Liquid-liquid equilibria for aromatic extraction systems using artificial neural networks

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Abstract

Phase equilibrium data regression is necessary to obtain the suitable values for modelling liquid-liquid extraction. It is important in equipment design, simulation, and optimization. Different factors may affect the accuracy of results. The present paper treats the nonlinear behavior of multicomponent liquid-liquid extraction using artificial neural networks. Two systems were studied, pentane-benzene-sulfolane system and heptane-benzene-tetraethylene glycol (TEG)-water system. Learning the first system in the neural networks doesn't gave good results while for the second equilibrium system, interaction terms were input, good values of mean square error and regression coefficient (R^2) values achieved.

Introduction

The monocyclic aromatic hydrocarbons like benzene, toluene, xylenes are valuable chemicals in petrochemical industry. They are produced by catalytic reforming of C6-C9 Naphtha's, followed by solvent extraction with selective solvents like Sulfolane or tetra ethylene glycol mixed with other solvents [1]. Sometimes the extraction process is important to reduce components like benzene for environmental requirements [2–5].

Phase equilibria and the interaction between phases are important issues in separation processes for both scientists and engineers. With multicomponent systems calculation is tedious, this is quite reason to use neural networks for evaluation of such systems. Sharifi and Abbasi et al., applied neural networks (NN) for perturbed-chain statistical associating fluid theory equation of state to estimate the cross-interaction parameters for each binary pair in multicomponent system, whereas phase equilibrium calculations require huge computation costs [6].

To understand the distribution of such components in an extraction process, their liquid-liquid equilibrium (LLE) behavior between the aqueous and organic phase must be known [1]. Experimental data are usually correlated in. There was extensive work on modelling and simulation methods on benzene recovery like universal quasichemical (UNIQUAQ), universal quasichemical functional activity coefficients (UNIFAC) and non-random two-liquid (NRTL) [7]. Modelling of phase equilibria is essential for simulation and optimization of separation processes in the chemical industry. The accurate phase equilibrium data regressions depend on different factors such as the experimental quality, the selected model used to describe the excess Gibbs energy, and the correlation algorithm [8, 9].

Theory

Artificial neural networks

The human brain processes a large amount of data in a parallel manner through neurons approximately in one millisecond after firing and multitasks are performed⁵. The brain neuron structure and the artificial neuron structures are shown in Fig. 1. The artificial neuron is a trial to mimic the behavior of biological

neurons [10]. Artificial neural network (ANN) and Biological neural network (BNN) can be interpreted in the similar methods [11].

In ANN, each input is multiplied by a factor called weight, the inputs multiplied by weights are summed and feed in a linear combination which makes the feed of hidden layer. The hidden layer neuron activates the input by an activation function (Fig. 2). Many types of activation functions existed [12], training p neuron sometimes called perceptron and updating the weights in many numbers of trials. The network consists of a number of neurons in the input layer, and hidden layers, the output layer gives the output of the neural network.

Neural networks applications in chemistry

There are some applications of artificial neural networks in chemistry field as well as in chemical engineering [13]; prediction of chemical patterns [9], prediction of organic chemistry reactions [9], proton design [14, 15] and phase equilibrium of binary system [16].

Building the neural network

Two systems were studied in this paper, the first one pentane-benzene-sulfolane, which was experimentally studied by Cassel et al., [17] (Table 1). The second system was heptane-*p*-xylene-TEG-water studied by Wang et al., [14] (Table 2). A neural network of 10 neurons in the hidden layer used for learning the Cassel et. al., [17] shown in Fig. 3 and the neural network trained by experimental work of Wang et. al., [14] is shown in Fig. 4.

Results And Discussion

Pentane-benzene-sulfolane

Training the neural network for the mixture of pentane-benzene-sulfolane gave a good result from the first attempts, on the program default settings. Better results achieved when increasing the training sets as shown in Table 3. An evident decrease in the mean square error achieved by increase the training set, and the correlation factor R^2 enhanced.

All data are tested in the neural network and the executed results were plotted for the pentane-benzene-sulfolane mixture as shown in Table 4 and Fig. 5. Figure 5 shows a zigzag shape line; the inflection could be attributed to the temperature factor.

Table 1
Experimental liquid-liquid equilibrium composition for pentane-benzene-sulfolane
system [16]

T(°C)	Raffinate			Extract		
	Pentane	Benzene	Sulfolane	Pentane	Benzene	Sulfolane
17	0.88	0.00	99.12	99.34	0.00	0.66
17	0.95	4.75	94.29	89.70	9.53	0.77
17	1.09	9.32	89.60	79.81	19.01	1.18
17	1.30	14.75	83.95	70.14	28.07	1.79
17	1.50	21.34	77.16	60.80	36.40	2.80
17	1.50	23.19	75.31	57.83	38.42	3.75
17	1.67	28.21	70.13	49.54	45.71	4.75
17	2.66	31.35	66.00	43.35	50.37	6.28
17	4.33	39.29	56.39	33.24	58.31	8.45
17	10.37	47.16	42.48	26.34	58.79	14.87
25	0.88	0.00	99.12	99.29	0.00	0.71
25	1.10	4.79	94.11	89.78	9.29	0.93
25	1.12	9.72	89.16	80.50	18.36	1.14
25	1.78	15.83	82.39	67.92	29.98	2.10
25	2.06	21.48	76.46	61.03	36.50	2.47
25	2.13	23.04	74.83	57.90	38.60	3.50
25	2.50	29.01	68.49	47.22	47.50	5.28
25	2.91	32.40	64.69	40.90	52.69	6.41
25	4.18	39.50	56.32	31.67	57.12	11.21
50	1.12	0.00	98.88	99.29	0.00	0.71
50	1.28	5.05	93.67	88.64	10.50	0.86
50	1.22	9.45	89.33	79.24	19.32	1.44
50	1.55	16.04	82.41	67.67	28.87	3.46
50	2.26	20.10	77.64	60.74	35.46	3.80
50	2.65	23.26	74.09	55.79	38.67	5.54

T(°C)	Raffinate			Extract		
	Pentane	Benzene	Sulfolane	Pentane	Benzene	Sulfolane
50	2.65	30.37	66.98	46.04	47.92	6.04
50	3.21	34.04	62.75	38.82	52.74	8.44
50	4.08	39.70	56.22	29.88	56.02	14.10

Table 2

Experimental liquid-liquid equilibrium composition for heptane-*p*-xylene-TEG-water system [17]

No.	Raffinate phase				Extract phase			
	Heptane	<i>p</i> -Xylene	TEG	Water	Heptane	<i>p</i> -Xylene	TEG	Water
1	0.7325	0.2614	0.0037	0.0024	0.0119	0.0347	0.5817	0.3716
2	0.4565	0.5268	0.0102	0.0065	0.00720	0.0531	0.5734	0.3663
3	0.4593	0.5284	0.0075	0.0048	0.0103	0.0741	0.5587	0.3569
4	0.6248	0.3692	0.0037	0.0024	0.00710	0.0473	0.557	0.3685
5	0.5492	0.4204	0.0185	0.0018	0.0102	0.0595	0.5677	0.3625
6	0.1617	0.8177	0.0125	0.008	0.00770	0.1255	0.5289	0.3379
7	0.4299	0.5504	0.016	0.0037	0.0172	0.1215	0.7014	0.1589
8	0.6172	0.3729	0.0077	0.0018	0.0227	0.0852	0.7265	0.1657
9	0.3121	0.678	0.0081	0.0018	0.0291	0.1795	0.6444	0.147
10	0.5442	0.412	0.0357	0.0064	0.0167	0.0877	0.7293	0.1663
11	0.5408	0.4259	0.0271	0.0055	0.0205	0.0939	0.7212	0.1645
12	0.4743	0.4911	0.0282	0.0077	0.0199	0.1071	0.7108	0.1525
13	0.4627	0.4955	0.034	0.0067	0.0199	0.0958	0.7201	0.1642

Heptane-*p*-xylene-TEG-water system

Training the neural network for the mixture of heptane-*p*-xylene-TEG-water system gave unsatisfied of mean square error and R^2 values even with increasing hidden layer neurons. The problem is solved by adding the interaction between the organic components and a quite reasonable result achieved. Further

enhancement achieved by increasing the input sets to the network. These trials are shown in Table 5. An evident decrease in the mean square error achieved by increase the training set, and the correlation factor R^2 enhanced.

Table 3
Values of MSE and R^2 for Training, Validation, and Testing

Results	Samples	MSE	R^2	Samples	MSE	R^2
Training	20	0.0259	0.9988	26	0.0852	0.9995
Validation	4	0.0119	0.9962	1	0.0291	0.9999
Testing	4	0.0199	0.9984	1	0.0632	0.9999

Adding term for the interaction between heptane and *p*-xylene and M2N refers to adding another terms for the interaction between heptane and TEG and the interaction between *p*-xylene and TEG. Values of R^2 enhanced from 0.9884 to 0.9978. This enhancement is important for good fitting. Experimental and neural network modeling results are plotted in Fig. 6.

Table 4
Experimental and model results for pentane-benzene-sulfolane system

T (°C)	ANN			Experimental		
	Pentane%	Benzene%	Sulfolane%	Pentane%	Benzene%	Sulfolane%
17	99.27155	-0.08375	0.860660	0.00	0.00	0.66
	89.76376	9.54814	0.726235	89.70	9.53	0.77
	79.78906	19.40284	0.852626	79.81	19.01	1.18
	69.94935	28.12000	1.897061	70.14	28.07	1.79
	60.69366	36.21941	2.997793	60.80	36.40	2.80
	57.99514	38.50091	3.409181	57.83	38.42	3.75
	49.54848	45.58060	4.803447	49.54	45.71	4.75
	43.69978	50.12378	6.135552	43.35	50.37	6.28
	33.60577	57.84664	8.637892	33.24	58.31	8.45
	26.38503	58.72607	14.82544	26.34	58.79	14.87
25	99.46963	0.18853	0.511169	99.29	0.00	0.71
	89.44026	9.46105	1.224706	89.78	9.29	0.93
	80.78937	18.26884	1.046113	80.50	18.36	1.14
	68.21736	29.65856	2.193729	67.92	29.98	2.10
	60.75473	36.87243	2.42173	61.03	36.50	2.47
	58.48516	38.75858	2.787028	57.90	38.60	3.50
	47.29987	47.24741	5.411572	47.22	47.50	5.28
	40.68110	51.79155	7.429843	40.90	52.69	6.41
	31.45940	57.07565	11.24073	31.67	57.12	11.21
50	99.30776	0.106843	0.589593	99.29	0.00	0.71
	88.48189	10.42493	1.073134	88.64	10.50	0.86
	79.46656	18.99350	1.538858	79.24	19.32	1.44
	67.69917	29.01656	3.302533	67.67	28.87	3.46
	60.91898	34.69250	4.377269	60.74	35.46	3.80
	56.13480	38.87121	4.990123	55.79	38.67	5.54
	45.96211	48.22792	6.076392	46.04	47.92	6.04

38.38134	52.63656	9.106049	38.82	52.74	8.44
29.01228	57.07592	13.80303	29.88	56.02	14.10

Table 5 The various network selected to enhance the values of MSE and R^2 for heptane-*p*-xylene-TEG-water system

		M1		M1N		M2N	
	No.	MSE	R^2	MSE	R^3	MSE	R^4
Training	9	0.00014	0.9884	0.00003	0.9974	0.00009	0.9993
Validation	2	0.01063	0.9518	0.00956	0.9133	0.00063	0.9964
Testing	2	0.00555	0.9334	0.00654	0.9468	0.00457	0.9664
Training	11	0.00124	0.9901	0.00003	0.9998	0.00005	0.9996
Validation	1	0.00438	0.9815	0.00013	0.9997	0.00008	0.9998
Testing	1	0.00492	0.9843	0.00026	0.9986	0.00041	0.9991

Conclusions

It is concluded that feed forward neural networks model is suitable for modeling experimental data of liquid-liquid equilibrium. A system pentane-benzene sulfolane was well simulated in the multilayer perceptron neural network. Neural network with Levenberg-Marquardt Algorithm to fit experimental data with 10 neurons in the input layer and 4 hidden layer neurons was efficient for the training. For the heptane-*p*-xylene-TEG-water system, the interaction between organic components of the mixture affects the simulation results by neural networks.

Declarations

Competing interests:

The authors declare no competing interests.

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Figures

Figure 1

The brain neuron structure and the artificial neuron structures

Name	Plot	Equation	Derivative
Identity		$f(x) = x$	$f'(x) = 1$
Binary step		$f(x) = \begin{cases} 0 & \text{for } x < 0 \\ 1 & \text{for } x \geq 0 \end{cases}$	$f'(x) = \begin{cases} 0 & \text{for } x \neq 0 \\ ? & \text{for } x = 0 \end{cases}$
Logistic (a.k.a Soft step)		$f(x) = \frac{1}{1 + e^{-x}}$	$f'(x) = f(x)(1 - f(x))$
TanH		$f(x) = \tanh(x) = \frac{2}{1 + e^{-2x}} - 1$	$f'(x) = 1 - f(x)^2$
ArcTan		$f(x) = \tan^{-1}(x)$	$f'(x) = \frac{1}{x^2 + 1}$
Rectified Linear Unit (ReLU)		$f(x) = \begin{cases} 0 & \text{for } x < 0 \\ x & \text{for } x \geq 0 \end{cases}$	$f'(x) = \begin{cases} 0 & \text{for } x < 0 \\ 1 & \text{for } x \geq 0 \end{cases}$
Parameteric Rectified Linear Unit (PReLU) [2]		$f(x) = \begin{cases} \alpha x & \text{for } x < 0 \\ x & \text{for } x \geq 0 \end{cases}$	$f'(x) = \begin{cases} \alpha & \text{for } x < 0 \\ 1 & \text{for } x \geq 0 \end{cases}$
Exponential Linear Unit (ELU) [3]		$f(x) = \begin{cases} \alpha(e^x - 1) & \text{for } x < 0 \\ x & \text{for } x \geq 0 \end{cases}$	$f'(x) = \begin{cases} f(x) + \alpha & \text{for } x < 0 \\ 1 & \text{for } x \geq 0 \end{cases}$
SoftPlus		$f(x) = \log_e(1 + e^x)$	$f'(x) = \frac{1}{1 + e^{-x}}$

Figure 2

The famous activation functions usually used

Figure 3

The neural network constructed for training mixture of pentane-benzene- sulfolane system

Figure 4

The neural network constructed for training mixture of heptane-*p*-xylene-TEG-water system

Figure 5

The ANN model and experimental values of (a) pentane, (b) benzene and (c) sulfolane

Figure 6

The ANN model and experimental values of (a) heptane, (b) p -xylene, (c) TEG and (d) water