



# Determination of mass transfer parameters in solvent-based oil recovery techniques using a non-equilibrium boundary condition at the interface



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## HIGHLIGHTS

- Dilute dissolution of gases into heavy oil was modeled accounting for 3 parameters.
- The unknown parameters were diffusion and mass transfer coefficients, and solubility.
- 3 Mass transfer parameters were measured through running one diffusion experiment.
- Sensitivity coefficients were applied to find the sensitivity of  $P$  to each unknown.
- Closer to onset of asphaltene precipitation, the interface resistance becomes larger.

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## ABSTRACT

Having a reliable estimate of gaseous-solvents molecular diffusion coefficients in heavy oil and bitumen is a requisite for analysis and design of gas injection and solvent-based recovery techniques. Nevertheless, diffusion coefficient is not measured accurately unless all other contributing mass transfer parameters are considered, included in the modeling, and estimated correctly. These other parameters are gas solubility and interface resistance, of which the latter is represented by mass transfer coefficient term. In this work, an analytical model is introduced in conjunction with an inverse technique to obtain these three abovementioned parameters using a single pressure decay data set. Sensitivity coefficient analysis is applied as an additional practical evaluation tool to display the sensitivity of the measured pressure to each of the unknown parameters. Characterization of the interface resistance as a physical phenomenon which hinders the molecular diffusion of gas through the interface and complicates the modeling is further investigated in this work. Incipient asphaltene precipitation in heptane-toluene-asphaltene mixture was chosen as a potential phenomenon which alters the interfacial resistance. It is shown that our proposed inverse analysis locates the unknown parameters correctly when history matching per se is not disclosing all the sufficient information for accurate parameter estimation.

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## 1. Introduction

In many oil sand reservoirs, bitumen viscosity is so high that it is not mobile at the reservoir condition. For this oil to be producible, viscosity must be reduced either by heat or by dilution. Using solvents not only reduces the viscosity by dilution, but it can also cause de-asphalting. This latter phenomenon upgrades the bitumen and leaves the heavier components inside the reservoir. In these processes, mass transfer parameters become important because they control the rate of dilution. Diffusion coefficient and solubility are two basic mass transfer parameters in dissolution of solvent gases into heavy oils. The first coefficient shows the rate of dissolution and the second one expresses the ultimate amount

of gas dissolution into heavy oil. Therefore, knowledge of these two parameters is necessary for designing solvent-based production schemes in these reservoirs. They could also be utilized in compositional reservoir simulators to forecast the recovery.

Among momentum, heat and mass transport processes, heat conduction and viscosity have standardized techniques for measurements. However, it is not the same for the diffusion coefficient; and measurements of mass transfer characteristics are often more challenging, specifically due to difficulties in measuring point values of concentration and other issues like: phase equilibrium, effect of convective transport and having a mixture rather than a pure fluid [1]. There are a few experimental methods to estimate the magnitude of gas diffusivity in a liquid. In one category of these techniques, the measurement is based on determination of the concentration of the diffusing gas along the diffusion path in the liquid with time. This technique needs compositional analysis and its downsides are numerous. They are system-intrusive, very

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**Nomenclature**

<i>A</i>	diffusion cell cross sectional area, m <sup>2</sup>
<i>C</i>	mass concentration, kg/m <sup>3</sup>
<i>D</i>	diffusion coefficient, m <sup>2</sup> /s
<i>E</i>	Objective function
<i>H</i>	Henry's law constant, MPa/(kg/m <sup>3</sup> )
<i>h</i>	height of bitumen column, m
<i>J</i>	sensitivity matrix
<i>k</i>	film mass transfer coefficient, m/s
<i>L</i>	vector of unknown values
<i>M</i>	group of coefficients
<i>M<sub>w</sub></i>	molecular weight, kg/(kg – mole)
<i>m</i>	mass of gas dissolved, kg
<i>N</i>	group of coefficients
<i>P</i>	pressure, MPa
<i>R</i>	universal gas constant, 0.0083144 MPa m <sup>3</sup> /kg – mol K
<i>T</i>	absolute temperature, K
<i>t</i>	time, s
<i>V</i>	volume, m <sup>3</sup>
<i>w</i>	gas mass fraction
<i>Z</i>	gas compressibility factor
<i>z</i>	vertical spatial coordinate, m

*Greek letters*

$\rho$	density of mixture, kg/m <sup>3</sup>
$\lambda$	eigen-value
$\rho$	damping parameter
$\Omega$	diagonal matrix

*Superscripts*

Asterisk chemical equilibrium condition

<i>n</i>	time step coefficient
<i>q</i>	iteration number
<i>T</i>	transpose

*Subscripts*

<i>b</i>	bitumen
<i>comp</i>	computed
<i>exp</i>	experimental
<i>eq</i>	equilibrium
<i>g</i>	gas
<i>gc</i>	gas cap
<i>i</i>	initial condition
<i>int</i>	interface
<i>m</i>	mass
<i>p</i>	eigen values index
<i>r</i>	relative

*Abbreviations*

BC	Boundary Condition
C7	heptane, C <sub>7</sub> H <sub>16</sub>
MM	Million
LM	Levenberg Marquardt
SAGD	Solvent Assisted Gravity Drainage
CSS	Cyclic Steam Stimulation
VAPEx	Vapour Extraction
EOS	Equation Of State
rms	root mean square

expensive, time consuming and labor intensive [2–4]. There are other methods that measure a dissolution-dependent property like pressure, solvent volume, or cumulative mass of dissolution and use this property to calculate diffusivity. These methods are referred to as indirect methods and have been referenced in many publications [1,5–11].

Within all these methods, the Pressure Decay technique introduced by Riazi [8] and Sachs [9] is a simple and highly reliable method. In this method, a high pressure constant volume diffusion cell is used in an isothermal condition. Heavy oil sample is placed at the bottom of the high pressure cell as a quiescent liquid column. At this time, the gas cap is pressurized to a certain pressure and then disconnected from the gas supply. Having constant gas-oil compositions in the cell, gas cap pressure starts to drop as a result of diffusion of the gas molecules into the heavy oil. This pressure drop is recorded with time and is used later in mathematical diffusion models to estimate mass transfer parameters. This technique has been applied by many authors to characterize mass transfer of gases into heavy oil and bitumen [5,8–10,12–21].

Different mathematical models have been introduced for modeling diffusion experiments and locating the unknown parameters using the pressure decay method. These models and their solutions are dissimilar in terms of the interface thermodynamic conditions, simplifying assumptions and parameter estimation algorithms. In addition to the diffusion coefficient and solubility, gas-oil interfacial resistance is another coefficient to be considered in the modeling and parameter estimation of the diffusion processes. There are many works published on determination of the first two assuming that no interfacial resistance exists [1,5,7,8,10,15,16,19]. However, only limited models [12–14,21–24] exist in the literature which considers the resistance at the interface of gas-oil as well. This

interfacial resistance is introduced in our calculations through reciprocal value of film mass transfer coefficient (*k*).

Tharanivasan et al. [16] studied different transport conditions at the interface based on the work of Zhang et al. [19], Upreti and Mehrotra [10] and Civan and Rasmussen [13] and categorized the boundary conditions used at the interface of these three works as equilibrium, quasi-equilibrium and non-equilibrium, respectively. When no gas concentration discontinuity (in the liquid phase) exists across the interface (right above and below the interface have the same concentration), the equilibrium term is used. Once passage of solvent molecules through the gas-liquid interface is hindered, the non-equilibrium term is applied. In the first case, the interfacial resistance term ( $1/k$ ) goes toward zero as the mass transfer coefficient (*k*) takes very large value. This allows removal of the resistance term from the whole modeling and simplifies the parameter estimation with one less parameter to be found. Otherwise, interfacial resistance should be included into the modeling and be estimated. Based on the preceding classification, the models of Riazi [8], Zhang et al. [19], Sheikha et al. [5], Etmnan et al. [1] would all be categorized as employing equilibrium boundary conditions.

On the other hand, from 2001 to 2009, Civan and Rasmussen model and their proposed inverse technique [12–14,22,23] remain the single diffusion model with non-equilibrium boundary condition in the related literature. They suggested a boundary condition which accounted for a possible hindrance in gas diffusion due to interfacial resistance and solved Fick's second law using their Robin-type boundary condition while keeping saturation concentration constant at the interface. Robin or third-type BC is a linear combination of a prescribed concentration and mass flux on the boundary of the domain. Their analytical solution is simplified to

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