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Viscosity Calculation at Moderate Pressure for Nonpolar Gases via Neural Network

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Abstract: A new method, based on Artificial Neural Networks (ANN) of Multi-Layer Perceptron (MLP) type, has been developed to estimate the viscosity at moderate pressure for pure nonpolar gases over a wide range of temperatures. An ANN was trained, using four physicochemical properties: Molecular weight (M), boiling point (T_b), critical Temperature (T_c) and critical Pressure (P_c) combined with absolute Temperature (T) as its inputs, to correlate and predict viscosity. A group of 52 nonpolar gases were used to train and test the performance of the ANN. The viscosity and input data for each individual gas was compiled on average at fifty different temperatures, ranging from the boiling points for each of the chosen gases to 1100 K. The maximum absolute error in viscosity, predicted by the ANN, was approximately 15%.

Key words: Artificial neural networks, nonpolar gases, physicochemical properties, viscosity

INTRODUCTION

Viscosity is indicated as being one of the most significant transport properties because it's related to the movement of molecular agitation. That means, the molecular transport of momentum is the corollary of the fluid forces of cohesion. Viscosity is required by chemical engineers involved in reactor applications, heat and mass transfer.

Accurate experimental measurements of viscosity, particularly at very high and/or very low temperature, are laborious and complex task. On the other hand, kinetic theory of gases made it possible to establish formulas for the calculation of gases viscosity of which have recently gained a wider acceptance, but very difficult to use because it comprises several parameters, which are often not easy to acquire.

After bibliographical synthesis, some empirical models were recapitulated among the most used in the calculation of nonpolar gases viscosity (Table 1).

At present, there is a considerable empirical models for estimating gases viscosity which have some limited success (Reid *et al*., 1977; Zhao, 1997; Adel Elsharkawy, 2004; Scalabrin *et al*., 2002; Maloka, 2005).

Table 1: Theoretical models for nonpolar gases viscosity calculation (Reid *et al*., 1977)

	Models
Chapman-d'Enskog	$\eta = 26.69\times10^{-6}\dfrac{\sqrt{MT}}{\sigma^2\Omega_v}$
Golubev	$\eta = \begin{cases}\eta_c^* T_r^{0.965} \text{ for } T_r<1\\ \eta_c^* T_r^{0.71+0.29/T_r} \text{ for } T_r>1\end{cases}$ $\eta_c^* = \dfrac{3.5M^{1/2}P_c^{2/3}}{T_c^{1/6}}$
Thodos	$\eta\xi = 4.610T_r^{0.618} - 2.04\exp(-0.449T_r) + 1.94\exp(-4.058T_r) + 0.1$ $\xi = T_r^{1/6}M^{-1/2}P_c^{-2/3}$
Reichenberg	$\eta = \dfrac{a^*T_r}{\left[1+0.36T_r(T_r-1)\right]^{1/6}}$ $a^* = \dfrac{M^{1/2}T_c}{\sum_i n_i C_i}$

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These approaches are typically limited to narrow ranges of compounds across narrow ranges of temperature.

The advances in Artificial Neural Networks (ANN) have provided a tool that may be used to avoid the shortcomings involved in empirical methods. Indeed, the ANN can approach uniformly any sufficiently regular bounded function, with an arbitrary precision, in a limited domain of its variables space, with faster speed of information processing, learning ability, fault tolerance and multi-output ability.

Although there are a few reports (Homer *et al*., 1999; Lee *et al*., 1994; Chang *et al*., 1995; Adel Elsharkwy and Gharbi, 2001) of using ANNs in the prediction of physicochemical properties, these reports have generally been restricted to liquid rather than gases. The present research presents the findings of a programme of work devoted to the application of ANNs gases viscosity.

PROCEDURE

Data base: The pool of compounds in this study consisted of 52 nonpolar gases (Table 2), because of experimental data deficiency over wide range of temperature, the viscosity were deduced by the first correlation shown in Table 1 and corrected by some experimental viscosity data (Reid *et al*., 1977; Division Scientifique de L'AIR LIQUIDE, 1976; Gosse *et al*., 1991) every 20 K ranging from the boiling point for all the compounds to 1100 K. This resulted in each gas being studied at approximately 50 different temperatures. The viscosity data obtained consist of 2652 vectors which were divided into two sets and used separately. One set of 1989 randomly selected vectors was used to train the ANN. The remaining 664 vectors, which contained approximately a third of the data base, were used as test set for checking the predictive performance of the ANN.

Table 2: List of gases used to provide training and test data of ANN (Reid *et al*., 1977; Division Scientifique de L'AIR LIQUIDE, 1976; Gosse, Déroulède *et al*., 1991)

Gas	M, g/mole	T_b, K	T_c, K	P_c, bar
1,3-Butadiene	54.091	268.65	425.15	43.22
1-butene	56.107	266.90	419.55	39.25
2,2-Dimethyl propane	72.15	282.65	433.78	31.96
Acetylene	26.038	189.20	308.33	61.91
Argon	39.948	87.29	150.86	48.98
Boron trichloride	117.17	285.65	451.95	38.70
Boron trifluoride	67.80	172.85	260.85	49.85
Bromine	159.808	331.90	584.15	103.35
Carbon dioxide	44.01	194.60	304.28	73.825
Carbon disulfide	76.131	319.40	552.000	78.000
Carbon monoxide	28.01	81.62	132.91	34.987
Carbon tetrachloride	153.823	349.70	556.40	45.00
Carbon tetrafluoride	88.01	145.21	227.70	37.43
Chlorine	70.906	239.05	417.15	77.00
Chloropentafluoroethane	154.48	235.15	353.15	31.60
Chlorotrifluoromethane	104.47	191.65	301.93	38.60
Cis-2-butene	56.107	276.87	433.15	42.07
Cyanogen	52.035	252.000	400.15	59.40
Cyclobutane	56.107	288.66	459.95	49.65
Cyclopropane	42.08	240.35	398.30	55.79
Deuterium	4.029	23.57	38.35	16.65
Diborane (B_2F_6)	27.67	180.65	289.80	40.53
Dichlorodifluoromethane	120.93	243.37	385.15	41.15
Dichlorotetrafluoroethane	170.93	276.75	418.85	32.63
Ethane	30.069	184.47	305.42	48.839
Ethylene	28.054	169.43	282.65	50.76
fluorine	37.997	84.95	144.30	52.15
Helium-4	4.0026	4.224	5.20	2.275
Hexafluoroethane	138.02	194.95	292.85	29.80
Hydrogen	2.016	20.384	33.24	12.98
Hydrogen iodide	127.912	237.75	423.95	83.000

Table 2: Continued

Gas	M, g/mole	T_b, K	T_c, K	P_c, bar
Isobutane	58.123	261.45	408.13	37.20
Isobutylene	56.107	266.03	417.85	40.01
Krypton	83.80	119.80	209.40	55.02
Methane	16.043	111.63	190.53	45.96
N-butane	58.123	272.65	425.18	37.96
Neon	20.179	27.10	44.40	27.56
Nitric oxide	30.006	121.40	180.15	64.85
Nitrogen	28.0134	77.347	126.20	33.999
Nitrous oxide	44.013	184.68	309.56	72.45
Octafluoropropane	188.02	236.45	345.05	26.80
Oxygen	31.9988	90.18	154.576	50.43
Perfluorobutane	238.028	271.45	386.45	24.27
Perfluorobutene	200.04	267.16	388.37	27.77
Propadiene	40.065	238.75	393.85	52.49
Propane	44.096	231.105	369.82	42.50
Propylene	42.08	225.43	364.75	46.10
Sulfur hexafluoride	146.05	209.50	318.69	37.59
Tetrafluoroethylene	100.016	197.53	306.45	40.50
Trans-2-butene	56.107	274.03	428.15	40.80
Trichlorodifluoromethane	137.38	296.92	471.15	43.74
Xenon	131.30	165.05	289.733	58.40

The inputs to the ANN consisted of absolute temperature and four physical properties (M, Tc, Tb, Pc). The choice of the nature and the number of ANN inputs has been done after bibliographical synthesis (Reid, Prausnitz and Sherwood, 1977; Gosse, Déroulède *et al*., 1991), particularly the model of Chapman-Enskog (Table 1).

Neural network design: Within the literature, ANNs which have been used for the estimation and the prediction of physicochemical properties have generally been multi-layered feedforward non-linear ANNs trained via the back-propagation rule to perform a function approximation. It has been shown that non-linear feed forward neural networks are capable of universal functional approximation and that a single hidden layer with sigmoid transfer function and one neuron in the output layer with linear transfer function is sufficient to uniformly approximate any continuous bounded function (Dreyfus *et al*., 2002).

ANNs are also sensitive to the number of neurons in their hidden layers. Too few neurons can lead to

underfitting. Too many neurons can contribute to overfitting. In the first case, the training points and the fitting curve points are all inaccurately estimated but in the second case, the training points are accurately estimated, however the fitting curve tasks wild oscillations between these points and this leads to poor generalization.

The choice of the number of neurons in the hidden layers is, therefore, a delicate compromise between providing sufficient neurons to adequately determine an approximate functional relationship and avoiding the use of too many neurons which can lead to overfitting.

After the evaluation of a considerable number of differently structured neural networks, the best ANN selected in this investigation had a single hidden layer with 30 neurons and an output layer with one neuron. The hidden layer had a tansig transfer function and the output layer had a purelin transfer function (Fig. 1).

The output viscosity of the designed ANN is given by this expression:

$$\eta = \sum_{i=1}^{30} W_{31,i} \left\{ \frac{1 - \exp\left(-2\left(\sum_{j=1}^{5} W_{i,j}X_j + b_i\right)\right)}{1 + \exp\left(-2\left(\sum_{j=1}^{5} W_{i,j}X_j + b_i\right)\right)} \right\} + b_{31} \quad (1)$$

where X_j represents the inputs variables (M, T_b, T_c, P_c and T) and W_{ij} being the weights from input (j) to neuron (i) with b_i and b_{31} representing bias of the neurons in hidden layer and bias of the neuron in output layer, respectively.

Normalization: As the values of the physical input properties to the ANN differed by several orders of magnitude, which may not reflect the relative importance of the properties in determining viscosity, all of the inputs matrix variables (X_i) were normalized by using

$$X_i^n = \frac{X_i}{\left(\sum_{i=1}^{n} X_i^2\right)^{\frac{1}{2}}} \quad (2)$$

Where X_i^n are the re-scaled input values and n=1,…,5 labels the input patterns. However, the target viscosity values weren't different by important orders of magnitude so there wasn't a need to be normalized.

RESULTS AND ANALYSIS

We have opted to use in this investigation the commercially available neural network toolbox supplied for the Matlab package due to its versatility. The ANN was trained using the Lavenberg-Marquadt back propagation algorithm.

The training algorithm used in the Matlab neural network toolbox was therefore trainlm which encompasses Lavenberg-Marquadt back propagation. To prevent over training, we have chosen to train the ANN until the minimum of the Mean of Squared Errors (MSE) performance function.

The weights and bias of the final trained ANN are summarized in the Table 3. Figure 2 shows a plot of target viscosity *vs* the correlated viscosity and Fig. 3 shows a plot of target viscosity vs. predicted viscosity by the ANN.

The statistical quality of the ANN for both training and test sets was then evaluated using following parameters: Squared correlation coefficient R,

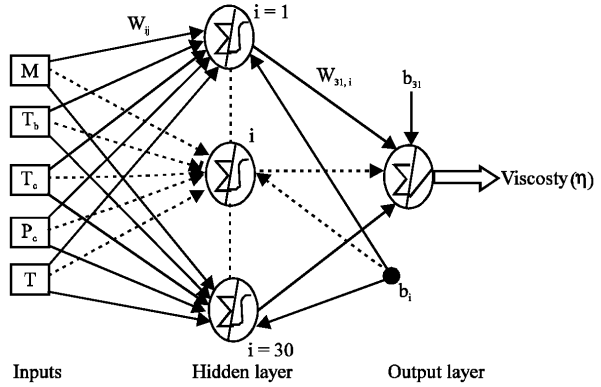

Fig. 1: Schematic operation of the ANN

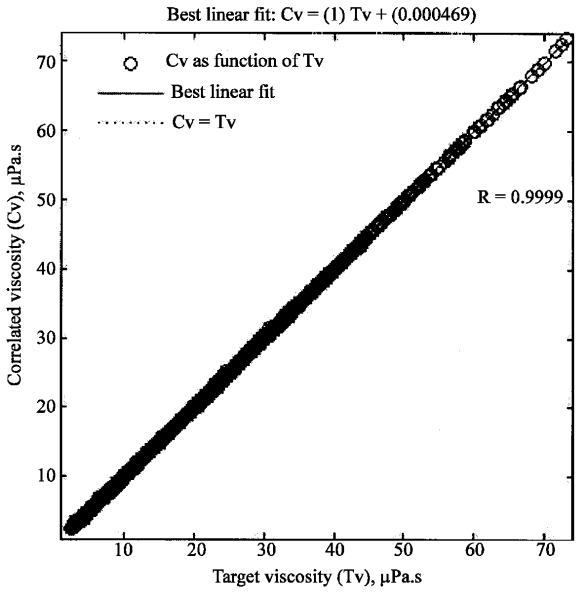

Fig. 2: Training or correlated results

Table 3: Weights and bias of the designed ANN

Hidden layer						Output layer	
Weights matrix					Bias vector	Weights vector	Bias vector
37.6468	-74.9423	95.8556	-63.9581	23.3281	-24.8066	84.7141	
236.3601	-166.0948	140.0404	-311.251	2.9123	-3.8596	17.6553	
-7.6105	-101.969	85.7236	-101.1029	0.99828	-1.1438	-21.0691	
-105.7294	-95.4821	71.2049	90.3356	0.99459	-0.98442	31.2072	
124.4201	-91.4223	16.3008	-9.5669	0.16856	-0.58225	-14.7499	
-113.7233	-375.5671	238.1092	7.5507	2.2856	-2.7206	-13.4755	
4.4242	29.8367	-8.8364	-26.0873	-1.2337	2.0842	-68.3284	
-4.6976	33.6818	-59.9495	43.6137	-20.079	20.5125	217.8401	
-211.4377	33.8831	11.8012	38.7715	-0.66586	0.27116	25.5537	
-9.5939	14.4785	15.8196	-60.3992	-0.061528	0.090345	39.9496	104.6895
-8.0103	18.8197	-8.4365	-0.39496	0.96619	0.19814	23.4808	
-103.1649	-252.3979	229.3485	-52.6815	-0.36015	0.75602	9.8745	
5.8245	9.4597	-8.6838	4.6144	-0.02635	-0.46247	-108.3947	
17.4142	-72.4752	35.8126	-19.8721	-0.10667	-0.45116	240.1328	
-100.2732	319.7116	-72.4102	-1071.1571	0.06508	-0.30041	8.3704	
145.2939	39.7231	-40.4825	-15.2097	-0.10385	0.42055	-32.6796	
26.7185	93.6339	2.9025	-192.1907	-1.9879	2.0489	-34.3943	
12.1399	-61.1697	14.3716	109.4575	0.12315	-0.4411	-29.0629	
14.3103	-25.3898	10.2232	-7.4421	-0.40892	0.58561	60.1941	
143.3846	133.608	38.795	-63.5322	1.1227	-0.44126	-36.8734	
56.2966	-21.921	2.0613	37.7494	0.86614	-0.38937	-115.9906	
129.868	-136.1961	94.0158	-199.7397	0.14226	0.025914	22.7025	
-5.2252	-5.3228	5.6516	-3.8152	-0.13704	0.053805	-108.7263	
-130.7224	-153.874	137.7436	-157.5938	0.67714	-0.93395	-28.8496	
20.3177	-46.4645	-42.8664	78.2359	22.1507	-20.6249	-34.9443	
64.6786	-77.8808	-9.1312	604.3145	0.26763	-0.61904	-29.3013	
54.4566	-22.6254	3.3718	35.0292	0.88896	-0.77337	135.8042	
-24.7407	58.0006	-35.8111	-44.8946	-0.44851	0.097837	-242.1439	
19.5423	-78.7647	47.8068	-62.4719	-0.54916	0.70418	-10.8486	
-161.575	-253.6512	122.2105	181.2687	40.254	-41.6271	-61.1156	

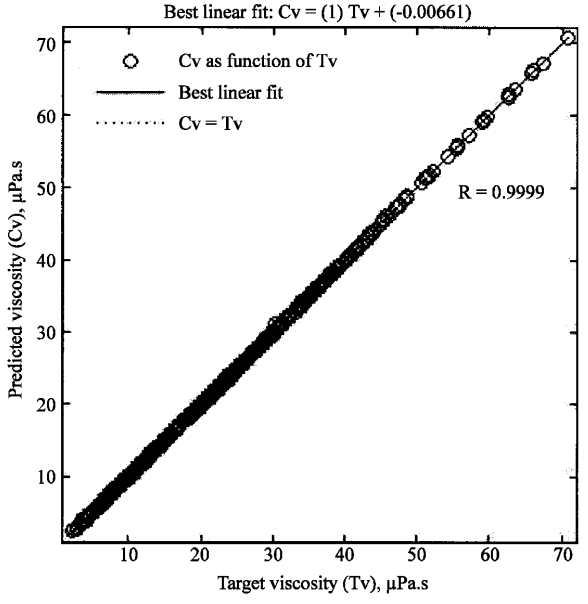

Fig. 3: Predicted results

$$R = 1 - \frac{\sum_{i=1}^{n}(y_i - y_i^t)^2}{\sum_{i=1}^{n}(y_i - y_0)^2} \quad (3)$$

where

$$y_0 = \frac{1}{n}\sum_{i=1}^{n}(y_i - y_i^t) \quad (4)$$

and the root-mean-square error, RMSE, is

$$RMSE = \left[\frac{1}{n}\sum_{i=1}^{n}(y_i - y_i^t)^2\right]^{1/2} \quad (5)$$

In order to compare the results of the ANN with the data base viscosity, we also evaluated the following parameters: Absolute Error (AE)

$$AE_i = \left(\frac{\left|y_i^t - y_i\right|}{y_i^t}\right)100 \quad (6)$$

Average Absolute Error (AAE),

$$AAE = \frac{1}{n}\sum_{i=1}^{n} AE_i \quad (7)$$

and Standard Deviation (STDEV),

Table 4: Statistical performance of the trained ANN for gas viscosity

Property	Correlated	Predicted
R	0.9999750	0.9999912
RMSE	0.1335	0.1370
AAE	0.69 (%)	0.704 (%)
STDEV	1.1626 (%)	0.5043(%)
AE (max)	15.1633 (%)	12.9961(%)

Table 5: Comparison between ANN and theoretical models according to the experimental values of low-pressure gas viscosity

			Absolute error [(calc. – exp.)/exp.]*100,%											
			Theoretical values σ and ε/k											
		Experimental value,	Available[2]		Estimated[2]		Thodos *et al.*[2]		Golubev[2]		Reichenberg[2]		Proposed ANN	
Compound	T, K	10^{-5} P[1]	η	AE	η	AE	η	AE	η	AE	η	AE	η	AE
1-butene	293.0	7.61	-	-	7.73	1.60	7.70	1.20	7.89	3.70	7.75	1.80	7.57	0.48
	333.0	8.39	-	-	8.53	1.70	8.75	4.30	8.93	6.40	8.78	4.60	8.61	2.62
	393.0	9.98	-	-	10.09	1.10	10.30	3.20	10.47	4.90	10.29	3.10	10.15	1.71
Acetylene	303.0	10.20	10.44	2.40	10.30	1.00	10.25	0.50	10.42	2.20	10.44	2.40	10.37	1.65
	374.0	12.60	12.70	0.80	12.88	2.20	12.70	0.80	13.02	3.30	12.69	0.70	12.68	0.63
	473.0	15.50	15.59	0.60	15.83	2.10	15.59	0.60	17.36	12.00	15.61	0.70	15.63	0.87
Carbon dioxide	303.0	15.10	15.31	1.40	15.24	0.90	15.28	1.20	15.54	2.90	-	-	15.04	0.37
	373.5	18.10	18.46	2.00	18.32	1.20	18.62	2.90	19.46	7.50	-	-	18.36	1.46
	473.1	21.90	22.45	2.50	22.62	3.30	22.95	4.80	26.06	19.00	-	-	22.78	4.00
Carbon disulfide	303.0	9.46	10.08	6.60	10.22	8.00	10.50	11.00	10.88	15.00	-	-	10.00	5.73
	371.2	11.90	12.48	4.90	12.65	6.30	12.99	9.20	13.33	12.00	-	-	12.34	3.69
	473.0	15.10	15.93	5.50	16.13	6.80	16.47	9.10	16.76	11.00	-	-	15.90	5.30
Carbon tetrachloride	398.0	13.30	13.54	1.80	13.38	0.60	13.58	2.10	13.91	4.60	13.65	2.60	13.19	0.83
	473.0	15.60	15.85	1.60	15.71	0.70	16.08	3.10	16.38	5.00	15.93	2.10	15.68	0.54
	573.0	19.00	19.36	1.90	19.44	2.30	19.80	4.20	20.16	6.10	19.27	1.40	18.94	0.34
Chlorine	293.0	13.30	13.51	1.60	13.43	1.00	13.47	1.30	13.81	3.80	-	-	12.90	2.98
	373.0	16.80	16.92	0.70	16.82	0.10	17.10	1.80	17.40	3.60	-	-	16.55	1.51
	473.0	20.90	20.96	0.30	21.19	1.40	21.51	2.90	22.11	5.80	-	-	20.91	0.06
Ethane	293.0	9.01	9.17	1.80	9.01	0.00	9.17	1.80	9.33	3.50	9.19	2.00	9.18	1.84
	323.0	9.98	10.14	1.60	10.00	0.20	10.16	1.80	10.37	3.90	10.20	2.20	10.05	0.68
	373.0	11.40	-	-	11.42	0.20	11.59	1.70	12.10	6.10	11.67	2.40	11.48	0.69
	523.0	15.30	15.41	0.70	15.44	0.90	15.67	2.40	18.82	23.00	15.62	2.10	15.41	0.73
Ethylene	323.0	11.10	11.20	0.90	11.40	2.70	11.23	1.20	11.30	1.80	11.12	0.20	10.97	1.19
	423.0	14.10	14.21	0.80	14.45	2.50	14.26	1.10	15.51	10.00	14.14	0.30	14.02	0.56
	523.0	16.80	16.90	0.60	17.19	2.30	16.95	0.90	21.17	26.00	16.87	0.40	16.72	0.50
Isobutane	293.0	7.44	7.50	0.80	7.50	0.80	7.58	1.90	7.76	4.30	7.60	2.10	7.54	1.32
	333.0	8.45	8.49	0.50	8.49	0.50	8.62	2.00	8.78	3.90	8.60	1.80	8.56	1.25
	393.0	9.95	9.97	0.20	9.96	0.10	10.13	1.80	10.31	3.60	10.08	1.30	10.03	0.84
Methane	293.0	10.90	10.98	0.70	11.11	1.90	10.95	0.50	-	-	-	-	11.08	1.67
	373.0	13.30	13.38	0.60	13.55	1.90	13.35	0.40	-	-	-	-	13.50	1.47
	473.0	16.00	16.06	0.40	16.32	2.00	16.03	0.20	-	-	-	-	16.06	0.37
	573.0	18.50	18.57	0.40	18.87	2.00	18.52	0.10	-	-	-	-	18.47	0.18
	773.0	22.70	22.95	1.10	22.97	1.20	22.86	0.70	-	-	-	-	22.85	0.64
Normal butane	293.0	7.39	7.49	1.30	7.57	2.40	7.43	0.50	7.61	3.00	7.48	1.20	7.46	0.99
	333.0	8.39	8.48	1.10	8.56	2.00	8.44	0.60	8.61	2.60	8.51	1.40	8.48	1.13
	393.0	9.98	10.12	1.40	10.23	2.50	10.03	0.50	10.10	1.20	10.26	2.80	9.99	0.11
Propane	293.0	8.06	8.08	0.30	8.11	0.60	8.21	1.80	8.37	3.80	8.11	0.60	8.04	0.22
	333.0	9.22	9.30	0.90	9.32	1.10	9.30	0.90	9.47	2.70	9.38	1.70	9.02	2.09
	398.0	10.70	10.80	0.90	10.90	1.90	11.09	3.60	11.33	5.90	10.75	0.50	10.56	1.35
	473.0	12.50	12.55	0.40	12.80	2.40	13.00	4.00	13.71	9.70	12.59	0.70	12.27	1.85
	548.0	14.20	14.24	0.30	14.56	2.50	14.78	4.10	16.47	16.00	14.29	0.60	13.92	1.96
Propylene	293.0	8.43	8.44	0.10	8.46	0.30	8.61	2.10	8.78	4.10	8.63	2.40	8.63	2.34
	423.0	12.10	12.11	0.10	12.14	0.30	12.26	1.30	12.66	4.60	12.21	0.90	12.16	0.50
	523.0	14.70	14.71	0.10	14.74	0.30	14.88	1.20	16.32	11.00	14.77	0.50	14.63	0.47
AAE,%				1.29		1.73		2.28		7.12		1.58		1.38

[1]10 poise (P) = 1Pa.s., [2]Values were obtained from Reid, Prausnitz and Sherwood (1977)

$$STDEV = \left[\frac{n\sum_{i=1}^{n} AE_i^2 - \left(\sum_{i=1}^{n} AE_i\right)^2}{n(n-1)} \right]^{1/2} \quad (8)$$

In these formulas, y_i represents either the ith trained or test viscosity value and y_i^t representing the corresponding target viscosity value, with n being the number of input vectors (1989 and 664 for the training and prediction sets, respectively). The results are summarized in Table 4.

Table 5 represents the results obtained by the designed ANN, these were compared with various theoretical models. The average absolute error for the estimated viscosity by the designed ANN is 1.38%, according to the experimental viscosity, for 13 different gases, at various temperatures. However, the AAE of other models, except the first one (1.29%), are all greater than the AAE value of the ANN.

CONCLUSIONS

The use of the designed ANN has been shown to accurately correlate and predict the nonpolar gases viscosity at moderate pressure (about 1 bar), over wide range of temperature (Temperature from boiling points of the chosen gases to T ≈ 2000K), for substantial number of variously gases (both organic and nonorganic compounds). The AAE in the predicted set of compounds was less than 1.39% for the gases in the Table 5. When the correlated (Fig. 2) and predicted values (Fig. 3 and Table 5) are considered jointly the AAE is approximately 0.93%, which is a serious competitor of commonly used method summarized in the Table 1. On the other hand, this method can be applied without depending on many complicated factors like σ, ω, ε/k and Ω_v, that are used in the almost other methods.

Nomenclature

b Bias
C_i Group contribution
k Boltzmann's constant
M Molecular weight, g/mol
n_i Number of atomic groups of ith type
P_c Critical pressure, bar
T Temperature, K
T_c Critical temperature, K
T_r Reduced temperature, T/Tc
W Weights

Greek

ε Energy-potential parameter
η Viscosity in Pa.s
σ Molecular diameter, Å
ω Acentric factor
Ω_v Collision integral for viscosity

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