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Determination of Diffusion Coefficient During Gas Injection in Heavy Oil Hydrocarbons

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Highlights

- Different methods of measuring diffusion coefficient in heavy oil were investigated.
- The best method was chosen according to the least error percentage.
- The diffusion coefficient behavior as a function of pressure was investigated in heavy oils and compared with that of intermediate to light oils.

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Abstract

An essential transport characteristic that links a substance's molar (mass) flux to its concentration gradient is the molecular diffusion coefficient. For modeling and performance forecasting of solvent-aided recovery processes of heavy oils such as VAPEX and SAGD; a reliable and accurate estimation of the molecular diffusion coefficient is a crucial input. Despite the importance of this parameter, there is no approved way to measure it, especially in systems with heavy oil and gaseous solvents that have limited solubility. This can be as a result of the intricacy of experimental measures and the challenge of analyzing experimental data. There are two direct and indirect methods for measuring the diffusion coefficient, the direct method has not been addressed because it is expensive and time-consuming. Indirect methods include Constant-Volume Methods (Pressure Decay), Constant-Pressure, Refractive Index, Nuclear Magnetic Resonance (NMR), X-ray Computer-Assisted Tomography (CAT), Pendent drop and Microfluidics. The advantage and disadvantages of these experimental methods established for diffusivity measurements of the gaseous solvent in heavy oil systems are discussed in this article. According to the investigations carried out in this study, the Constant-Volume Methods (Pressure Decay) with the least error percentage (1.05%) was chosen as the best method for measuring the diffusion coefficient. The diffusion coefficient of light and heavy oil was compared, and light oil has a higher diffusion coefficient.

Keywords: Heavy oil, Diffusion coefficient, Diffusivity, Gas injection

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1. Introduction

The molecular diffusion coefficient is a transport parameter that connects a substance's molar (mass) flow to its concentration gradient [1]. Diffusional mass transfer occurs in mixing solvents (like CO2/CH4- Bitumen, CO2/n-hexadecane, CO2-Heavy oil) or dissolving size of different instruments affects the rate of molecular diffusion in mass transfer. However, it is still a natural effect that must be calculated for accurate prediction. In oil recovery applications, molecular diffusion happens when

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soluble/miscible substances are injected into the reservoir to enhance production rates and ultimate recoveries through dilution [2] and/or wettability alteration [3].

The main categories of thermal EOR methods are in situ combustion (ISC), combustion assisted gravity drainage (CAGD) and steam injection in various forms include: Cyclic steam stimulation (CSS), steam flooding, and steam-assisted gravity drainage (SAGD). In situ combustion involves injecting air into viscous oil reservoirs to burn some of the existing oil, which produces heat. The combustion front largely determines whether the ISC is a forward or reverse process. The novel in-situ combustion method known as Combustion Assisted Gravity Drainage (CAGD) employs a horizontal injector at the reservoir's top and a horizontal producer parallel to and close to the reservoir's bottom. With the help of an electric heater, combustion is started after air is injected. By gravity drainage, the heated oil starts to descend to the beneath horizontal producer. Then, as it develops and spreads laterally, the combustion front moves toward the heel-end of the injector. By producing oil directly and injecting air only a short distance away, thermal upgrading is preserved, and oxygen is consumed effectively [4]. Cyclic stimulation of steam (CSS) is carried out in three stages (steam injection, soaking period to maximize heat transfer to the oil and oil viscosity reduction, and finally production period). Initially, a high oil flow rate is obtained. However, it gradually decreases. In steam flooding, steam is injected into dedicated injection wells, the introduction of steam into the reservoir efficiently displaces oil by heating the oil to a temperature at which its viscosity is sufficiently decreased [5]. In steam-assisted gravity drainage (SAGD), steam is continuously injected using two horizontal wells: the lower producer well (which collects the heated water and oil) and the upper steam injector well. The heated oil and condensed water being drained by gravity has an impact on SAGD.

Co-injection of hydrocarbon solvents with steam [6] has been proposed as a method of enhancing bitumen production rate [7]. Regardless of the injected concentration of the heated solvent, condensation and release of heat of vaporization at its dew point due to temperature difference between the injected vapour and bitumen on vapour-bitumen interface, results in heating and slight viscosity reduction in the bitumen, and forming a drainage bank of condensed solvent on interface, which can significantly improve the dispersive and diffuse mixing of solvent with bitumen that enhances the production rate of the process compared to lower operating temperatures [1].

Because the molecular diffusion of injected solvents into the bitumen has a great potential to contribute to overall mixing and mobilization of viscous bitumen, it must be included in the design and performance prediction of new solvent-aided bitumen recovery systems. This diffusive mass transport phenomenon is theoretically modeled using the diffusivity or diffusion coefficient. The rate of diffusional mass transfer is related to the concentration gradient across the substances. Despite the relevance of gaseous solvent diffusivity in engineering applications, measuring and precisely estimating it in bitumen has proven difficult.

It's worth noting that there isn't a widely accepted and universally applicable method for determining the molecular diffusion coefficient. Unlike measurements of viscosity or thermal conductivity, which can be done with standardized techniques and equipment, mass transfer characteristics are often more challenging to measure due to difficulties in measuring point values of concentration and other issues that complicate the transport process. Experiments were conducted to measure the diffusion coefficient for solvent diffusion in the oil. Direct and indirect approaches can be used to categorize various experimental methods. There are also several empirical correlations based on experimental data that could be used to calculate the dispersion coefficient under certain conditions [8].

The diffusion coefficient may be calculated by direct measurement and compositional analysis of the concentration distribution across the species during the tests. Sigmund [9] employed a Loschmidt diffusion cell technique, which involved stacking two gas chambers on top of one other and separating them with an isolation valve. In his tests, the gas chambers were pre-filled with various gases in various

volumes and allowed to attain thermal equilibrium at the test temperature. After the isolation ball valve between the gas chambers was opened, the diffusion tests commenced. The isolation valve was intermittently closed throughout the experiment to collect gas samples from each gas chamber for gas chromatography analysis. Because of the retrograde condensation and normal vaporization that occurred during the sampling, the model acquired for analytical testing differed from the actual gas composition in the chamber. The diffusion process and gas sampling were continued until the ultimate concentration change (at equilibrium) reached 30 to 50 percent.

Direct approaches are well-known for being time-consuming, costly, and system-intrusive [9-11]. Instead, the bulk of the proposed methods tries to determine the diffusion coefficient indirectly by measuring other quantifiable system characteristics that change as a result of the diffusional mass transfer of solvent into the bitumen. The two most common indirect methods are: constant-volume methods, in which the rate of pressure drop in a confined diffusion cell is measured and analyzed; and constant-pressure methods, in which the diffusing gas is continuously supplied into the diffusion cell to maintain the pressure across the gas/liquid interface while the rate of gas supply and/or speed of interface movement (swelling) is recorded and analyzed. Other approaches based on NMR spectra change, CAT scanning signal, volume change data of a pendant drop, and microfluidic techniques have also been presented. The advantages and disadvantages of several experimental approaches established for diffusivity measurements of the gaseous solvent in bitumen systems are discussed in this article. Despite substantial study in diffusivity measurement, additional research is needed to produce more viable, efficient, and trustworthy methods.

2. Diffusion coefficient measurement methods

The diffusion coefficient was first analytically defined by contrasting heat transfer via conduction with mass transfer by random molecular mobility [12]. In mass transfer studies, the diffusion coefficient is used to describe dimensionless numbers such as Schmidt number (Sc), Prandtl number (Pr), Lewis number (Le) and Peclet number (Pe) to characterize various processes. In fluid dynamic processes, the Schmidt number is the ratio of momentum diffusivity (ν: kinematic viscosity) to mass diffusivity which used to characterize fluid flows in processes with simultaneous momentum and mass diffusion. The Prandtl number defined as the ratio of momentum diffusivity to thermal diffusivity. It therefore assesses the relation between momentum transport and thermal transport capacity of a fluid. In fluid dynamics and thermodynamics, the Lewis number is a dimensionless number defined as the ratio of thermal diffusivity to mass diffusivity. It is used to characterize fluid flows where there is simultaneous heat and mass transfer. The Lewis number puts the thickness of the thermal boundary layer in relation to the concentration boundary layer. In most conditions, mixing in a reservoir is due to a combination of advective transport and diffusive transport. The ratio of the rate of these two transport mechanisms, known as the Peclet number, can be used to characterize flow in a reservoir and in Interface's microfluidic systems. There are no approved tools and methods for calculating and measuring the diffusion coefficient. This problem is due to technical issues, such as counting the concentration at the interface of chemicals. Due to the importance of diffusion coefficient in petroleum engineering, several people conducted laboratory and experimental studies to measure the diffusion coefficient of gases in fluids. However, most of these people made assumptions that made many errors in measuring the diffusion coefficient.

Direct and indirect approaches can be used to categorize various experimental methods:

- Direct Methods: The diffusion coefficient is calculated using direct methods by measuring the solvent concentration and the penetration depth. Such methods, encompassing a wide range of physicochemical processes such as mass spectrometry, radio-active tracer technology, spectrophotometry, and others, are often more reliable. The diffusivity is calculated using

compositional analysis techniques [13]. Direct methods are very expensive and require a long time to perform the test, so these methods are not popular.

- Indirect Methods: Indirect approaches track changes of the system's diffusion rate dependent properties. Indirect methods include Pressure Decay, Constant-Pressure, Refractive Index, NMR, CAT, Pendent drop and Microfluidics. These approaches have the advantage of not requiring the change in composition to be determined.

In the following, each of the indirect methods is examined separately. In each method, previous studies have been reviewed and experimental and calculated data have been compared. Finally, each method has been compared according to the error percentage, and the method with the least error has been selected as the best method.

3. Indirect methods

3.1. Constant-volume methods (pressure decay)

The pressure decay approach has gotten increased attention among indirect methods because of its simplicity in terms of experimental observations. Gas (as a solvent) and oil are injected into a cell (Figure 1). The content of the cells is initially out of balance. The gas dissolves into the oil as the experiment goes, lowering the pressure inside the cell. The amount of gas transported into the oil may be calculated by recording the pressure and level of the liquid in the cell. The diffusion coefficient is derived using this information. The pressure decay method fails when dealing with complex hydrocarbon mixtures with multiphase behavior. Luo and Gu [14], on the other hand, had disproved this strategy. They demonstrated that slight modifications in boundary condition assumptions resulted in orders of magnitude discrepancies in reported values.

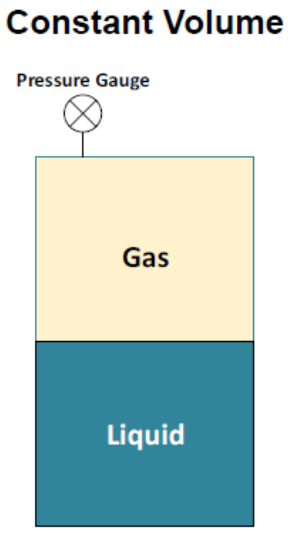

Figure 1

Experiment design of a constant-volume setup [54]

The advantages and disadvantages of this method:

- Advantage: This test is easy to do.

- Disadvantage: The properties of the gas (such as density and viscosity) change during the test. The correctness of the state equation determines the outcome. Selecting adequate boundary conditions for the gas/liquid interface is required.

Riazi [15] proposed this method, which gained popularity due to its ease of use in experimental measurement. In this method, known as a pressure-decay, the rate of pressure decreases in the cell (due to molecular diffusion) and the velocity of the gas/liquid interface was recorded using a constant-volume constant-temperature visual cell. To compute the equilibrium concentration at the interface and fugacity for each time step, a semi-analytical technique combined with the Peng-Robinson equation of state (EoS) was used. The slightly change of fluid characteristics and binary interactions coefficients was selected to match the final equilibrium concentration of species in the system and to reduce the error encountered with EoS. Moreover, Riazi suggested a correlation to predict interface velocity, which could then be compared to the rate of interface movement observed experimentally to confirm the diffusion coefficient value. Because the pressure changes throughout a pressure-decay test, the diffusion coefficient of gas and liquid phases at different pressures and concentrations might measure by performing single diffusion test.

Extension of Rasmussen and Civan [16] work and introducing an instantaneous solvent concentration in the interface rather than the concentration at equivalent pressure, Etminan et al. [17, 18] proposed an analytical solution to the diffusion problem. They investigated a broader time-dependent Robin type boundary condition that takes into consideration possible interface resistance, which is anticipated to block immediate equilibrium. Etminan et al. employed Henry's rule to explain the instantaneous concentration (which should be established quickly if the contact is free of resistance), making the solution more applicable to dilute liquids. The pressure could be predicted and history matched using the proposed analytical solution in the Laplace domain. However, because there was no analytical closed-form Laplace inverse solution, the inverse transform was numerically computed using the Stehfest approach.

Yang et al. [19] used Cauchy's residue theorem to find an exact solution to the quasi-equilibrium model, assuming that mass transfer resistance at the contact is negligible. To include a restricted number of infinite series terms in the exact solution, an approximate method was devised. The finite-acting problem was similar to Sheikha et al.'s [20] work in the Laplace domain, or Pacheco-Roman and Hejazi's approximation solution for estimating solubility and diffusion coefficient at the same time. The approximation solution, according to their findings, deviates from the observed pressure profile early on. This variation was more severe in systems with a significant pressure drop. However, when compared to results obtained using the integral heat method, their approximation solution performed better. By assuming a somewhat large gas cap, Yang et al.'s technique might provide an excellent match with the early-time pressure profile (such that the pressure loss in the gas cap is minimal). Minimizing the pressure degradation in the gas cap, on the other hand, would diminish the overall accuracy of the diffusion coefficient estimation.

Sheikha et al [20] in their work, presented a mathematical model to calculate the diffusion coefficient. The values of additional variables, including the diffusion coefficient, are needed to get the forward solution to Equation 1 for the gas phase pressure as a function of time. An inverse solution method is used to obtain the diffusion coefficient from Equation 1.

$$P(t) = P_i \exp\left(\frac{\sqrt{D}ZRT\sqrt{t}}{LMK_h}\right)^2 erfc\left(\frac{\sqrt{D}ZRT\sqrt{t}}{LMK_h}\right) \quad (1)$$

Where P is pressure (Pa), P_i is initial pressure, D is diffusivity coefficient (m2/s), Z is gas compressibility factor, R is universal gas constant; (8314 J kmol-1 K-1), T is temperature (°C or K), t is time (s), L is height of gas zone in the pressure cell, M is molar mass of gas (kg/kmol) and K_h is Henry's constant (Pa m3/kg).

3.2. Constant-pressure methods

For decades, until discovering the pressure-decay method, constant-pressure diffusion measuring techniques were the primary indirect means of getting diffusion coefficient. The simplicity with which these techniques may be quantitatively evaluated is their principal benefit over pressure-decay procedures. The assumption of equilibrium at the interface is valid when the pressure in the gas cap is maintained, and so the diffusion difficulties are greatly simplified. The measurement error and degree of freedom are reduced due to this simplification. To put it another way, the inaccuracies and ambiguities that come with characterizing the change in pressure-dependent attributes over time are eliminated. Due to the simplicity of mathematical modeling and the correctness of the assumptions, this approach has been used to study substantial relationships between diffusion coefficient and fluid characteristics for decades. The difficulty in accounting for liquid swelling, as well as the technical issues connected with maintaining pressure, might be mentioned as limitations of these systems. Figure 2 shows a schematic of this method.

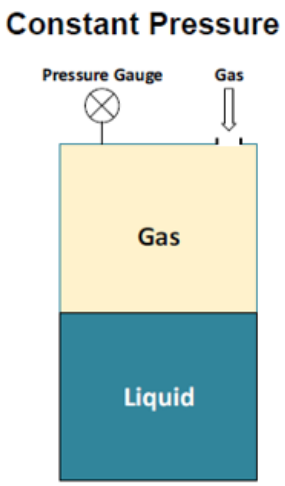

Figure 2

Experiment design of a constant-pressure setup [54]

The advantages and disadvantages of this method:

- Advantage: Changes in pressure-dependent characteristics are no longer a problem. Does not necessitate the use of a state equation
- Disadvantage: Maintaining pressure in a high-temperature experiment while monitoring gas injection rate is difficult. For the diffusion of low solubility gases in liquids, the interface velocity can be minimal.

Pomeroy et al. [21] were among the first to propose the measurement of diffusion coefficient by constant pressure testing. The gas and the quiescent liquid were brought into contact at the requisite pressure in their configuration. Following that, a continuous supply of the diffusing gas is employed to compensate for the pressure reduction in the gas cap induced by molecular diffusion of gas into the bulk liquid. The method of Pomeroy et al. used simplified initial and boundary conditions, resulting in an analytical solution. When differentiated concerning the length of the liquid column gave an expression relating the cumulative volume of diffused gas to the diffusion coefficient. They incorporated a constant variable

that reflected the change in fluid volume per unit amount of gas injected during the experiment to account for fluid swelling (due to gas dissolution and density reduction). The change-of-volume device had to be used to independently measure this value. Pomeroy et al used the slope of the cumulative volume diagram of the amount of injected gas versus the square of time to calculate the diffusion coefficient.

For the finite domain moving boundary diffusion problem, Do et al. [22] established an approximate analytical solution. Swellings are substantial in carbon dioxide and light oil systems, making infinite-domain validity difficult. Furthermore, in the finite domain, the moving-boundary diffusion problem has no closed-form solution. As a result, Do et al. [22] proposed a solution based on assuming a variable power profile for the concentration distribution across the liquid column as an approximation (parabolic and polynomial). The assumption reduces the problem to a single parametric beginning value problem with parameters based on the diffusion coefficient, density, and solubility of the mixture. This study also took into account the constant diffusion coefficient. To validate the model, the diffusion coefficient (obtained using the finite domain method) was compared to that published by Grogan et al. [23].

Sabet et al. [24] developed a method for determining the diffusion coefficient using the interface velocity during an infinite-acting diffusion experiment using the similarity solution methodology. To bring gas and liquid into contact, the researchers employed a high-pressure visual diffusion chamber. A continual supply of diffusing gas maintained the cell pressure constant throughout their experiment. A high-resolution digital camera was used to capture the motions of the gas/liquid interface, which was then used to graphically compute the diffusion coefficient. They utilized their model to account for swelling and discovered that in some circumstances, non-swell models might overestimate the diffusion coefficient by up to two times. Sabet et al. [24] have presented a graphical technique for calculating the diffusion coefficient from the gas uptake rate. The rate of gas absorption solution was later utilized by Khalifi et al. [25] to estimate the diffusivity of the gaseous DME in Athabasca bitumen at extreme temperatures and pressures. Sabet et al.'s [24] solution was expanded by Khalifi et al. [1, 26, 27] to include the influence of the initial solvent concentration on the diffusion coefficient. Meng et al. [28] proposed an alternative to Sabet et al.'s [24] evolved solution, in which they considered the impact of density change throughout the diffusion experiment. The diffusion coefficient D can be calculated with equation 2 based on Fick's second law of diffusion:

$$ln\left[\frac{dm(t)}{dt}\right] = -\frac{\pi^2 D}{4\hat{z}^2}t + ln(\frac{2A\rho_{eq}D}{\hat{z}}) \tag{2}$$

Where D is the ith gas component's diffusion coefficient (m2/s), t is the passage of time (s), $\hat{z}$ thickness of liquid layer/film (m), A area of the gas–liquid interface (m2) and ρ_{eq} is equilibrium mass concentration of gas in liquid, (kg/m3).

3.3. Refractive index method

The refractive index is the ratio of the velocity of wave propagation in a reference phase to the velocity of wave propagation in the phase of interest [8]. In diffusion experiments, the refractive index is defined as:

$$n = \frac{c}{v} \tag{3}$$

Where c represents the speed of light in vacuum, v represents the speed of light in the appropriate phase, and n represents the refractive index (see Figure 3).

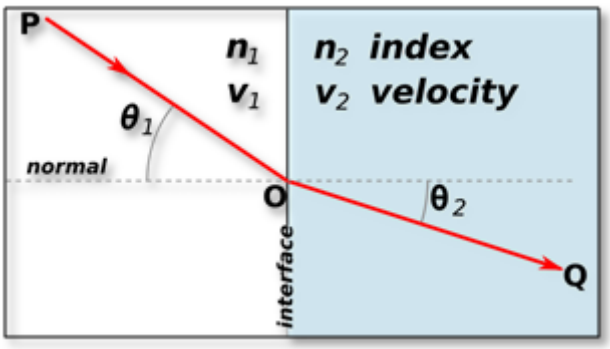

Figure 3

Light Refraction at the Intersection of Two Media [8]

When the velocity of light in vacuum is used as a reference, the refractive index is always greater than 1. Because water has a refractive index of 1.33, light travels 1.33 times quicker in vacuum than it does in water. When light moves from one media to another, such as air, water, or glass, its propagation direction may shift in proportion to the refractive index difference [8].

The refractive indices of a solution will change depending on the concentration of a sample component. As a result, the angle of refraction may be used to calculate the attention of a solution phase. A laser beam is usually emitted through the diffusion cell when measuring refractive indices experimentally. Based on the concentration of the solution at each height, the related refractive angle of the laser beam is calculated. As a result, the point at which the laser beam is caught by a CCD camera will represent the concentration inside the diffusion cell at that precise height. A CCD picture captured during the diffusion process is shown in Figure 4 [8].

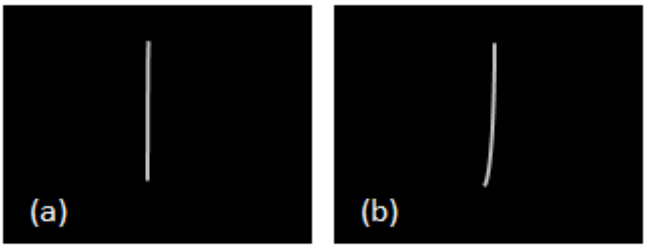

Figure 4

Sample of Light Refraction Results: a) Initial Time, b) After Diffusion Occurred [8]

It should be emphasized that the approach outlined above applies only to clear fluids. This method cannot be employed since heavy oil is opaque even when diluted.

The advantages and disadvantages of this method:

- Advantage: Simplicity in experimental application
- Disadvantage: Suitable for transparent fluids only and not for heavy oil.

Because the diffusion process is one-dimensional, Crank [29] presented Equation 4, in which the least squares method is used to solve Fick's second law.

$$\frac{C(x,t)}{C_0} = 1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1}.\exp\left[-\frac{D(2n+1)^2\pi^2 t}{4l^2}\right] \times \cos\frac{(2n+1)\pi x}{2l} \tag{4}$$

Here, l is the liquid length and $C(x,t)$ represents the concentration of the diffusing solute at distance x and time t in the gel.

3.4. Nuclear magnetic resonance (NMR)

Nuclear Magnetic Resonance (NMR) was created with chemical, physical, and therapeutic applications in mind. The density of hydrogen protons is calculated using this approach.

NMR happens when the nuclei of certain atoms are bathed in a static magnetic field and then exposed to an oscillating magnetic field. Specific nuclei experience this effect depending on their atomic makeup, whereas others do not. NMR measurements may be done on any nucleus with an odd number of protons, neutrons, or both, such as hydrogen (H), carbon (C), and sodium (Na) due to the nature of magnetic resonance (Na). Most nuclei present in earth formations are too faint to be identified using an NMR equipment such as a borehole NMR logging tool because the nuclear magnetic signal induced by external magnetic fields is too weak. However, the main atom in water and other hydrocarbon compounds emits a strong signal. As a result, the signal's strength might be utilized to gauge the amount of hydrogen on hand [8].

For various solvent and bitumen combinations [1, 30], the NMR approach has been effectively used to measure reservoir fluid characteristics [31], water-cut [1], solvent concentration, and viscosity reduction. Solvent dissolving in heavy oil produces a drop in viscosity and, subsequently, a shift in the mobility of the hydrogen molecules in the mixture during a molecular diffusion experiment.

In an MRI study of the diffusivity of gases in bitumen, Fayazi and Kantzas [32] established a numerical solution that accounts for liquid swelling. An indirect solvent concentration profile was created using the connection between the magnetic resonance signal strength and the amount of existing hydrogen protons to analyze the diffusivity of different gaseous solvents in bitumen. Their numerical solution used a moving mesh finite difference configuration similar to Etminan et al. [33]. The liquid column was divided into a particular and fixed number (100) of layers with constant masses, and the height increased as the liquid density decreased. They obtained density from a linear relationship between density and solvent. The height of all layers is added together to determine the interface location at each period. They may solve the diffusivity value by fitting the infinite dilution diffusivities of Vignes [34] model over the empirically "measured" concentration data using projected liquid heights and the interface equilibrium mass faction.

According to Fayazi [32], the equilibrium pressure and the total mass of the solvent were used to determine the solvent concentration or the mass fraction of the solvent dissolved in the oil at each phase (Equation 5,6).

$$M_s^{dis}[n] = M_s^{total}[n] - \frac{P[n] \times A \times (H_c - H_o[n]) \times MW_s}{Z[n] \times R \times T} \tag{5}$$

$$w_s[n] = \frac{M_s^{dis}[n]}{M_s^{dis}[n] + M_o} \tag{6}$$

Where w_s is the solvent concentration (mass fraction), M_o is the mass of oil (g), n is the step number, $H_o$is the height of swollen oil (cm), $H_c$is the height of the cell (cm), and $M_s^{dis}$is the mass of dissolved solvent (g).

The advantages and disadvantages of this method:

- Advantage: It's possible to use it on a tiny volume of fluids. It is not necessary to make a concentration profile assumption.
- Disadvantage: Converting the NMR spectrum to concentration creates a significant error. Experiments can be pretty costly to carry out.

Figure 5 depicts a sample diffusion coefficient estimated using the NMR experiment [8].

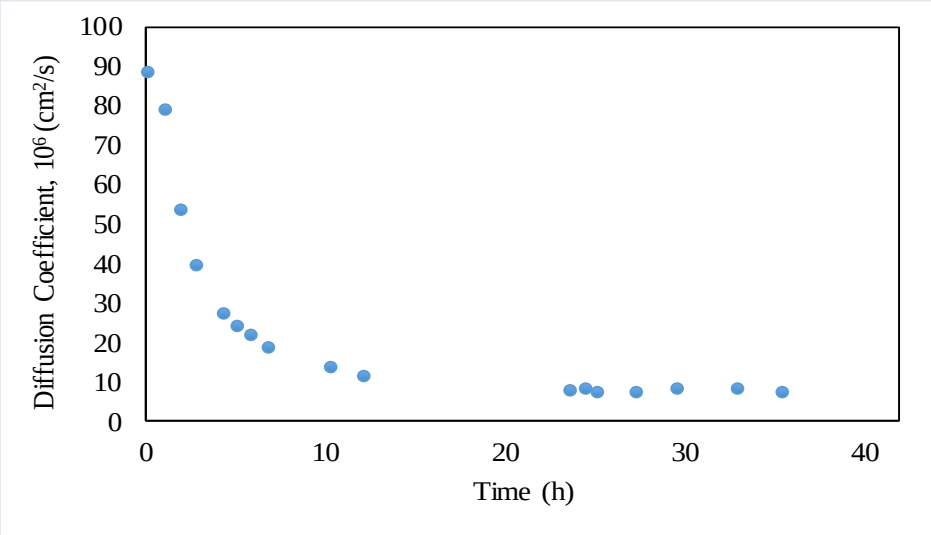

Figure 5

NMR Experimental Results: Diffusion Coefficient as a Function of Time [8]

3.5. X-ray computer-assisted tomography (CAT)

Computer-assisted Tomography (CAT) scanning using X-ray has been widely employed at research centers across the world for reservoir rock characterization and fluid flow visualization [35, 36].

The amount of energy lost by X-rays when they travel through a medium depends on the density of the substance and the length of the passage through it. CAT generates x-rays from a source that surrounds the item of interest. A detector on the opposite side of the source, on the other hand, captures one-dimensional projections of attenuated x-rays. As the sample moves longitudinally through the scanner, these projections are gathered and used to rebuild a two-dimensional image of the object [37-40].

Exposing a medium to x-rays, gathering the departing x-rays from the medium (Figure 6), and averaging the power at each cross-section can be used to create a transmitted intensity vs. elevation curve. According to the x-ray intensity and density connection, the resulting curve might be translated into a density curve [8].

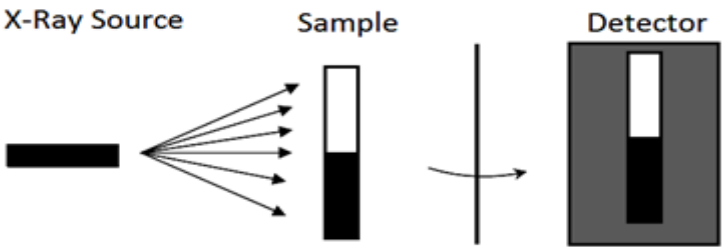

Figure 6

CAT Scanning using X-Rays as a Schematic [8]

The advantages and disadvantages of this method:

- Advantage: This test can also be done with a small amount of fluid.
- Disadvantage: The precision of the planned connection between density and concentration is relied upon. Experiments can be pretty costly to carry out.

Wen and Kantzas [37] used this approach to monitor concentration profiles at a bitumen-solvent contact. The solvent dissolves in the oil, diluting it and changing the coefficient of linear attenuation. This is the basic idea behind using CAT scanning to study the diffusion process. In a diffusion experiment, heavy oil is placed adjacent to the solvent in a fixed volume cell as the first step (Figure 7). Due to its greater oil gravity, the solvent sits on top of the heavy oil in the cell. A set vertical-sectional location of the diffusion cell is scanned at a given frequency during the diffusion process. Figure 10 shows a typical CT scan picture of solvent spreading into heavy oil.

The diffusivity of dimethyl ether and propane in different viscosity Canadian heavy oil samples were then investigated using the CAT scanning results of Diedro et al. [41]. Their study, however, was predicated on the false premise that density and concentration are linearly related, which is especially true for systems combining gases and bitumen. According to their observations, diffusional mass transfer is quicker in low viscosity liquids.

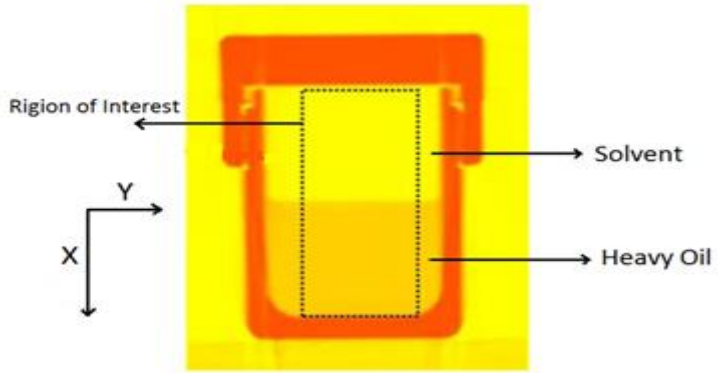

Figure 7

Diffusion Process Illustration [8]

3.6. Pendant drop

Several authors have devised methodologies that use the volume-change data of a spherical gas bubble surrounded by liquid [42] or a liquid drop hanging in a gas-filled environment [43], similar to one-dimensional swelling-based measurements [24, 26, 32, 44] at constant pressure settings. The schematic of the experimental setup is shown in Figure 8.

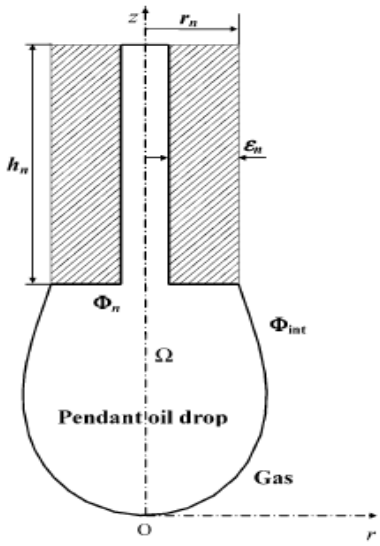

Figure 8

Schematic diagram of a pendant oil drop surrounded by a gas [45]

The advantages and disadvantages of this method:

- Advantage: The time of the trial can be drastically decreased. Can be done with a standard high-pressure IFT measuring device.
- Disadvantage: It necessitates arduous numerical analysis. For low solubility gases in bitumen, the change in liquid volume owing to molecular diffusion can be negligible.

Yang and Gu [45] suggested a finite-element numerical solution for gaseous solvent-bitumen systems that detect changes in the pendant drop volume produced by gaseous solvent diffusion in bitumen. The difference in drop volume is tracked throughout the experiment using a sophisticated computer-aided image analysis technique. The volume results from the experiment are then compared to the volumes predicted by the time-consuming numerical computation of the concentration distribution in the drop. Diffusion coefficient and swelling factor were the modifiable factors in their depreciation, with the minimization function being the disparities between the actual and anticipated volumes. The diffusivity of Light gases in bitumen was then determined using their previously developed technique [46].

The pendant drop technique includes calculating the profile of a liquid droplet suspended in another phase at a mechanically stable state, which is controlled by the interaction of surface forces and gravity. The Kravanja [47] equation, which is based on Laplace's equation, establishes a nonlinear differential equation that connects the droplet profile to the interfacial tension.

$$D_{Ai} = \frac{(1 - x_i)}{8(y_i - 1)} \frac{D^2}{t} \tag{7}$$

Where subscript A denotes oil component, subscript i denotes the i gas, x_i is the molar fraction of i-th gas contained in oil droplets solute, y_i is the molar fraction of i-th gas contained in the dense gas phase at the phase interface, D is the equivalent diameter of droplet, and t is the time.

3.7. Microfluidics

Microfluidics has gained widespread acceptance and usage in various technological and health-related disciplines, with the significant benefits being a significant reduction in experimental time and the use

of far fewer materials. With the purpose of reducing experimental time, Fadaei et al. [48] were the first to use microfluidic experiments to measure the diffusion coefficient in a gas and bitumen system (to less than 10 minutes). Despite the changes in implementation technique, they took a similar strategy to Grogan et al. [23]. However, in Fadaei et al.'s [48] microfluidic approach, the bitumen plug was injected into a microfluidic channel pre-filled with carbon dioxide, rather than the carbon dioxide bubble being delimited by bitumen [23]. The expansion of the bitumen was documented during the diffusion experiment using time-lapse imagery taken every 5 seconds. To analyze their data, they used Jamialahmadi et al.'s [44] method with minor changes to account for the unique aspect of their experiment, in which bitumen swelling occurred in both directions of the bitumen plug. Talebi et al. [49] expanded their study by combining their microfluidic technology with fluorescence microscopy to calculate the diffusion coefficient and concentration in the mixing. In both infinite and finite domains, their diffusivity estimates computed from one-dimensional bitumen swelling data were in good agreement with published data. The schematic of the experimental setup is shown in Figure 9.

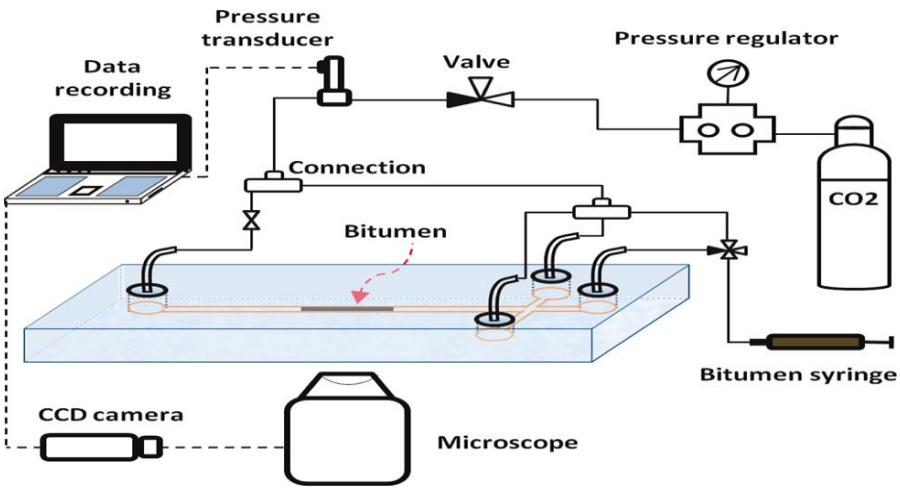

Figure 9

CO2 heavy oil diffusion coefficient measured using an experimental microfluidic system [48]

The advantages and disadvantages of this method:

- Advantage: The experiment's cost and length can be significantly decreased. There is a significant reduction in the amount of fluid required.
- Disadvantage: In a micro-sized channel, mass transfer caused by the capillary effect is unavoidable.

In summary, the basis of mathematical formulation and simplification assumptions of the mentioned methods are listed in Table 1.

Table 1

The basis of mathematical formulation and simplification assumptions of the presented methods

Method	Equation	Assumptions	Reference
Constant-Volume (Pressure Decay)	$P(t) = P_i \exp\left(\frac{\sqrt{D}ZRT\sqrt{t}}{LMK_h}\right)^2 erfc\left(\frac{\sqrt{D}ZRT\sqrt{t}}{LMK_h}\right)$	1- No chemical reaction between the bitumen and the gas 2- Isothermal conditions throughout the	[20]

		pressure-decay experiment 3- A constant value of diffusion coefficient 4- Thermodynamic equilibrium at the gas/bitumen interface	
Constant-Pressure	$\ln\left[\frac{dm(t)}{dt}\right] = -\frac{\pi^2 D}{4\hat{z}^2}t + \ln(\frac{2A\rho_{eq}D}{\hat{z}})$	1- System has a finite domain. 2- One-dimensional and one-way diffusion 3- The equilibrium boundary condition at the gas–liquid interface at constant-temperature 4- A constant effective diffusion coefficient for simplicity of the mathematical manipulation	[55]
Refractive Index	$\frac{C(x,t)}{C_0} = 1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{2n+1}.\exp\left[-\frac{D(2n+1)^2\pi^2 t}{4l^2}\right] \times \cos\frac{(2n+1)\pi x}{2l}$	1- System is finite. 2- Initial gas concentration is zero at the start of the diffusion process. 3- The gas concentration at the gas-liquid interface is taken to be the equilibrium concentration $c_{eq.}$ 4- $\frac{\partial c}{\partial z} = 0$ at the thickness of liquid layer/film (the impermeable boundry) at all times	[29]
Nuclear Magnetic Resonance (NMR)	$M_s^{dis}[n] = M_s^{total}[n] - \frac{P[n] \times A \times (H_c - H_o[n]) \times MW_s}{Z[n] \times R \times T}$	1- Isothermal diffusion process 2- No chemical reaction or asphaltene precipitation during the process 3- There is no resistance to mass transfer at the oil and gas phases interface 4- One-way mass transfer from the gas phase to the liquid phase 5- Diffusion in 1-D system with a constant cross section	[32]
Pendant drop	$D_{AB} = \frac{(1-x_B)}{8(y_B-1)}\frac{D^2}{t}$	1- Drop diameter decreases with time	[47]

		2- Mass transfer is carried out with equimolar countercurrent diffusion

4. Result and discussion

The key challenge is determining the molecular diffusion coefficient of gases in heavy oil/bitumen. Because the diffusivity of gases in heavy oil is very low, the testing time is long; Low mass transfer rate due to low solubility of gases in heavy oil; It is challenging to obtain data with high accuracy for a long time; and difficulties in dissociating the effect of various phenomena can all cause difficulties in conducting the experiments.

In pressure-decay method, by replacing all parameters in equation 1, the calculated values were obtained. The experimental and calculated pressure-decay profiles for CO2 gas in heavy oil at 75°C are plotted in Figure 10. Pressure-decay experimental data from Sheikha [20] study were used. Figure 10 shows that the results obtained from the calculated method provide reasonable agreement with the pressure-decay experimental data. In this method, the average error percentage was 1.05%.

In the Constant-Pressure method, the volume changes calculated from equation 2 and the experimental data of the gas phase at 21 oC are plotted in Figure 11. There is a good agreement between the experimental and calculated volumes. Experimental data are used from Li et al. [50] study. The average error of this method was estimated to be 1.51%.

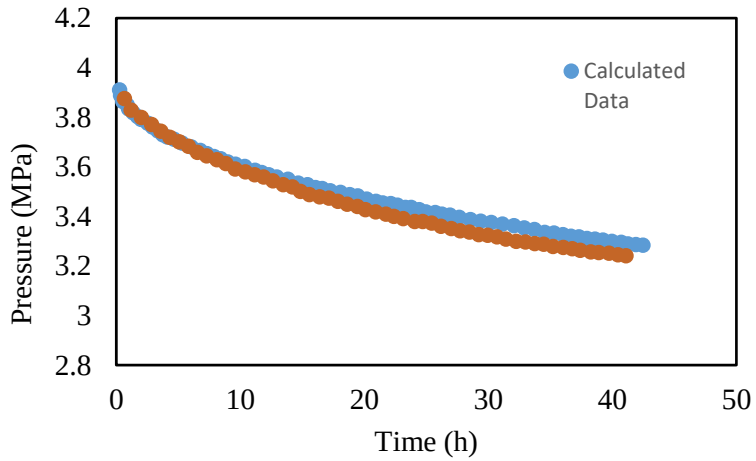

Figure 10

Constant-Volume data using experimental and calculated data of CO2 in heavy oil

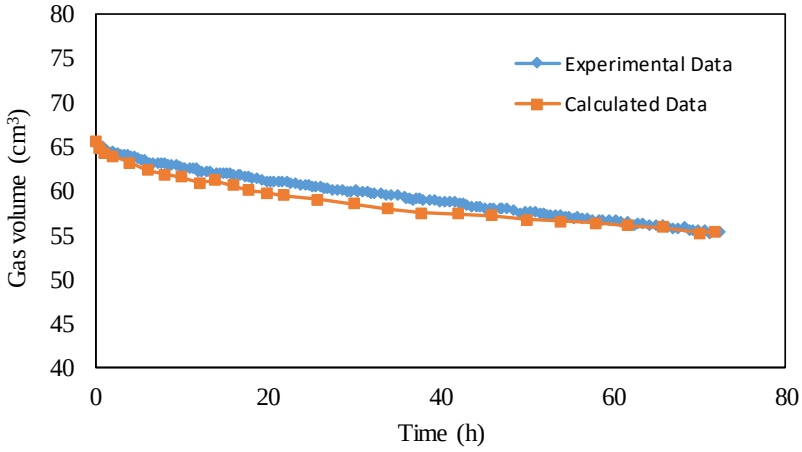

Figure 11

Constant-Pressure data using experimental and calculated data of CO2 in heavy oil

Refractive Index method as a function of diffusion distance, experimental data of solution concentration and calculated from equation 4 was presented in Figure 12. As the distance increases, the concentration of the solution in the diffusion distance decreases. Although the calculated results are higher than the experimental results of Liang [51] study, they are reasonable due to the adoption of different diffusion conditions including low concentration and high diffusion temperature. This method has caused up to 15% error.

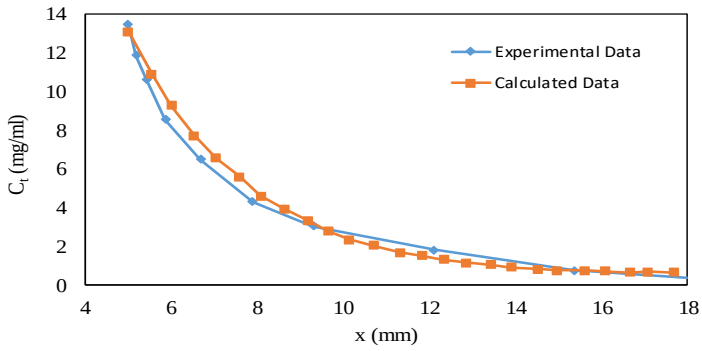

Figure 12

Distribution of solute concentration against length after 32 hours of conducting the experiment

For NMR method, equations 5 and 6 are used to calculate CO2 concentration. This method was applied to CO2 solvent and the concentration profile of the experimental sample [32] and the calculated data at a pressure of 615 psi after 48 hours are compared in Figure 13. The average error calculated for this method was estimated to be 12%.

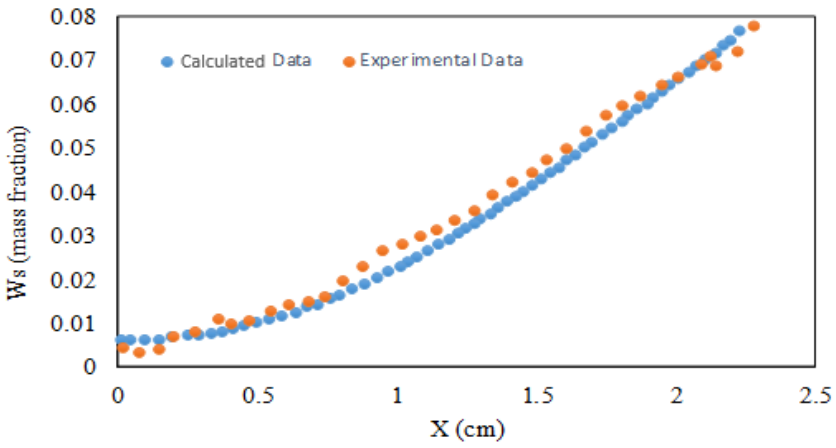

Figure 13

Difference between the experimental and calculated data of CO2 concentration

From the CAT method in Figure 14, the diffusion coefficient is plotted as a function of the diffusion distance at a pressure of 130 psi and after 72 hours. As clearly shown, the profile shows the same trend: close to the interface, there is a sharp decrease in the diffusion coefficient, and the diffusion coefficient gradually decreases to an almost constant value along the diffusion distance. The reason for this result could be that, close to the carbon dioxide/heavy oil interface, the concentration of carbon dioxide is higher. The average error calculated for this method is estimated to be 11%.

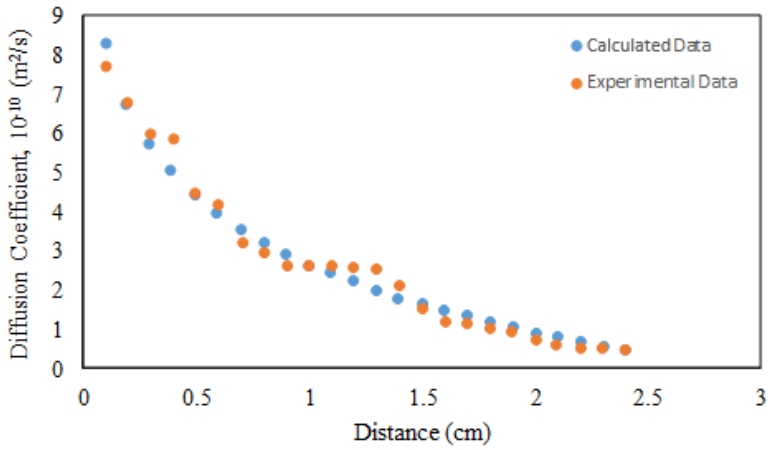

Figure 14

CO2 diffusion coefficient in bitumen at a pressure of 130 psi

In the pendent drop method, the correspondence of the experimental diffusion coefficients from Kravanja [47] studies of water in CO2 with the coefficients calculated by equation 7 under similar conditions at 45 °C is presented in Figure 15. The average error for this method was estimated to be 14%.

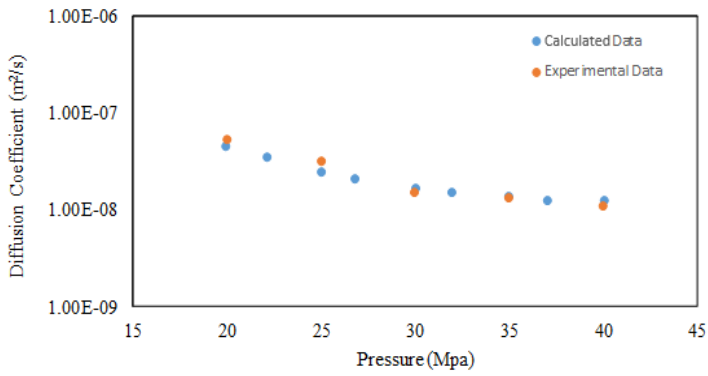

Figure 15

Comparison of experimentally obtained and calculated diffusion coefficients of water in supercritical CO2

According to the available experimental and calculated data from each method, the Constant-Volume Methods (Pressure Decay) with the least error was selected as an easy and effective method. Table 2 shows the average error percentage of each method.

Table 2

The average error percentage of determine diffusion coefficient methods

Methods	%Error
Constant-Volume Methods (Pressure Decay)	1.05
Constant-Pressure Methods	1.51
Refractive Index Method	15
Nuclear Magnetic Resonance (NMR)	12
X-ray Computer-Assisted Tomography (CAT)	11
Pendent drop	14
Microfluidics	-

There are influential factors to measure the diffusion coefficient that must be considered. The diffusivity coefficient is influenced by several factors, the most important of which are:

- Type of gas and porous medium interface: Because of the constraints imposed by the solid matrix, the rate of diffusion into a porous media is much slower than in an empty area.
- Reservoir pressure: Diffusion coefficient is generally directly related to pressure. Because the density of a gas is precisely proportional to pressure at constant temperature, the product of diffusivity and gas density is often found to remain constant throughout a wide pressure range.
- Reservoir temperature: Diffusivity rises with increasing temperature.

Using the Pressure Decay method, the diffusion coefficient of carbon dioxide (CO2) in heavy oils with different API (under certain temperature and pressure conditions) has been investigated in the laboratory. CO2 is one of the most common gases used for miscible injection in the reservoir. For this reason, many authors have studied the CO2 diffusion coefficient in heavy oil. Briefly, the CO2 diffusion coefficient measured by different authors is shown in Table 3. Table 3 shows that the CO2 diffusion coefficient varies in different heavy oils (different oil components, temperatures, pressures) [52].

Table 3

CO2 diffusion coefficient in heavy oils

T (°F)	P (psi)	API	D ($\times 10^{-8}\ \frac{m^2}{s}$)	Reference
70	450-812	6.4	0.012-0.024	[48]
77-194	580	9.1	0.016-0.047	[56]
81	198-685	11.1	0.022-0.036	[57]
77	250-651	14.1	0.0453-0.0595	[58]
59-86	116-290	15.4	0.0171-0.0641	[59]

Figure 16 shows the diffusion coefficient values of Table 3 against API and its curve fitting equation. It should be noted that the diffusion coefficient of heavy oils has been obtained at different pressures. This is why the diffusion coefficient of 15.4 API has decreased instead of increasing. As shown in the figure, the CO2 diffusion coefficient is higher when the API is higher. This is because at a higher API, there are more oil-light components. The higher viscosity of the heavy oil, the lower CO2 diffusion coefficient in the heavy oil.

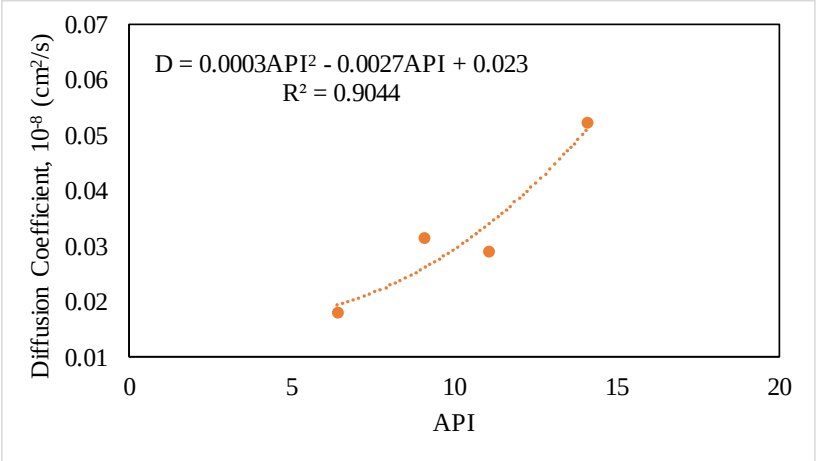

Figure 16

Variation of CO2 diffusion coefficient as a function of 'API gravity

According to studies on diffusion coefficients in light and heavy oils, results have been obtained. Under similar conditions, the diffusion coefficient in light oils is larger than in heavy oils. The reason for this phenomenon is that it is more accessible to diffusion of gas in light oils. Table 4 shows the diffusion coefficient of CO2 in light and heavy oil under the same conditions [53].

Table 4

CO2 diffusion coefficient in light and heavy oil

T (°F)	P (psi)	API	D ($\times 10^{-8}\ \frac{m^2}{s}$)
122	408	11.5	0.061
122	408	29.3	3.9-4
122	708	11.5	0.07-0.073
122	708	29.3	3.5-5.5
122	1134	11.5	0.075-0.078
122	1134	29.3	6.7-6.9
122	1459	11.5	0.15-0.16
122	1459	29.3	9.6-9.7

Figures 17 and 18 show the diffusion coefficient against pressure for 11.5 API and 29.3 API, respectively. According to these two Figures, the effect of pressure on the diffusion coefficient for a specific oil can be clearly seen. As shown in figures, the CO2 diffusion coefficient decreases with decreasing pressure at constant temperature. CO2 diffusion coefficient is less sensitive at low temperatures. As a general result, the CO2 diffusion coefficient in heavy oil decreases with decreasing pressure and temperature.

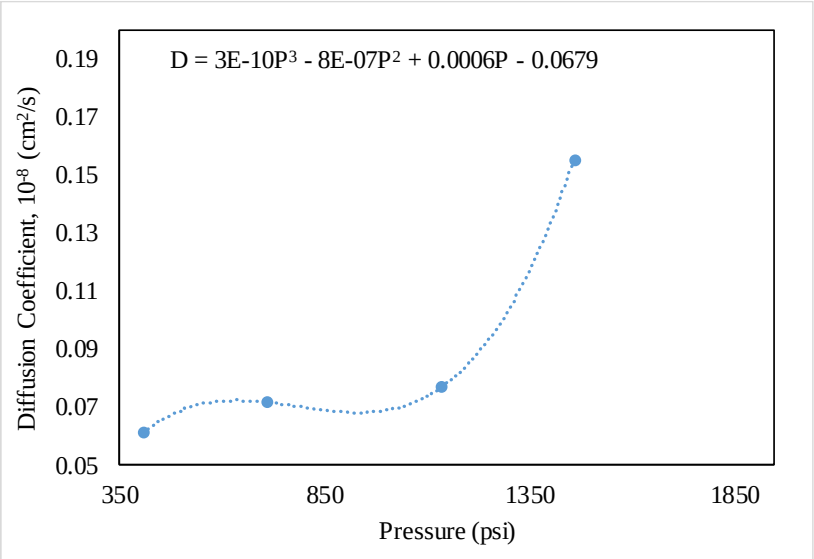

Figure 17

Diffusion coefficient of 11.5 API at different pressures

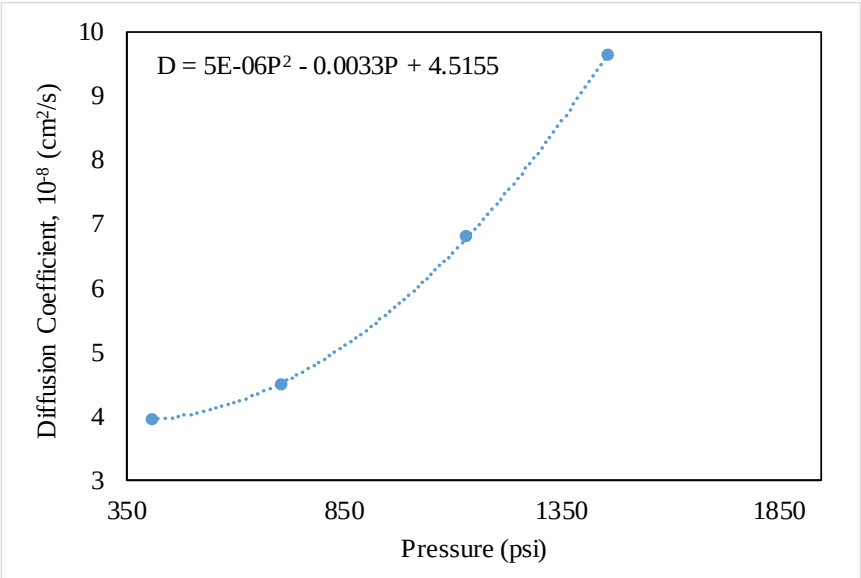

Figure 18

Diffusion coefficient of 29.3 API at different pressures

5. Conclusions

According to the investigation of different laboratory methods of diffusion coefficient in heavy oils and examination of the laboratory and calculated data of each method, the following results were obtained:

1. Direct diffusivity measurements that rely on continuous species concentration measurements are time-consuming and system-intrusive [9].
2. Since the pressure decay method is more accessible than other methods and has a lower percentage of error (1.05%), it has become a standard method for measuring the diffusion coefficient. However, additional errors due to inaccurate boundary conditions should be considered.
3. Although fixed pressure methods are more challenging to perform, they eliminate errors due to boundary conditions. Furthermore, for systems with low solubility gases in heavy oil, the movement of the gas/liquid interface in constant-pressure approaches can be minimal and thus unmeasurable.
4. While both NMR and X-ray Computer-Assisted Tomography (X-ray CAT) are rapid and straightforward to use, accuracy in translating observed viscosity (in NMR) and density (in X-ray CAT) to solvent concentration is crucial.
5. Pendent-drop measurements can be completed in a fraction of the time while consuming the least amount of the material. However, analyzing the recorded volume-change data is numerically difficult.
6. CO2 diffusion coefficient in heavy oil increases with increased pressure and temperature.
7. Type of gas and porous medium interface, reservoir pressure and reservoir temperature are influential factors for measuring the diffusion coefficient.

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