of this heavy oil obtained by using the simulated distillation is given in Table 1. It can be seen from this table

Table 1
Compositional analysis result of the original heavy oil with the asphaltene content of $w_{asp} = 14.5$ wt.% (*n*-pentane insoluble)

Carbon number	wt.%
C_1	0.00
C_2	0.00
C_3	0.00
C_4	0.00
C_5	0.00
C_6	0.00
C_7	0.00
C_8	0.00
C_9	0.00
C_{10}	0.00
C_{11}	0.00
C_{12}	0.86
C_{13}	1.14
C_{14}	1.33
C_{15}	1.74
C_{16}	1.75
C_{17}	2.01
C_{18}	2.08
C_{19}	2.13
C_{20}	1.87
C_{21}	2.28
C_{22}	1.50
C_{23}	2.24
C_{24}	2.08
C_{25}	1.66
C_{26}	1.56
C_{27}	1.80
C_{28}	1.85
C_{29}	1.56
C_{30}	1.50
C_{31}	1.93
C_{32}	1.78
C_{33}	1.04
C_{34}	1.02
C_{35}	1.47
C_{36}	1.40
C_{37}	0.87
C_{38}	0.85
C_{39}	1.35
C_{40}	1.23
C_{41}	0.62
C_{42}	0.61
C_{43}	1.10
C_{44}	0.80
C_{45}	0.80
C_{46}	0.57
C_{47}	0.72
C_{48}	0.72
C_{49}	0.68
C_{50+}	47.50
Total	100.00

that there are no light components under C_{12} and that there is a large fraction of heavy components with $C_{50+} = 47.50$ wt.%. Given the measured asphaltene content of $w_{asp} = 11.5$ wt.% (*n*-heptane insoluble) [18] or $w_{asp} = 14.5$ wt.% (*n*-pentane insoluble) [19], it is obvious that the carbon numbers of most asphaltenes in this heavy oil are much larger than C_{50}. The purities of propane (Praxair, USA) and *n*-pentane (Fisher Scientific, USA) used are 99.5 and 99.9%, respectively.

2.2. Oil sample preparation

The experimental procedure for preparing the reconstituted heavy oil sample with a known asphaltene content is described below. In the experiment, first, asphaltenes are precipitated from the original heavy oil by using the standard ASTM D2007 method. More specifically, $100 \, cm^3$ of heavy oil is mixed with $4000 \, cm^3$ of n-pentane that is used as a precipitant. The mixture is agitated by using a magnetic stirrer (SP46925, Barnstead/Thermolyne Corporation, USA) for 12 h and then filtered through a $2.5 \, \mu m$ filter paper (Whatman, England). The filter cake, which is mainly composed of precipitated asphaltene particles, is kept rinsing with n-pentane until it remains colorless. On the other hand, the filtrate is distilled to recover the dissolved n-pentane and the remainder is the maltenes. Such separated asphaltene particles and maltenes are slowly dried on the heated magnetic stirrer to remove any dissolved n-pentane until their weights do not change from the readings of an electric balance (PM 4600, Mettler Toledo, Canada). The density of the maltenes is measured by using a digital densitometer (DMA 512P, Anton Paar, USA) and found to be $\rho_{mal} = 962 \, kg/m^3$ at the atmospheric pressure and $23.9 \, °C$. Then the asphaltene particles are ground and sifted through a $75 \, \mu m$ sieve (RX-29, Fisher Scientific, USA). Finally, the reconstituted heavy oil sample is prepared by dispersing 7.0 wt.% of sifted asphaltene particles into the maltenes and agitating the mixture for 1 h at $T = 80 \, °C$. In this study, the following three heavy oil samples with different asphaltene contents are tested: maltenes ($w_{asp} = 0.0$ wt.%), reconstituted oil ($w_{asp} = 7.0$ wt.%), and original oil ($w_{asp} = 14.5$ wt.%).

2.3. Solubility measurement

In this study, three different heavy oils are saturated with propane in the pressure range of $P = 200$–800 kPa and at $T = 23.9 \, °C$, respectively. The detailed experimental procedure for measuring propane solubility in each heavy oil sample at an equilibrium pressure is described below. First, $100 \, cm^3$ of heavy oil sample is introduced into a vacuumed high-pressure saturation cell (P/N, HTPO198-P20, Chandler Engineering, USA) with a total net volume of $780 \, cm^3$. Then propane is injected into the saturation cell, which is kept rotating continuously for 1 week to mix the heavy oil sample with the solvent. It should be noted that additional propane may be added into the saturation cell in order to reach the pre-specified equilibrium pressure. After the heavy oil–propane system reaches the equilibrium state as indicated by a constant cell pressure, approximately $15 \, cm^3$ of the solvent-saturated heavy oil is taken from the saturation cell and flashed under the atmospheric conditions. The volume of propane released during this flash vaporization process is measured by using a JEFRI gasometer (GOR-10-S, DBR, Canada) with a maximum gas volume measurement relative error of 0.5%. The measured solvent volume is finally converted into solvent mass by using the P–R EOS, which results in a relative error of approximately 1.0% under the atmospheric conditions. The flashed-off heavy oil is weighed on an electric balance (PM 4600, Mettler Toledo, Canada) with an accuracy of 0.1% in the

range of 0–1000 g. In this study, solubility is defined as the ratio of the mass of the dissolved propane to that of the heavy oil sample with no solvent dissolution, i.e., $g \, C_3H_8/100 \, g$ oil. Therefore, the maximum relative error for such determined solubility is estimated to be 1.6%.

2.4. Viscosity measurement

After the solubility measurement, approximately $70 \, cm^3$ of the remaining propane-saturated heavy oil is transferred from the saturation cell to a high-pressure high-temperature rolling-ball viscometer (P/N 1602-830, Chandler Engineering, USA) for viscosity measurement. This viscometer is capable of measuring liquid viscosity from 0.2 to 20,000 mPa s in large ranges of pressure (up to 69 MPa) and temperature (15–150 °C).

2.5. Measurements of molecular diffusivity and oil-swelling factor

In this work, a newly developed dynamic pendant drop volume analysis (DPDVA) method is applied to measure the molecular diffusivity and oil-swelling factor of the heavy oil–propane system simultaneously. A brief description of the DPDVA method is given below and its technical details can be found elsewhere [20,21]. Fig. 1 shows the schematic diagram of the experimental setup for applying the DPDVA method. The major component of the setup is a see-through windowed high-pressure cell, which is filled with a test solvent at the pre-specified pressure and temperature. Then a heavy oil sample is introduced to form a pendant oil drop inside the pressure cell. Subsequent solvent dissolution into the pendant oil drop causes its volume to increase (i.e., the oil-swelling effect) until it is completely saturated with the solvent. The sequential digital images of the dynamic pendant oil drop are acquired and analyzed to measure the oil drop volumes at different times. Theoretically, a mass-transfer model is formulated to describe the molecular diffusion process of the solvent in the pendant oil drop. This model is solved numerically by applying the semi-discrete Galerkin finite element method. With the guessing values of solvent molecular diffusivity and oil-swelling factor, the volume of the dynamic pendant oil drop at any time is calculated from the predicted transient solvent concentration distribution inside the pendant oil drop. Mathematically, an objective function is constructed to express the overall discrepancy between the theoretically calculated and experimentally measured volumes of the dynamic pendant oil drop at different times. The solvent molecular diffusivity and oil-swelling factor of the heavy oil–solvent system are used as adjustable parameters and thus determined once the objective function is minimized.

3. EOS modeling

In phase behaviour studies, the vapor-liquid equilibrium (VLE) of reservoir fluids under the actual reservoir conditions is usually modeled by using an equation of state (EOS). The EOS modeling requires the critical pressure (P_c), critical temperature (T_c), and Pitzer acentric factor (ω) for each component of the
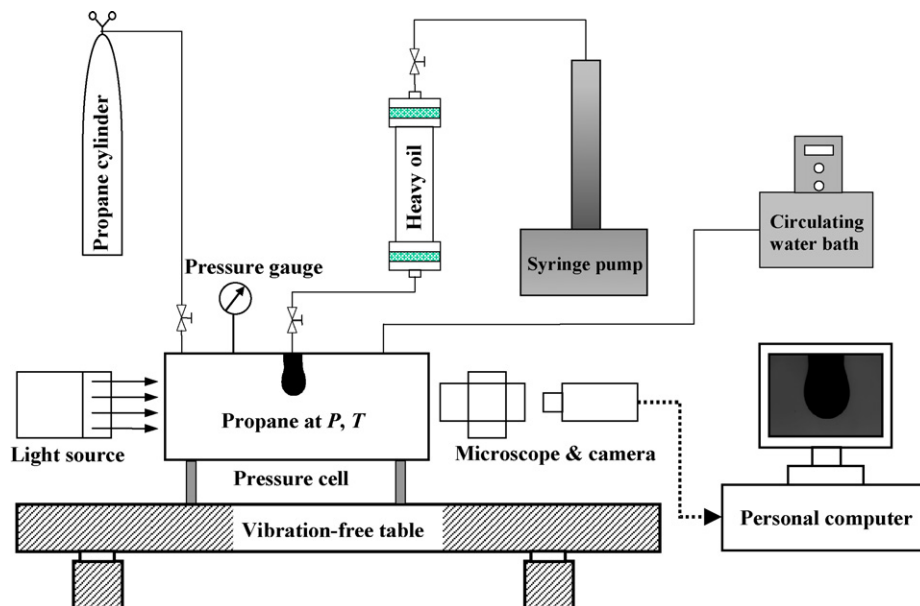
Fig. 1. Schematic diagram of the experimental setup used for measuring the solvent molecular diffusivity and oil-swelling factor of the heavy oil–solvent system by applying the dynamic pendant drop volume analysis (DPDVA) method [20,21].

fluids. These requirements are difficult to meet for heavy oil due to its extremely complicated composition. In practice, heavy oil is often represented by a series of pseudo components, each of which is treated as a single component with definite P_c, T_c and ω. In this study, with the compositional analysis result given in Table 1, the original heavy oil is roughly subdivided into the following four pseudo components: component #1 (C_{12}–C_{30}), component #2 (C_{31}–C_{45}), component #3 (C_{46}–asphaltenes), and component #4 (asphaltenes). In this lumping scheme, the first three pseudo components altogether represent the maltenes, whereas the fourth pseudo component alone represents the asphaltene content in the heavy oil. The reconstituted heavy oil consists of the first three pseudo components (i.e., maltenes) with 7.0 wt.% of component #4 (asphaltenes).

Some important properties of the four pseudo components are determined by using the CMG Winprop module (Version 2004.12, Computer Modelling Group Ltd., Canada) with the P–R EOS [22]. More specifically, the molecular weight M, specific gravity γ, P_c, T_c, and ω for each of components #1 and #2 are calculated from the Winprop's hydrocarbon component library up to C_{45}. The molecular weight M and specific gravity γ for each of components #3 and #4 are used as adjustable parameters to tune the P–R EOS and thus determined once the calculated densities of the maltenes and the original heavy oil are equal to their measured data, respectively [23]. For each of components #3 and #4, its P_c, T_c, and ω are calculated from its determined molecular weight M and specific gravity γ by using some existing correlations. These properties of the four pseudo components are needed in predicting the propane solubilities in the three heavy oil samples at different equilibrium pressures and listed in Table 2. It is seen from the molecular weight M and specific gravity γ that the pseudo component with larger carbon numbers has larger molecular weight and becomes heavier. In general, its critical temperature T_c and Pitzer acentric factor ω

increase, whereas its critical pressure P_c decreases as its carbon numbers increase. With the determined molecular weights and mole fractions of the four pseudo components, the apparent molecular weight of each heavy oil sample is calculated to be $M_{oil} = 393$ kg/kmol (maltenes), 411 kg/kmol (reconstituted oil), and 432 kg/kmol (original oil), respectively.

In order to predict the propane solubilities in three heavy oil samples with different asphaltene contents at different equilibrium pressures and $T = 23.9\,^\circ C$, propane in each heavy oil–solvent system is considered as component #5, in addition to the above-mentioned four pseudo components. With the above-calculated P_c, T_c, and ω values for the four pseudo components and those for propane, at a given pressure and $T = 23.9\,^\circ C$, two-phase flash calculations are performed to determine the fractions and compositions of the liquid and gas phases and predict the propane solubility in each heavy oil sample. The so-called binary interaction coefficients δ_{ij} ($i, j = 1, 2, 3, 4, 5, i < j$) for each pair of the five components are used as adjustable parameters and thus determined once the predicted propane solubilities best match the measured data for the three different heavy oil samples at five equilibrium pressures and $T = 23.9\,^\circ C$.

In particular, the binary interaction coefficient δ_{i5} ($i = 1, 2, 3, 4$) for each of the four pseudo components with propane (component #5) is listed in Table 2. It should be pointed out that the binary interaction coefficients δ_{ij} ($i, j = 1, 2, 3, 4, i < j$) among the four pseudo components are found to be approximately one order smaller than that between each of the first three pseudo components and propane and thus are not included in the table. It is clearly seen from Table 2 that asphaltenes (component #4) have significantly larger binary interaction coefficient δ_{45} with propane than the first three pseudo components, δ_{i5} ($i = 1, 2, 3$). Physically, the binary interaction coefficient accounts for the interactions between two different components [24,25]. These results indicate that asphaltenes have the strongest interactions

Table 2
Some important properties of the four pseudo components of the original heavy oil calculated by using the CMG Winprop module with the P–R EOS

	Pseudo component			
	#1 (C_{12}–C_{30})	#2 (C_{31}–C_{45})	#3 (C_{46}–asphaltenes)	#4 (asphaltenes)
Weight percentage (wt.%)	32.94	16.87	35.69	14.50
Molecular weight, M (kg/kmol)	273	460	590	1072
Specific gravity, $\gamma = \rho_o/\rho_w$	0.8679	0.9234	1.1075	1.1779
Mole fraction (mol.%)	52.13	15.86	26.16	5.85
Critical pressure, P_c (MPa)	1.458	0.860	1.210	0.863
Critical temperature, T_c (K)	781.6	914.5	1125.8	1293.8
Pitzer acentric factor, ω	0.810	1.200	1.164	1.381
Binary interaction coefficient δ_{i5} with propane	0.0127	0.0216	0.0196	0.2347

Table 3
The detailed experimental results of three heavy oil–propane systems at P = 200, 350, 500, 650 and 800 kPa at T = 23.9 °C

Experimental data	Heavy oil sample	Equilibrium pressure (kPa)				
		200	350	500	650	800
Solubility (g C_3H_8/100 g oil)	Maltenes	1.69	6.02	13.19	19.58	52.39
	Reconstituted oil	1.58	5.61	9.42	15.00	42.09
	Original oil	1.28	5.00	6.20	9.29	21.28
Oil-swelling factor	Maltenes	1.060	1.114	1.177	1.303	1.537
	Reconstituted oil	1.060	1.086	1.167	1.256	1.497
	Original oil	1.047	1.081	1.141	1.205	1.325
Viscosity (mPa s)	Maltenes	272.39	83.37	23.66	12.94	4.65
	Reconstituted oil	824.71	393.72	133.31	39.08	6.22
	Original oil	2493.23	1574.11	156.02	38.88	22.84
Molecular diffusivity (10^{-9} m²/s)	Maltenes	0.083	0.270	0.395	0.758	1.610
	Reconstituted oil	0.077	0.175	0.266	0.649	0.904
	Original oil	0.053	0.077	0.144	0.300	0.490

with propane, in comparison with the first three pseudo components in the original heavy oil. Hence, in the present case, the asphaltene content in heavy oil affects its propane solubility to the largest extent [26].

4. Results and discussion

In this study, the solubility, oil-swelling factor, viscosity, and molecular diffusivity of three propane-saturated heavy oil samples with different asphaltene contents are measured at P = 200, 350, 500, 650 and 800 kPa and T = 23.9 °C, respectively. This temperature is the same as that of the actual reservoir from which the original heavy oil is collected. The equilibrium pressures are selected below the vapour pressure (P_v = 923 kPa) of propane at T = 23.9 °C. Table 3 lists all the measured experimental results for three different heavy oil–propane systems.

4.1. Propane solubility in heavy oil

Fig. 2 shows the measured (symbols) and calculated (lines) solubilities of propane in three different heavy oil samples at five different equilibrium pressures. In general, the propane solubilities calculated from the Winprop module with the P–R EOS well match the measured data at lower equilibrium pressures. It is noted that at P = 800 kPa, however, the measured solubilities for the maltenes and reconstituted oil (w_{asp} = 7.0 wt.%)

are much higher than their respective calculations. In this study, asphaltene content is determined by using n-pentane as a precipitant. Hence, when propane is made in contact with the heavy oil samples, it expectedly precipitates more asphaltenes than n-pentane does [13,27]. Once a heavy oil is saturated with propane
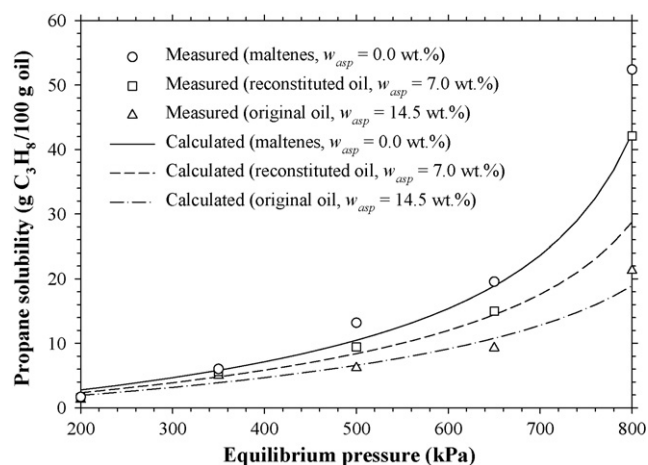

Fig. 2. Comparison between the measured and calculated solubilities of propane in three different heavy oil samples at P = 200, 350, 500, 650 and 800 kPa and T = 23.9 °C. The symbols represent the measured propane solubilities, whereas the lines represent the propane solubilities calculated by applying the Winprop module with the P–R EOS.
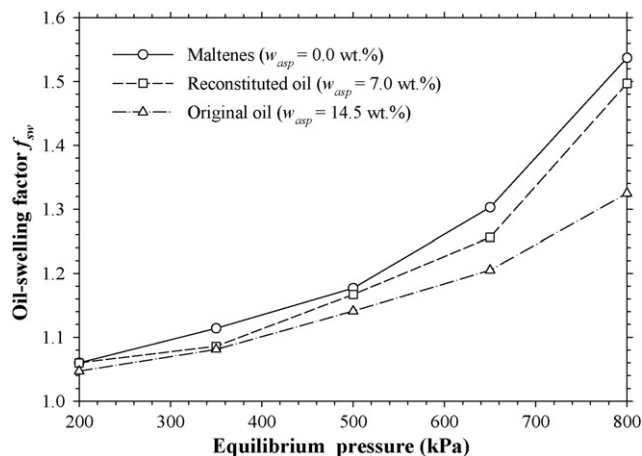
Fig. 3. Measured oil-swelling factors for three different heavy oil samples at $P = 200, 350, 500, 650$ and 800 kPa and $T = 23.9\,°C$.

Fig. 4. Measured viscosity versus the equilibrium pressure for three propane-saturated heavy oil samples at $T = 23.9\,°C$.

at a sufficiently high pressure, additional asphaltenes are precipitated so that the heavy oil is in-situ upgraded. In this study, it has been observed during the saturation test that for the three heavy oil–propane systems, some asphaltenes are precipitated to the bottom of the saturation cell at $P = 800$ kPa. Consequently, the measured solubility of propane in this upgraded heavy oil increases due to its reduced asphaltene content.

Also, it is seen from Fig. 2 that the measured propane solubilities in three different heavy oil samples increase with the equilibrium pressure, at a low equilibrium pressure, the propane solubilities are rather low and remain almost the same in three different heavy oil samples. For example, at $P = 200$ kPa, the propane solubility in the maltenes ($w_{asp} = 0.0$ wt.%) is 1.69 g $C_3H_8/100$ g oil, which is about 1.32 times of 1.28 g $C_3H_8/100$ g oil in the original heavy oil ($w_{asp} = 14.5$ wt.%). On the other hand, at a high equilibrium pressure, the propane solubilities in three heavy oil samples are much higher. It is also noted that the propane solubility in the maltenes is the highest at the same equilibrium pressure. The asphaltene content affects the propane solubility in heavy oil to a larger extent at a higher equilibrium pressure. When the equilibrium pressure is increased to $P = 800$ kPa, the propane solubility in the maltenes is increased to 52.39 g $C_3H_8/100$ g oil, which is about 2.46 times of 21.28 g $C_3H_8/100$ g oil in the original heavy oil. This may be due to in-situ upgrading of the heavy oil (i.e., asphaltene precipitation) at such a high pressure. The solvent has a higher solubility in such an upgraded heavy oil. In a field application, if propane is injected into a heavy oil reservoir at a sufficiently high pressure, asphaltene precipitation occurs so that the heavy oil is in-situ upgraded. Therefore, additional propane can be dissolved into the upgraded heavy oil under the practical reservoir conditions and the viscosity of the in-situ upgraded heavy oil can be further reduced.

4.2. Oil-swelling factor

The well-known oil-swelling effect is caused by the dissolution of solvent into the heavy oil and thus it increases as the solvent solubility in the heavy oil increases. Fig. 3 shows
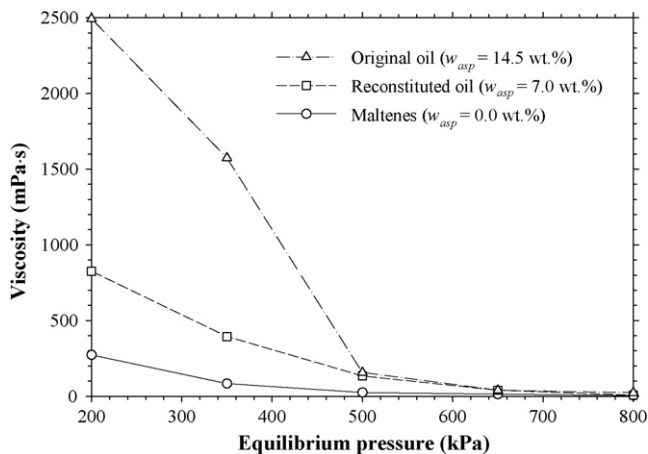
the measured oil-swelling factors for three different heavy oil samples at $P = 200, 350, 500, 650$ and 800 kPa and $T = 23.9\,°C$. It is found that the volume increase of the maltene–propane system is the largest at the same equilibrium pressure. This is because propane has the largest solubility in the maltenes. Also, it is noted that the oil-swelling factors increase quickly for three heavy oil samples when the equilibrium pressure increases from $P = 650$–800 kPa. For the three heavy oil–propane systems tested in this study, some asphaltene precipitates are found at the bottom of the saturation cell at $P = 800$ kPa. Therefore, the large increase of the oil-swelling factor from $P = 650$–800 kPa is attributed to the asphaltene precipitation in this pressure range. In this case, as described previously, additional propane is further dissolved into the in-situ upgraded heavy oil [18].

4.3. Propane-saturated heavy oil viscosity

Fig. 4 shows the measured viscosities of three different heavy oil–propane systems at $P = 200, 350, 500, 650$ and 800 kPa and $T = 23.9\,°C$. It can be seen from this figure that after propane is dissolved into a heavy oil sample, the viscosity of the heavy oil–propane system is significantly reduced, even at low equilibrium pressures. For example, at the equilibrium pressure of $P = 200$ kPa, the viscosity of the original heavy oil–propane system is 2493 mPa s, which is about one tenth of the viscosity of the original heavy oil with no solvent dissolution ($\mu_{oil} = 24{,}137$ mPa s). At an intermediate equilibrium pressure of $P = 500$ kPa, the original heavy oil–propane system has the viscosity of 156 mPa s and thus it is mobile enough to flow in a heavy oil reservoir. When the equilibrium pressure is increased to $P = 800$ kPa, the viscosity of the original heavy oil–propane is only 23 mPa s, which is in the same order of magnitude as that for a light or medium oil. Such propane-saturated heavy oil can be easily recovered from the heavy oil reservoir.

The previous PVT tests of this original heavy oil–propane system showed that asphaltene precipitation was not observed up to $P = 500$ kPa [19]. Hence, viscosity reduction of the propane saturated-heavy oil at the equilibrium pressures of $P \leq 500$ kPa is caused by solvent dissolution alone. In addi-

tion, at low equilibrium pressures, the viscosities of these three heavy oil–propane systems differ substantially due to their rather different asphaltene contents and thus propane solubilities. Higher asphaltene content in heavy oil causes a lower propane solubility and a higher oil viscosity. When the equilibrium pressure is high enough ($P \geq 650$ kPa), however, the three propane saturated-heavy oil systems have considerably low viscosities ($\mu < 40$ mPa s). In this case, both solvent dissolution and asphaltene precipitation contribute to significant heavy oil viscosity reduction.

Accurate determination of the solvent-saturated heavy oil viscosity is of practical significance during the solvent-based heavy oil recovery processes. In this study, the following one-parameter Lederer equation [28] is used to calculate the viscosities of the three heavy oil–propane systems tested:

$$\mu = \mu_o^{f_o} \mu_s^{f_s}, \tag{1}$$

$$f_s = \frac{c_s}{\lambda c_s + c_o}, \qquad f_s + f_o = 1, \qquad c_s + c_o = 1, \tag{2}$$

where μ_o and μ_s are the viscosities of the heavy oil sample with no solvent dissolution and the liquid solvent, respectively, f_s and f_o are the weighted volume fractions, and c_o and c_s are the volume fractions of the heavy oil sample and solvent in the heavy oil–solvent system, respectively. The weighting factor λ is to be determined experimentally [29]. In this study, propane is assumed to be in a liquid phase once it is dissolved into the heavy oil. The viscosity of the liquid propane is found to be $\mu = 0.1012$ mPa s at its vapor pressure $P_v = 923$ kPa and $T = 23.9\,^\circ$C by applying the Jossi–Stiel–Thodos correlation [30]. Also, the volume fraction of dissolved liquid propane c_s in the heavy oil–propane system can be related to the oil-swelling factor f_{sw} measured by using the DPDVA method [20,21]:

$$c_s = 1 - \frac{1}{f_{sw}}, \tag{3}$$

where the oil-swelling factor f_{sw} is defined as the ratio of the volume of the solvent-saturated heavy oil to that of the heavy oil with no solvent dissolution.

In this study, the weighting factor λ is determined by finding the best fit between the calculated and measured viscosity data of each heavy oil–propane system with the same asphaltene content at different equilibrium pressures and $T = 23.9\,^\circ$C. The numerical procedure for determining the weighting factor λ is described below. For each propane saturated-heavy oil tested at five different equilibrium pressures, an objective function is defined to quantify the overall discrepancy between the theoretically calculated viscosity μ_{ci} from the Lederer equation with a guessing value of the weighting factor λ and the experimentally measured viscosity μ_{mi} at different equilibrium pressures or propane volume fractions c_{si}, $i = 1, 2, \ldots, 5$:

$$E(\lambda) = \sqrt{\frac{1}{5} \sum_{i=1}^{5} \left| \frac{\mu_{ci} - \mu_{mi}}{\mu_{mi}} \right|^2} \times 100\%. \tag{4}$$

Mathematically, the objective function $E(\lambda)$ is equal to the root-mean-squared relative error between the theoretically calculated
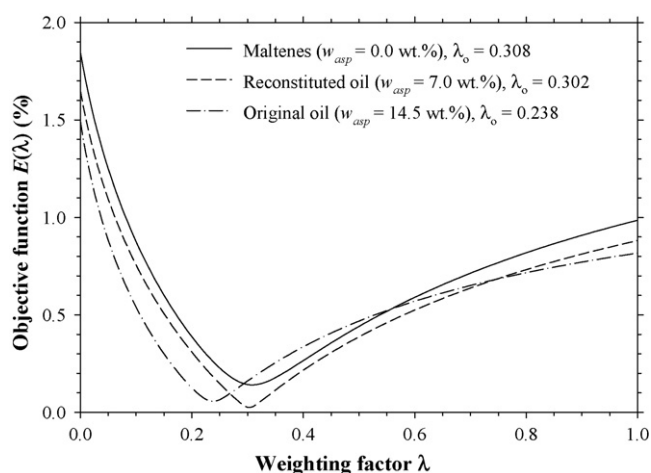

Fig. 5. Variations of the objective function $E(\lambda)$ with the weighting factor λ for three propane-saturated heavy oils with different asphaltene contents.

and experimentally measured viscosities for each propane saturated-heavy oil tested at five different equilibrium pressures. Hence, the objective function $E(\lambda)$ is minimized by using the weighting factor λ as an adjustable parameter to find the best fit of the theoretically calculated viscosities to the experimentally measured data. Once the minimum objective function is found, the corresponding λ_o is the determined weighting factor for the solvent saturated-heavy oil in the pressure range tested.

Fig. 5 shows the variations of the objective function $E(\lambda)$ with the weighting factor λ for three propane saturated-heavy oils with different asphaltene contents. The optimum weighting factors for the maltenes ($w_{asp} = 0.0$ wt.%), reconstituted oil ($w_{asp} = 7.0$ wt.%), and original oil ($w_{asp} = 14.5$ wt.%) are found to be $\lambda_o = 0.308$, 0.302, and 0.238, respectively. Eq. (2) shows that the smaller the weighting factor λ is, the stronger effect the solvent has on the viscosity of the heavy oil–solvent system. As the above-determined optimum weighting factors λ_o for the three propane saturated-heavy oils with different asphaltene contents are far less than unity, i.e., $f_s > f_0$, the dissolved propane has much stronger influence than the heavy oil composition (mainly asphaltene content) on the viscosity of each heavy oil–propane system in the pressure range tested. By using the obtained λ_o, the viscosities of each heavy oil–propane system at different equilibrium pressures are calculated from Eq. (2). Fig. 6 shows the comparison between the calculated (lines) and measured (symbols) viscosities of three different heavy oil–propane systems at different propane volume fractions and $T = 23.9\,^\circ$C. It can be seen from this figure that the viscosities calculated from the Lederer equation agree well with the measured data.

4.4. Propane molecular diffusivity in heavy oil

Molecular diffusion of a solvent in heavy oil is the most important physical process involved in the solvent-based heavy oil recovery process. In this study, the propane molecular diffusivity in each heavy oil sample is measured by using the DPDVA method [20,21] and the measured data for the three
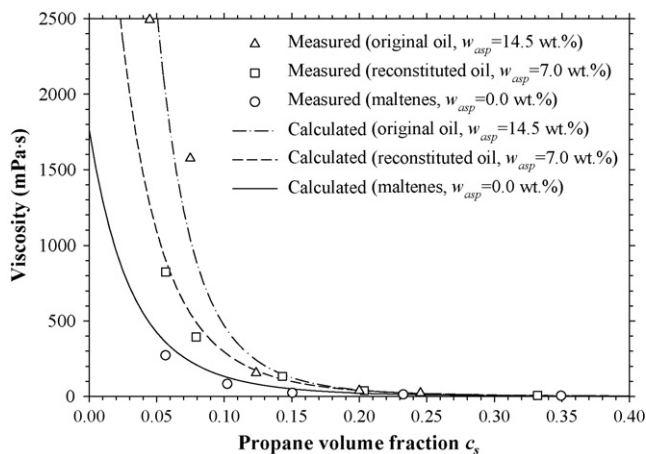
Fig. 6. Comparison between the calculated and measured viscosities of three different heavy oil–propane systems at different propane volume fractions and $T = 23.9\,^{\circ}C$. The symbols represent the measured viscosities, whereas each line represents the viscosities calculated from the Lederer equation with the determined optimum weighting factor λ_o obtained in Fig. 5.

heavy oil–propane systems are shown in Fig. 7. It can be seen from this figure that the propane molecular diffusivity increases with pressure but decreases with asphaltene content. In particular, the propane molecular diffusivity in the maltenes is the largest among the three heavy oil samples tested at the same equilibrium pressure.

The classical Stokes–Einstein's equation indicates that the molecular diffusivity of rigid spherical particles in an infinitely diluted liquid is inversely proportional to the liquid viscosity. It has been found that this equation is inaccurate in predicting the molecular diffusivity of a gas in a liquid [31]. In the literature, Hayduk and Cheng [32] proposed the following correlation between the molecular diffusivity of a gas in a liquid and the gas-saturated liquid viscosity:

$$D = \alpha\mu^{-\beta}, \qquad (5)$$

where α and β are two to-be-determined constants. In this correlation, the composition, temperature, and pressure effects on the
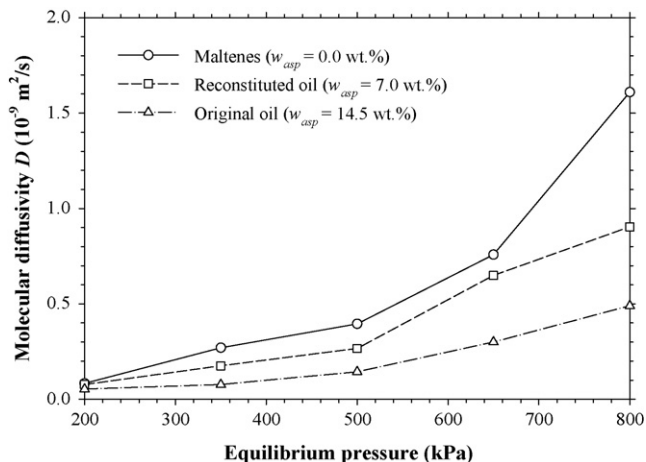

Fig. 7. Measured molecular diffusivities of propane in three different heavy oil samples at $P = 200, 350, 500, 650$ and $800\,kPa$ and $T = 23.9\,^{\circ}C$.
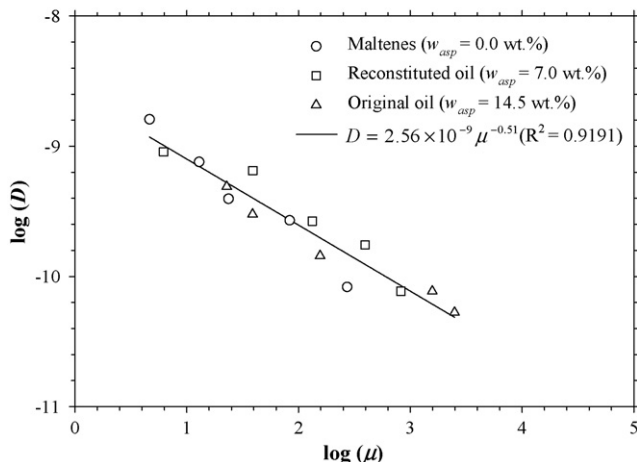

Fig. 8. Logarithmic relation between the measured propane molecular diffusivities in heavy oil D (m^2/s) and the measured viscosities μ $(mPa\,s)$ of three propane-saturated heavy oil samples at $P = 200, 350, 500, 650$ and $800\,kPa$ and $T = 23.9\,^{\circ}C$.

molecular diffusivity are taken into account implicitly through the gas-saturated liquid viscosity.

Fig. 8 shows the logarithmic relation between the measured diffusivities and viscosities of three heavy oil–propane systems at five different equilibrium pressures. Based on the measured data (symbols) in this figure, a linear relation (the solid line) between $\log(D)$ and $\log(\mu)$ is obtained by applying the linear regression and the corresponding correlation coefficient is found to be $R^2 = 0.9191$. The molecular diffusivity of propane in heavy oil D (m^2/s) is correlated to the propane-saturated heavy oil viscosity μ $(mPa\,s)$ by

$$D = 2.56 \times 10^{-9}\mu^{-0.51} \quad (R^2 = 0.9191). \qquad (6)$$

It is worthwhile to emphasize that this correlation is independent of the asphaltene content in the heavy oil and valid in the pressure range tested. This correlation indicates that when in-situ upgrading occurs during a solvent-based heavy oil recovery process, i.e., the viscosity of solvent-saturated heavy oil is reduced significantly, the molecular diffusivity of solvent in an in-situ upgraded heavy oil increases substantially.

5. Conclusions

In this paper, three heavy oil samples with different asphaltene contents, i.e., maltenes ($w_{asp} = 0.0\,wt.\%$), reconstituted oil ($w_{asp} = 7.0\,wt.\%$), and original oil ($w_{asp} = 14.5\,wt.\%$), are tested to model the heavy oils that are in-situ upgraded to different extents during a solvent-based heavy oil recovery process. These three heavy oil samples are saturated with propane at $P = 200, 350, 500, 650$ and $800\,kPa$ and $T = 23.9\,^{\circ}C$. The detailed experimental results show that the propane solubility is the largest in the maltenes, which results in the largest oil-swelling factor. The viscosity of the heavy oil–propane system with less asphaltene content is lower, which leads to a larger molecular diffusivity of propane in the heavy oil. Therefore, in-situ upgrading of heavy oil enhances further solvent dissolution into the upgraded heavy oil. As the equilibrium pressure increases,

the solubility, oil-swelling factor, and molecular diffusivity of each heavy oil–propane system increase, whereas its viscosity is dramatically reduced. In the EOS modeling, the two-phase flash calculations indicate that asphaltenes have the largest binary interaction coefficient with propane and the strongest effect on propane solubility in the heavy oil, in compassion with the other three lighter pseudo components. In addition, two specific correlations are applied to analyze the quantitative relationships among the propane concentration, oil-swelling factor, viscosity, and molecular diffusivity. It is found that propane dissolution significantly reduces the heavy oil viscosity even at low pressures. It is also found that the viscosity of the heavy oil–solvent system at a low pressure is strongly dependent on the asphaltene content in the heavy oil. Finally, the molecular diffusivity of each heavy oil–propane system can be fairly correlated to its viscosity, irrespective of the heavy oil composition (mainly asphaltene content) and the equilibrium pressure (or propane concentration).

Acknowledgments

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