MODULAR NEURAL NETWORK MODELING FOR REFRACTIVE INDICES OF SOME BINARY SYSTEMS

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Received: 09/06/2009
Accepted after revision: 20/07/2009

Abstract: In this work, a modular neural network (MNN) model for prediction of the thermodynamic properties was established. The numbers of patterns used in this study were 175, 160 for training and 15 for validation phases. After evaluating a large number of trials with various MNN architectures, the optimal model was a network with two hidden layers, with 8 neurons in the first and, also, in the second layer, the slabs having the possibility of three activation functions and a jump connection. The mean percentage errors obtained with the best MNN model was of 0.0817%. This result implies that the designed MNN model was properly capable of learning the relationship between the input and output parameters and also confirms that the neural network was able to reproduce simultaneously more that one system, unlike traditional models where one mathematical model was required for each system.

Keywords: *refractive indices, prediction, modular neural networks*

INTRODUCTION

This paper, as part of a continuing study in our laboratory, presents experimental densities, refractive indices, viscosities and the surface tensions for the binary mixtures [1-5]. Experimental measurements of these properties for binary mixtures have gained much importance in many chemical industries and engineering disciplines.
In the present work, refractive indices, *n* for the binary systems: water – propionic acid, toluene - propionic acid, water – *n*-propanol and toluene – *n*-propanol at different temperatures, and normal atmospheric pressure, are presented. This property provides important information about intermolecular interactions in liquid mixtures because is indirectly related to the dispersion forces between molecules [6-8].
For years, many researchers made good use of various methods for predicting properties of concrete. One of these methods is artificial neural networks (ANN). Mehlman, Wentzell and McGuffin [9] used ANN to predict simple physical properties of several quaternary solvent systems based on training data from binary systems. Lisa, Curteanu and coworkers introduced new applications of the artificial neural network (ANN) to predict excess molar volumes from experimental refractive index of binary mixtures [10] and to predict density or viscosity of some binary mixtures [11, 12].
In this paper we used modular neural network modeling for prediction of refractive indices of some binary systems.

MATERIALS AND METHODS

Data bases

The fluids used for the experiments, *n*-propanol, propionic acid and toluene, have a p.a. purity. The samples were prepared using a Mettler Toledo XP 105 balance with an accuracy of $\pm 10^{-4}$ g, covering the whole composition range of the mixtures. The accuracy in the determination of the mole fraction of the samples used was $\pm 10^{-4}$. The refractive indices were measured with Kruss (digital ABBE refractometer) and a thermostat Lauda E100 designed to maintain a constant temperature with a precision of 0.01 ºC. This device was calibrated by using double-distilled water. The experimental refractive indices, *n*, obtained from the measurements of the pure compounds and binary mixtures at all the investigated temperature are summarized in table 1.

Neural Network Modeling

The NeuroSolution 4.0 program was used for all neural network operations. Unless otherwise stated, the modular neural networks with one, two or three hidden layers were generated. Modular feed-forward networks (MNN) are a special class of multilayer perceptrons which process their input using several parallel multilayer perceptrons, and then recombine the results (figure 1).
In this work, modular multilayer feed forward neural networks, trained with a back propagation learning algorithm, were used to predict the refractive indices of some binary mixtures. Different neural network architectures (figure 1) were automatically built as implemented in the *NeuroSolution* program.

Table 1. *The experimental refractive indices values, n*

Systems	X_2	T, K				
		292.15	**299.15**	**305.15**	**311.15**	**317.15**
water – propionic acid	0.0000	1.3331	1.3324	1.3319	1.3312	1.3306
	0.1249	1.3631	1.3612	1.3596	1.3580	1.3563
	0.2334	1.3841	1.3729	1.3711	1.3692	1.3674
	0.3789	1.3873	1.3815	1.3793	1.3770	1.3748
	0.5121	1.3881	1.3848	1.3826	1.3804	1.3782
	0.6259	1.3887	1.3856	1.3835	1.3814	1.3792
	0.7608	1.3889	1.3860	1.3836	1.3813	1.3790
	1.0000	1.3863	1.3861	1.3808	1.3783	1.3758
toluene – propionic acid	0.0000	1.4958	1.4923	1.4892	1.4862	1.4832
	0.1198	1.4859	1.4819	1.4784	1.4750	1.4715
	0.2474	1.4746	1.4706	1.4672	1.4637	1.4603
	0.3754	1.4630	1.4588	1.4551	1.4515	1.4478
	0.4955	1.4494	1.4457	1.4425	1.4393	1.4361
	0.6243	1.4352	1.4316	1.4285	1.4253	1.4222
	0.7478	1.4199	1.4166	1.4137	1.4108	1.4079
	0.8737	1.4041	1.4008	1.3980	1.3952	1.3923
	1.0000	1.3863	1.3861	1.3808	1.3783	1.3758
water – *n*-propanol	0.0000	1.3331	1.3324	1.3319	1.3312	1.3306
	0.1236	1.3581	1.3563	1.3547	1.3531	1.3516
	0.2451	1.3688	1.3668	1.3650	1.3632	1.3614
	0.3673	1.3784	1.3751	1.3722	1.3693	1.3664
	0.5028	1.3802	1.3773	1.3749	1.3724	1.3700
	0.6286	1.3823	1.3797	1.3774	1.3752	1.3729
	0.7542	1.3843	1.3815	1.3790	1.3766	1.3742
	0.8788	1.3853	1.3825	1.3801	1.3776	1.3752
	1.0000	1.3851	1.3825	1.3804	1.3782	1.3760
toluene – *n*-propanol	0.0000	1.4958	1.4923	1.4892	1.4862	1.4832
	0.1184	1.4865	1.4826	1.4793	1.4759	1.4726
	0.2479	1.4759	1.4722	1.469	1.4658	1.4627
	0.3753	1.4647	1.458	1.4523	1.4466	1.441
	0.4979	1.4502	1.447	1.4442	1.4415	1.4387
	0.6220	1.436	1.4331	1.4306	1.4281	1.4256
	0.7483	1.4239	1.4178	1.4126	1.4074	1.4022
	0.8758	1.4041	1.4014	1.3991	1.3968	1.3945
	1.0000	1.3851	1.3825	1.3804	1.3782	1.376

In this study, there were a total 175 patterns, each with 4 components (sample code, molar fraction X_2, temperature and refractive indices) which were used for training and validation the neural networks. Three of the components were the input variables, whereas the last components were the output variables.

The 175 patterns were randomly divided into 160 and 15 data set for the training and validation the neural networks, respectively. The first (learning) partition was used to perform the training of the network. In order to estimate the performance of the trained network on new data not used in the training, a second validation partition was used.

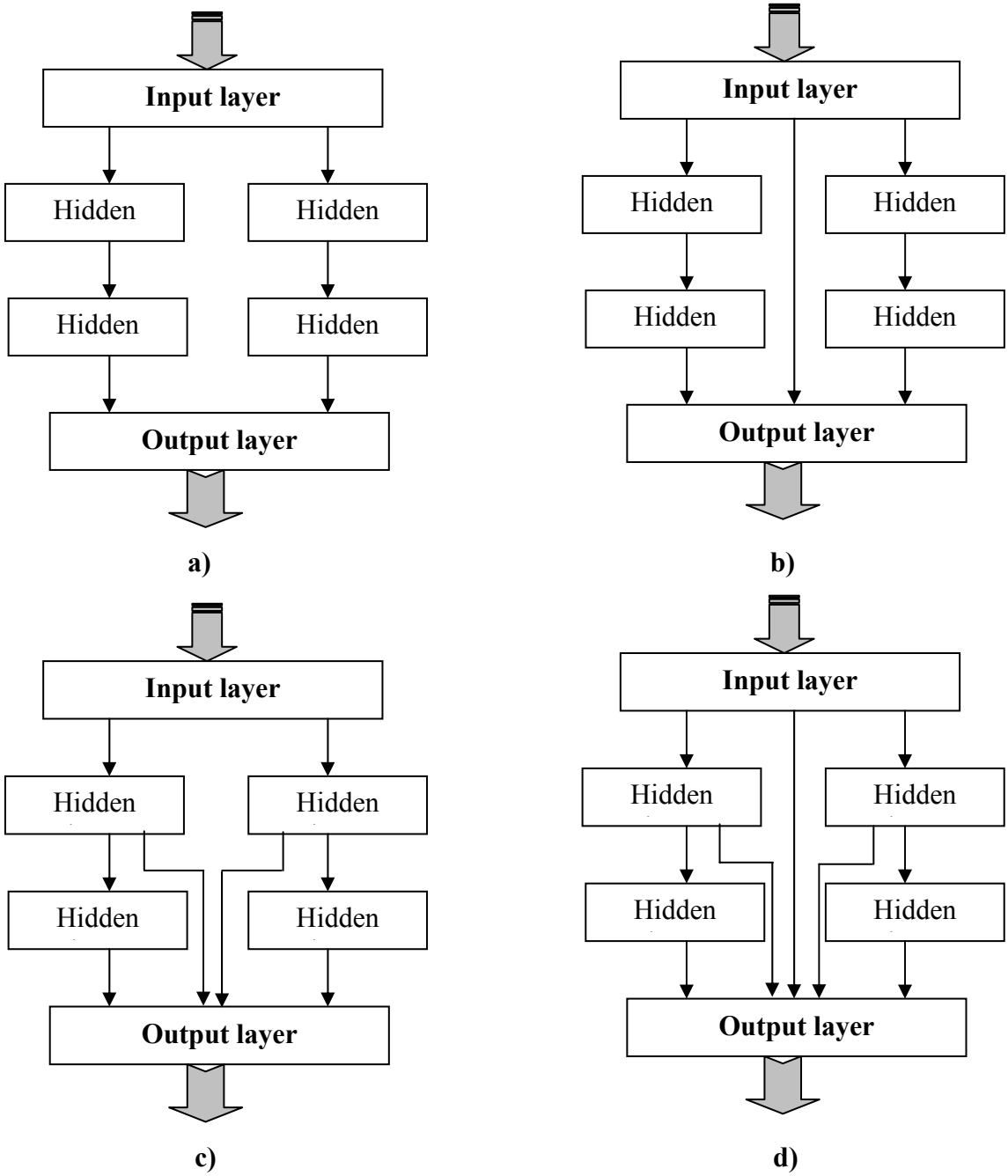

Figure 1. *Modular Neural Network architectures based on the standard backpropagation algorihm:*
a) Two hidden slabs with possibility of two activation functions;
b) Two hidden slabs with possibility of three activation functions, and a jump connection;
c) Two hidden slabs with possibility of four activation functions and a jump connection between hidden layer, and output layer;
d) Two hidden slabs with possibility of five activation functions and a jump connection between hidden layer and output layer.

RESULTS AND DISCUSSIONS

The first step of the study was to establish the "input" parameters for the ANN algorithms. The parameters considered as inputs of the neural model were: sample code (S), molar fraction (X_2) and temperature (T). The value of the refractive indices, n is the output variable for neural model.

In this study a number of different neural network architecture of a single, or two and three hidden layer and topology of 3 inputs, 4…16 hidden neurons and 1 output were used since a number of different experiments were performed (table 2). The best network topology was determined based upon the mean squared errors (MSE) on the training data and percent error E_p (%).

The mean squared error was computed using the following formula [13, 14]:

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

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 the mean absolute percentage error are calculated by using equations (2) and (3), respectively.

$$r^2 = 1 - \left(\frac{\sum_i (d_i - y_i)^2}{\sum_i (y_i)^2} \right) \tag{2}$$

$$E_p = \left| \frac{d_i - y_i}{y_i} \right| \cdot 100 \tag{3}$$

For al the built neural models, the number of training epochs was 100,000.

In order to determine the optimal number of hidden neurons in hidden layers and the number of hidden layers, training was performed for 3:x:y:z:1 architectures. Firstly, the number of hidden neurons was chosen equal to one-half the total number of inputs plus outputs. Then the numbers of nodes were increased by an increment of 4 in each step, to improve the model performance. Among the various structures, models of good performance were produced by the 3:12:12:1 type a), 3:8:8:1 type b), 3:16:16:1 type c) and 3:16:16:1 type d) (Table 2). The aim of this work was to obtain an MNN model with a minimal dimension and minimum errors in training and validation phases.

Figure 2 shows the testing results of the final trained the best MNN model. On the y-axis, the network output is represented and compared with the experimental data (on the x-axis). The scatter plot showed that the predictive capability of the neural network was satisfactory and the data points were well concentrated around the ideal unity-slope line selected. The linear adjustment between measured and estimated values gives almost a slope practically equal to 1.

The built modular neural models were tested during the validation stage by using 15 pairs of input-output data that were not employed during the training stage.

Table 2. Performances of the modular neural networks in the training phase

No.	Modular Neural Network type	Topology	MSE	r^2	E_p (%)
1.	(**a**)	MNN(3:4:1)	0.0019	0.995	0.235
2.	(**a**)	MNN(3:8:1)	0.0014	0.996	0.198
3.	(**a**)	MNN(3:12:1)	0.0019	0.9959	0.233
4.	(**a**)	MNN(3:4:4:1)	0.0003	0.9993	0.091
5.	(**a**)	MNN(3:8:8:1)	0.0002	0.9994	0.072
6.	(**a**)	MNN(3:12:12:1)	0.0001	0.99958	0.060
7.	(**a**)	MNN(3:16:16:1)	0.0002	0.99949	0.071
8.	(**a**)	MNN(3:4:4:4:1)	0.0057	0.98824	0.344
9.	(**a**)	MNN(3:8:8:8:1)	0.0018	0.996	0.222
10.	(**a**)	MNN(3:12:12:12:1)	0.0015	0.9968	0.198
11.	(**b**)	MNN(3:4:1)	0.0054	0.988	0.348
12.	(**b**)	MNN(3:8:1)	0.0040	0.9914	0.309
13.	(**b**)	MNN(3:12:1)	0.0046	0.990	0.350
14.	(**b**)	MNN(3:16:1)	0.0045	0.990	0.330
15.	(**b**)	MNN(3:4:4:1)	0.0007	0.998	0.146
16.	(**b**)	MNN(3:8:8:1)	0.0002	0.99953	0.066
17.	(**b**)	MNN(3:12:12:1)	0.0004	0.99947	0.073
18.	(**b**)	MNN(3:4:4:4:1)	0.0058	0.988	0.345
19.	(**b**)	MNN(3:8:8:8:1)	0.0035	0.9926	0.284
20.	(**b**)	MNN(3:12:12:12:1)	0.0019	0.99607	0.214
21.	(**c**)	MNN(3:4:1)	0.0019	0.9958	0.235
22.	(**c**)	MNN(3:8:1)	0.0014	0.9968	0.198
23.	(**c**)	MNN(3:12:1)	0.0019	0.9959	0.233
24.	(**c**)	MNN(3:16:1)	0.0018	0.9960	0.223
25.	(**c**)	MNN(3:4:4:1)	0.0011	0.9975	0.173
26.	(**c**)	MNN(3:8:8:1)	0.0002	0.99950	0.068
27.	(**c**)	MNN(3:12:12:1)	0.0002	0.99952	0.067
28.	(**c**)	MNN(3:16:16:1)	0.0002	0.99953	0.067
29.	(**c**)	MNN(3:4:4:4:1)	0.0029	0.99385	0.281
30.	(**c**)	MNN(3:8:8:8:1)	0.0024	0.9948	0.254
31	(**c**)	MNN(3:12:12:12:1)	0.0028	0.9939	0.263
32.	(**d**)	MNN(3:4:1)	0.0276	0.939	0.830
33.	(**d**)	MNN(3:8:1)	0.0049	0.989	0.340
34.	(**d**)	MNN(3:12:1)	0.0045	0.990	0.329
35.	(**d**)	MNN(3:16:1)	0.0045	0.9903	0.330
36.	(**d**)	MNN(3:4:4:1)	0.0007	0.9983	0.145
37.	(**d**)	MNN(3:8:8:1)	0.0003	0.9992	0.094
38.	(**d**)	MNN(3:12:12:1)	0.0003	0.99927	0.089
39.	(**d**)	MNN(3:16:16:1)	0.0002	0.99951	0.073
40.	(**d**)	MNN(3:4:4:4:1)	0.0022	0.9953	0.241
41.	(**d**)	MNN(3:8:8:8:1)	0.0019	0.9958	0.222
42.	(**d**)	MNN(3:12:12:12:1)	0.0022	0.9952	0.228

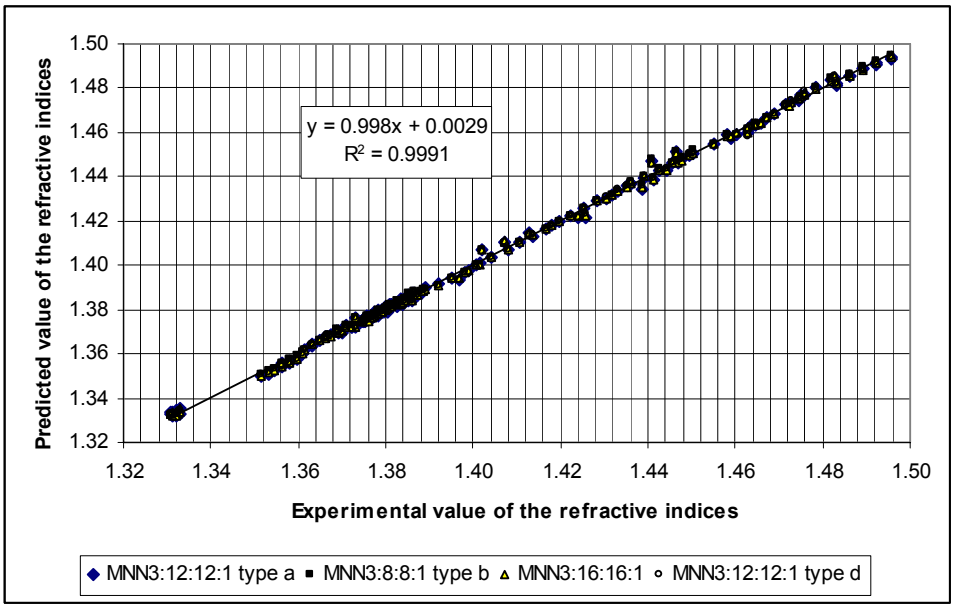

Figure 2. *Correlation between the experimental data and the predicted values of the MNN model for refractive indices in the training stage*

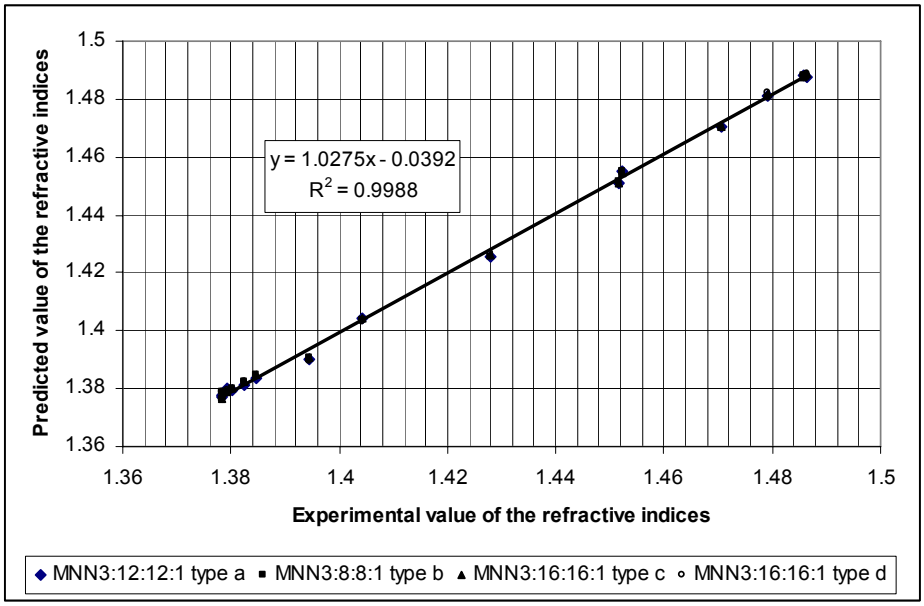

Figure 3. *Correlation between the experimental data and the predicted values of the MNN model for prediction of refractive indices in the validation stage*

Figure 3 presents comparatively the experimental values and the predictions obtained during the validation stage for the modular neural networks with the best performances of each type. The mean percentage error obtained with the 3:12:12:1 type a) modular network was 0.094%, for the 3:8:8:1 type b) was 0.0817%, for 3:16:16:1 type c) was 0.0855% and for 3:16:16:1 type d) was of 0.14038%.

These results imply that the designed MNN model was properly capable of learning the relationship between the input and output parameters. These results also confirm that a

properly trained neural network was able to produce simultaneously more that one system, unlike traditional models where one mathematical model was required for each system [15, 16]. The final selected MNN model provided satisfactory results over the whole set of values of the refractive indices.

CONCLUSIONS

The main quality indicator of a neural network is the generalization capacity, which means its ability to predict accurately the output to validation (unseen) data. In this study, we have benefited from this feature of ANN. The results show that the predicted refractive indices are in good agreement with those of the experimental work. Therefore, the modular neural network models can be used to predict accurately this property. The mean percentage errors obtained with the best MNN model (3:8:8:1 type b) was of 0.0817%.

Acknowledgements: This research occurred in the framework of the Project PN II ID_600/ no.64/1.10.2007, for which the authors acknowledge financing authority.

REFERENCES

1. Lisa, G., Lisa, C.: *Rev. Roum. Chim*., **2007**, **52**, 647.
2. Lisa, G., Bolat, G., Popa, I.M.: *Rev. Roum. Chim.*, **2005**, **50**, 699.
3. Lisa, G., Lisa, C., Apreutesei, D., Barabulică, I., Gâlea, D.: *Rev. Roum. Chim.*, **2004**, **49**, 911.
4. Lisa, G.: *Rev. Chim.*, **2004**, **55**, 121.
5. Bolat, G., Ortega, J., Lisa, G., Popa, I.M.: *Buletinul Institutului Politehnic Iaşi,* **2006**, **LII**, 30.
6. Lugo, L., Comunas, M.J.P., Lopez, E.R., Fernandez, J.: *J. Thermal. Anal. Cal*., **2007**, **87**, 179.
7. Zadjia, A.: *J. Chem. Thermodynamics*, **2006**, **38**, 201.
8. Resa, J.M., Gonzalez, C., Juez, M., Ortiz de Landuluce, S., *Fluid Phase Equilibria*, **2004**, **217**, 175.
9. Mehlman, S.A., Wentzell, P.D., McGuffin, V.L.: *Analytica Chimica Acta*, **1998**, **371**, 117.
10. Lisa, G., Curteanu, S., Lisa, C.: *Rev. Roum.Chim.*, **2008**, **53**, 859.
11. Lisa, G., Curteanu, S.: *Studii şi Cercetări Ştiinţifice – Chimie şi Inginerie Chimică, Biotehnologii, Industrie Alimentară*, **2005**, **VI**(1), 65.
12. Curteanu, S., Lisa, G.: *Buletinul Institutului Politehnic Iaşi*, **2004**, **XL**, 96.
13. Razmi-Rad, E., Ghanbarzadeh, B., Mousavi, S.M., Emam-Djomeh Khazaein, J.: *Journal of Food Engineering*, **2007**, **81**, 728.
14. Topcu, I.B., Saridemir, M., *Computational Materials Science*, **2007**, **41**, 117.
15. Lisa, G., Apreutesei, D., Lisa, C., Aioanei, R.: *Conferinţa Internaţională de Ştiinţe Aplicate,* Chimie şi Inginerie Chimică CISA, **2009**.
16. Lisa, G., Huhurez, C., Matran, R.M., Lisa C.: *Environmental Engineering and Management Journal*, **2008**, **7**, 309.