Modeling the Interfacial Tension of Water-Based Binary and Ternary Systems at High Pressures Using a Neuro-Evolutive Technique

Yasser Vasseghian, † Alireza Bahadori, ‡ Alireza Khataee, *,§∥⊥ Elena-Niculina Dragoi, *,# and Masoud Moradi†

†Research Center for Environmental Determinants of Health (RCEDH), Health Institute, Kermanshah University of Medical Sciences, 6715847141 Kermanshah, Iran
‡School of Environment, Science and Engineering, Southern Cross University, Lismore, NSW 2480, Australia
§Research Laboratory of Advanced Water and Wastewater Treatment Processes, Department of Applied Chemistry, Faculty of Chemistry, University of Tabriz, 51666-16471 Tabriz, Iran
∥Department of Environmental Engineering, Gebze Technical University, 41400 Gebze, Turkey
⊥Institute of Research and Development, Duy Tan University, 550000 Da Nang, Vietnam
#Faculty of Chemical Engineering and Environmental Protection “Cristofor Simionescu”, “Gheorghe Asachi” Technical University, Bld Mangeron no 73, 700050 Iasi, Romania

3 Supporting Information

ABSTRACT: In this study, artificial neural networks (ANNs) determined by a neuro-evolutionary approach combining differential evolution (DE) and clonal selection (CS) are applied for estimating interfacial tension (IFT) in water-based binary and ternary systems at high pressures. To develop the optimal model, a total of 576 sets of experimental data for water-based binary and ternary systems at high pressures were acquired. The IFT was modeled as a function of different independent parameters including pressure, temperature, density difference, and various components of the system. The results (total mean absolute error of 3.34% and a coefficient of correlation of 0.999) suggest that our model outperforms other habitual models on the ability to predict IFT, leading to a more accurate estimation of this important feature of the gas mixing/water systems.

INTRODUCTION

The interfacial tension (IFT) is a parameter defined for fluids in pure and mixed conditions, and in addition to the surface of the fluid, it depends on the soluble mass. These properties of fluids cause their outer layer to act as elastic sheets, which can lead to the adhesion of two levels of fluid.1 The IFT plays an important role in processes such as extraction, distillation, absorption, heat transfer in welding conditions, and mass transfer during extraction.2 Although IFT measurements are of interest, measuring it in a laboratory setup requires an expensive experimental device and a complex interpretation method. Therefore, it is important to apply a reliable method that provides an accurate estimate of IFTs. So far, many efforts have been made to predict the IFT of pure mixtures and fluids based on statistical thermodynamics, including the parachor method,3–5 the corresponding states principle,6,7 perturbation theory,8,9 density functional theory,10,11 and gradient theory.2,12–14 However, it was proven that some precise mathematical models are able to predict IFT with higher accuracy than conventional methods.15,16 Therefore, the necessity of using accurate mathematical models to predict the IFT of different types of fluids is undeniable. A significant number of previous studies were devoted to the modeling of IFT between H2O and other compounds in binary and...
Table 1. Previous Modeling Studies for the Interfacial Tension between H₂O and Other Compounds at High Pressures

| system                  | T (K) | p (MPa) | approach | equation of state | year | ref |
|-------------------------|-------|---------|----------|-------------------|------|-----|
| H₂O + CO₂               | 313   | 0–25    | LGT      | PR                | 1993 | 17  |
| H₂O + CO₂               | 298.15–318.15 | 0–5.9  | Cahn-Type | PR                | 2007 | 19  |
| H₂O + CO₂               | 297.9–373.3  | 1–60   | DFT      | SAFT-VR           | 2010 | 20  |
| H₂O + CO₂               | 287–313    | 0.1–25 | SGT      | SAFT-VR Mie 2006  | 2010 | 21  |
| H₂O + CO₂               | 298–374    | 1–60   | DSA      |                   | 2010 | 22  |
| H₂O + CO₂               | 300–383    | 0–30   | simulation |            | 2012 | 23  |
| H₂O + CO₂               | 298.2–333.2 | 0–5    | SGT      | PCP-SAFT          | 2012 | 24  |
| H₂O + CO₂               | 298–448    | 2–50   | simulation | SW              | 2012 | 25  |
| H₂O + CO₂               | 343–423    | 2–50   | simulation | SW              | 2012 | 26  |
| H₂O + CO₂               | 297.8–374.3 | 1.01–60 | LGT      | CPA               | 2013 | 27  |
| H₂O + CO₂               | 298.15–398.15 | 0.1–60  | SGT      | sPC-SAFT         | 2014 | 28  |
| H₂O + CO₂               | 298.15–303.15 | 0–25   | simulation | SAFT-γ Mie     | 2014 | 29  |
| H₂O + CO₂               | 284.15–312.15 | 1–6    | SGT      | CK-SAFT          | 2014 | 30  |
| H₂O + CO₂               | 298–448    | 2–60   | simulation | SAFT-VR Mie     | 2016 | 31  |
| H₂O + CO₂               | 278.2–469.2 | 0.1–69.1 | simulation | PR              | 2018 | 32  |
| H₂O + CO₂               | 298–373    | 3–15   | ADSA     |                   | 2019 | 33  |
| H₂O + N₂ + CO₂          | 298.15–373.15 | 1–30  | LGT      | CPA               | 2013 | 27  |
| H₂O + N₂ + CO₂          | 298–373    | 1–30   | LGT      | SRK               | 2001 | 18  |
| H₂O + CO₂ + N₂          | 298–448    | 2–40   | SGT      | SAFT-VR Mie      | 2016 | 31  |
| H₂O + CO₂ + Ar          | 298–473    | 2–60   | SGT      | SAFT-VR Mie      | 2016 | 34  |
| H₂O + CO₂ + CO₂         | 298.15–448.87 | 0.5–45 | simulation | PR-NRTL          | 2018 | 35  |

*SGT: square gradient theory; LGT: linear gradient theory; DFT: density functional theory; DSA: drop shape analysis; ADSA: axisymmetric drop shape analysis; GDT: density gradient theory; PR: Peng–Robinson; PR-NRTL: Peng–Robinson nonrandom two liquid; SRK: Soave–Redlich–Kwong; SAFT-VR: statistical associating fluid theory for variable range; SAFT-VR Mie: statistical associating fluid theory for variable range potentials of the Mie form; PCP-SAFT: perturbed-chain polar statistical associating fluid theory; SW: Span–Wagner; CPA: cubic-plus-association; sPC-SAFT: simplified perturbed-chain statistical associating fluid theory; PC-SAFT: perturbed-chain statistical associating fluid theory; SAFT-γ Mie: statistical associating fluid theory for γ Mie form; CK-SAFT: original statistical associating fluid theory.

Table 2. Interfacial Tension γ for Water-Based binary and Ternary Systems at Temperatures T and Pressures p, Where Δρ is the Calculated Density Difference

| system type                      | p (MPa) | T (K)  | Δρ (kg m⁻³) | γ (mN m⁻¹) | ref |
|----------------------------------|---------|--------|-------------|-----------|-----|
| (30 mol % CO₂ + 70 mol % H₂) + H₂O | 0.50–45.10 | 298.03–448.87 | 762.5–994.7 | 33.3–72.0 | 35  |
| H₂O + H₂                         | 0.50–45.20 | 298.03–448.87 | 890.40–996.80 | 42.90–73.00 | 35  |
| pure N₂ + H₂O                    | 1.00–30.00 | 298.15–373.15 | 711.18–986.13 | 51.11–71.43 | 18  |
| (23.6 mol % CH₄ + 76.36 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 732.62–987.24 | 50.49–71.28 | 18  |
| (50.09 mol % CH₄ + 49.91 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 755.49–988.48 | 49.17–71.12 | 18  |
| (74.93 mol % CH₄ + 25.07 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 775.76–989.67 | 48.54–71.30 | 18  |
| (24.97 mol % CO₂ + 75.03 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 626.33–985.58 | 41.64–69.33 | 18  |
| (50.72 mol % CO₂ + 49.28 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 484.55–985.23 | 33.61–67.96 | 18  |
| (75.85 mol % CO₂ + 24.15 mol % N₂) + H₂O | 1.00–30.00 | 298.15–373.15 | 297.77–984.69 | 29.23–65.85 | 18  |
| H₂O + CO₂                        | 1.00–60.05 | 297.80–374.30 | 41.00–979.00 | 19.69–66.00 | 22  |
| H₂O + CO₂                        | 1.00–60.05 | 297.80–374.30 | 103.80–981.50 | 23.10–65.90 | 31  |
| H₂O + N₂                         | 2.00–40.00 | 298.15–448.05 | 670.40–974.10 | 38.90–71.10 | 31  |
| (51.20 mol % CO₂ + 48.80 mol % N₂) + H₂O | 2.00–40.00 | 298.15–448.03 | 399.00–969.00 | 28.10–64.00 | 31  |
| H₂O + CO₂                        | 5.00–45.00 | 307.40–382.90 | 38.60–866.40 | 22.30–45.00 | 52  |
| H₂O + CO₂                        | 1.10–22.45 | 322.80–322.90 | 196.40–969.67 | 29.10–63.70 | 53  |

*Expanded uncertainties at 95% confidence are U(T) = 0.05 K, U(p) = 70 kPa, and U(γ) = 0.017γ.*
of polymerization processes, removal of toxic compounds, pharmaceutical freeze-drying modeling and monitoring, thermal characteristics of PVC/clay nanocomposite foams, protein secondary structure prediction, and design of optimally loaded supercavitating hydrofoils.

Based on the advantages that ANN can bring, the focus of this work is on accurately and efficiently predicting IFT from the available literature. Thus, the necessity of performing complex and difficult laboratory experiments is reduced. For this purpose, a series of 576 IFT data for different water-based binary and ternary systems were extracted from the literature and used to determine IFT as a function of pressure, temperature, density difference, and various components of the system being modeled. To the authors knowledge, this is the first study about tackling the issue of IFT prediction in water-based binary and ternary systems using ANNs determined by a neuro-evolutive approach combining DE and CS.

## STUDY FRAMEWORK

### Database

To model the IFT in water-based binary and ternary systems at high pressures, the available literature was investigated. The search was carried out on Google Scholar, Web of Science, ACS, RSC, Scopus, Pubmed, ScienceDirect, and Springer Link, and 41 articles were identified with the search words “interfacial tensions”, “IFT”, “binary system”, and “ternary system”. After a careful analysis of the published materials, a set of binary and ternary systems containing water were identified, and the experimental conditions under which the IFT was measured, collected, and processed were obtained. Table 2 presents a summary of the gathered data that were further used for modeling purposes. The supplementary Excel file (Table S1) contains the total experimental data employed in this work.

### Modeling Procedure

In this work, the models were determined based on a neuro-evolutive technique that combines ANNs with DE and CS. ANNs represent the model of the process and the role of DE and CS was to determine the optimal architecture (number of layers and number of neurons in each layer) and the optimal parameters (weights, biases and type of activation function for each neuron). Although inspired from different sources, both DE and CS have a similar structure: a population of potential solutions is updated/evolved (though a series of steps) until a stop criterion is reached. In this work, for both DE and CS, the stop criterion is represented by the number of iterations reaching a prespecified value.

The main difference between the two algorithms is represented by the steps used to evolve the individuals in the population. In addition, while the population of DE is fixed, the CS population varies depending on the number of cloned solutions added and on the number of weak individuals eliminated. The strongness of the individuals is measured by a function which in the DE case is named the fitness function and in the CS case the affinity function. As it represents the same aspect and it is implemented using the same relation (eq 1), it will be further referred to as the fitness function.

\[
\text{fitness} = \frac{1}{\text{MSE}_{\text{train}} + 10^{10}} 
\]  

where MSE\text{train} represents the mean squared error computed in the ANN training phase.

The steps used by DE to evolve the individuals are mutation, crossover, and selection. In the mutation phase, a mutation operator is applied to each individual to generate a mutated one. In the crossover phase, characteristics from the parents (current and mutated individuals) are combined to form a child. All of the generated children form the trial population. In the selection phase, the trial population is compared with the current one and the individuals with the best fitness are selected to form the new generation. The mutation variant used in the current DE version is rand/2 (2 differential terms with the base vector randomly chosen) and the crossover version is binomial. To eliminate the need for manually setting the control parameters (that direct the magnitude and direction of search), an auto-adaptation approach is employed.

The steps used by CS to evolve the individuals are selection, cloning, hypermutation, and receptor editing. In the selection step, the best \( n \) individuals are selected to be cloned. After that, the selected individuals are cloned several times (set based on the parameters of the algorithm). In the hypermutation step, some parameters (genes) are mutated. This introduces new genetic material and creates diversity. In the reception editing phase, the worst solutions are removed and a set of newly generated individuals is added to the population.
To raise the quality of the initial solutions, the optimizers start with, and after the individuals are randomly generated, the opposition based principle is applied. In the current approach, the individuals represent ANNs in an encoded form. The encoding is necessary to transform the ANN into a structure that the optimizers can work with: vector with real numbers. In this work, a direct encoding, where a one-to-one mapping between the genotype (individual) and phenotype (ANN) exists, is applied.

Distinctively from other approaches, where multiple optimizers are included in the neuro-evolutive process, in this work, the algorithms are run in the serial mode. First, DE is applied to determine a set of solutions. After that, the best solution is selected and it is used as a starting point for the CS algorithm. In this manner, CS performs a local search around a single solution and not around multiple solutions as it would happen when it is included in the DE algorithm (hybridization). Figure 1 presents a general schema of the overall modeling approach.

To model the considered process, the main important characteristics of the systems must be identified and included as inputs. The independent variables of the process are pressure, temperature, and density difference. As the gathered data included both binary and ternary systems with various components (H2O, CO2, H2, N2, CH4) and in various proportions, additional inputs for the ANN were added. In all of the systems, the aqueous phase is represented by H2O and therefore, this information is not included in the model. As a result, only the information about the gaseous phase was added (substance type and quantity).

After that, all of the gathered data was processed (normalized, randomized and split into groups) to be included in the neuro-evolutive approach. The normalization step was based on the min–max approach and has the role of scaling all of the features and speeding up the training time. The randomization implies setting a random order for all of the data so that the training and testing are applied on randomly chosen points. From the available data (576 points), 75% were selected for training and the rest (25%) for testing.

DE and CS optimizations are procedures that simulate the natural processes by using random numbers. Therefore, a set of 25 simulations were performed with each algorithm. The settings used for the DE algorithm are as follows: number of iterations: 500, number of individuals in the population: 40 and for CS, number of iterations: 100, number of individuals in the population: 20, number of clones for each individual: 10, percent of newly added individuals: 10, mutation factor: 80. For DE, the control parameters are automatically adjusted using a self-adaptive procedure. Regarding the ANN, the following limits were imposed: the maximum number of hidden layers: 2, the maximum number of neurons in each hidden layer: 20. All of these parameters were set based on a series of preliminary tests.

### RESULTS

The statistics of the simulations performed with the proposed approach are listed in Table 3, where the architecture is indicated using the following notation inputs: neurons_hidden_layer: outputs.

The mean absolute error (MAE) computed from the testing data for the best solution is 5.34% (in the case of DE) and 3.34% (in the case of CS). This indicates that CS was able to perform a good local search and to improve the DE intermediary solution. A point-by-point comparison between the experimental data and outputs generated by DE and CS solutions for a small set of testing data (25 points) is given in Figure 2.

![Figure 2](image-url)

**Figure 2.** Point-by-point comparison between the experimental data and the outputs generated by the DE and CS solutions for 25 testing points.

As can be observed, there are very small differences between the experimental data and the values generated by the ANN found by the CS algorithm. Due to the flexibility of the ANN model and its generalization capabilities, it can be efficiently used to determine IFT for a variety of binary and ternary systems containing water. The mathematical relations describing the ANN are represented by eqs 2–15. These equations can be used to predict IFT for any conditions considered as inputs in the model. Although the ANN has a good generalization capability, the error of the predictions (in comparison with the real experimental values) depends on whether an interpolation or extrapolation is performed. In the case of interpolation (when the inputs values are in the same interval as the training data), the error is similar to the reported testing error. In the case of extrapolation (when the input values are outside the interval of the training data), the predictions are prone to higher errors.
In eqs 2–5, the CO$_2$ value, H$_2$ value, N$_2$ value, and CH$_4$ value indicate the quantity of each compound in the system. The sum of all of the quantities for these compounds must be 1.$^{22}$
Given that the coefficient of interaction in binary and ternary systems is dependent on temperature, it is necessary to investigate IFT at various temperatures. For this purpose, different isotherms have been measured in binary and ternary systems in the range of pressures 0.5−45 MPa. The results for the binary and ternary systems are displayed in Figures 3−8. The relative standard deviation (σ(γ)/γ) of the IFT at each instance was 0.3%; in all instances, it was <0.9%. Generally, the relative uncertainty of IFT of all instances is 0.8%, and the relative uncertainty for combining all instances at 95% confidence is 1.7%. As shown in Figures 3−8, the IFT for all binary and ternary studied systems is reduced as the temperature and pressure increase. These results agree with previous studies of binary and ternary systems,56−58 and the developed ANN model accurately predicts this behavior. For the (H2O + CO2) system, in the studies by Chiquet et al. (2007), Kvamme et al. (2007), Georgiadis et al. (2010), and Chow et al. (2016), a sudden change in gradient is observed and this is due to IFT dependence on pressure,27 the aspect which is fully gathered by the current model that accurately predicts this change in gradient.

The dependence of pressure, temperature, and IFT of the (CH4 + N2) + H2O system is relatively simple. At constant gas composition and temperature, the reduction of IFT by increasing the pressure with different slopes at all pressures can be observed. At constant gas composition and pressure, the IFT decreases by increasing the temperature. At constant pressure and temperature, increasing the methane content results in the reduction of IFT18 and the current model accurately captures this behavior. The current model also accurately predicts the dependence of IFT on component concentration in ternary systems. At constant pressure and temperature, enhancing the carbon dioxide and nitrogen content leads to IFT reduction so that this behavior is properly predicted by the model. Also, the current model accurately predicts the complex temperature and pressure dependence of ternary systems with high CO2 concentration.

The MAE between the experimental data and the outputs of the determined ANN model (described by eqs 2−15) is 3.34% for all of the 576 instances; this aspect can also be observed from Figures 3−8, and the model closely follows the experimental data. Table 4 summarizes the IFT values using empirical and ANN models in this study and previous studies. As can be seen, compared with previous studies, the current ANN generates more accurate IFT values. Therefore, it can be concluded that the ANN model is able to describe IFT in water-based binary and ternary systems with various components. One of the main advantages of the proposed model is simplicity and lower parameters compared to other models. For example, Georgiadis et al.22 employed more than one model to express the dependence of pressure on the temperature in the (H2O + CO2) system, while in the present study, one model is used for predicting the dependence of pressure on temperature in the mentioned system and in the other systems studied. In addition, the current model considers a greater variety than other models.
because it can cover a wide range of variables, systems, and components.

**CONCLUSIONS**

An optimal ANN determined using DE and CS algorithms is proposed for the IFT determination of water-based systems.

A total amount of 576 IFT values for 15 water-based binary and ternary systems at high pressure were considered for correlating the pressure, temperature, density difference, and various components of the system with IFT. 75% of the data was employed for training the ANN and 25% for testing the predictive ability of the model. This chosen model can

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*Figure 7.* Interfacial tensions $\gamma$ of the ternary systems as a function of pressure at different isotherms: ○ at 298 K; □ at 313 K; △ green at 333 K; ◊ at 353 K; and △ orange at 373 K; ----, calculated values using the ANN model.

*Figure 8.* Interfacial tensions $\gamma$ of the ternary systems as a function of pressure at different isotherms: ○ at 298 K; □ at 323 K; △ at 373 K; and ◊ at 448 K; ----, calculated values using the ANN model.
predict the data with a MAE of 3.34% and a coefficient of correlation of 0.999. The results indicate that the determined model is a very accurate tool that can correlate the IFT of a wide variety of compounds such as CO₂, H₂, N₂, and CH₄ in a wide range of pressure, temperature, and density difference values.

**ASSOCIATED CONTENT**

* Supporting Information*

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b03518.

Reference values from the literature (xlsx)

**AUTHOR INFORMATION**

Corresponding Authors
*E-mail: a_khataee@tabrizu.ac.ir, alirezhakhataee@duytan.edu.vn (A.K.).

*E-mail: elenan.dragoi@gmail.com (E.-N.D.).

**ORCID**

Alireza Khataee: 0000-0002-4673-0223

**Notes**

The authors declare no competing financial interest.

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