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Prediction of Sulfur Solubility in Sour Gases and Pure H_2S Using A GA-RBF Model

Bowen Sun[1, a, *], Ping Guo[1, b], Zhouhua Wang[1], Guocai Wu[2], Yin Bai[3]

[1]State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation, Southwest Petroleum University, Chengdu 610500, China;

[2]Second Oil Production Plant, PetroChina Changqing Oilfield Company, Qingyang, Gansu 745106, China;

[3]Research Institute of Exploration and Development, PetroChina Tarim Oilfield Company, Korla, Xinjiang 841000, China.

[a]sbwswpu@163.com, [b]guopingswpi@vip.sina.com

Abstract

Accurate estimation of sulfur solubility in super-critical fluids is of significance for preventing deposition of elemental sulfur in sour gas reservoir and successful exploitation of sour gases. Laboratory experiments of sulfur solubility are expensive, sophisticated, and time-costly. For this task, a radial basis function model combined with genetic algorithm was suggested in estimating the solubility of sulfur in super-critical fluids. 239 sets of investigational data composed of sour gases as well as pure H2S were obtained from the open literature so as to advance the model. Six important aspects were assumed as independent variables in estimating sulfur solubility basing on the Grey Analysis System: temperature, pressure, density of super-critical fluid, critical temperature, critical pressure of gas and the mole fraction of H2S. The result proposes that the performance of the suggested model demonstrated excellent conformity with the laboratory data. The absolute average relative deviation for sour gases and pure H2S datasets were calculated to be 4.98% and 3.31%, respectively, and compared with two commonly used empirical correlations (Chrastil, Hu et al.). The compared results indicate that the performance of the proposed model outperformed the two existing approaches which are regarded in the present work in prediction of sulfur solubility in super-critical fluids. Moreover, outlier analysis was carried out on the entire datasets to recognize the doubtful experimental data and applicable range of the suggested model.

Keywords

Sulfur; sour gas; solubility; genetic algorithm; radial basis function; prediction.

1. Introduction

In the past two decades, several gas reservoirs containing high H2S content are identified in the world, in which the H2S content are found to even reach up to 95%. Nevertheless, the most noticeable tickler for such gas reservoir is the sulfur deposition in gas well or formation [1-2]. It is well-acknowledged that deposition of sulfur would lead to a considerable and radical reductions of gas production and even block production facilities [2-3]. The occurrence of sulfur precipitation has as well been reported in the transportation of natural gas [4]. Accurate knowledge of sulfur solubility in super-critical sour gases is prerequisite to preventing sulfur from deposition. Sulfur may occur as a number of the polymeric species which range up to S8 in the gas as well as mixes with some other gases to give out poly-sulfides like H2S9 [5-6], resulting in the complex calculation of sulfur solubility in super-critical sour gases. The

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available methods for estimation of sulfur solubility can be categorized into the following: experiment, semi-empirical correlation, thermodynamic models, and artificial neural networks.

The solubility of sulfur in super-critical fluids can experimentally be acquired through the use of super-critical extraction method [2, 7-15]. Several laboratory measurements have been conducted to obtain sulfur solubility in super-critical fluids. Nevertheless, experimental determinations are usually expensive, sophisticated, and time-costly. Consequently, semi-empirical correlations have been suggested to predict sulfur solubility in super-critical fluids to avoid the demand of experimental measures.

Chrastil [16] (1982) suggested a correlation for the solubility of solids and liquids in super-critical gases depending on three distinct parameter equations. Chrastil associated solubility openly to the density or concentration of a gas. This avoided the complication of the equations of state, showing important variations from the investigational findings. In 1997, by analyzing experimental data of Brunner and Woll [10], Roberts [17] built a theoretical formula of sulfur solubility on the basis of Chrastil's solubility model. Since it is convenient and practical, this solubility model is widely used in sour gas reservoir advancement. Nevertheless, even if the data acquired is predicted, the model still exists with significant errors, while the application conditions of this model are not given. Hu et al. [18] (2014) brought in additional recent experimental data and showcased a tailored Chrastil correlation building a relationship between sulfur solubility and sour gases properties, which they found that the density of 200 kg/m3 was a good demarcation point. Thus, the density of solubility log-log association is considerably linear before and after this. Later, Guo and Wang [19] (2016) also proposed a new sulfur solubility model that predicted sulfur solubility in sour gases and a method based on the experimental data to fit co-efficients of the new model. They transferred the constant correlation co-efficient k in Chrastil's model to be a variable as a function of temperature. They also introduced the M by weakening the effects of the gas density and used a cycle approach to attain the value of M in minimizing the error. However, certain M values of the formula in their model are over 256 (S8). According to the open literature [3,5-6], sulfur may occur in the quantity same as species of polymeric which goes up until S8 within the gas. It merges together with others to bring about poly-sulfides or sulfanes like H2S9.

A majority of sulfur solubility thermodynamic models depicted through these researchers actually utilized PR-EOS - Peng-Robinson Equation of State. Tomcej et al. [20] (1998) utilized sulfur's critical temperature together with its pressure in describing the behavior of sulfur. From this, they achieved success in comparing data for experimentation at pressures between 8 and 69 MPa as well as at varied temperatures of between 305 and 480K for mixes spanning the whole H2S extent. Later, Gu et al. [2] also suggested experimental vital co-ordinates meant for S8 molecule. These parameters were decided upon from regressing vapor or steam pressures under minimal heat or temperature. Accordingly, Karan et al. [6] (1998) performed a regression of the pretty and repellent PR-EOS parameters for the S8 types out of steam pressure as well as for the pure sulfur's fluid density data. Overall, forming sulfur and sour gas stage stability through conservative thermodynamic representations can obviously necessitate using of a lot of unknown parameters.

In the recent past, Amir H. Mohammadi et al. [3] (2008) proposed artificial neural network algorithm in predicting sulfur content of pure H2S at increased temperatures as well as at great pressure, which was successful in correlating the data for experimental by use of pressures from 7 to 60 MPa as well as at high temperatures of 316.3K to 433.15K. Mehdi Mehrpooya et al. [21] (2010) noted that an extensive artificial neural network algorithm in estimating the sulfur solubility of sour gases basing on variables like temperature, pressure, sour gas-free gas gravity as well as the equal mole portion of H2S. Due to the fact that traditional BP feed-forward neural network is prone to fall to local optimality and the slowness of convergence rate, and the complexity of sulfur behavior in super-critical sour gases, to some extent, affecting the

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performance of prediction. The mean total relative deviation out of all the forecasted and experimental data is 17%.

Aimed at getting over the disadvantages of the former calculation methods, the GA-RBF algorithm is used in extending the capability for estimating sulfur solubility in sour gases as well as in pure H2S. In Part 2, an in-depth RBF algorithm together with GA-RBF model is supplied. In Part 3, the whole experimental database and analysis of main issues influencing sulfur solubility in sour gases are presented. In Part 4, the Exactness of GA-RBF model is analyzed, and GA-RBF model additionally evaluated against two empirical correlations. Lastly, the conclusions were presented in Part 5.

2. Methodology

2.1. RBF Neural Network Algorithm

Measured against conventional artificial neutral network of BP, the RBF neural network may change and improve to hidden layer in the training phase following specified challenges, and allotment of hidden layer's neurons which might be determined through volume, group and allocation of training models [22]. The central positions, the hidden layer's width may be animatedly recognized by great rate of learning. After recognizing BP's network structural design, it is expected to fail in transforming during training. Thus, it is hard establishing the hidden layer as well as hidden layers' neurons. Union's rate related to the system or network is also sluggish while the training got more connections with the samples. The compliment of RBF neural network is seemingly greater as compared to neural network of BP.

RBF network involves of 3 feed-forward layers which are diagrammatically displayed in Fig. 1. It shows one input layer, one hidden layer and one output layer, all of which have neuron numbers of r, i, and k correspondingly. The neuron found in input layer spreads input characteristics to following layers, with each neuron in the layer which is hidden linked to a function of kernel Φi(x) (taking account of the function of Gaussian), featured by Ci and (center) σi (width). Every output neuron yk calculates simply subjective summing up for answers of the neurons that are hidden for a specific pattern of input, xi

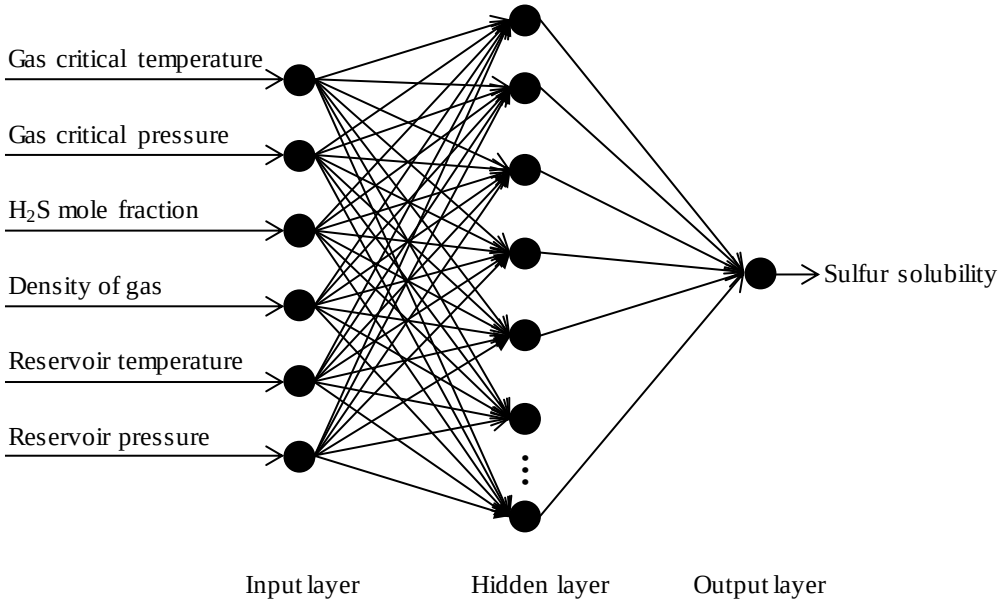

Fig 1. Topology of RBF neural network.

RBF network input layer recognizes non-linear mapping from the x to Φ(x), the equation is distinct as follows:

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$$\Phi_i(X) = \exp\left[-\frac{\|X-C_i\|^2}{2\sigma_i^2}\right]; i = 1,2 \dots, p. \quad (1)$$

Where Ci and σi are the center and width of the ith hidden neuron.

RBF network output layer finds out linear mapping from the Φ(x) to y, the equation is presented as follows:

$$y = \sum_{i=1}^{r} w_{ik}\, \Phi_i(x)\; ; k = 1,2, \dots, m. \quad (2)$$

Where k is the output layer node. wik is the weight connecting the ith hidden neuron to the output neuron. Φi(x) is the response of the ith hidden neuron for an input vector.

The error function can be illustrated by the actual network output tk:

$$e = \sum_{k=1}^{m}(t_k - y_k)^2 \quad (3)$$

Where e is the mean square error (MSE). tk and yk are predicted and target value.

2.2. Optimized GA-RBF Algorithm

Genetic algorithm utilizes the group explore technique, in which the population can be picked as the result. Through selection, cross-over, alteration and further heritable processes, modern era populace is definable. They may steadily evolve to best possible condition with estimate possible result. Genetic Algorithm is essentially an iterative procedure. Here, each iteration tends to hold some candidate solutions arranged by their advantages and disadvantages [22]. Certain solutions are picked based on certain pointers and calculated to give a new origination of contestant results with heritable operative being the intention union arrived at.

Another point which requires further instruction is changing the traditional GA-fitness function (MSE) into Absolute Average Relatively Deviation (AARD), and turn the error function (MSE) of RBFNN to Absolute Average Relatively Deviation (AARD).

$$MSE = \frac{\sum_{i=1}^{N}(\lambda_{pre}(i)-\lambda_{exp}(i))^2}{N} \quad (4)$$

$$AARD = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{C_i^{exp}-C_i^{pre}}{C_i^{exp}}\right| \quad (5)$$

Where N is the number of experimental data points. Cexp iand Cpre I are the experimental and calculated data for sulfur solubility. Cexp ave is the average value of all the experimental sulfur solubility.

The corresponding parameters and optimized results are reported in Table 1. RBF model optimized through GA methodology is designated as GA–RBF as briefly described in flowchart in Fig. 2. The criterion of GA convergence to the optimum condition is depicted in Fig. 3. To advance the efficiency of training of the RBF neural net, the inputs in addition to intentions were standardized so as to be within the range of [-1, 1] by utilizing:

$$P_n = \frac{2(P-P_{min})}{P_{max}-P_{min}} - 1 \quad (6)$$

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Where Pn and P are the normalized and actual parameters. Pmax and Pmin are the maximum and minimum values of P.

Table 1. Parameters of the GA-RBF model created in this task to estimate the solubility of sulfur in sour gases and pure H2S.

Parameters	Sour gases	Pure H2S
Input data form	[-1,1]	[-1,1]
Generation	200	200
Population size	20	20
Crossover fraction	0.8	0.8
Migration fraction	0.2	0.2
Best spread	2.18350185687041	6.5129006389426
Best MN	86	37
Target error	1.0e-6	1.0e-6

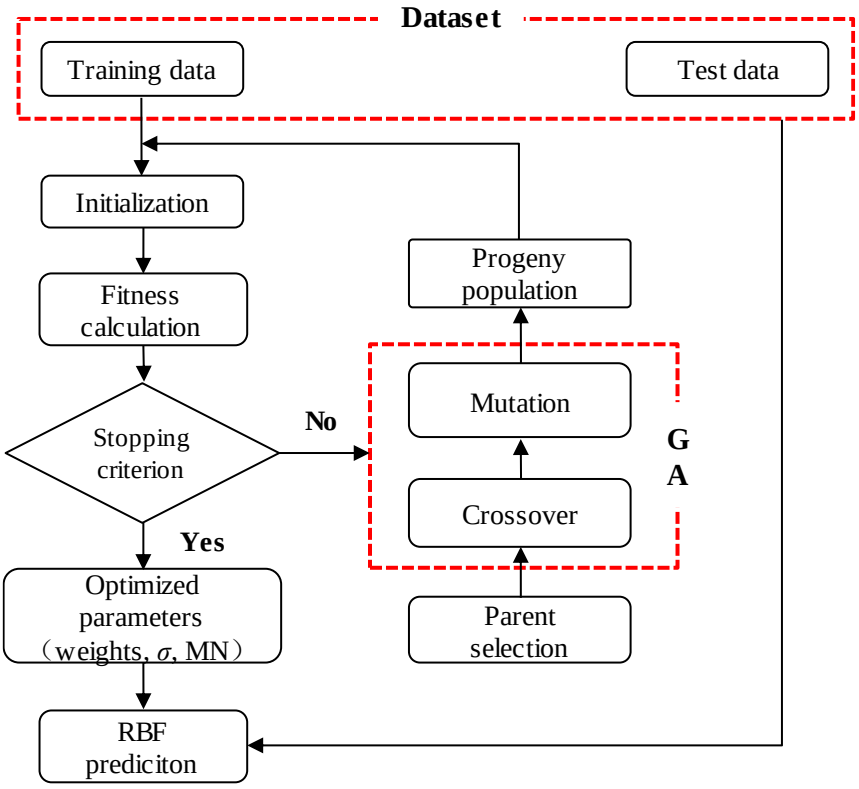

Fig 2. Flowchart of the newly proposed GA-RBF model.

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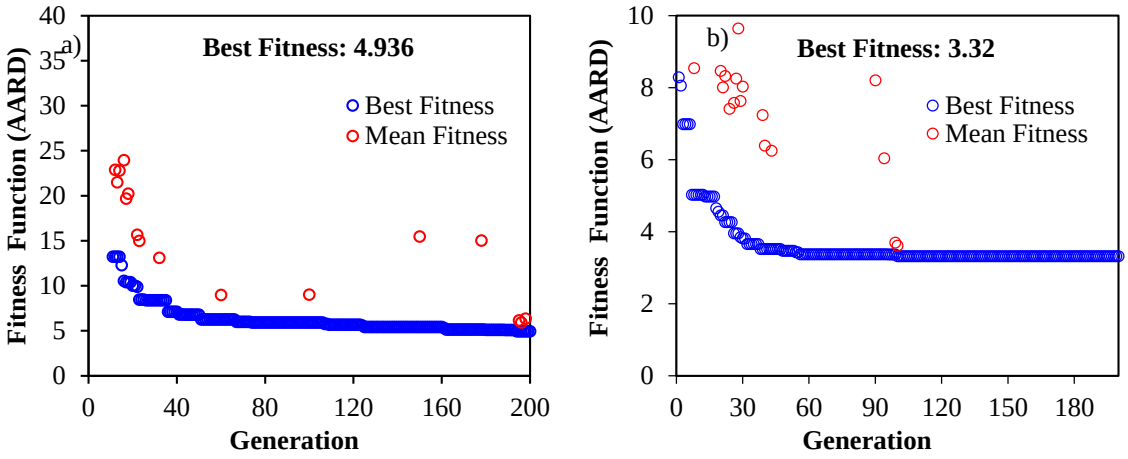

Fig 3. Performance of RBF in converging to optimum condition for (a) sour gas; (b) pure H2S.

3. Data Collection and Analysis

3.1. Experimental Database

A brief description of the total sulfur solubility experiments is presented in Table 2. To further the diversity of solubility of sulfur and authenticate the model, we introduced sulfur solubility of sour gases data that was done in 2010.

For sour gases, excluding the doubtful sulfur solubility data, and the data of high H2S content (H2S content >40%), we adopted the experimental data of Brunner and Woll [10] (1980), Sun and Chen [13] (2003), Yang et al. [14] (2009) and Bian et al. [15] (2010). The data covers wide ranges of temperature (303-433) K, pressure (10-60) MPa, fraction of CH4 (0.65-0.89), fraction of H2S (0.0495-0.2), and fraction of CO2 (0.008-0.2). To develop RBF model, the selected data were partitioned randomly into training (129sets), and testing (55sets). For pure H2S, we utilized the experimental data of Roof [8] (1971), Brunner and Woll [10] (1980), and Gu et al. [2] (1993). The data cover ranges of temperature (316-433) K, and pressure (7.03-60) MPa. The selected data were partitioned randomly into training (39 sets), as well as testing (16 sets). For this, the set of training is utilized to find the best RBF parameters and tuning network weights and biases. The testing data can be said to be an autonomous set which is not visible by network in training hence being utilized to evaluate the capability of generalization of network that is trained.

Table 2. Summary of the database of the solubility of sulfur in sour gases and pure H2S.

Author	System	Mole fraction of H2S (%)	T (K)	P (MPa)
Roof [8] (1971)	Pure H2S	100	317-394	7.03-31.17
Brunner and Woll [10] (1980)	Pure H2S and mixtures of (H2S+ CO2+CH4+N2)	100 6-20	373-433	10-60
Gu et al. [2] (1993)	pure H2S	100	363	11.83-36.21
Sun and Chen [13] (2003)	mixtures of (CH4 + H2S + CO2)	4.95-26.62	303-363	20-45
Yang et al. [14] (2009)	Sour gas	6.86	373	16-36
Bian et al. [15] (2010)	Sour gas	13.79	336-396	10-55.2

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3.2. Selection of Independent Variables on the Solubility Of Sulfur

It is renowned that good variety of independent variables is important in improving correctness of models of regression. Availability of outmoded or immaterial independent(s) for resultant type can reduce directness, scalability, as well as correctness of the model, hence ought to be abolished.

The grey relational analysis methodology has widely been utilized in recent years due to its applicability to the study of small samples and few data [23]. For this task, the independent variables going into the inference model for sulfur's solubility in sour gases were chosen through this methodology. Considering the 70 datasets of Brunner and Woll [10] (1980) which include a broad range of all independent variables, we chose it as the sample for examination. As illustrated in Fig. 4, the grey relation analysis screens out a lot of impact aspects as probable for the solubility of sulfur in super-critical sour gases. Basin on the principle of the least variable for predictive model, we in the end adopted six independent variables (Critical temperature and pressure of gas, mole fraction of H2S, density of sour gas, temperature, pressure) for sour gases, and two independent variables (temperature, pressure) for the pure H2S.

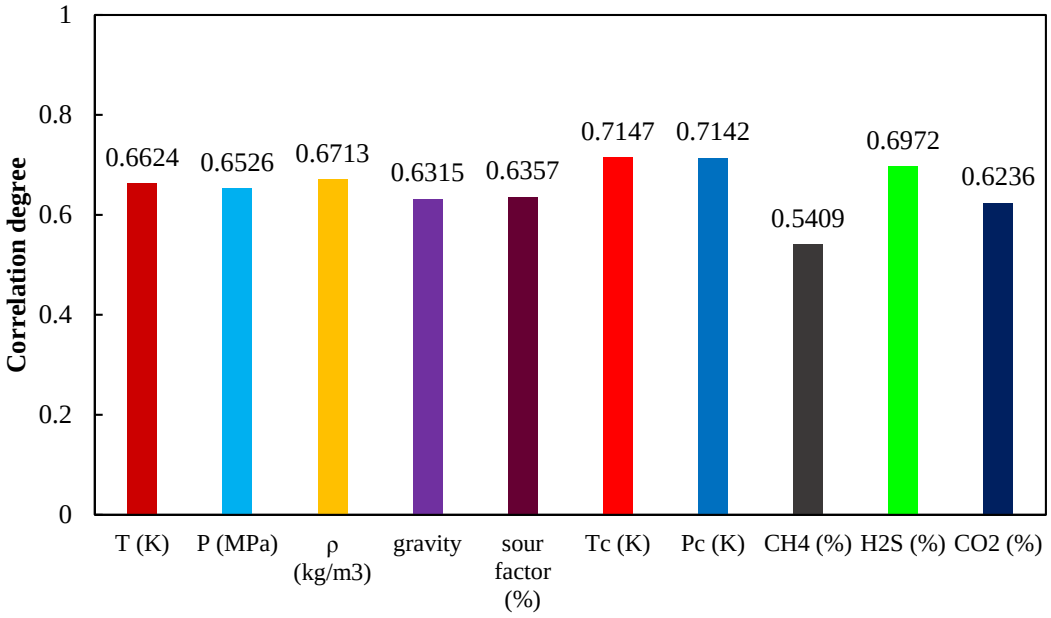

Fig 4. Grey correlation degree for the solubility of sulfur in sour gases.

4. Results and Discussions

4.1. Accuracy of the Proposed Model and Validation

In showing correctness of this contemporary model, distinction amid estimated and experimental sulfur solubility values is displayed in Fig. 5. As illustrated, a firm cloud of positions, around 45° line in the datasets show vigor of projected model. Outstanding accord is existent between the estimation of the GA-RBF model as well as the data on experimental sulfur solubility. For a clearer contrast between sulfur solubility computed by eminent models and experimental data, the Average Absolute Relative Deviation (AARD), Root Mean Square Error (RMSE), Standard Deviation (SD), and Co-efficient of Determination (R2) were computed by this equation:

$$AARD = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{C_i^{exp} - C_i^{pre}}{C_i^{exp}}\right| \quad (7)$$

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$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(C_i^{exp} - C_i^{pre}\right)^2} \quad (8)$$

$$SD = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N}\left(\frac{C_i^{exp} - C_i^{pre}}{C_i^{exp}}\right)^2} \quad (9)$$

$$R^2 = 1 - \frac{\sum_{i=1}^{N}\left(C_i^{exp} - C_i^{pre}\right)^2}{\sum_{i=1}^{N}\left(C_{ave}^{exp} - C_i^{pre}\right)^2} \quad (10)$$

Where N is the number of experimental data points. Cexp iand Cpre I are the experimental and calculated data for sulfur solubility. Cexp ave is the average value of all the experimental sulfur solubility.

The evaluation outcomes in estimate of the sulfur solubility of sour gases between GA-RBF and other models examined in this work are illustrated in Table 3. Out of all the models, the GA-RBF outperforms all the other correlations considered in this work and is intended at estimating sulfur solubility. Table 3 emphasize that the suggested model predictions are in outstanding agreement with the experimental data and with AARD of 4.98% in sour gases, 3.31% in pure H2S. Meanwhile, AARD for the best overtly published correlation in the present work is 17% [21] and 6.1% [3]. The correlation co-efficient (R2) of the GA-RBF model is near 1 (0.9999 in sour gas and 0.9971 in pure H2S) and is more than the same co-efficient calculated for other correlations.

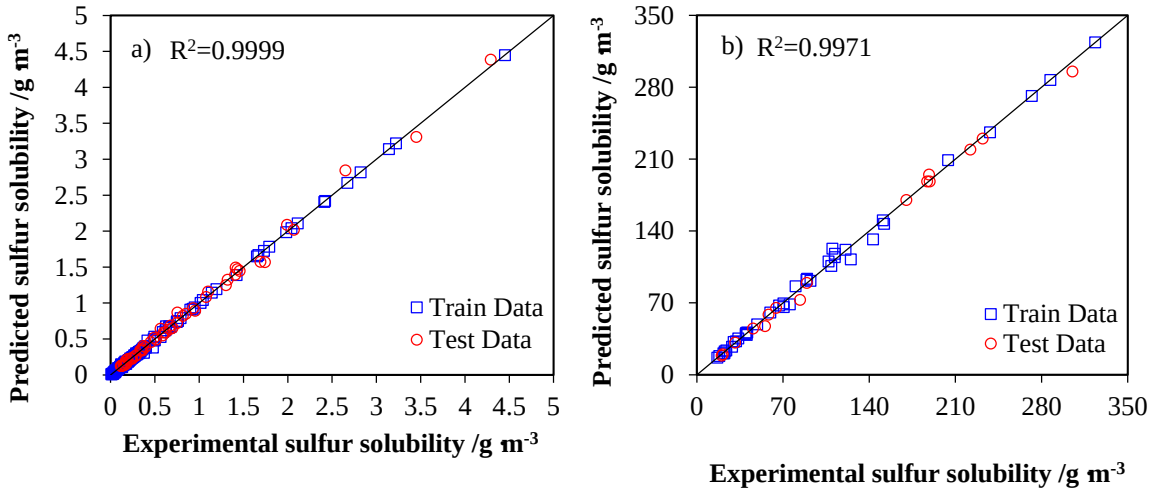

Fig 5. Estimated the solubility of sulfur values against experimental data of (a) sour gas; (b) pure H2S.

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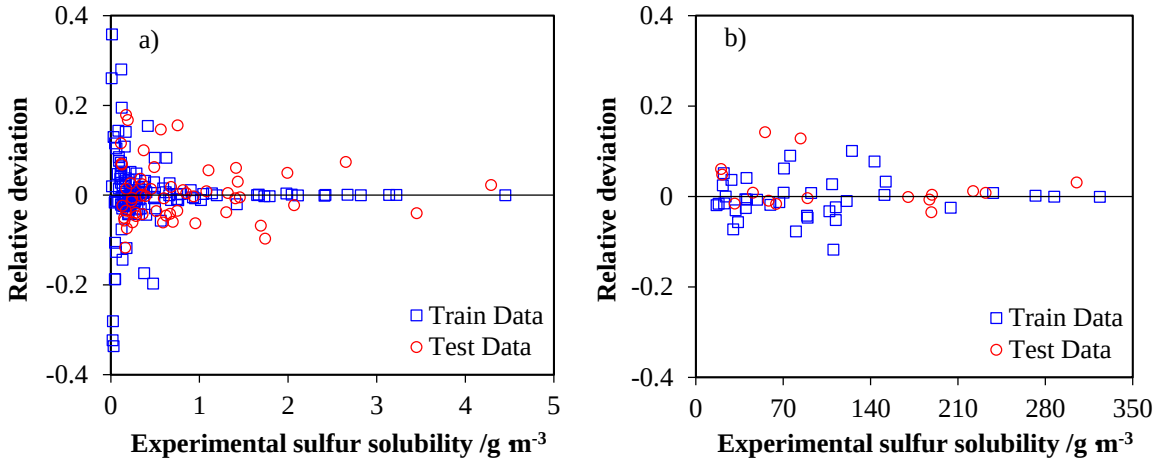

Fig 6. Relative deviations of GA-RBF model outputs versus target data of (a) sour gas; (b) pure H2S.

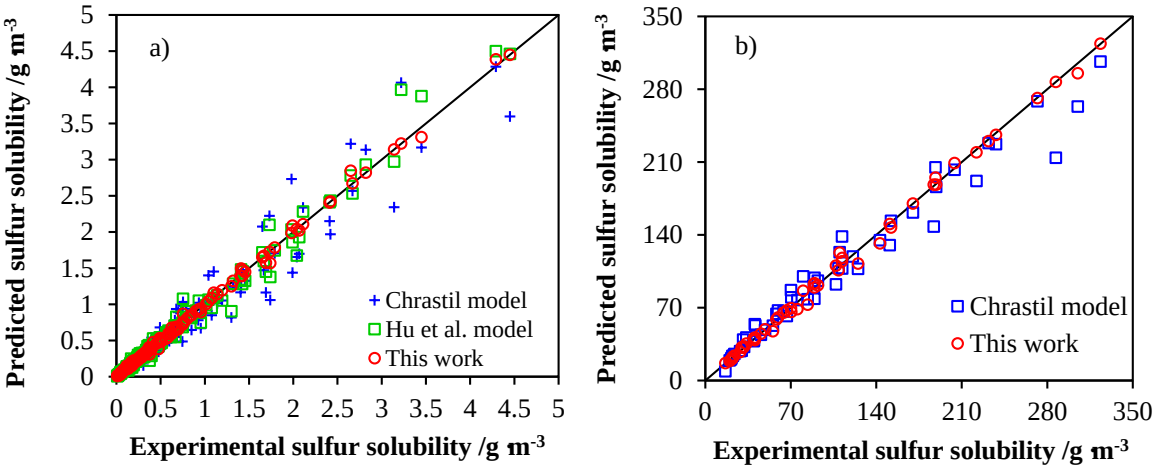

Fig 7. Comparison of the proposed GA-RBF with other models considered in this work to estimate the solubility of sulfur in (a) sour gas; (b) pure H2S.

Table 3. Comparison between GA-RBF model proposed in this task and literature models.

Evaluation matrices	Chrastil	Hu et al.	GA-RBF	N
Sour gases				184
AARD (%)	15.90	9.00	4.98	
SD	0.21	0.13	0.09	
RMSE	0.19	0.10	0.03	
R2	0.9386	0.9958	0.9999	
Pure H2S				55
AARD	9.70	9.70	3.31	
SD	0.14	0.14	0.05	
RMSE	15.87	15.87	4.37	
R2	0.9531	0.9531	0.9971	

4.2. Outlier Diagnostics

Outlier diagnostics is of immense importance in advancing mathematical models for intentions of discovering applicability field of the model as well as attributes of obtainable experimental data. The influence of statistical algorithm is practical for this intention.

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Three components, which are standardized residual (R), Hat indices (H) and critical Leverage limit (H*), are utilized for Leverage method. Computation particulars of these structures are established from references [24-25]. Outliers are determined by drafting William's diagram (by plotting of R versus H). Valid data ranges from -3≤R≤3 and H≤H*. The positions of data exceeding domain can be recognized to being outliers. Meanwhile, GHL positions are placed in domain of -3≤R≤3 and H≥ H*, while BHL positions range from R<-3 to R>3. The GHL positions might as well be chosen as those that might not be symbolized and approximated through this model. BHL positions are, on the other side, probably connected with experimental errors.

To perform outlier analytics, the William Diagram is plotted as shown in Fig. 8. On the sour gas data, the critical Leverage limit (H*) is computed to be 0.1141. As seen from this diagram, a great part of data points fall in suitable field of-3≤R≤3 and H≤H*. 2 data positions fall in the domain of -3≤R≤3 and H≥H*, which proposes that these 2 datasets are out of the applicability field or area of the practical model. Considerably, the model cannot symbolize or forecast the data that follows. Moreover, 6 data points fall in the area of R<-3 (even when they are larger or smaller than the (H*), they are well thought-out as outliers of the model points. These BHL (Bad High Layer) points are placed in the series of comparatively higher solubility. Considering that these calculations, more so at great temperatures pressures, are intensely improved the difficulty and danger of the experiment, more errors prone, for pure H2S, the critical Leverage limit (H*) is computed as 0.1636. All data points placed in the range of GHL have no outlier in test. Due to the outers concerning the sulfur solubility data were calculated at circumstances that were not replicated by other researchers. It is now hard to investigate whether the discovery is suitable and whether more endeavors are probable in testing the results.

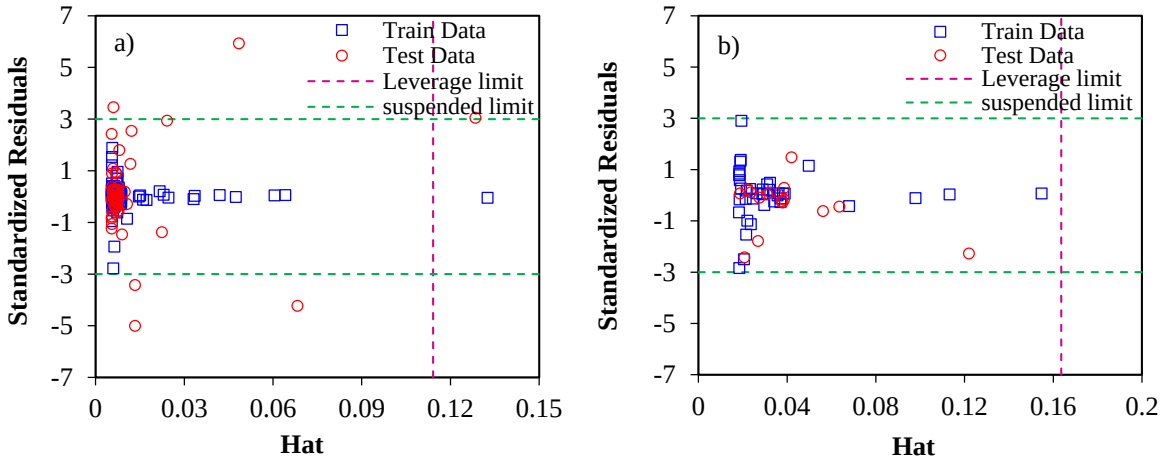

Fig 8. Detection of outlier data and applicability field of the models related in this task to estimate the solubility of sulfur in (a) sour gas; (b) pure H2S.

5. Conclusions

In this task, GA-RBF model was created to approximate the solubility of sulfur in super-critical fluids. A record containing 239 sets of experimental data was gathered from preceding literature before the advancement of regression model. Finally, a 3 layers RBF model optimized by genetic algorithm was proposed for regression. Outstanding conformity was attained between experimental sulfur solubility as well as approximated sulfur solubility in sour gases and pure H2S by use GA-RBF model. The statistical indices of AARD, SD, RMSE and R2 were 4.98%, 0.09, 0.03 and 0.9999, respectively, in sour gases. It was also computed as 3.31%, 0.05, 4.37, and 0.9971, respectively, in pure H2S. These outcomes show high predict correctness and strong generalization capacity of the suggested model. Furthermore, the outlier diagnostics

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offer an alternative for data testing. Comparison of the results with that attained from two generally used correlations showed that important advancement in the approximation of correctness which may be accomplished through the use of GA-RBF model.

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