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Dedicated to the memory of
Professor Cristofor I. Simionescu (1920–2007)

PREDICTION OF EXCESS THERMODYNAMIC PROPERTIES FROM EXPERIMENTAL REFRACTIVE INDEX OF BINARY MIXTURES 2. ARTIFICIAL NEURAL NETWORK MODELLING

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This paper introduced new applications of the artificial neural networks (ANNs) to prediction excess molar volumes from experimental refractive index of binary mixtures. Densities (ρ) and refractive index (n) for water – propionic acid, toluene – propionic acid, water – n-propanol and toluene – n-propanol mixtures have been measured over the whole composition range at normal atmospheric pressure and different temperatures. The excess molar volumes V^E were calculated. The most common type of neural network, feed-forward network with multiple layers, has been used. The input variables were system type, temperature, molar fraction and refractive index of the solution and the output variable was the solution excess molar volume. Good results are obtained both in training and validation phases, emphasizing the generalization capabilities of the neural models.

INTRODUCTION

This paper, as part of a continuing study in our laboratory, presents experimental densities and refractive index for binary mixtures.[1-5] Experimental measurements of these properties for binary mixtures have gained much importance in many chemical industries and engineering disciplines.

The experimental values obtained for the various physical properties may be used for establishing other parameters as well, characterizing the binary mixtures.[6] In order to achieve this goal, one should use theoretical, empirical or half-empirical methods.[7] Many of these methods are based on relations which are well accounted for from the physical viewpoint concerning the dependence between the molecular and the macroscopic parameters.

Based on the easiness of determination of the refractive index, as well as the rather good measurement accuracy and the low material consumption, this parameter can be used in establishing theoretical or empirical correlations with other properties, more difficult to determine, like density dielectric permittivity and surface tension. Efforts in this area have included correlation of refractive index with density, surface tension and dielectric permittivity, the use of several well-known mixing rules for refractive index, *n*, that allow more or less accurate prediction[8-10] of excess molar volume V^E.[11,12] In this work, we have tested different artificial neural networks in order to predict the excess molar volume according to the data on the refractive indices.

In recent years, artificial neural networks (ANNs) have become a powerful tool for chemical applications. The use of ANNs ranges from interpreting sequences of DNA and predicting molecular properties to the classification and identification of compounds from spectral data. The wide applicability of ANNs stems from their flexibility and ability to model non-linear systems without prior knowledge of an empirical model. This gives ANNs an advantage over traditional fitting methods for some chemical applications. Determining of the physical properties of organic compounds based on their structures, quantitative

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structure-property relationship, is a major research subject in computational chemistry. Using ANNs, many authors have discussed the estimation of compound properties based on topological indices with great success.[13-15] The normal boiling point, density and refractive index of alkene compounds with the number of carbons atoms varying from 4 to 20 have been successfully used to train an ANNs by Zhang R. et al.[13] Good prediction results were obtained.

In this work, the new application of ANNs to the prediction of excess thermodynamic properties of the binary mixtures is considered. A general modeling methodology based on ANNs is presented and successfully applied to the estimation of some properties.

EXPERIMENTAL

The liquids involved in the experiments were of analytical purity and preparation of the solutions was performed by weighing, with an accuracy of ±0.0001 g.

The densities of the pure compounds and of the binary mixtures were experimentally determined at various temperatures of 292.15, 299.15, 305.15, 311.15 and 317.15 K. The values of excess molar volumes, V^E, were calculated from the experimental data according to the following equations:

$$V^E = V_m - \sum_{i=1}^{2} V_i x_i \quad (1)$$

where x_i, and V_i represent the mole fraction and molar volume of the i pure component of the mixture, respectively. The molar volume of the mixture, V_m, was calculated from the following equation:

$$V_m = \sum_{i=1}^{2} x_i M_i / \rho_m \quad (2)$$

where M_i is the molecular weight of component i in the mixture and ρ_m density of the mixture. The accuracies of the excess molar volumes are estimated to be ±0.002cm^3·mol^{-1}.

The densities of the pure compounds and of the binary of mixtures were measured using digital densimeters from Anton Paar, model DMA58 with a digital reading of ±0.2 kg·m^{-3}. The refractive index of the pure liquids and solutions were measured to an accuracy of the ±2·10^{-4} with RA refractometer from Melter Toledo at the wavelength of sodium D line at 589.3 nm. The refractometer was calibrated using twice distilled and deionised water, and calibration was checked every few measurements.

The values of the excess molar volumes and refractive index for the pure compounds and water-propionic acid (S=1), toluene-propionic acid (S=2), water-n-propanol (S=3) and toluene-n-propanol (S=4) solutions, for different molar fractions (X_2 – mole n-propanol or propionic acid/mole mixtures) and temperatures (T) are given in Table 1. S is the codification of the system type.

Table 1

Experimental data for the refractive index and excess molar volume V^E, cm^3·mol^{-1}

X_2	T, K									
	292.15		299.15		305.15		311.15		317.15	
	n	V^E	n	V^E	n	V^E	n	V^E	n	V^E
water-propionic acid (S=1)										
0	1.3310	0	1.3324	0	1.3319	0	1.3312	0	1.3306	0
0.1249	1.3631	-0.5572	1.3612	-0.5524	1.3596	-0.5483	1.3580	-0.5443	1.3563	-0.5403
0.2334	1.3841	-0.8021	1.3729	-0.7983	1.3711	-0.7953	1.3692	-0.7924	1.3674	-0.7896
0.3789	1.3873	-1.0040	1.3815	-0.9844	1.3793	-0.9673	1.3770	-0.9501	1.3748	-0.9327
0.5121	1.3881	-1.0370	1.3848	-1.0158	1.3826	-0.9974	1.3804	-0.9788	1.3782	-0.9599
0.6259	1.3887	-0.9275	1.3856	-0.9206	1.3835	-0.9148	1.3814	-0.9089	1.3792	-0.9030
0.7608	1.3889	-0.6849	1.3860	-0.6840	1.3836	-0.6833	1.3813	-0.6826	1.3790	-0.6819
1	1.3863	0	1.3861	0	1.3808	0	1.3783	0	1.3758	0
toluene-propionic acid (S=2)										
0	1.4958	0	1.4923	0	1.4892	0	1.4862	0	1.4832	0
0.1198	1.4859	0.4709	1.4819	0.4104	1.4784	0.3571	1.4750	0.3024	1.4715	0.2463
0.2474	1.4746	0.5305	1.4706	0.4565	1.4672	0.3914	1.4637	0.3246	1.4603	0.2562
0.3754	1.4630	0.5274	1.4588	0.4641	1.4551	0.4083	1.4515	0.3512	1.4478	0.2926
0.4955	1.4494	0.5247	1.4457	0.4697	1.4425	0.4214	1.4393	0.3718	1.4361	0.3210
0.6243	1.4352	0.4634	1.4316	0.4135	1.4285	0.3696	1.4253	0.3246	1.4222	0.2785
0.7478	1.4199	0.3114	1.4166	0.2784	1.4137	0.2494	1.4108	0.2197	1.4079	0.1893
0.8737	1.4041	0.1116	1.4008	0.1145	1.3980	0.1170	1.3952	0.1195	1.3927	0.1220
1	1.3863	0	1.3861	0	1.3808	0	1.3783	0	1.3758	0
water-n-propanol (S=3)										
0	1.3331	0	1.3324	0	1.3319	0	1.3312	0	1.3306	0
0.1236	1.3581	-0.4684	1.3563	-0.4632	1.3547	-0.4163	1.3531	-0.4547	1.3516	-0.4507
0.2451	1.3688	-0.4551	1.3668	-0.4660	1.3650	-0.4604	1.3632	-0.4859	1.3614	-0.4965
0.3673	1.3784	-0.3726	1.3751	-0.4164	1.3722	-0.4536	1.3693	-0.4944	1.3664	-0.5347
0.5028	1.3802	-0.3989	1.3773	-0.4477	1.3749	-0.4906	1.3724	-0.5345	1.3700	-0.5793

X_2	T, K									
	292.15		299.15		305.15		311.15		317.15	
	n	V^E	n	V^E	n	V^E	n	V^E	n	V^E
0.6286	1.3823	-0.4699	1.3797	-0.4858	1.3774	-0.4986	1.3752	-0.5144	1.3729	-0.5297
0.7542	1.3843	-0.4027	1.3815	-0.3947	1.3790	-0.3728	1.3766	-0.3811	1.3742	-0.3749
0.8788	1.3853	-0.1310	1.3825	-0.1463	1.3801	-0.1169	1.3776	-0.1737	1.3752	-0.1881
1	1.3858	0	1.3828	0	1.3804	0	1.3782	0	1.3760	0
toluene-n-propanol (S=4)										
0	1.4958	0	1.4923	0	1.4892	0	1.4862	0	1.4832	0
0.1184	1.4865	0.4920	1.4826	0.3824	1.4793	0.2860	1.4759	0.1872	1.4726	0.0861
0.2479	1.4759	0.3494	1.4722	0.2749	1.4690	0.2092	1.4658	0.1420	1.4627	0.0730
0.3753	1.4647	0.2853	1.4580	0.1995	1.4523	0.1240	1.4466	0.0468	1.4410	-0.0324
0.4979	1.4502	0.3418	1.4470	0.2401	1.4442	0.1506	1.4415	0.0591	1.4387	-0.0346
0.6220	1.4360	0.3473	1.4331	0.2685	1.4306	0.1992	1.4281	0.1282	1.4256	0.0557
0.7483	1.4239	0.2035	1.4178	0.1544	1.4126	0.1113	1.4074	0.0671	1.4022	0.0219
0.8758	1.4041	0.0083	1.4014	-0.0594	1.3991	-0.1186	1.3968	-0.1793	1.3945	-0.2415
1	1.3851	0	1.3825	0	1.3804	0	1.3782	0	1.3760	0

ARTIFICIAL NEURAL NETWORK MODELLING

Working principle and background

Artificial Neural Networks (ANNs) are model free formulators that have exceptional ability to perform complex multidimensional, nonlinear vector mappings and complex pattern recognition or classification. The artificial neural networks, inspired by the biological nervous system, are the simple clustering of the primitive artificial neurons. These clusters are in turn connected to one another, the topology of which also varies. Basically there are three classes of neurons. One which takes input from the user, second is the hidden neuron (which in turn forms the hidden layer) and the third one being the ones which give the output. The network function is determined mainly by the interconnections between the neurons and they need not be linear in nature. Each input to a neuron has attached to it as synaptic weight factor that determines the contribution of that connection to the overall result. In fact, the training of these ANNs takes place by the adjustment of these weights so as to get optimum result in the training set performing well with the actual test data. This is the key behind ANNs self learning and memory.

In most cases a multi-layered neural network is preferred. A backpropagation algorithm can be used to train these multilayer feed-forward networks with differentiable transfer functions, to perform function approximation, pattern association, and pattern classification. The term backpropagation refers to the computation and correction of network error with respect to weights and biases. The training of ANNs by backpropagation involves three stages: 1) the feed forward of the input training pattern, 2) the calculation and backpropagation of the associated error and 3) the adjustment of the weights. The process has to be optimized for the application and this can be achieved using various optimization strategies.

Fig. 1 shows schematic drawing of a neural network topology. As explained earlier, the neural network is made up of three basic layers: input, hidden and output layers. Each layer has different number of neurons and the behavior of the network depends on the interconnection between the elements. The hidden layers consist of one or more number of hidden neurons, depending on the application.

The set of known inputs/outputs patterns is usually divided in two subsets (training and validation subset) to train the MLP (multilayer perceptron) neural networks and quantify the prediction performance.

The training set is used to train the multilayer neural network (called also multilayer perceptron, MLP), that is to minimize the errors. In the training phase, the neural network learns the behavior of the process. The training data set contains both input patterns and the corresponding output patterns (also called target patterns). Neural training leads to finding values of connection weights that minimize differences between the network outputs and the target values. During the learning phase, the optimization scheme updates all weights and biases. The training phase is considered complete when the error of all the training patterns is less than a prespecified error criterion or a maximum number of epochs had been reached. If, after the entire set of training patterns was presented, the overall error is still unacceptable, the neural network would be

returned to the beginning of the training patterns and the process would be repeated.

The validation set is not explicitly presented during the training stage. The purpose of developing a neural model is to devise a network (set of formulae) that captures the essential relationships in the data. These formulae are then applied (after training phase) to new sets of inputs to produce corresponding outputs. This is called generalization and represents subsequent step after training - validation phase. Since a neural network is a nonlinear optimization process made up of learning and testing phases, the initial data set must be split into two subsets: one for training and one for testing. A learning algorithm should lead to a good fit to the training samples and, simultaneously, to a network that has a good generalization capability. A network is said to generalize well when the input-output relationship, found by the network, is correct for input/output patterns of validation data which were never used in training the network (unseen data).

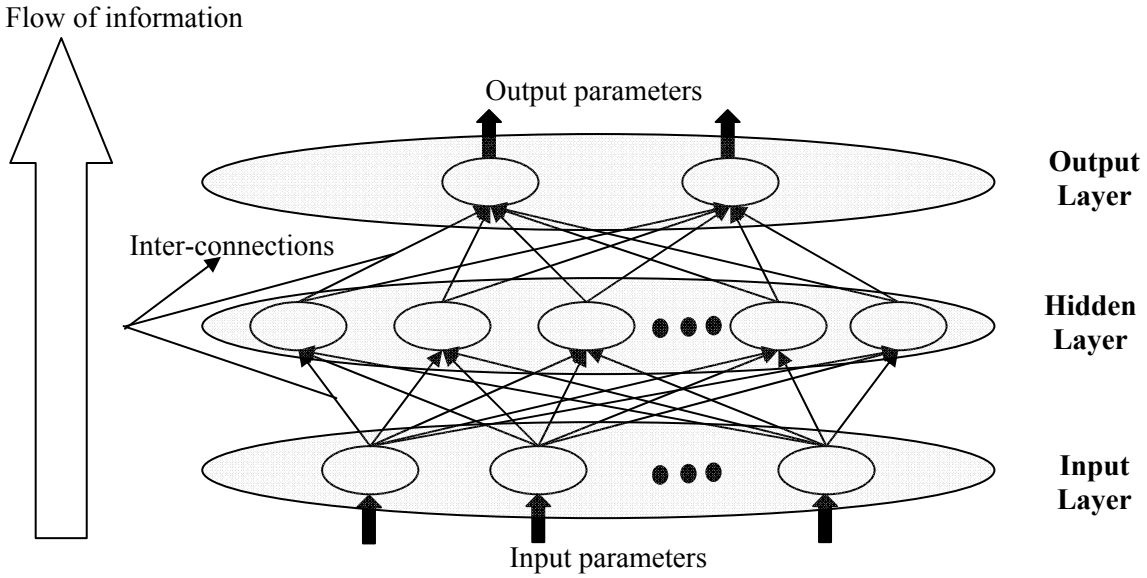

Fig. 1 – Schematic drawing of a neural network topology.

RESULTS AND DISCUSSION

In this work, the neural models are devoted to the computation of the excess molar volume (V^E) (output neuron) as function of system code (**S**), temperature (**T**), molar fraction ($\mathbf{X_2}$) and refractive index of the solution (**n**) (input neurons). Firstly, the experimental data (**175**) were split into training and validation sets, with **125** and **50** experimental points, respectively.

We developed and trained many neural networks and then we selected the best one that balances the size and the performance. After establishing input and output variables, one important problem in the developing of a neural network is the determining the network architecture, that is the number of hidden layers and the number of neurons in each hidden layer. A special software application – NeuroSolutions – was used in this paper in order to project and obtain predictions of neural networks. In this program, the following specifications are necessary: the network type (MLP in our case), the input and desired output values, the stop condition of the training, the number of hidden layers, the number of processing elements in hidden layers, the activation functions, the learning rule, the maximum number of epochs and some configuration parameters to display the neural model development. The input variables are: S, T, X_2, n and output variable is V^E. The backpropagation algorithm was used for training, the activation function was hyperbolic tangent, the learning rule was momentum and the stop criterion was associated with an imposed number of iterations (epochs) established in connection with the training error.

In this study a number of different neural network architectures of a single, two or three hidden layers and topologies of **4** inputs, **8**…**62**

hiddens and **1** outputs were used since a number of different experiments were performed (Table 2).

The best network topology was determined based upon the mean squared error (MSE), the absolute fraction variance (R^2) and mean absolute percentage error (MAPE %) on the training data.

The mean squared error was computed using the following formula:[16]

$$\mathrm{MSE} = \frac{\sum_{j=1}^{P}\sum_{i=1}^{N}(d_{ij} - y_{ij})^2}{N \cdot P} \qquad (3)$$

where P is the number of output processing elements (in this case, P = 1), N is the number of exemplars in the data set, y_{ij} is the network output for exemplar i at processing element j, and d_{ij} is the desired output for exemplar i at processing element j.

In addition, the absolute fraction variance (R^2) and mean absolute percentage error are calculated by using equations (4) and (5), respectively.

$$R^2 = 1 - \left(\frac{\sum_i (d_i - y_i)^2}{\sum_i (y_i)^2} \right) \qquad (4)$$

$$\mathrm{MAPE} = \left| \frac{d_i - y_i}{y_i} \right| \cdot 100 \qquad (3)$$

Good predictions are obtained with the following neural models: MLP (4:48:1) of a single hidden layer with 48 intermediate neurons, MLP (4:60:30:1) of a two hidden layers with 60 and 30 intermediate neurons, respectively, and MLP (4:40:30:20:1) of a three hidden layers with 40, 30 and 20 intermediate neurons, respectively. The performances of these models are marked in grey in Table 2.

Table 2

Different topologies tested for the feed forward neural networks

No.	Network topology	MSE	R^2	MAPE %
1	MLP(4:8:1)	0.0019	0.9957	9.35
2	MLP(4:12:1)	0.00057	0.9987	8.34
3	MLP(4:14:1)	0.00049	0.9989	6.84
4	MLP(4:16:1)	0.00058	0.9987	7.66
5	MLP(4:20:1)	0.00039	0.9991	5.03
6	MLP(4:24:1)	0.00030	0.9993	4.45
7	MLP(4:28:1)	0.00036	0.9992	6.05
8	MLP(4:32:1)	0.00025	0.9994	3.62
9	MLP(4:36:1)	0.00040	0.9991	5.29
10	MLP(4:40:1)	0.00021	0.9995	4.56
11	MLP(4:44:1)	0.00029	0.9993	4.38
12	MLP(4:48:1)	0.00019	0.9996	2.71
13	MLP(4:52:1)	0.00027	0.9994	4.37
14	MLP(4:56:1)	0.00028	0.9993	3.97
15	MLP(4:60:1)	0.00021	0.9995	3.82
16	MLP(4:10:10:1)	0.00074	0.9984	8.15
17	MLP(4:12:4:1)	0.00078	0.9983	9.17
18	MLP(4:20:20:1)	0.00022	0.9995	4.64
19	MLP(4:24:8:1)	0.00026	0.9994	4.85
20	MLP(4:30:30:1)	0.000075	0.9998	2.01
21	MLP(4:36:12:1)	0.00015	0.9996	2.97
22	MLP(4:50:30:1)	0.000066	0.9998	1.80
23	MLP(4:52:12:1)	0.00020	0.9995	3.92
24	**MLP(4:60:30:1)**	**0.00001**	**0.9999**	**0.56**
25	MLP(4:62:24:1)	0.00004	0.9999	1.77
26	MLP(4:40: 20:10:1)	0.00013	0.9997	3.43
27	MLP(4:40: 30:20:1)	0.000016	0.9999	0.72
28	MLP(4:45: 35:25:1)	0.000049	0.9998	2.08
29	MLP(4:50: 30:20:1)	0.000049	0.9998	1.71
30	MLP(4:40: 20:10:1)	0.000072	0.9998	2.23

Fig. 2 contains a comparison between experimental data and training results developed by ANN model of MLP(4:60:30:1). The good concordance of the two sets of data proves that the neural network have learned well the behavior of the system. The validation stage is presented in Fig. 3. The linear least square fit line, its equation and the R^2 value, are shown in Figs. 2 and 3 for the training and validation data. The generalization capability is the most important feature of a neural model, so the validation of the best neural models with one, two or three hidden layers are given in Table 3.

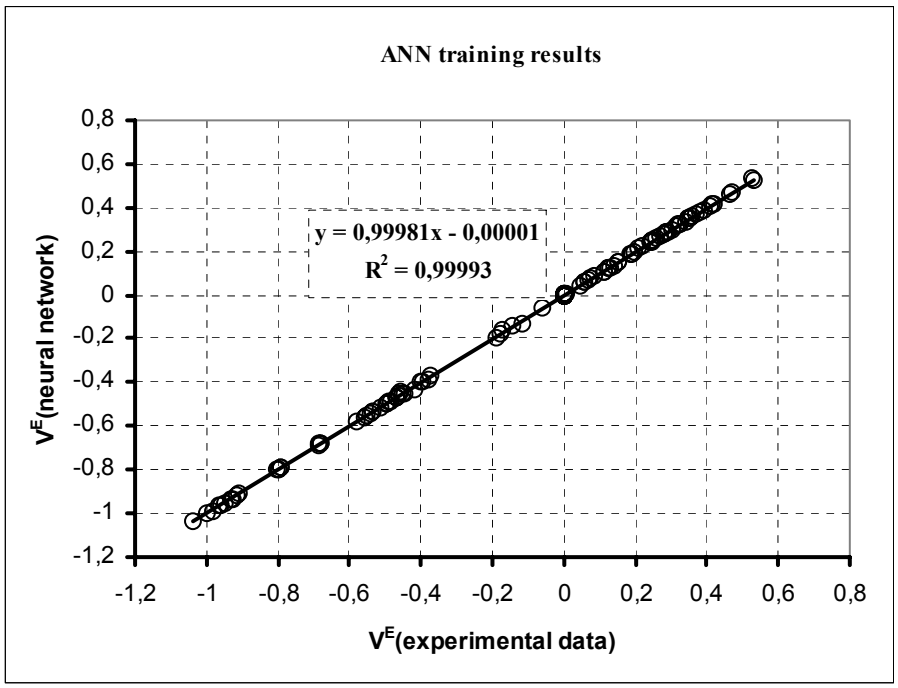

Fig. 2 – Comparison of the experimental data of V^E with the training results of MLP(4:60:30:1).

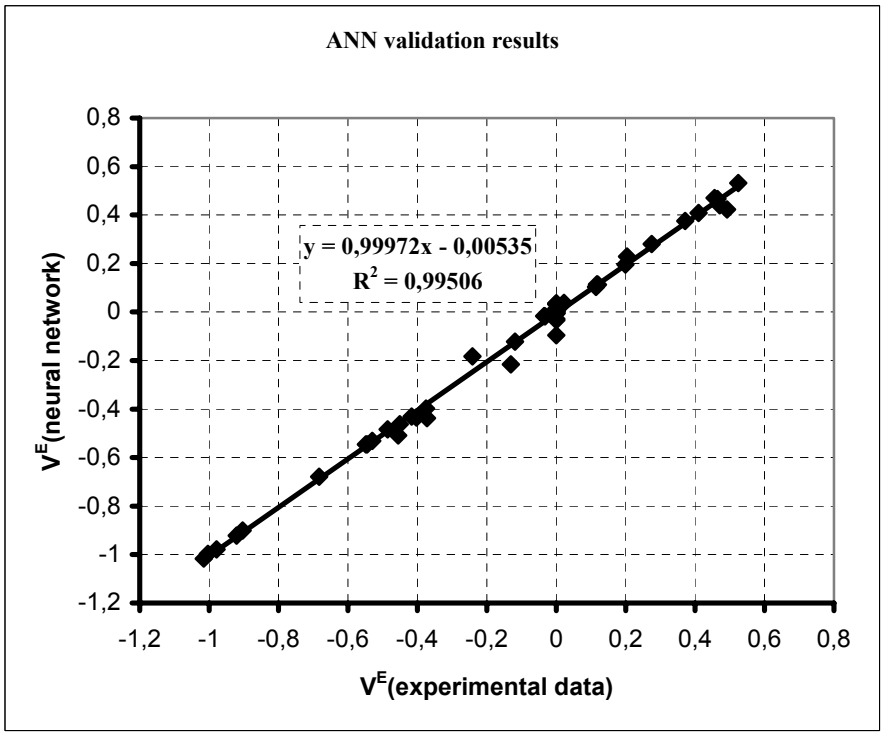

Fig. 3 – Comparison of the experimental data of V^E with the validation results of MLP(4:60:30:1).

The best predictions on validation data are obtained with MLP(4:60:30:1) neural model. Figs. 4 and 5 contain the experimental conditions (temperature and molar fraction), experimental excess molar volume and neural network predicted excess molar volume. A good agreement between the two categories of data is observed.

These results imply the fact that the designed ANN model was properly able of learning the relationship between the input and output parameters. The final selected ANN model provided satisfactory results over the whole set of values for all the four binary systems.

CONCLUSIONS

Conventional methods to predict the excess thermodynamic properties from experimental refractive index in binary mixtures involve a step by step procedure based on a complex mathematical model with large time and memory complexity. Hence the application of neural networks leads to effective route to model complex systems without much of mathematical complexities. Further advantages with neural networks are that they represent an automated learning process by the adjustment of the synaptic weights and they save the valuable experimentation time.

This work has shown the excellent capability of a neural network approach for prediction of the excess thermodynamic properties from experimental refractive index in binary mixtures. Simple architecture neural networks and simple methods of establishing the networks' structure are proposed for process modeling: feed-forward networks with a single, two or three hidden layers and reasonable number of neurons in these layers.

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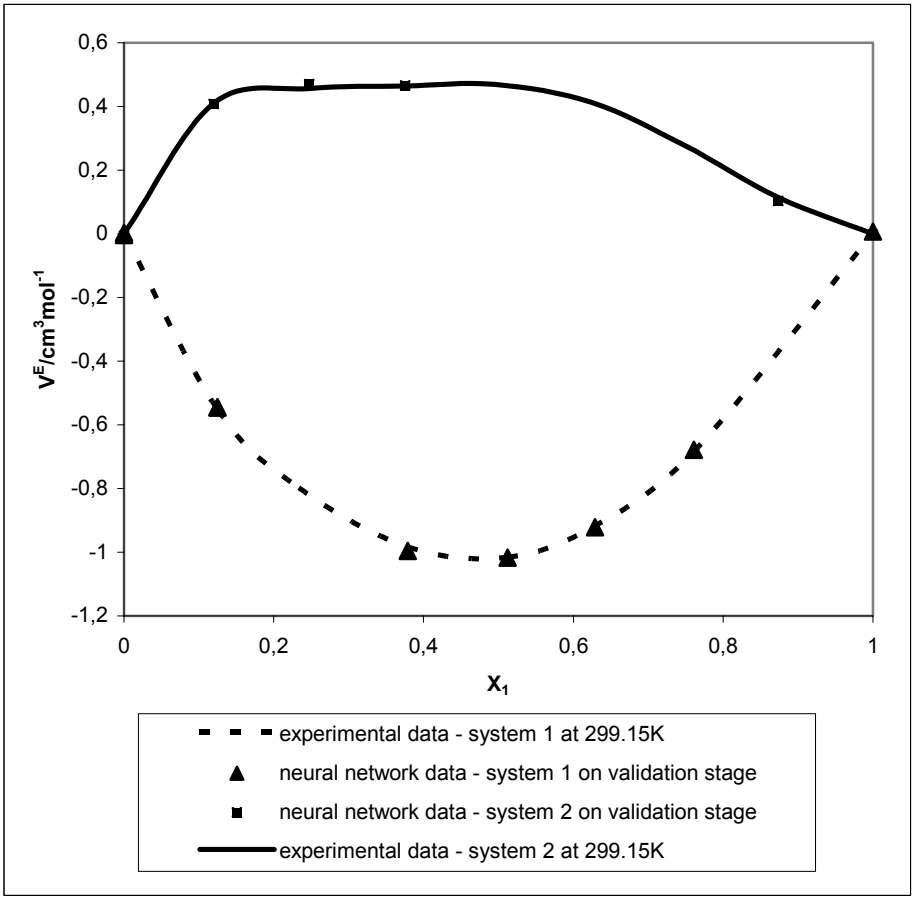

Fig. 4 – Prediction of MLP(4:60:30:1) on validation data for water –propionic acid (system 1) and toluene propionic acid mixtures (system 2).

Table 3

Validation of the best neural models of MLP type

System code (S)	X_2 (mol/mol solution)	T (K)	n	V^E (cm^3/mol)	V^E MLP(4:48:1)	MAPE%	V^E MLP(4:60:30:1)	MAPE%	V^E MLP(4:40:30:20:1)	MAPE%
1	0.6259	299.15	1.3856	-0.9206	-0.9444	0.0251	-0.9220	0.0015	-0.9274	0.0073
3	0.2451	292.15	1.3688	-0.4550	-0.4578	0.0060	-0.5089	0.1057	-0.4958	0.0821
4	0.1184	292.15	1.4865	0.4919	0.3725	0.3205	0.4228	0.1634	0.4168	0.1802
4	0.4979	317.15	1.4387	-0.0345	0.0209	2.6492	-0.0164	1.0980	-0.0046	6.4857
2	0.8737	311.15	1.3952	0.1194	0.1146	0.0426	0.1120	0.0667	0.1328	0.1003
3	0.7542	292.15	1.3843	-0.4026	-0.3924	0.0259	-0.4342	0.0726	-0.4567	0.1183
2	0.1198	299.15	1.4819	0.4104	0.3981	0.0309	0.4076	0.0067	0.3993	0.0276
1	0.1249	311.15	1.3580	-0.5442	-0.5233	0.0399	-0.5450	0.0013	-0.5351	0.0170
2	0.8737	305.15	1.3980	0.1169	0.1217	0.0392	0.1143	0.0228	0.1309	0.1066
4	0.7483	317.15	1.4022	0.0218	0.0120	0.8211	0.0371	0.4113	0.0222	0.0152
1	0.1249	305.15	1.3596	-0.5482	-0.5276	0.0390	-0.5460	0.0041	-0.5405	0.0143
2	0.3754	299.15	1.4588	0.4640	0.4786	0.0305	0.4661	0.0045	0.4730	0.0189
3	0.6286	317.15	1.3729	-0.5297	-0.5312	0.0029	-0.5310	0.0025	-0.5299	0.0004
4	0.2479	299.15	1.4722	0.2748	0.3103	0.1143	0.2805	0.0202	0.2847	0.0347
1	0.7608	305.15	1.3836	-0.6832	-0.6854	0.0031	-0.6788	0.0065	-0.6798	0.0049
4	0.7483	292.15	1.4239	0.2035	0.2490	0.1829	0.2284	0.1090	0.2193	0.0721
1	0.5121	299.15	1.3848	-1.0157	-1.0075	0.0081	-1.0172	0.0013	-1.0169	0.0011
2	0.1198	292.15	1.4859	0.4709	0.4302	0.0945	0.4395	0.0713	0.4310	0.0926
3	0.3673	299.15	1.3751	-0.4164	-0.3907	0.0657	-0.4308	0.0335	-0.4140	0.0057
4	0.8758	317.15	1.3945	-0.2414	-0.1843	0.3100	-0.1826	0.3222	-0.2021	0.1942
3	0.1236	317.15	1.3516	-0.4506	-0.4408	0.0223	-0.4626	0.0258	-0.4627	0.0261
4	0.8758	305.15	1.3991	-0.1186	-0.1104	0.0736	-0.1222	0.0293	-0.1266	0.0635
1	0.6259	317.15	1.3792	-0.9029	-0.9003	0.0029	-0.9001	0.0031	-0.8935	0.0105
1	0.3789	292.15	1.3873	-1.0040	-0.9849	0.0193	-0.9962	0.0077	-0.9963	0.0077
2	0.8737	299.15	1.4008	0.1144	0.1166	0.0185	0.1036	0.1038	0.1143	0.0006
3	0.2451	311.15	1.3632	-0.4858	-0.4813	0.0094	-0.4831	0.0056	-0.4723	0.0285
4	0.3753	317.15	1.4410	-0.0324	-0.0081	2.9792	-0.0181	0.7857	-0.0039	7.1708
1	0.5121	311.15	1.3804	-0.9787	-0.9731	0.0057	-0.9780	0.0007	-0.9771	0.0016
2	0.4955	292.15	1.4494	0.5246	0.5045	0.0399	0.5327	0.0151	0.5201	0.0086

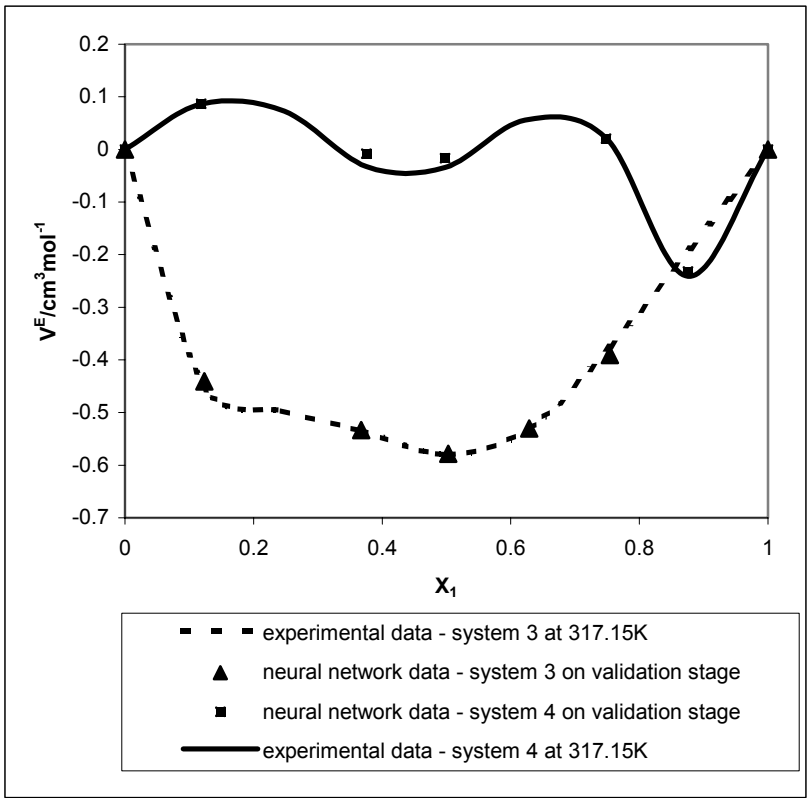

Fig. 5 – Prediction of MLP(4:60:30:1) on validation data for water –n-propanol (system 3) and toluene- n- propanol mixtures (system 4).

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