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Accurate prediction of miscibility of CO_2 and supercritical CO_2 in ionic liquids using machine learning

Mohammad Mesbah[a,*], Shohreh Shahsavari[b], Ebrahim Soroush[c], Neda Rahaei[d], Mashallah Rezakazemi[e,*]

[a] Young Researchers and Elites Club, Science and Research Branch, Islamic Azad University, Tehran, Iran
[b] Young Researchers and Elites Club, Shiraz Branch, Islamic Azad University, Shiraz, Iran
[c] Young Researchers and Elites Club, Ahvaz Branch, Islamic Azad University, Ahvaz, Iran
[d] Islamic Azad University, Science and Research Branch, Chem. Eng. Dept., Hesarak, Tehran, Iran
[e] Faculty of Chemical and Materials Engineering, Shahrood University of Technology, Shahrood, Iran

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ABSTRACT

In this study, the solubility of CO_2 and supercritical (SC) CO_2 in 20 ionic liquids (ILs) of different chemical families over a wide range of pressure (0.25–100.12 MPa) and temperature (278.15–450.49 K) were predicted, using a robust machine learning method of multi-layer perceptron neural network (MLP-NN). The developed model with the R^2 of 0.9987, MSE of 0.6293 and AARD% of 1.8416 showed a great accuracy in predicting experimental values. In another approach for predicting the CO_2 solubility, an empirical correlation with several constants was developed. With the R^2 of 0.9922, MSE of 3.7874 and AARD% of 3.5078 the empirical correlation showed acceptable results; nevertheless weak compared to the ANN. The significance of this correlation is that it needs no physical property of the ILs or their mixture, and for its estimation, even a simple calculator is sufficient. A comprehensive statistical assessment conducted to assure the robustness and generality of the model. In addition, the applicability of the model and quality of experimental data was fully investigated by Leverage approach.

1. Introduction

In the recent decade, CO_2-Ionic Liquid (IL) systems have been the subject of many research studies [1,2]. Due to the high solubility of CO_2 in ILs and insolubility of ILs in pure CO_2, these systems seem very interesting for numerous industrial applications. CO_2-IL systems could be effectively used in many chemical, petrochemical, and pharmaceutical applications; nevertheless, the most important applications of these systems that put them in the spotlight are CO_2 capture and recovery of compounds from an IL media.

The global warming and climate change are undeniable results of using fossil fuels as the main energy source. The necessity to prevent CO_2 emissions in widely accepted and currently is a common step in many industries [3,4]. This operation could be performed in many stages of the process. Generally, different types of CO_2 capture technologies could be categorized as pre-combustion, post-combustion and oxy-combustion. In pre-combustion processes, the CO_2 is separated from the fuel before combustion. A common pre-combustion CO_2 capture method in the gas industry is the absorption of CO_2 by aqueous amine solutions [5]. In the post-combustion processes, the CO_2 is separated from the flue gas components [6,7]. Chemical absorption, physical absorption, adsorption [8], membrane separation, and cryogenic separation are the most common post-combustion CO_2 capture processes [9]. In the oxy-combustion process, the combustion is performed in a pure O_2 environment.

Among these processes, amine scrubbing has been used since the 1930s and is very effective. Nevertheless, organic solvents are highly pollutant and volatile. In recent years, room temperature ionic liquids (RTILs) have been considered as great alternatives for conventional organic solvents [10]. As green substances, they have the capability to dissolve a wide range of inorganic, organic, and organometallic compounds, have a low vapor pressure, high polarity, and high thermal conductivity. They are immiscible with various organic solvents and have the possibility of tuning their properties by changing the combination of cation and anion [11].

In addition, ILs have been used to increase the selectivity of membranes for CO_2 [12–14]. In fact, by immobilizing an IL with high solubility of CO_2 inside the pores of a polymeric or ceramic membrane,

* Corresponding authors.
E-mail addresses: m_mesbah@alum.sharif.edu (M. Mesbah), mashalah.rezakazemi@gmail.com (M. Rezakazemi).

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Nomenclature

F	Activation function
ANN	Artificial neural network
AARD	Average absolute relative deviations
BP	Back-propagation
b_a	Bias of neuron a
R^2	Correlation coefficient
E	Cost function
Pci	Critical pressure of Component i (bar)
Tci	Critical temperature of Component i (K)
FFNN	Feed-forward neural network
GC	Group contribution
IL	Ionic liquid
x_i	ith input
LM	Levenberg–Marquardt
r_a	Linear combiner output
MSE	Mean square error
x_{CO2}	Mole percent of CO_2
Mw_i	Molecular weight of component i (g/mol)
MLP	Multilayer perceptron
y_a	Neuron output
N	Number of data point
PR	Peng–Robinson
PC-PSAFT	Perturbed chain polar statistical associating fluid theory
P	Pressure (KPa)
RK	Redlich–Kwong
SWCF	Square Well Chain Fluid
SC	Supercritical
w_{ia}	Synaptic weight of the ith input of neuron a
T	Temperature (K)

the permselectivity of CO_2 separation process increases. These kinds of membranes are known as a supported ionic liquid membrane (SILM). Besides, another family of ILs including metals in their anion (also known as Magnetic Ionic Liquids, MILs) has been applied to enhance CO_2 solubility and transport properties. Using these kinds of ILs in SILMs for CO_2 separation has also been studied [15–17].

One of the most important deficiencies in using ILs for extraction process is recovering the compounds form IL media. The distillation or evaporation processes would not be an option for thermally sensitive or volatile products. Using ILs miscibility with water for liquid-liquid extraction is also not an option due to cross-contamination between the phases presents [18].

Supercritical CO_2 (SC-CO_2) is proposed as the key to this problem [18]. In order to facilitate extraction SC-CO_2 dissolves in the liquid, but as mentioned before ILs are insoluble in CO_2 and subsequently, the product can be recovered. The SC-CO_2 extraction of anticipated solute from an IL solvent without contamination seems a reasonable solution. Cheap, non-toxic, non-flammable, moderate critical conditions, abundant, and easy to remove from extracted products is may be the most suitable fluid in supercritical processes [19].

In order to design processes involving CO_2-IL mixtures, comprehensive knowledge of operational condition is required. The complications in experimental measurements and their costly and time-consuming nature increase the necessity of developing prediction methods for prognosticating phase behavior of CO_2-ILs mixtures. In this manner, several attempts have been made by the researchers to use thermodynamic based models such as Peng–Robinson (PR) equation of state [20], group contribution (GC) method [21–24], Redlich–Kwong (RK) [25], truncated perturbed chain polar statistical associating fluid theory PC-PSAFT [26], Square Well Chain Fluid (SWCF) [27]. The asymmetric nature and the large dipole moment of ILs and quadruple moment of CO_2 make a strong molecular interaction that in the event of supercritical operations is very complicated to show by thermodynamic models. Due to this problem, each of the aforementioned thermodynamic based models could be used for a specific type of system and not for a wide range of mixtures. In addition, these thermodynamic models are complex, time-consuming, require advance mathematical derivations, and require several parameter adjustments. Therefore, a general unifying accurate model is required to predict the phase behavior of different types of CO_2-IL systems.

In this study, following our previous works [19,28], we attempt to predict the solubility of CO_2 and SC-CO_2 in ILs at a wide range of pressure (0.25–100.12 MPa) and temperature (278.15–450.49 K) with neural networks. The neural networks as a robust mathematical algorithm can provide an accurate model without requiring the physical aspects of the problem. This offers an opportunity to get rid of the complications in thermodynamic modeling caused by the quadruple moment of CO, the asymmetric nature, and large dipole moment of ILs. The systems under consideration involve 20 ionic liquids with different chemical structures and many of the systems contain SC-CO_2. In addition, for hand calculation purposes, a simple empirical correlation with several constant for predicting CO_2 solubility in each IL will be presented. The suggested correlation is based on multiple regression method. This simple, accurate, and straightforward correlation is a handy tool in engineering applications. At the end, the quality of experimental data and applicability of the proposed model would be fully

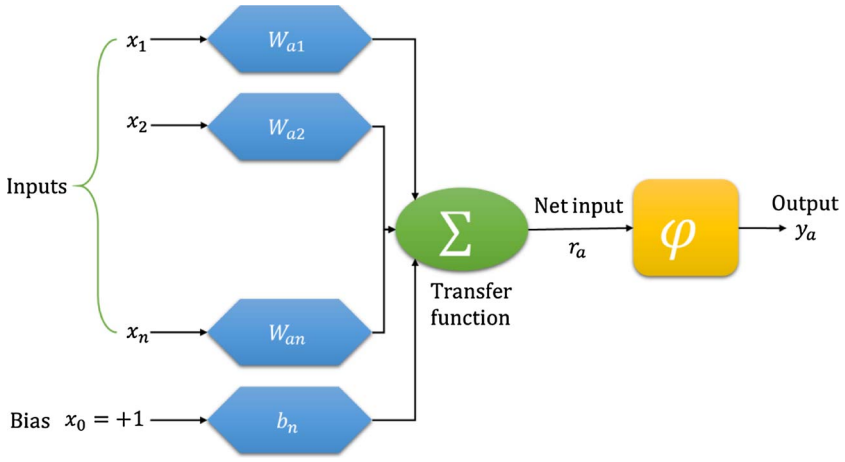

Fig. 1. A simple graphical illustration of an artificial neuron.