Excess Molar Enthalpies for the Binary and Ternary Mixtures of Cyclohexane, Tetrahydropyran, and Piperidine at 308.15 K and Atmospheric Pressure: Experimental Measurements and Correlations

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Excess molar enthalpies for the binary and ternary mixtures of cyclohexane, tetrahydropyran, and piperidine have been measured using a Calvet microcalorimeter at 308.15 K and atmospheric pressure. All of the binary and ternary systems show endothermic behavior. The experimental binary results are correlated using the Redlich−Kister equation. The excess molar enthalpies for the ternary mixture are correlated using the Cibulka equation. The capability of an artificial neural network algorithm to model these data is finally studied.

Introduction

Excess thermodynamic properties of liquid mixtures are of great interest to conveniently design industrial processes and also to provide useful information on the molecular interactions required for optimizing thermodynamic models. In comparison with the experimental data reported in the literature for binary systems, the experimental data for ternary mixtures remain quite scarce. Cyclic ethers and amines represent two particularly interesting families of molecules for the purpose of testing models. Some hexacyclic aliphatic compounds, such as piperidine, tetrahydropyran, 1,4-dioxane, and cyclohexane, which are of very similar size and shape, differ in only one functional group (NH, O, CH_2) and are therefore of interest to study excess molar enthalpies of their binary and ternary mixtures. In our previous papers,[1,2] we have reported experimental excess molar enthalpies for binary and ternary mixtures of cyclohexane, with cyclic ethers (tetrahydropyran and 1,4-dioxane). In this communication, we continue our previous study[2] and report experimental excess molar enthalpies at 308.15 K and atmospheric pressure, for the binary and ternary mixtures of cyclohexane with tetrahydropyran (a cyclic monoether) and piperidine (a self-associated secondary cyclic amine). Few excess molar enthalpy data, for these binary systems, have been reported in the literature, at (293.15 to 298.15) K.[3−7] To our knowledge, there is no other literature data on the excess molar enthalpy of the ternary mixture. Our experimental data have been measured using a Calvet microcalorimeter. The Redlich−Kister equation[8] is used to correlate the experimental binary data. The experimental ternary data are correlated using the Cibulka equation.[9] The capability of artificial neural network (ANN) algorithm[10] as an alternative method to model these (binary and ternary) data is finally investigated.

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Table 1. Purities and Suppliers of Materials

chemical	CAS number	supplier	purity (GC)
cyclohexane	110-89-4	Merck	> 0.99
tetrahydropyran	142-68-7	Fluka	> 0.99
piperidine	110-89-4	Merck	> 0.99

Experimental Section

Materials. Purities and suppliers of materials are provided in Table 1.

Experimental Apparatus and Procedure. The experimental enthalpy data were measured at atmospheric pressure by means of a flow calorimeter (Calvet microcalorimeter, model C80, Setaram, Lyon, France). The temperature was maintained constant at (308.15 ± 0.02) K. The mixtures were prepared by mass using a Mettler PE 160 balance (precision of ± 0.1 mg), ensuring a probable uncertainty in the mole fraction less than 10^{-4}. The apparatus and procedures were tested by determining excess enthalpies for the standard system benzene + cyclohexane, and the results were found to differ by less than 3 % from those of Marsh.[11]

In our study of the ternary system, the excess molar enthalpy $H^E_{m,12+3}$ was determined for several pseudobinary mixtures in which component 3 (piperidine) was added to binary mixtures of components 1 (cyclohexane) and 2 (tetrahydropyran). For this purpose, binaries with fixed mole ratios, x_1/x_2, were prepared by mass. The excess molar enthalpy $H^E_{m,123}$ of the ternary mixture was then obtained from the relationship

$$H^E_{m,123} = H^E_{m,12+3} + (1 - x_3)H^E_{m,12} \quad (1)$$

where x_3 is the mole fraction of piperidine and $H^E_{m,12}$ is the excess molar enthalpy of the particular binary mixture.

Results and Discussion

The experimental excess molar enthalpies, for binary mixtures of cyclohexane, tetrahydropyran, and piperidine, are reported in Table 2 and plotted in Figure 1 along with some available literature data, at (283.15, 293.15, and 298.15) K,[3−6] for

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Table 2. Experimental and Calculated or Predicted Excess Molar Enthalpies, H^E, for the Binary Mixtures at 308.15 K and Atmospheric Pressure

x	experimental H^E	calculated H^E (using the Redlich−Kister equation)[8]	100 AD[b] (using the Redlich−Kister equation)[8]	calculated/predicted H^E (using ANN)	100 AD (using ANN)
mole fraction	J·mol^{-1}	J·mol^{-1}		J·mol^{-1}	
			x cyclohexane + (1 − x) piperidine		
0.101	258[a]	258	0.0	243	5.8
0.201	476	475	0.2	475	0.2
0.300	650	651	0.2	661	1.7
0.400	780	779	0.1	788	1.0
0.499	845	845	0.0	856	1.3
0.601	847[a]	846	0.1	861	1.7
0.679	795	798	0.4	798	0.4
0.801	635	633	0.3	623	1.9
0.901	391	392	0.3	395	1.0
			x cyclohexane + (1 − x) tetrahydropyran[a]		
0.101	157	156	0.6	165	5.1
0.200	294	295	0.3	298	1.4
0.300	412	411	0.2	402	2.4
0.400	490	490	0.0	486	0.8
0.499	526	528	0.4	545	3.6
0.606	519	517	0.4	521	0.4
0.699	461	462	0.2	442	4.1
0.800	355	354	0.3	365	2.8
0.900	198	199	0.5	190	4.0
			x tetrahydropyran + (1 − x) piperidine		
0.102	104	105	1.0	108	3.8
0.196	182	181	0.5	192	5.5
0.301	243[a]	243	0.0	245	0.8
0.397	279	278	0.4	273	2.2
0.497	292	293	0.3	286	2.1
0.598	284	284	0.0	281	1.1
0.700	251	251	0.0	253	0.8
0.801	194[a]	193	0.5	194	0.0
0.893	116	117	0.9	113	2.6

[a] From ref 2. [b] AD = |(experimental value − predicted value)/(experimental value)|. AAD = $(1/N)\sum_{i=1}^{N}$|(experimental value − predicted value)/(experimental value)|), where N represents number of experimental data. Using the ANN algorithm, the ADD for the cyclohexane + piperidine mixture is 1.7 %; the ADD for the cyclohexane + tetrahydropyran mixture is 2.7 %; the ADD for the tetrahydropyran + piperidine mixture is 2.1 %. All of these data were used for developing the ANN[10] except for the data indicated by footnote *a*, which were used for validation. Using Redlich−Kister equation,[8] the ADD for the cyclohexane + piperidine mixture is 0.2 %; the ADD for the cyclohexane + tetrahydropyran mixture is 0.3 %; the ADD for the tetrahydropyran + piperidine mixture is 0.4 %. All of these data were used for developing the Redlich−Kister equation.[8]

comparison. The experimental results, for the ternary mixture, are reported in Table 3 and plotted in Figure 2.

Correlation of Data. The experimental binary data were correlated using the Redlich−Kister equation:[8]

$$H^E_{m,jk}/J\cdot mol^{-1} = x(1-x)\sum_{i=0}^{n} A_i(2x-1)^i \qquad (2)$$

The coefficients A_i obtained by an unweighted least-squares regression method are listed in Table 4 together with the standard deviation s, for the three binary systems. In the above equation, i represents the number of parameters, and x stands for mole fraction of the first component in the binary mixture. The results of the correlation are reported in Table 2.

Several correlations for ternary excess molar enthalpies are available in the literature. In the present work, the experimental ternary data were correlated using Cibulka equation:[9]

$$H^E_{m,123}/J\cdot mol^{-1} = H^E_{m,bin}/J\cdot mol^{-1} + x_1x_2(1-x_1-x_2)(B_0 + B_1x_1 + B_2x_2) \qquad (3)$$

where $H^E_{m,bin}$, known as the binary contribution to the excess molar enthalpy, is given by:

$$H^E_{m,bin} = H^E_{m,12} + H^E_{m,13} + H^E_{m,23} \qquad (4)$$

where $H^E_{m,jk}$ is the excess enthalpy calculated from the correlated data of the $j-k$ pair (eq 2) using the ternary mole fractions x_j and x_k. This simplest method assumes that there are no ternary effects; that is, the ternary excess enthalpy is just a sum of the binary enthalpies of mixing. The values of $H^E_{m,12}$ at three specified constant ratios x_1/x_2 (approximate ratio values were 0.25, 1, and 4) of mole fractions of cyclohexane and tetrahydropyran in their binary mixtures were interpolated using the Redlich−Kister equation.[8]

The parameters B_i of eq 3 determined by the unweighted least-squares regression method are listed in Table 5. The results of the Cibulka equation[9] are reported in Table 6.

As can be observed in Table 2 and Figure 1, all of the binary mixtures are formed endothermally, suggesting that N−H···O hydrogen bonds formed in piperidine + tetrahydropyran are weaker than the N−H···N bonds solely present in piperidine + cyclohexane, which has the largest excess enthalpy of the three systems. All of the binary mixtures show symmetric H^E behavior. In Figure 1, our H^E results are reported, for the binary systems x cyclohexane + (1 − x) tetrahydropyran, x cyclohexane + (1 − x) piperidine, and x tetrahydropyran + (1 − x) piperidine, as a function of the mole fraction, x, and compared to literature data, at (283.15, 293.15, and 298.15) K.[3−6] As can be seen, our data, for the system cyclohexane + piperidine, are close to those of Woycicki and Sadowska[5] and Cabani and Ceccanti,[6] at 298.15 K, and lower than those of Moelwyn-Hughes and Thorpe,[3] at 293.15 K. For cyclohexane + tetrahydropyran, our data are lower than those of Moelwyn-Hughes and Thorpe,[3] at 293.15 K, and higher than those of Brocos et al.,[4] at 298.15 K. For tetrahydropyran + piperidine, our data

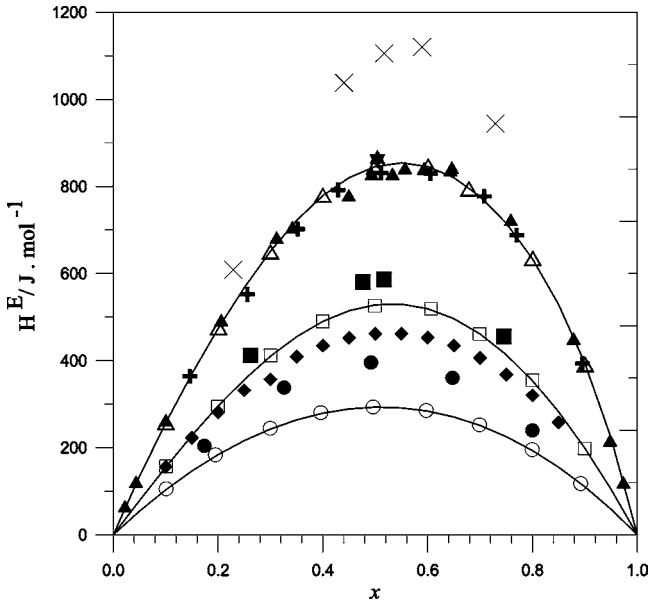

Figure 1. Excess molar enthalpies, H^E/J·mol^{-1}, for the three binary systems investigated. {x cyclohexane + (1 − x) piperidine}, [△, our data at 308.15 K; ×, Moelwyn-Hughes and Thorpe[3] at 293.15 K; + and ★, Woycicki and Sadowska,[5] at 298.15 and 283.15 K, respectively; ▲, Cabani and Ceccanti[6] at 298.15 K]; {x cyclohexane + (1 − x) tetrahydropyran}, [□, our data at 308.15 K; ◆, Brocos et al.,[4] at 298.15 K; ■, Moelwyn-Hughes and Thorpe[3] at 293.15 K]; {x tetrahydropyran + (1 − x) piperidine}, [○, our data at 308.15 K; ●, Moelwyn-Hughes and Thorpe[3] at 293.15 K]. Solid lines: fit our experimental results with the Redlich−Kister equation.[8]

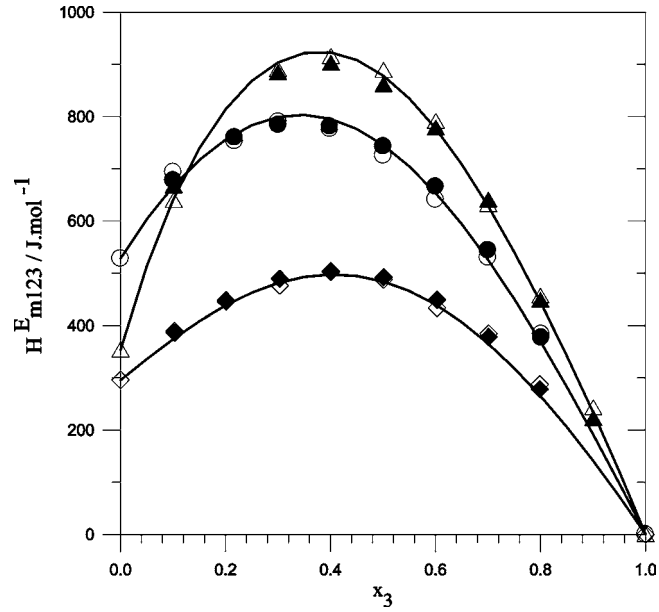

Figure 2. Excess molar enthalpies $H^E_{m,123}$, for the ternary mixtures of {cyclohexane (1) + tetrahydropyran (2) + piperidine (3)}, at 308.15 K. (◇, ◆), $x_1/x_2 = 0.25$; (○, ●), $x_1/x_2 = 1$; (△, ▲), $x_1/x_2 = 4$. Hollow symbols: experimental data. Solid symbols: ANN calculations. Solid lines: calculated values using Cibulka equation.[9]

Table 3. Measured Molar Enthalpies of Mixing, $H^E_{m,12+3}$, and Excess Molar Enthalpies, $H^E_{m,123}$/J·mol^{-1}, for the Ternary Mixture of Cyclohexane (1) + Tetrahydropyran (2) + Piperidine (3) at 308.15 K and Atmospheric Pressure

x_1	x_2	x_3	$H^E_{m,12+3}$	$H_{m,123}{}^E$ / J·mol^{-1}
	$x_1/x_2 = 0.25$; $H^E_{m,12}$/J·mol^{-1} = 296[a]			
0.179	0.717	0.104	124	389
0.160	0.639	0.201	209	446
0.139	0.557	0.303	270	476
0.120	0.480	0.401	325	502
0.100	0.399	0.501	340	488
0.079	0.320	0.603	316	434
0.060	0.239	0.701	296	385
0.040	0.161	0.799	228	288
	$x_1/x_2 = 1.00$; $H^E_{m,12}$/J·mol^{-1} = 528[a]			
0.450	0.449	0.101	219	695
0.392	0.391	0.217	340	753
0.350	0.350	0.301	421	791
0.301	0.300	0.399	459	777
0.250	0.250	0.500	462	726
0.200	0.200	0.600	430	642
0.150	0.150	0.700	372	531
0.099	0.099	0.802	279	384
	$x_1/x_2 = 4.00$; $H^E_{m,12}$/J·mol^{-1} = 354[a]			
0.719	0.180	0.101	322	641
0.560	0.140	0.300	643	891
0.480	0.120	0.400	704	917
0.399	0.100	0.501	713	890
0.320	0.080	0.600	650	792
0.240	0.060	0.700	526	632
0.160	0.040	0.800	386	457
0.080	0.020	0.900	208	243

[a] The values of $H^E_{m,12}$ at specified constant ratios x_1/x_2 were calculated using eq 2.

Table 4. Values of Coefficients A_i in Equation 2 and Standard Deviation s for the Binary Mixtures of Cyclohexane (1), Tetrahydropyran (2), and Piperidine (3)

system	A_0	A_1	A_2	A_3	A_4	s / J·mol^{-1}
1,2	2112.2	303.6	−230.5	0	0	1.3
1,3	3383.4	678.9	26.5	428.9	528.6	1.5
2,3	1172.1	48.4	19.3	0	0	1.0

Table 5. Values of Coefficients B_i in Equation 3 for the Ternary Mixture of Cyclohexane (1) + Tetrahydropyran (2) + Piperidine (3)

coefficient	value
B_0	3578.0
B_1	1724.8
B_2	−7434.6

are also lower than those of Moelwyn-Hughes and Thorpe,[3] at 293.15 K. As can be seen in Table 3 and Figure 2, the ternary system also shows an endothermic behavior in the whole range of composition, and the maximum $H^E_{m,123}$ is 917 J·mol^{-1}. No data of the same system were found in the literature for comparison.

ANN Algorithm. ANN algorithms are known to be effective to model complex systems. These models have large numbers of computational units connected in a massively parallel structure and do not require an explicit formulation of the mathematical or physical relationships of the handled problem.[2,10] The ANNs are first subjected to a set of training data consisting of input data together with corresponding outputs. 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 it uses to make predictions for new inputs.[2,10]

Feed-forward neural networks are the most frequently used and are designed with one input layer, one output layer, and hidden layers.[2,10] The number of neurons in the input and output layers is equal to the number of inputs and outputs, respectively.[2,10] The accuracy of model representation depends on the architecture and parameters of the neural network.[2,10]

To develop the ANN, the data sets are generally subdivided into three groups corresponding to the following three steps:

Table 6. Comparison between Measured and Calculated/Predicted Values of Excess Molar Enthalpies, $H_{m,123}^E$/J·mol^{-1}, for the Ternary Mixture of Cyclohexane (1) + Tetrahydropyran (2) + Piperidine (3) at 308.15 K and Atmospheric Pressure

x_1	x_2	x_3	experimental $H_{m,123}^E$	calculated $H_{m,123}^E$ (using the Cibulka equation)[9]	100 AD[a] (using the Cibulka equation)[9]	$H_{m,123}^E$ (using ANN)	100 AD[b] (using ANN)
mole fraction	mole fraction	mole fraction	J·mol^{-1}	J·mol^{-1}		J·mol^{-1}	
0.179	0.717	0.104	389	376	3.3	386	0.8
0.160	0.639	0.201	446	439	1.6	448	0.4
0.139	0.557	0.303	476	482	1.3	489	2.7
0.120	0.480	0.401	502	498	0.8	504	0.4
0.100	0.399	0.501	488	484	0.8	492	0.8
0.079	0.320	0.603	434*	438	0.9	449	3.5
0.060	0.239	0.701	385	365	5.2	378	1.8
0.040	0.161	0.799	288	266	7.6	278	3.5
0.450	0.449	0.101	695	667	4.0	678	2.4
0.392	0.391	0.217	753	767	1.9	760	0.9
0.350	0.350	0.301	791	799	1.0	784	0.9
0.301	0.300	0.399	777	796	2.4	781	0.5
0.250	0.250	0.500	726	746	2.8	743	2.3
0.200	0.200	0.600	642*	654	1.9	666	3.7
0.150	0.150	0.700	531	526	0.9	544	2.4
0.099	0.099	0.802	384	367	4.4	377	1.8
0.719	0.180	0.101	641	643	0.3	668	4.2
0.560	0.140	0.300	891	871	4.6	885	3.1
0.480	0.120	0.400	917	923	0.7	903	1.5
0.399	0.100	0.501	890	878	1.3	862	3.1
0.320	0.080	0.600	792	777	1.9	779	1.6
0.240	0.060	0.700	632	628	0.6	641	1.4
0.160	0.040	0.800	457*	445	2.6	449	1.8
0.080	0.020	0.900	243	233	4.1	223	8.2

[a] ADD using Cibulka equation 9: 2.4 %. All of these data were used for developing the Cibulka equation.[9] [b] ADD using ANN:[10] 1.9 %. All of these data were used for developing the ANN except for the star-marked data, which were used for validation.

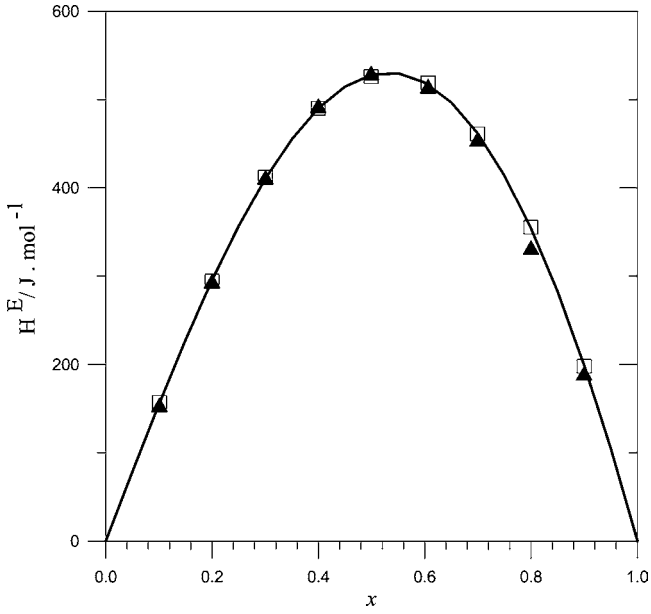

Figure 3. Excess molar enthalpy H^E for (x) cyclohexane + (1 − x) tetrahydropyran binary mixtures at 308.15 K.[2] □, experimental data; ▲, ANN results. Solid line: calculated using Redlich−Kister equation.[8]

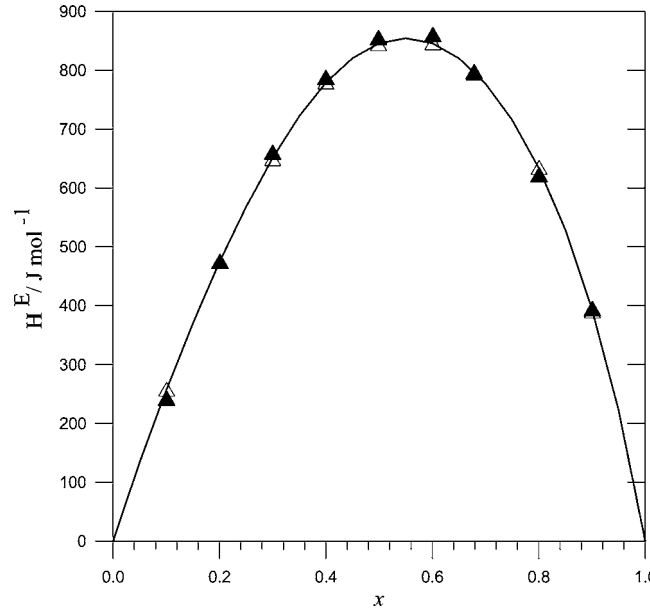

Figure 4. Excess molar enthalpy H^E for (x) cyclohexane + (1 − x) piperidine binary mixture at 308.15 K and atmospheric pressure. △, experimental data; ▲, ANN results. Solid line: calculated using Redlich−Kister equation.[8]

training, testing, and validation.[2,10] 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 the optimization algorithm, until it correctly emulates the input/output mapping, by minimizing the average root-mean-square error.[2,10] The testing set is used during the adjustment of the network's synaptic weights to evaluate the algorithm's 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 model after the fitting process.[2,10]

Table 7 reports a summary of the feed-forward ANN used in this work along with the number of neurons, hidden layers, experimental data, and type of activation function. Tables 2 and 6 compare the measured and calculated/predicted values of excess molar enthalpies for the binary and ternary mixtures of cyclohexane, tetrahydropyran, and piperidine. For more clarity, the comparison was also given in a graphical way by plotting, along with the experimental and ANN calculated/predicted excess enthalpy data in Figure 2, for the ternary system, and in Figures 3, 4, and 5, for the binary systems. Considering that not all of the experimental data in Tables 2 and 6 were used to develop the ANN, the ANN[10] can be regarded a useful tool for

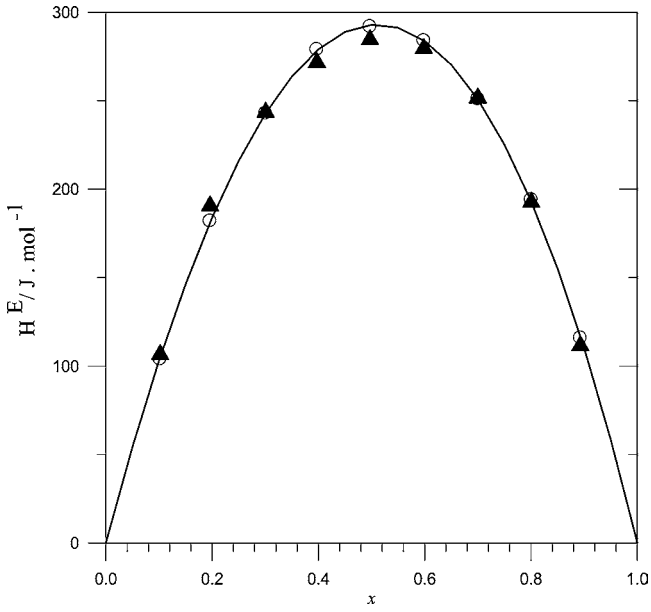

Figure 5. Excess molar enthalpy H^E for (x) tetrahydropyran + (1 − x) piperidine binary mixtures at 308.15 K. ○, experimental data; ▲, ANN results. Solid line: calculated using Redlich−Kister equation.[8]

Table 7. Number of Neurons, Hidden Layers, Experimental Data, and the Type of Activation Function Used in This Algorithm[a]

layer	number of neurons
1	2
2	5
3	1

[a] Number of hidden layers = 1; number of data used for training (and testing) = 47; number of data used for validation = 7; type of activation function: tangent sigmoid; input neurons: mole fractions of cyclohexane (1) and tetrahydropyran (2); output neuron: $H^E_{m,123}$/(RT). In developing the ANN, three data corresponding to $H^E_{m,123}$ = 0 for pure compounds were also considered.

modeling these systems with encouraging results with respect to the Cibulka equation,[9] as shown in Table 6.

Conclusions

We reported experimental excess molar enthalpy data for the binary and ternary mixtures of cyclohexane, tetrahydropyran, and piperidine at 308.15 K and atmospheric pressure, which were measured using a Calvet microcalorimeter. All of the binary systems are formed endothermally and show symmetric H^E behavior. The binary piperidine-containing mixture has the largest H^E value with cyclohexane and the lowest H^E value with tetrahydropyran. The ternary system also shows an endothermic behavior in the whole range of composition with a maximum $H^E_{m,123}$ value equal to 917 J·mol^{-1}. The Redlich−Kister equation[8] was used to correlate the binary H^E data, while the Cibulka equation[9] was employed to correlate the ternary $H^E_{m,123}$ data. A feed-forward ANN algorithm[2,10] was then used to model satisfactorily the above-mentioned experimental data with respect to the Cibulka equation.[9] Furthermore, the ANN[10] presented in this work has shown good predictive power for both ternary and binary systems.

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Supporting Information Available:

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