Full Length Article

A new analytical model for estimation of the molecular diffusion coefficient of gaseous solvents in bitumen – Effect of swelling

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A B S T R A C T

Diffusion of gaseous solvents in bitumen is one of the imperative factors in studying solvent-aided thermal recovery processes. Experiments have shown significant swelling of bitumen upon diffusion of some gaseous solvents. Among different techniques for measurement of diffusion coefficient, Pressure Decay methods are the most popular. In one approach, liquid is placed in a cell overlaid by a gas cap. Diffusion of gas into the liquid at constant cell pressure can be achieved by continuously supplying the makeup gas into the gas cap. Different analytical and numerical models to estimate the molecular diffusion based on the mass of the injected gas have been proposed in the literature. However, there are two drawbacks associated with the previous models: 1) most of these models have been developed based on a no-swelling assumption while experiments have shown the significant swelling of bitumen upon diffusion of some solvents; 2) the models that incorporate the effect of swelling are numerical or semi-analytical and they are computationally-expensive, not straightforward, and prone to numerical error. Therefore, there is need for a simple analytical model that not only includes the swelling effect, but also is easy to implement. This study presents a novel analytical model obtained based on similarity solution along with a simple graphical method to estimate diffusion coefficients of gases in liquids. The experimental data of a moving interface as a result of swelling is used as an input to the developed model to estimate the diffusion coefficient. The developed model is applied to the available data in the literature and the predicted diffusion coefficients are compared to the previously reported ones. Moreover, the presented model is used to interpret new experimental data of nC_4-nC_{10} system to estimate the molecular diffusion coefficient. The developed model and the experimental data find applications in design and optimization of solvent-aided thermal recovery of bitumen and heavy oils.

1. Introduction

The steady energy demand and lack of an adequate substitute for petroleum products, along with the recent concerns about greenhouse gas emissions, have driven the industry toward utilizing solvents in bitumen/heavy oil recovery processes. The high viscosity of bitumen makes it immobile at reservoir temperature imposing challenges in production and processing of bitumen. However, light gases or the so-called solvents can be dissolved in bitumen to significantly increase its mobility [1]. The concentration of the injected solvent in bitumen recovery processes can vary from low such as Solvent-Assisted SAGD to very high such as Heated VAPEX [2]. In general, thermal-solvent assisted gravity drainage processes aim at increasing oil production rate and reducing energy usage (i.e. steam-oil-ratio) compared to the conventional SAGD process [3] and thus reduce energy intensity and water consumption and treatment. Solvent addition in a thermodynamically optimized recovery process can also achieve minimum energy

requirement, which translates into less greenhouse gas emissions [4,5].

Molecular diffusion coefficient is used for calculating the rate of solvent dissolution in bitumen and it is a key parameter in design and optimization of oil recovery processes involved in solvent injection. Despite the importance of diffusion coefficient, there is no standard or universally acceptable technique used for its measurement. Molecular diffusion coefficient measurement experiments are often associated with convective mass transfer, phase equilibrium, and variation of mixture compositions [6]. Such challenges necessitate use of various simplifying assumptions for interpreting the experimental measurements. However, some of these assumptions may result in unreliable estimation [7]. The molecular diffusion of gaseous solvents in bitumen can be experimentally estimated using methods that can be broadly classified into direct and indirect techniques. In the direct methods, analytical tools like chromatography are used to determine the composition of the diffusing species in the bitumen column with time. However, such methods can be very tedious and relatively expensive.

Alternatively, indirect methods overcome many of these challenges by measuring one or more of the system parameters that alter due to the diffusion phenomenon. Such parameters include gas-liquid interface velocity or change of solution volume [8–10], rate of gas pressure drop in a confined PVT cell also referred to as pressure-decay [11–14], rate of gas supply (makeup) from the top of a PVT cell into a stagnant liquid mixture with constant volume [15–17] or a modified pressure decay method at which the interface pressure and consequently solvent concertation at interface are kept constant [6]. Other recent and more sophisticated techniques that can be used for indirect measurement of diffusivity are NMR spectra change and CAT-scanning method [18], and lately the method of dynamic pendent drop analysis [19]. All of the early works involved with the indirect measurements of diffusivity were done based on the movement of the interface [8–10] or gas injection rate into the liquid [15–17].

In 1996, Riazi [11] developed a mathematical model and introduced the concept of pressure-decay in diffusivity measurement. His semi-analytical method could obtain the concentration-dependent diffusivity from the position of gas/liquid interface and rate of gas pressure drop in a confined volume PVT cell for a system of methane (C_1) and n-pentane (nC_5). Zhang et al. [14] later simplified Riazi's approach by eliminating the effect of the moving interface and were able to determine the diffusion coefficient in heavy oil using a simple analytical model. Their method relied on calculating the diffusion coefficient using an equilibrium concentration which was determined at an estimated value of the saturation pressure. Upreti and Mehrotra [12,13] used a time-dependent boundary condition to propose a numerical approach that could solve for the concentration-dependent diffusion coefficient while counting for bitumen swelling. Civan and Rasmussen [20–23] developed an analytical solution for the experimental setup of Zhang et al. [14] by introducing a mass transfer coefficient as an extra unknown to the problem. Tharanivasan et al. [24,25] studied various interface assumptions used in the previous studies. They classified the boundary conditions as equilibrium, quasi-equilibrium, and non-equilibrium, and concluded that depending on the type of the dissolved gas, different boundary conditions should be used. Sheikha et al. [7,26] proposed an analytical approach for measurements reported by Upreti and Mehrotra [12,13] to calculate the diffusion coefficient. Their method assumed negligible swelling of the bitumen and used Henry's constant to estimate the solvent concentration at the gas-bitumen interface. Jamialahmadi et al. [10] proposed an approximate solution for the moving interface behavior for prediction of the diffusion coefficient. Ghaderi et al. [27] proposed a graphical approach based on Integral Method (IM) that estimated the concentration-dependent diffusion coefficient for a synthetic set of data. In 2010, Etminan et al. [6] introduced a modified pressure decay technique in which pressure of the gas cap in the diffusion cell is kept constant. They related the gas cap pressure to a Dirichlet type boundary condition at the gas/liquid interface and proposed a mathematical solution that could find both the saturation concentration and the diffusion coefficient. Etminan et al. later works [28–30] have focused on the presence of resistance to mass transfer at the interface and proposed numerical solutions for estimation of diffusion coefficient, mass transfer coefficient, and gas solubility. In 2014, Etminan et al. [31] proposed a numerical approach that takes the liquid swelling into account and uses a front-tracking moving boundary algorithm for estimating the diffusion coefficient. Roman and Hejazi [32] proposed a graphical method that used the late time data of the pressure decay experiment to determine the diffusion coefficient and Henry's constant. Recently, Talebi et al. [33] used a swelling-based microfluidic technique to determine the propane diffusion coefficient in bitumen based on the mathematical model developed by Jamialahmadi et al. [10]. The microfluidics-based measurement of diffusion coefficient has also been applied to CO_2-bitumen [34] and CO_2-oil [35] systems. Table 1 summarizes a list of studies related to diffusion coefficient measurement and estimation.

The objective of this work is to provide a reliable and straightforward analytical approach that can accurately predict the molecular diffusion coefficient using the variation in interface level measured through the modified pressure decay experiment. The proposed technique utilizes the transient time data along with the equilibrium mass fraction of the solvent at the solvent/bitumen interface for estimation of the molecular diffusion coefficient. To the best of our knowledge, such an analytical model that considers the swelling effect for estimation of the molecular diffusion coefficient is lacking in the literature. The developed model in this work fills the gap by introducing a simple tool for interpretation of experimental data of diffusion of gases into liquids.

2. Analytical model

Fig. 1 depicts the schematic of the system considered in this study. In this figure, h_{oi} is the initial height of the bitumen column and h_{gi} is the initial height of the gas phase (gas cap). The vertical axis is pointed downward, and its origin is selected at the initial location of the gas-liquid interface. The location of the interface at each time is shown by $s(t)$. The gas phase pressure is kept constant by injecting the makeup gas into the cell. The assumptions of the isothermal system, no chemical reaction between the gas and the liquid, non-volatile liquid, no mass transfer resistance at the interface, no natural convection, and constant unidirectional diffusion coefficient are used in this study.

The general form of Fick's second law is given by:

$$\frac{\partial(\rho\omega_g)}{\partial t} = \frac{\partial}{\partial z}\left(\rho D \frac{\partial\omega_g}{\partial z}\right), \tag{1}$$

where ρ is the mixture mass density, ω_g is the mass fraction of gas in bitumen column, and D is the molecular diffusion of gas into bitumen. Since the solubility of gases into bitumen are low especially at the early time of the experiments, one may safely assume that the density variations are negligible. This assumption particularly seems reasonable since our model relies on the early time experimental data of gas diffusion in bitumen. Also, the molecular diffusion coefficient is assumed to be independent of the concentration. These assumptions turn the final form of the governing PDE tractable. Therefore, the governing PDE will change to:

$$\frac{\partial\omega_g}{\partial t} = D \frac{\partial^2\omega_g}{\partial z^2}. \tag{2}$$

Furthermore, since we are interested only in early time data of the diffusion process, h_{oi} is assumed to be large enough such that the diffusion front does not reach the bottom of the cell. Therefore, the system can be assumed to be infinite acting with the following initial and boundary conditions:

$$\omega_g(z, t = 0) = 0, \tag{3}$$

$$\omega_g(z = s(t) \leq 0, t) = \omega^*, \tag{4}$$

$$\omega_g(z \to +\infty, t) = 0, \tag{5}$$

where ω^* is the equilibrium mass fraction of gas in bitumen. We need another condition for the motion of the interface to close the formulation of the problem. The following approach is used to obtain the Stefan boundary condition. Eq. (2) is integrated from $s(t)$ to $+\infty$:

$$\int_{s(t)}^{+\infty} \frac{\partial\omega_g}{\partial t} dz = D \int_{s(t)}^{+\infty} \frac{\partial^2\omega_g}{\partial z^2} dz. \tag{6}$$

Applying the Leibniz integration rule, we get:

$$\frac{\partial}{\partial t}\left[\int_{s(t)}^{+\infty} \omega_g dz\right] + \omega^* \frac{ds}{dt} = D \int_{s(t)}^{+\infty} \frac{\partial^2\omega_g}{\partial z^2} dz. \tag{7}$$

The right-hand-side of Eq. (7) can be integrated as: