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RESEARCH NOTES

Extension of an Artificial Neural Network Algorithm for Estimating Sulfur Content of Sour Gases at Elevated Temperatures and Pressures

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In this communication, we report an extended artificial neural network algorithm to estimate sulfur content of sour/acid gases. The main advantage of this algorithm is that it eliminates any need for characterization parameters, due to the tendency of sulfurs to react, required in thermodynamic models. To develop this tool, reliable experimental data found in the literature on sulfur content of various gases are used. To estimate the sulfur content of a gas, the information on temperature, pressure, gravity of acid gas free gas, and the concentrations of hydrogen sulfide and carbon dioxide in the gas are required. The developed algorithm is then used to predict independent experimental data (not used in its development). It is shown that the artificial neural network algorithm can be used as an efficient tool to estimate sulfur content of various gases.

1. Introduction

Sulfur deposition is a serious problem that can affect sour natural gas production, transportation and processing.^{1–29} Deposition of sulfur can block production facilities and cause a substantial and drastic reduction in the permeability of the formation near the wellbore.^{1,2} The occurrence of sulfur precipitation has also been reported in natural gas transportation and processing.^{1–5} Accurate knowledge of sulfur + sour/acid gas phase behavior is therefore important to avoid sulfur deposition problems.¹ One of the main difficulties in describing the phase behavior of sulfur-containing systems is the lack of suitable characterization parameters as the tendency of sulfurs to react takes their characterization difficult.^{1–29} Sulfur may exist as a number of polymeric species ranging up to S₈ in the gas and combines with other gases to produce polysulfides or sulfanes such as H₂S₉.^{1,14,15} The amount of each molecule depends both on pressure and temperature.^{1,10–13} Modeling sulfur + sour/acid gas phase equilibrium by conventional thermodynamic models therefore requires the use of many unknown parameters considering the above-mentioned reactions should be taken into account.^{1–29} The various thermodynamic models reported in the literature typically use the Peng–Robinson equation of state (PR-EoS)³⁰ and concern ranges of pressure, temperature, and hydrogen sulfide amount at gas production conditions typically greater than those of gas transportation and processing conditions.

Artificial neural network (ANN) algorithms are known to be effective to model complex systems. These tools are first subjected to a set of training data consisting of input data together with corresponding outputs.^{1,31,32} After a sufficient number of training iterations, the neural network learns the patterns in the data fed to it and creates an internal model, which is used to make predictions for new inputs.¹

The aim of this work is to extend a previously reported feed-forward (back-propagation) neural network (FNN) algorithm¹ for estimating the sulfur content of hydrogen sulfide to various sour gases. In this method, the sulfur content of a gas is estimated from the information on temperature, pressure, acid

gas free gas gravity, and concentrations of hydrogen sulfide and carbon dioxide in the gas. The optimization algorithm chosen in this work is a modified Levenberg–Marquardt algorithm^{33,34} with Bayesian regularization technique, which is specially indicated to optimize ANNs using small learning sample size. The reliable experimental data reported in the literature are used to develop and then validate this model.

2. Feed-Forward Neural Network Algorithm

Feed-forward neural networks are the most frequently used and are designed with one input layer, one output layer, and hidden layers.^{1,31,32,35–39} The number of neurons in the input and output layers is equal to the number of inputs and outputs, respectively.^{1,31,32,35–39} The accuracy of the model representation depends on the architecture and parameters of the neural network.^{1,31,32,35–39} In the FNN algorithm, the input layer of the network receives all the input data and introduces scaled data to the network.^{1,39} The data from the input neurons are propagated through the network via weighted interconnections.^{1,39} Every *i* neuron in a *k* layer is connected to every neuron in adjacent layers.^{1,39} The *i* neuron within the hidden *k* layer performs the following tasks: summation of the arriving weighted inputs and propagations of the resulting summation through an activation function, *f*, to the adjacent neurons of the next hidden layer or to the output neuron(s). In this work, the activation function is tangent sigmoid:^{1,39}

$$f(x) = \frac{1 - e^{-x}}{1 + e^{-x}} \quad x \in [-\infty, +\infty] \text{ and } f(x) \in [-1, +1] \quad (1)$$

where *x* stands for parameter of activation function. A bias term, *b*, is associated with each interconnection in order to introduce a supplementary degree of freedom. The expression of the weighted sum, *S*, to the *i*th neuron in the *k*th layer (*k* ≥ 2) is^{1,39}

$$S_{k,i} = \sum_{j=1}^{N_{k-1}} [(w_{k-1,j,i} I_{k-1,j}) + b_{k,i}] \quad (2)$$

where *w* is the weight parameter between each neuron–neuron interconnection and *I*_{*i*} = [*I*_{1,*i*}, ..., *I*_{*N_{k-1}*},*i*] represents an input

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Table 1. Literature Review of Experimental Data for Sulfur Content of Various Gases¹

author(s)	system	<i>T</i> /K	<i>p</i> /MPa	remarks
Sun and Chen ²⁹	7 (CH ₄ + H ₂ S + CO ₂) ternary mixtures	303–363	20–45	The mole fraction of H ₂ S in sour gas mixture ranges from 0.0495 to 0.2662 and that of CO ₂ ranges from 0.0086 to 0.1039.
Migdisov et al. ²⁸ Gu et al. ²⁷	gaseous hydrogen sulfide two rich-H ₂ S natural gases	323–563 363	up to 20 up to 34.7 and 32.8	The mole fractions of H ₂ S are 0.4411 and 0.9509.
	pure H ₂ S, pure CO ₂ and pure methane	363 (for the solubility of sulfur in pure H ₂ S) 363 and 383 (for the solubility of sulfur in pure CO ₂) and 383 (for the solubility of sulfur in pure methane)	up to 36.2 (for the solubility of sulfur in pure H ₂ S) up to 40.5 (for the solubility of sulfur in pure CO ₂) up to 50.2 (for the solubility of sulfur in pure methane)	
Davis et al. ²⁶	sour gases	333, 363, 393, and 423 K	5–55	high hydrogen sulfide content (up to 90%)
Brunner et al. ²⁴	sour gas mixtures of various compositions	394–486	up to 155	
Brunner and Woll ²¹	pure hydrogen sulfide and four gas mixtures composed of H ₂ S, CO ₂ , CH ₄ , and N ₂	373–433	up to 60	
Swift et al. ²³ Roof ²⁰ Kennedy and Wieland ¹⁹	hydrogen sulfide hydrogen sulfide pure methane, carbon dioxide, hydrogen sulfide, and mixtures of these gases	390–450 317–394 339, 367, and 394	35–140 up to 31 6.9–41.3	

Table 2. Experimental and Calculated/Predicted Sulfur Content of Various Gases (mass fraction) Using ANN Algorithm

ref	gas	<i>T</i> /K	<i>p</i> /MPa	acid gas free gas gravity	equivalent mole fraction of H ₂ S	mass fraction of sulfur	number of experimental data	AAD% ^{a,b}
29	gas mixture 1	303–363	30–45	0.553	0.0495	6.655×10^{-6} – 3.342×10^{-5}	8	13.3
	gas mixture 2				0.0993	1.005×10^{-5} – 4.016×10^{-5}	8	7.1
	gas mixture 3 ^c				0.1498	1.272×10^{-5} – 7.183×10^{-5}	8	30.6
	gas mixture 4 ^c				0.1771	1.274×10^{-6} – 8.626×10^{-5}	9	25.6
	gas mixture 5 ^c				0.2662	1.897×10^{-5} – 1.430×10^{-4}	8	8.9
	gas mixture 6 ^c				0.1000	1.010×10^{-5} – 3.928×10^{-5}	8	8.0
	gas mixture 7 ^c				0.1003	9.826×10^{-6} – 3.951×10^{-5}	8	8.0
27	gas mixture 1 ^c	363	11.5–32.8	0.650	0	1.729×10^{-2} – 5.798×10^{-2}	6	12.1
	gas mixture 2 ^c		18.4–34.7	0.641		2.691×10^{-4} – 2.267×10^{-3}	6	101.1
24	gas mixture 1	398	12.9–48.8	0.553	0.9075	1.800×10^{-4} – 5.970×10^{-2}	10	66.2
	gas mixture 2	473	8–155		0.4350	1.24×10^{-3} – 8.50×10^{-2}	9	8.0
	gas mixture 3	485	7.5–76		0.1275	0.00128 – 1.40×10^{-2}	9	6.0
	gas mixture 4	398–458	6.7–134.2		0.4100	6.49×10^{-4} – 5.090×10^{-2}	11	11.8
	gas mixture 5	398	7–43	0.655		1.80×10^{-5} – 6.670×10^{-3}	8	20.6
	gas mixture 6		7–43.5	0.859		4.0×10^{-5} – 8.180×10^{-3}	8	26.0
	gas mixture 7	398–408	7–43	0.863		3.0×10^{-5} – 6.390×10^{-3}	9	32.2
21	gas mixture 1	373–433	10–60	0.577	0.2750	1.120×10^{-2} – 4.33×10^{-1}	19	11.7
	gas mixture 2			0.598	0.2200	1.010×10^{-4} – 3.230×10^{-3}	24	9.4
	gas mixture 3			0.573	0.1275	4.7×10^{-5} – 3.570×10^{-3}	28	11.7
	gas mixture 4				0.1150	4.7×10^{-5} – 7.340×10^{-3}	22	18.7
27	CH ₄	383	20.5–50.2	0.553	0	2.799×10^{-5} – 1.295×10^{-4}	4	54.6
21	H ₂ S	373–433	10–60	0	1	1.060×10^{-2} – 1.760×10^{-1}	37	11.7 (5.7)
20	H ₂ S	316–383	7.0–31.1	0	1	1.243×10^{-2} – 7.227×10^{-2}	21	7.4 (6.4)
27	H ₂ S	363	11.8–36.2	0	1	3.526×10^{-2} – 7.423×10^{-2}	6	12.5
	CO ₂	363–383	12.1–40.5	0	0.75	3.112×10^{-5} – 1.170×10^{-3}	12	7.3

^a AAD: average absolute deviation = $(1/N) \sum_{i=1}^N |(\text{experimental value} - \text{predicted/calculated value})/(\text{experimental value})|$. ^b The values in parentheses show the results obtained from the previous ANN algorithm. ^c Data were used for validation, the rest of data were used for training (and testing).

vector. Using this feed-forward network algorithm with tangent sigmoid activation function, the output, O , of the i neuron within the hidden k layer is¹

$$O_{k,i} = \frac{1 - e^{-\sum_{j=1}^{N_{k-1}} [(w_{k-1,j,i}I_{k-1,j}) + b_{k,i}]}}{1 + e^{-\sum_{j=1}^{N_{k-1}} [(w_{k-1,j,i}I_{k-1,j}) + b_{k,i}]}} \quad (3)$$

The Levenberg–Marquardt algorithm^{33,34} is used for optimization purposes, as mentioned earlier.

To develop the ANN, the data sets are generally subdivided into three groups corresponding to the following three steps: training, testing, and validation.^{1,39} After partitioning the data sets, the training set is used to adjust the parameters. All synaptic weights and biases are first initialized randomly. The network is then trained; its synaptic weights are adjusted by optimization algorithm, until it correctly emulates the input/output mapping, by minimizing the average root-mean-square error.^{1,39} The

Table 3. Detail of ANN Algorithm Used in This Work

layer	number of neurons
1	4
2	17
3	1
number of hidden layers	1
number of parameters	103
number of data used for training (and testing)	260
number of data used for validation	46
type of activation function	tan-sigmoid
output neuron	natural logarithm of the calculated sulfur content of gas (in mass fraction)
input neurons	temperature (K), pressure (MPa), gravity of acid gas-free gas, and equivalent concentration of hydrogen sulfide (mole fraction) in the gas (eq 4)

Table 4. Values of Weights for Each Layer

layer 2			layer 3	
0.58274	5.13869	-1.49169	3.18662	-2.35474
-0.75830	-0.73549	-0.85845	2.36560	-4.25981
-4.85564	1.87309	4.17176	1.76611	-0.22616
-2.13779	-0.82793	1.87449	2.03593	-3.56093
0.04096	-1.16610	4.46781	4.76739	4.49547
2.49628	-0.47425	-1.09815	3.65959	1.12266
-0.64781	-0.25767	-0.63557	-1.90440	7.01662
-2.21274	-1.44727	-0.46426	3.76886	3.47944
-0.51147	-11.6307	2.83739	-10.0780	-6.57318
0.98058	5.64753	0.51279	-2.74405	4.21609
-1.44949	2.30670	-4.87516	-1.86728	3.23409
-1.17176	-1.99992	3.245994	-2.21420	1.65367
-0.69644	-0.44335	-10.3004	-4.59895	4.60172
-1.34977	1.28707	-1.30199	1.64366	-8.39672
-0.32533	-0.24922	-3.56770	-7.46831	-5.23477
-0.59265	-1.28936	-9.17508	-6.60693	-4.15341
1.05091	-2.92450	1.042373	1.66293	1.29322

Table 5. Values of Bias for Each Layer

layer 2	layer 3
-0.95553	-0.42778
1.45527	
5.52016	
3.15303	
1.97285	
-2.16296	
-0.80335	
-1.84660	
-4.91712	
5.49252	
0.07095	
1.09827	
-1.10336	
0.46998	
-2.42865	
-3.48609	
1.75044	

testing set is used during the adjustment of the network's synaptic weights to evaluate the algorithms performance on the data not used for adjustment and stop the adjusting if the error

on the testing set increases. Finally, the validation set measures the generalization ability of the algorithm after the fitting process.^{1,39}

3. Results and Discussion

Most of the reported experimental data on the solubility of sulfur in sour/acid gases are in the high temperature ranges, typically about 373 K. Table 1 summarizes literature review of experimental data for sulfur content of various gases.¹ The experimental data reported in Table 2 were used to develop our algorithm. As can be observed, this table reports sulfur content of various gases including pure methane, hydrogen sulfide, and carbon dioxide at different temperatures and pressures. The ANN algorithm detailed in Tables 3–5 with one hidden layer was devoted to compute the natural logarithm of sulfur content (in mass fraction) of gases (output neuron) in function of temperature, pressure, gravity of acid gas-free gas, and equivalent concentration of hydrogen sulfide in the gas. The equivalent concentration of hydrogen sulfide in the gas is calculated using the following equation:⁴⁰

$$z_{\text{H}_2\text{S}}^{\text{equi}} = z_{\text{H}_2\text{S}} + 0.75z_{\text{CO}_2} \quad (4)$$

where z is the mole fraction in the gas and the superscript *equi* stands for equivalent concentration of hydrogen sulfide in the gas phase. The optimum number of neurons in the hidden layer according to both the accuracy of the fit (minimum value of the objective function) and the predictive power of the neural network was found to be 17.

Table 2 and Figure 1 compare the results of ANN algorithm with the experimental data.^{20,21,24,27,29} As can be seen, acceptable agreement has been achieved and the average absolute deviation (AAD) among all the experimental and calculated/predicted data is 17%. Considering the fact that such measurements, especially at high temperatures and high pressures, are indeed not easy and some experimental data may have errors, therefore the AAD up to 17% can be regarded acceptable. It should be mentioned

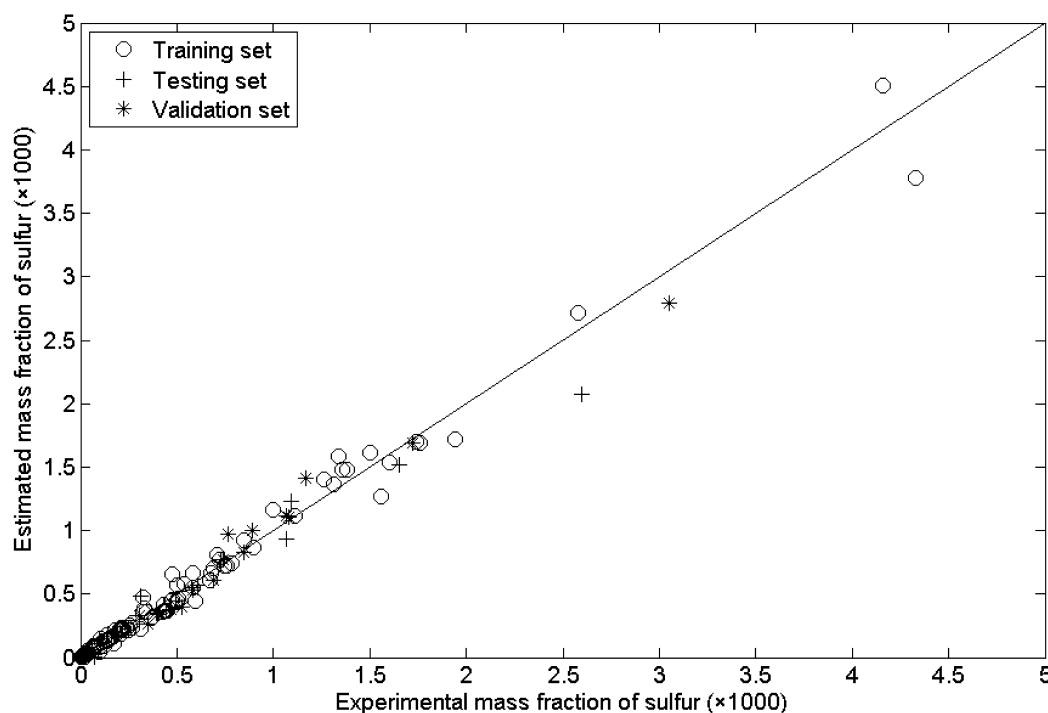


Figure 1. Estimated sulfur content of gases reported in Table 2 using the ANN algorithm versus corresponding experimental values.

that the ANN algorithm introduced in this study was developed using the most reliable experimental data found in the literature on sulfur content of various gases. We are fully aware that the ANN procedure was applied to low number of data and this procedure would largely gain in accuracy by developing slightly broader database constituted of well distributed data. This is a remark to be taken into account for further work. By generating reliable experimental data on sulfur content of various gases with different concentrations of acid gases in the future and readjusting the model parameters, more reliable predicted data would be expected.

4. Conclusions

A previously reported feed-forward artificial neural network algorithm¹ for estimating sulfur content of hydrogen sulfide was extended to estimate sulfur content of various gases at high temperatures and high pressures. To estimate sulfur content of a gas, information on temperature, pressure, gravity of acid gas-free gas, and equivalent concentration of hydrogen sulfide in the gas phase are required. The algorithm was developed using the most reliable experimental data found in the literature. The agreement between the experimental and calculated/predicted data was generally acceptable. This study showed a need to generate more reliable experimental data for developing the predictive tools.

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