MODELLING OF CO₂ AND GREEN-HOUSE GASES (GHG) MISCIBILITY AND INTERACTIONS WITH OIL TO ENHANCE THE OIL RECOVERY IN GAS FLOODING PROCESSES

By
Mohammed Kamal Emera

THESIS SUBMITTED FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Australian School of Petroleum
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To my father

To my mother

To my son (Ahmed)

To my wife, my brothers, and my sisters

To Prof. Hemanta Sarma

To Prof. Helmy Sayyouh

To Prof. Saad Eid

To all my family, friends, and professors
I feel privileged to have **Prof. Hemanta Sarma** as my principal supervisor on this research. I have been deeply educated from his professional experience and his outstanding kind personality. I appreciate all what he has done for me professionally and personally. Prof. Sarma is not only my supervisor but also he is a kind friend and well wisher. He always a good listener for me and for my problems even the personal problems. I cannot find how I can reward him but I will keep all what he has kindly done for me for all my life. Generally, he is one of the few people who have positively affected my life very much. I am deeply thankful to him for his support through these years. Also, I wish him all the best for his life, his career, and for his family especially his wife and his lovely daughter. In addition, I am deeply thankful to **Dr. Seung Kam**, my co-supervisor for all of his advices and support. Dr. Kam is a very nice, helpful, and knowledgeable person.

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Abstract

1. Objective

The objective of this research has been to develop more reliable models to predict the miscibility and interactions between CO$_2$ or green-house gas (GHG) and oil (dead and live oils) over a wider range of conditions, based on data from different site sources, considering all the major variables affecting each modelled parameter, and for different injected gas compositions. The Genetic algorithm (GA), an artificial intelligence technique based on the Darwinian theory of evolution that mimics some of the natural processes in living organisms, was used to develop these models, based on GA software that has been developed in this work (as a modelling technique). While applications of GA have been used recently in the mathematical and computer sciences, its applications in the petroleum engineering, especially EOR research, have been limited.

2. Motivation to Investigate the Potential of GA-based Models

The detrimental effects of CO$_2$ and/or GHG emissions from various industrial and human-activity sources on the environment are a major concern worldwide. This has resulted in an intensive global R&D effort to lower or mitigate the damaging impact of GHG on the environment. One potentially attractive and effective means of lowering the GHG emissions could be to capture them from their major sources of emissions and then sequester them in depleted oil and gas reservoirs while also enhancing oil recovery.

Typically, a GHG stream, also referred to as “flue gas”, contains high percentages of CO$_2$ in addition to other gases, notably, N$_2$, NO$_x$ and SO$_x$. The presence of high CO$_2$ content in the flue gas, in particular, could make this option potentially viable, provided the miscibility and interaction properties between the injected gas and reservoir fluids are favorable. Therefore, it is critical to ascertain the likely miscibility and interactions parameters between the injected gas (CO$_2$ or flue gas) and oil at different conditions to
determine the optimal miscibility and interaction conditions that contribute to oil viscosity reduction and oil swelling. They in turn enhance oil recovery through improved gas flooding process performance due to higher oil mobility, volumetric sweep efficiency, and relative permeability to oil.

Often miscibility and interactions between injected gases and oils are established through “experimental methods”, “new mathematical models” based on phase equilibria data and equations of state (EOS), and available “published models”. Experimental methods are time-consuming and costly. Moreover, they can handle only limited conditions. Mathematical models require availability of a considerable amount of reservoir fluid composition data, which may not be available most of the time. Although, the published models are simpler and faster to use, one must however recognise that most of these models were developed and validated based on limited data ranges from site-specific conditions. Therefore, their applications cannot be generic. Another noteworthy point is that most of the interactions models have been developed using dead oil data and pure CO$_2$ as an injected gas. Hence, they do not perform well for a wider range of live oils, as well as injected flue gases, which contain different components besides CO$_2$.

Consequently, there is a need to have more reliable miscibility and interaction models, which can handle a much wider range of conditions and different data sources. Also, these models should be able to consider all the major variables, different injected gas compositions, and live oil in addition to dead oil.

3. GA-based Models Developed in This Research

- **GA-based model for more reliable prediction of minimum miscibility pressure (MMP) between reservoir oil and CO$_2$:** This model recognised the major variables affecting MMP (reservoir temperature, MW$_{C5+}$, and volatiles and intermediates compositions). It has been successfully validated with published experimental data and compared to common models in the literature. It is noted that GA-based CO$_2$-oil MMP offered the best match with the lowest error and standard deviation.

- **GA-based flue gas-oil MMP model:** For this model, the MMP was regarded as a function of the injected gas solubility into oil, which in turn is related to the injected gas critical properties (pseudocritical temperature and pressure) besides reservoir temperature and oil composition. A critical temperature modification factor was also used in developing this model. The GA-based model has also been successfully validated against published experimental data and compared to several models in the literature. It yielded the best match with the lowest average error and standard
deviation. Moreover, unlike other models, it can be used more reliably for gases with higher N₂ (up to 20 mole%) and different non-CO₂ components (e.g., H₂S, N₂, SOₓ, O₂, and C₁-C₄) with higher ratio (up to 78 mole%).

- **GA-based CO₂-oil physical properties models**: These models have been developed to predict CO₂ solubility, impact on the oil swelling factor, CO₂-oil density, and CO₂-oil viscosity for both dead and live oils. These models recognised the major variables that affect each physical property and also properly address the effects of CO₂ liquefaction pressure and oil molecular weight (MW). These models have been successfully validated with published experimental data and have been compared against several widely used models. The GA-based CO₂-oil properties models yielded more accurate predictions with lower errors than other models that have been tested. Furthermore, unlike the other tested models, which are applicable to only limited data ranges and conditions, GA-based models can be applied over a wider data range and conditions.

- **GA-based flue gas-oil physical properties models**: These models predict flue gas-oil properties such as, flue gas solubility, impact on the oil swelling factor, and flue gas-oil density and viscosity while recognising all the major variables affecting each property. Also, the GA-based models recognised the different injected flue gas compositions. These models have been successfully validated with published experimental data and have also been compared against other commonly reported CO₂-oil models, which are often used for flue gas-oil physical properties prediction. The GA-based models consistently yielded a lower prediction error than the models that have been tested. Furthermore, unlike other models, which are applicable only over limited data ranges and conditions, GA-based models can be valid over a wider range of data under various conditions.

All the above-mentioned models, developed in this research, are particularly useful when experimental data are lacking and the project financial situation is a concern. In addition, these models can be useful as a fast track gas flooding project screening guide. Also, they can easily be incorporated into a reservoir simulator for CO₂ or flue gas flooding design and simulation. Furthermore, they can serve as yet another useful tool to design optimal and economical experimental test protocols to determine the miscibility and interactions between the injected CO₂ or flue gas and oils in gas flooding processes.
Publications and Awards Based on This Research  
(2003-2006)

The following are the awards I have received and papers I have authored and/or co-authored based on my PhD research at The University of Adelaide, Australia:

1. Scholarships and Awards
   - Santos Ltd International Post-Graduate Scholarship, Australia, 2003-2006.

2. Publications:
   - Refereed Journal Papers


- **Refereed Journal Papers (passed the pre-screening for publications in the specified journals)**


- **Papers Submitted to Refereed Journals**


- **Conference Papers**


Statement of Originality

This work contains no material which has been accepted for the award of any other degree or diploma at any university or any tertiary institution and, to best of my knowledge and believe, this thesis contains no material previously published or written by another person, except where due reference has been made in the text.

I give consent to this copy of my thesis, when deposited in the University Library, being available for loan and photocopying.

Signed……………………………………………….   Date……………………………..
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Table A-9-1. Experimental data used for testing of the GA-based gas-oil mixture density model for flue gas-oil (dead and live oils) density data.

Table A-10-1. Experimental data used for developing and testing of the GA-based flue gas-oil viscosity models and the models prediction results.
Nomenclature

α  
Empirical parameter presented as a function of density and temperature and can be obtained by fitting the experimental data

β  
Mutation factor (0-1)

γ  
Oil specific gravity (oil density at 15.56°C), γ_o  
Oil gravity, °API

Δ  
Difference operator

ΔP  
Additional pressure, MPa

ξ  
Viscosity parameter of a mixture

θ  
Ratio between gas volume at standard conditions to the volume at system T and P

λ  
Mutation factor (0-1)

µ  
Gas (CO₂ or flue gas)-oil viscosity, mPa.s

µ*  
Viscosity of a gas mixture at low pressure, mPa.s

µ_d  
Viscosity of dead oil at the specified temperature, mPa.s

µ_i  
Initial oil viscosity at the specified temperature, mPa.s

µ_j*  
Viscosity of component j as a gas at low pressure, mPa.s

µ_m  
Mixture viscosity, mPa.s

µ_o  
Oil viscosity, mPa.s

µ_s  
Solvent viscosity, mPa.s

ρ  
Gas (CO₂ or flue gas)-oil density, g/cm³

ρ_c'/  
Pseudocritical density of a mixture, g/cm³

ρ_i  
Initial oil density at the specified temperature, g/cm³

ρ_MMP  
CO₂ density at the MMP, g/cm³

ρ_r  
Reduced density, fraction

Σ  
Sum operator
ANN  Artificial Neural Network
API  Oil gravity °API
Av.  Average
B  Ratio between oil volume at system T, and 1 atm to the volume at system T and P
b  Characterization parameter obtained by measuring the viscosity of the oil at 30° C and 1 atm. (μ<sub>30</sub>)
B<sub>0</sub>  Oil expansion factor
C<sub>g</sub>  GA evaluation constant = 1.0 for gas (CO<sub>2</sub> or flue gas) solubility, oil swelling factor, and gas (CO<sub>2</sub> or flue gas)-oil density models and =100 for gas (CO<sub>2</sub> or flue gas)-oil viscosity models and =5000 for MMP models
C<sub>i</sub>  Carbon number
Dev.  Deviation (error), %
EOS  Equation of state
exp  Exponential factor
Exp.  Experimental
F  Weighting composition parameter
F<sub>impure</sub>  Impurity correction factor
Fit (i)  Average fitness of chromosome i, where the chromosome has many fitness values based on the number of data available (j)
F<sub>R</sub>  Mole percentage of C<sub>2</sub> through C<sub>6</sub> in the reservoir fluid, %
GA  Genetic algorithm
GHG  Green-house gas
I  Oil characterization index
Interm.  Intermediates components, C<sub>1</sub>-C<sub>4</sub>, H<sub>2</sub>S, and CO<sub>2</sub>, fraction
K<sub>i</sub>  Normalized partition coefficient for carbon number i
m  Exponent
MF<sub>i</sub>  Critical temperature modification factor of injected gas component i
M<sub>inj</sub>  Molecular weight of the injected gas
M<sub>j</sub>  Molecular weight of gas component j
MMP  Gas-oil Minimum Miscibility Pressure, MPa
MMP<sub>CO2</sub>  CO<sub>2</sub>-oil Minimum Miscibility Pressure, MPa
MMP<sub>flue gas</sub>  Flue gas-oil Minimum Miscibility Pressure, MPa
MW  Average molecular weight of the oil
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>MW_{C5+}</td>
<td>Molecular weight of the C_{5+} components</td>
</tr>
<tr>
<td>MW_{C7+}</td>
<td>Molecular weight of the C_{7+} components</td>
</tr>
<tr>
<td>MW_{CO2}</td>
<td>CO_{2} molecular weight</td>
</tr>
<tr>
<td>MW_{flue gas}</td>
<td>Flue gas molecular weight</td>
</tr>
<tr>
<td>MW_{i}</td>
<td>Molecular weight of component i</td>
</tr>
<tr>
<td>n</td>
<td>Non-CO_{2} components, components, or the GA population size</td>
</tr>
<tr>
<td>nn</td>
<td>Number of available data points</td>
</tr>
<tr>
<td>NPV</td>
<td>Net present value</td>
</tr>
<tr>
<td>P (c)</td>
<td>Crossover probability</td>
</tr>
<tr>
<td>P (m)</td>
<td>Mutation probability</td>
</tr>
<tr>
<td>P_{b}</td>
<td>Bubble point pressure, MPa</td>
</tr>
<tr>
<td>P_{c}</td>
<td>Gas critical pressure, MPa</td>
</tr>
<tr>
<td>P_{C, CO2}</td>
<td>CO_{2} critical pressure, MPa</td>
</tr>
<tr>
<td>P_{C, inj.}</td>
<td>Injection gas critical pressure, MPa</td>
</tr>
<tr>
<td>P_{Ci}</td>
<td>Critical pressure of the gas component i, MPa</td>
</tr>
<tr>
<td>P_{cj}</td>
<td>Critical pressure of component j</td>
</tr>
<tr>
<td>P_{cm}</td>
<td>Mole average pseudocritical pressure, MPa</td>
</tr>
<tr>
<td>P_{CW}</td>
<td>Weight average pseudocritical pressure, MPa</td>
</tr>
<tr>
<td>pen</td>
<td>GA penalty value</td>
</tr>
<tr>
<td>P_{fit(i,j)}</td>
<td>Fitness function of data number j of chromosome i, fraction</td>
</tr>
<tr>
<td>P_{liq}</td>
<td>Gas (CO_{2} or flue gas) liquefaction pressure at the specified temperature, MPa</td>
</tr>
<tr>
<td>P_{pc}</td>
<td>Flue gas weight average pseudocritical pressure, MPa</td>
</tr>
<tr>
<td>P_{r, CO2}</td>
<td>Reduced CO_{2}-oil MMP, fraction</td>
</tr>
<tr>
<td>P_{r, flue gas}</td>
<td>Reduced flue gas-oil MMP, fraction</td>
</tr>
<tr>
<td>PR-EOS</td>
<td>Peng-Robinson equation of state</td>
</tr>
<tr>
<td>P_{s}</td>
<td>Saturation pressure, MPa</td>
</tr>
<tr>
<td>PV</td>
<td>Pore volume</td>
</tr>
<tr>
<td>RBA</td>
<td>Rising bubble apparatus</td>
</tr>
<tr>
<td>SA</td>
<td>Simulated annealing</td>
</tr>
<tr>
<td>SF</td>
<td>Oil swelling factor, fraction</td>
</tr>
<tr>
<td>SF_{i}</td>
<td>Dong (1999) factor representing the strength of species i in changing the apparent critical temperature of the mixture relative to the critical temperature of CO_{2}</td>
</tr>
<tr>
<td>Sol</td>
<td>Flue gas or CO_{2} solubility, mole fraction or scf/bbl or sm^{3}/m^{3}</td>
</tr>
<tr>
<td>Term</td>
<td>Definition</td>
</tr>
<tr>
<td>------------</td>
<td>-----------------------------------------------------------------------------</td>
</tr>
<tr>
<td>STDEV</td>
<td>Standard deviation, %</td>
</tr>
<tr>
<td>$T_{ac}$</td>
<td>Mole average pseudocritical temperature with using factor $SF_i$, K</td>
</tr>
<tr>
<td>$T_c$</td>
<td>Gas critical temperature, °C</td>
</tr>
<tr>
<td>$T_{c, CO2}$</td>
<td>Critical temperature of pure CO$_2$ gas, °C (31.1°C)</td>
</tr>
<tr>
<td>$T_{c, inj}$</td>
<td>Injected gas critical temperature, K</td>
</tr>
<tr>
<td>$T_{c, flue gas}$</td>
<td>Flue gas critical temperature, °C</td>
</tr>
<tr>
<td>$T_{ci}$</td>
<td>Critical temperature of gas component i, °C</td>
</tr>
<tr>
<td>$T_{Ci}$</td>
<td>Critical temperature of gas component i, K</td>
</tr>
<tr>
<td>$T_{cj}$</td>
<td>Critical temperature of component j</td>
</tr>
<tr>
<td>$T_{CM}$</td>
<td>Mole average critical temperature, K</td>
</tr>
<tr>
<td>$T_{cm}$</td>
<td>Mole average pseudocritical temperature, °C</td>
</tr>
<tr>
<td>$T_{CW}$</td>
<td>Weight average pseudocritical temperature with using the critical temperature modification factor ($MF_i$), °C</td>
</tr>
<tr>
<td>$T_{cw}$</td>
<td>Weight average pseudocritical temperature, °C</td>
</tr>
<tr>
<td>$T_{pc}$</td>
<td>Pseudocritical temperature (may be weight average or mole average), °C</td>
</tr>
<tr>
<td>$T_{pc}$</td>
<td>Flue gas weight average or mole average pseudocritical temperature, °C</td>
</tr>
<tr>
<td>$T_r$</td>
<td>Reduced reservoir temperature, °C</td>
</tr>
<tr>
<td>$T_R$</td>
<td>Reservoir temperature, °C</td>
</tr>
<tr>
<td>$T_{Res.}$</td>
<td>Reservoir temperature, K</td>
</tr>
<tr>
<td>Value$_{cal.}$</td>
<td>MMP, solubility, swelling, density, and viscosity predicted value</td>
</tr>
<tr>
<td>Value$_{exp.}$</td>
<td>MMP, solubility, swelling, density, and viscosity experimental value</td>
</tr>
<tr>
<td>$V_{c/m}$</td>
<td>Pseudocritical volume of a mixture, cm$^3$/g</td>
</tr>
<tr>
<td>$V_{cC7+/m}$</td>
<td>Critical volume of the C$_7+$ fraction, cm$^3$/g</td>
</tr>
<tr>
<td>$V_{cj}$</td>
<td>Critical volume of component j, cm$^3$/g</td>
</tr>
<tr>
<td>$V_o$</td>
<td>Oil volume fraction</td>
</tr>
<tr>
<td>Vol.</td>
<td>Volatiles (C$_1$ and N$_2$) mole percentage, %</td>
</tr>
<tr>
<td>Volatiles</td>
<td>Volatile components, C$_1$ and N$_2$, fraction</td>
</tr>
<tr>
<td>$V_s$</td>
<td>Solvent volume fraction</td>
</tr>
<tr>
<td>$w_i$</td>
<td>Weight fraction of component i, fraction</td>
</tr>
<tr>
<td>$w_{ic2+}$</td>
<td>Component i normalized weighting fraction in the C$_2+$ fraction of oil</td>
</tr>
<tr>
<td>$X$</td>
<td>Gas concentration, mole %</td>
</tr>
<tr>
<td>$x_{CO2}$</td>
<td>CO$_2$ mole percentage in the injection gas, %</td>
</tr>
<tr>
<td>$x_i$</td>
<td>Mole fraction of gas component i</td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
</tr>
<tr>
<td>--------</td>
<td>-------------</td>
</tr>
<tr>
<td>$x_j$</td>
<td>Mole fraction of component $j$, fraction</td>
</tr>
<tr>
<td>$y$</td>
<td>Mole fraction of diluted component</td>
</tr>
<tr>
<td>$y_{CO_2}$</td>
<td>Mole fraction of CO$_2$ in the injected flue gas, fraction</td>
</tr>
<tr>
<td>$y_i$</td>
<td>Mole fraction of the gas component $i$ in the injected gas, fraction</td>
</tr>
</tbody>
</table>