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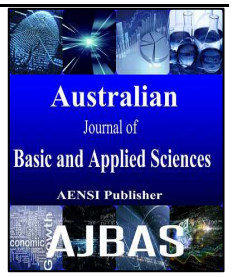

A Neural Network Approach to Predict the Density of Aqueous MEA Solution

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ABSTRACT

Background: Aqueous solution of alkanolamines such as MEA, DEA and MDEA has been used as the potential solvents for the removal of acidic gases like CO_2 and H_2S from power plants and various other industries**. Objective:** A computational model based on artificial neural networks (ANNs) has been developed to predict the density of aqueous MEA solution. Statistical analysis of the predicted data has also been performed. **Results:** A good agreement was found between the predicted and literature data**. Conclusion:** The value of low predicted error via statistical analysis showed that the applied model can successfully predict the density of the aqueous MEA solution**.**

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INTRODUCTION

Lately, it has been shown that global warming is one of the main environmental issues due to the increasing concentration of carbon dioxide (CO_2) (Mondal *et al*., 2015). It is subsequently important to develop and utilize efficient techniques for CO_2 gas removal. Post-combustion capture using absorption is one of the most widely used technique to remove CO_2 and alkanolamines have long been recognized as viable solvents for CO_2 removal in industries and power plants. Due to the strong base nature and fast reactivity with CO_2, aqueous monoethanolamine (MEA) is still considered as the most popular solvents for CO_2 capture in industries (Wong *et al*., 2015).

Physiochemical properties such as density of aqueous MEA is very important for the analysis of absorption and desorption processes (Shaikh *et al*., 2014; Teng *et al*., 1994). Many studies have been done to find out the density of aqueous MEA at different concentration and temperature conditions. But the measurement of density of aqueous MEA at different other temperatures and concentrations is very costly, time-consuming and difficult. So, lately, a new computational prediction technique named artificial neural network (ANN) has attracted great attention. For example, Golzar *et al*. (2014) has used multilayer perceptron ANN model to predict the thermophysical properties of binary mixtures of common ionic liquids with water or alcohol.

In this study, a computational model based on ANNs was developed to predict the density of aqueous MEA solution. All the data used in this work was taken from literature (Han *et al*., 2012). A statistical analysis of the data was performed in terms of least square correlation (R^2), root mean square error (RMSE), average relative deviate (ARD), standard deviation (SD) and average absolute deviation (AAD).

MATERIALS AND METHODS

The following subsections provide information about the density database utilized, ANN, and the model used, as well as the parameters and optimization processes.

Density database of aqueous MEA solution:

The data used in this work for the prediction of density of aqueous MEA solution was taken from Han *et al*., (2012). A total of 180 data sets were taken and used to develop a computational model.

Artificial Neural Network Model:

The prediction of the density data of aqueous MEA solution was done through ANN-based model. Neural networks are valuable numerical strategies motivated by the function of human brain. Neural networks are computational structures comprising of substantial quantities of basic process units connected on an enormously parallel scale. These units are similar to neurons existing in human brain and the synapse they form.

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Each neuron is associated with a set of some input connections as the processing elements and as well as the single output, shown in Fig. 1.

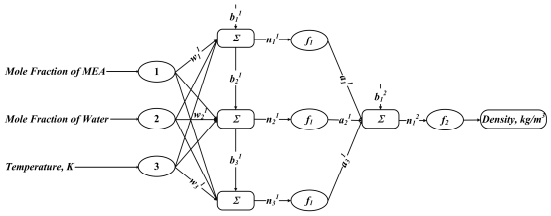

Fig. 1: Schematic of components of simple neuron.

The expression for output activation can be described as follows:

$$h(x) = g(a(x)) = g\left(b + \sum_i w_i x_i\right)$$

The function *g(a(x))* is denoted as the activation function, where *b* is the neuron bias present in the neural network, it is biased because in case of no inputs then *b* would be the pre-activation.

w is the connection weight vector, described as:

$$w = [w_1, w_2, w_3, ..., w_N]$$

And *x* is the input vector, described as:

$$x = [x_1, x_2, x_3, ..., x_N]$$

The specific model proposed in this work for the prediction of density data is multilayer perceptron (MLP), the most common type of ANN (Torrecilla *et al*., 2007).

Multilayer perceptron model (MLP):

A multilayer perceptron (MLP) is feed-forward neural network which needs a preferred output in order to learn. A MLP neural network comprises of input layer, hidden layer and output layer. The point of MLP neural network is to make a model that appropriately maps a set of input information onto a set of suitable outputs utilizing historic information so that the model can be used to generate an output when the preferred output is vague (Nikravesh and Aminzadeh, 2001). In order to train the network, MLP utilizes a supervising learning technique known as back-propagation. In this technique, at first, all the weights are adjusted arbitrarily, then the output is compared with the targeted values in the training sets and the desired error is propagated back into the network. This procedure is repeated continuously until the outputs are acceptably close to the target values (Celikoglu, 2006; Wu and Liu, 2012).

Training neural networks:

When a neural network is formed for a specific application, it is prepared to be trained. Training is the methodology by which connection weights are assumed. Initially, the weights are picked up haphazardly. There are two types of training methodology: supervised and un-supervised training. Supervised training (ST) is refined by providing neural network a set of sample data alongside the expected output from each of these samples. ST is the most widely recognized type of neural network training. As the ST starts, the neural network is taken through a series of iterations or epochs till the time the output data matches with the targeted data with a sensibly small error. Unsupervised training (UST) is similar to ST with the exception that no targeted values are provided. UST generally arises when neural network characterize the inputs into some groups (Mirarab *et al*., 2014).

Validating neural networks:

After the neural network is trained, it must be assessed to check whether it is prepared for genuine use. This step is vital to be deliberately performed to figure out whether additional training is needed. To accurately validate a neural network, different data sets for validation must be kept aside from training data sets. It is exceptionally essential that different data sets may be for validation. Training the neural network with a desired set and furthermore utilizing the same set to predict the error of the whole neural network will most likely prompt to bad results. The error obtained utilizing the training sets will considerably be lower than the error on the remaining data sets of neural network. The reliability of the validated data must be maintained continuously. In the event that validation is performed poorly, this probably implies that there was data present in the validation set that was not accessible in the training set. The way that this condition should be resolved is by attempting an alternate, more arbitrary, method for differentiating the data into training and validation sets. If it fails, the validation and training sets must be merged together as one huge training set. At that point, new data must be obtained to serve as the validation data. In some situations, it may be difficult to acquire additional data for either training or validation. While this methodology will do without the security of a proper validation, if additional data cannot be acquired, this may be the only alternative (Mirarab *et al*., 2014).

Testing neural networks:

The test error is not utilized in the training sets, but is merely used once the network has been established, to measure the model accuracy. It is also

helpful to plot the test error during the training process. In the event that the error in the test set reaches a minimum value at a different iteration number than the validation set error, this may show that the data sets are poorly divided. The existence of irregular noise can also have a significant impact on the capability of neural network to separate between cases near decision limits which will lead to inaccurate predictions (Mirarab *et al*., 2014).

Optimization procedure:

The optimal design of the artificial neural network (ANN) was figured out by trial and error method. In this study, the number of neurons in the hidden layers was found out by optimizing the mean-squared error (MSE). Actually, a low number of neurons may not result in enabling a network to achieve the anticipated error, while a large number may cause over fitting. The performance of ANN was assessed by calculating the MSE versus number of neurons, as shown in Fig. 2. It is apparent from Fig. 2 that the lowest MSE was obtained with nine neurons. Therefore, nine neurons were selected as the optimum condition for the present study.

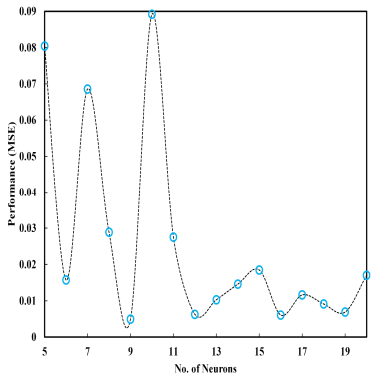

Fig. 2: Variation of Performance (MSE) with number of neurons.

RESULTS AND DISCUSSION

Prediction of density using ANNs:

In this study, a total of 180 data points were utilized from literature to analyze the prediction performance of ANN technique. Mole fraction of water, mole fraction of MEA and operational temperature was taken as input variables, while density was assigned as output. All the 180 data points were randomly divided into three data sets, i.e. 70% for training (126 data points), 15% for validation (27 data points) and 15% for testing (27 data points).

For density prediction of aqueous MEA, a multilayer perceptron (MLP) model of ANN was utilized with back propagation algorithm. This specific model was chosen based on their capability to represent the non-linear relationship between input and output data. Levenberg-Marquardt was utilized as a training function for MLP neural network due to its good performance in non-linear regression problems (Mirarab *et al*., 2014). The optimum number of neurons in hidden layer was chosen by optimization procedure. Tan-sigmoid transfer function was used to train the MLP neural-network (Fig. 3). Therefore, in our study, nine neurons were selected in the hidden layers.

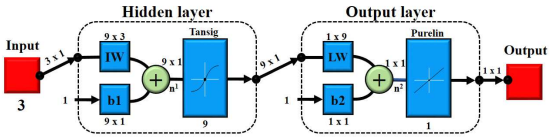

Fig. 3: Artificial Neural Network (ANN) design for prediction of density.

The performance analysis of prediction data for training, validation and testing data-sets was done in terms of mean square error (MSE). So, a graph of MSE vs epochs was plotted for training, validation and testing data-sets to get the best neural network configuration as shown in Fig. 4. It is clearly seen from Fig. 4 that the MSE values decreased as the number of epochs increased because the weights have been updated after each epoch.

A comparison of predicted density results from the proposed MLP neural network model with literature data is shown in Fig. 5 for training, validation and testing respectively. It is evident from Fig. 5 that the predicted results were in very good

agreement with the literature data. Table 1 summarizes the literature and predicted data of aqueous MEA with corresponding data sets used in the ANN modeling.

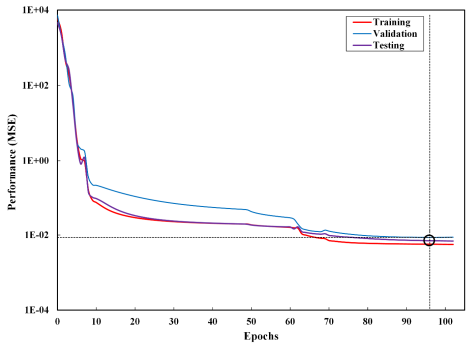

Fig. 4: Variation of Performance (MSE) vs Epochs for training, validation and testing data sets.

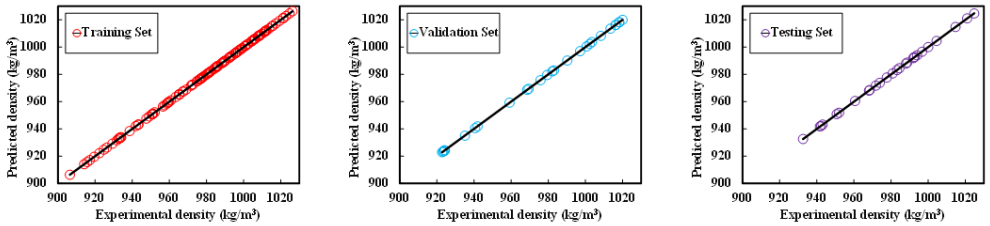

Fig. 5: Evaluation of ANN predicted and experimental data of density for training, validation and testing data sets.

Validation performed in the developed model is done by means of external test based on the following statistical quantities:

Least-squared correlation (R^2):

$$R^2 = \frac{\sum_{i=1}^{i=n}\left(x_i - \bar{x}\right)^2 - \sum_{i=1}^{i=n}\left(x_i - y_i\right)^2}{\sum_{i=1}^{i=n}\left(x_i - \bar{x}\right)^2}$$

Root mean square error (RMSE):

$$RMSE = \sqrt{\frac{\sum_{i=1}^{i=n}\left(y_i - x_i\right)^2}{n}}$$

Average relative deviation (ARD):

$$ARD = \frac{\sum_{i=1}^{i=n}\left|\frac{x_i - y_i}{x_i}\right|}{n}$$

Standard deviation (SD):

$$SD = \sqrt{\sum_{i-1}^{i=n}\frac{1}{n-1}\left(x_i - \bar{x}\right)^2}$$

Average absolute deviation (AAD):

$$AAD = \frac{\sum_{i=1}^{i=n}\left|x_i - y_i\right|}{n}$$

Table 1: Experimental and predicted data of density with corresponding data sets used in ANN modeling.

Run No.	Temperature (K)	Mole Fraction			Density (kg/m^3)	
		x[MEA]	x[H_2O]		Experimental	Predicted
Training Set						
1	298.15	0	1		997	996.99270
2	298.15	0.1122	0.8878		1010.9	1010.83227
3	298.15	0.2278	0.7722		1021.3	1021.16469
4	298.15	0.4077	0.5923		1026.3	1026.29901
5	298.15	0.5412	0.4588		1024.7	1024.72866
6	298.15	1	0		1011.9	1011.85753
7	303.15	0	1		995.6	995.60765
8	303.15	0.1122	0.8878		1008.4	1008.59162
9	303.15	0.1643	0.8357		1013.8	1013.69935
10	303.15	0.3067	0.6933		1021.4	1021.45642
11	303.15	0.4077	0.5923		1022.8	1022.82980
12	303.15	1	0		1008	1007.98017
13	308.15	0.1122	0.8878		1006.2	1006.20062
14	308.15	0.1643	0.8357		1011	1011.01756
15	308.15	0.2278	0.7722		1015.2	1015.25074
16	308.15	0.4077	0.5923		1019.3	1019.26992
17	308.15	0.5412	0.4588		1017.3	1017.33607
18	308.15	0.7264	0.2736		1012.3	1012.37012
19	308.15	1	0		1004	1004.04496
20	313.15	0	1		992.2	992.19475

21	313.15	0.2278	0.7722		1012.1	1012.14469
22	313.15	0.4077	0.5923		1015.7	1015.65006
23	313.15	0.7264	0.2736		1008.5	1008.45972
24	318.15	0.1122	0.8878		1001.1	1001.02991
25	318.15	0.1643	0.8357		1005.3	1005.32451
26	318.15	0.2278	0.7722		1009	1008.95781
27	318.15	0.3067	0.6933		1011.4	1011.34403
28	318.15	0.4077	0.5923		1012	1011.99048
29	318.15	0.5412	0.4588		1009.7	1009.71062
30	318.15	1	0		996	996.02352
31	323.15	0	1		988	988.00682
32	323.15	0.1122	0.8878		998.1	998.26626
33	323.15	0.2278	0.7722		1005.6	1005.69385
34	323.15	0.3067	0.6933		1007.8	1007.85711
35	323.15	0.4077	0.5923		1008.3	1008.29865
36	323.15	0.5412	0.4588		1005.9	1005.86738
37	328.15	0	1		985.7	985.65316
38	328.15	0.1122	0.8878		995.5	995.39171
39	328.15	0.1643	0.8357		999.2	999.23367
40	328.15	0.2278	0.7722		1002.4	1002.35352
41	328.15	0.3067	0.6933		1004.4	1004.31514
42	328.15	0.4077	0.5923		1004.6	1004.57272
43	328.15	0.5412	0.4588		1002	1002.01007
44	333.15	0.1643	0.8357		996.1	996.04696
45	333.15	0.2278	0.7722		999	998.93687
46	333.15	0.3067	0.6933		1000.7	1000.71468
47	333.15	0.4077	0.5923		1000.8	1000.80631
48	333.15	0.5412	0.4588		998.2	998.13104
49	333.15	0.7264	0.2736		992.7	992.62845
50	333.15	1	0		983.9	983.83242
51	338.15	0	1		980.5	980.49582
52	338.15	0.1122	0.8878		989.5	989.33373
53	338.15	0.2278	0.7722		995.4	995.44392
54	338.15	0.4077	0.5923		997	996.99162
55	338.15	0.5412	0.4588		994.2	994.21871
56	338.15	1	0		979.8	979.77774
57	343.15	0.1122	0.8878		986.1	986.16073
58	343.15	0.1643	0.8357		989.4	989.40504
59	343.15	0.2278	0.7722		991.9	991.87467
60	343.15	0.3067	0.6933		993.2	993.32364
61	343.15	0.4077	0.5923		993.1	993.12119
62	343.15	0.5412	0.4588		990.2	990.26102
63	343.15	0.7264	0.2736		984.6	984.64266
64	348.15	0	1		974.8	974.81668
65	348.15	0.1643	0.8357		985.9	985.95350
66	348.15	0.2278	0.7722		988.3	988.22877
67	348.15	0.3067	0.6933		989.5	989.52662
68	348.15	0.4077	0.5923		989.2	989.18869
69	348.15	0.5412	0.4588		986.2	986.24754
70	348.15	0.7264	0.2736		980.6	980.59060
71	348.15	1	0		971.6	971.64969
72	353.15	0.1643	0.8357		982.4	982.41521
73	353.15	0.3067	0.6933		985.6	985.65857
74	353.15	0.4077	0.5923		985.2	985.18966
75	353.15	0.7264	0.2736		976.5	976.48317
76	353.15	1	0		967.5	967.54891
77	358.15	0.1122	0.8878		976.1	976.09108
78	358.15	0.1643	0.8357		978.7	978.78866
79	358.15	0.3067	0.6933		981.8	981.71790
80	358.15	0.4077	0.5923		981.2	981.12180
81	358.15	0.5412	0.4588		978	978.02690
82	358.15	0.7264	0.2736		972.3	972.31411
83	358.15	1	0		963.4	963.40991
84	363.15	0	1		965.3	965.41159
85	363.15	0.1122	0.8878		972.5	972.54587
86	363.15	0.1643	0.8357		975	975.07149
87	363.15	0.2278	0.7722		976.9	976.82083
88	363.15	0.3067	0.6933		977.7	977.70396
89	363.15	0.4077	0.5923		977.1	976.98525
90	363.15	1	0		959.2	959.22865
91	373.15	0	1		958.6	958.53860
92	373.15	0.1122	0.8878		965.3	965.15308
93	373.15	0.1643	0.8357		967.2	967.35688
94	373.15	0.2278	0.7722		969	968.81277
95	373.15	0.5412	0.4588		965.3	965.21466
96	373.15	0.7264	0.2736		959.3	959.45391
97	373.15	1	0		950.9	950.74753
98	383.15	0	1		951.2	951.16862
99	383.15	0.1122	0.8878		957.4	957.34783
100	383.15	0.3067	0.6933		960.8	960.94596
101	383.15	0.4077	0.5923		959.8	959.82462
102	383.15	0.5412	0.4588		956.5	956.42573
103	383.15	0.7264	0.2736		950.6	950.67065
104	393.15	0	1		943.4	943.32275
105	393.15	0.1122	0.8878		949.1	949.14641
106	393.15	0.3067	0.6933		952.1	952.19584
107	393.15	0.4077	0.5923		950.9	950.94956
108	393.15	0.5412	0.4588		947.6	947.50731
109	393.15	1	0		933.5	933.48524
110	403.15	0.1643	0.8357		941.9	942.06811
111	403.15	0.2278	0.7722		943.1	942.99736
112	403.15	0.5412	0.4588		938.6	938.45977
113	403.15	0.7264	0.2736		932.7	932.84226
114	403.15	1	0		924.7	924.71148

115	413.15	0	1		926.3	926.33830
116	413.15	0.1122	0.8878		931.7	931.62617
117	413.15	0.1643	0.8357		932.9	932.96594
118	413.15	0.2278	0.7722		934	933.81835
119	413.15	0.3067	0.6933		933.8	933.97909
120	413.15	0.5412	0.4588		929.3	929.18718
121	413.15	1	0		915.7	915.69527
122	423.15	0	1		917.1	917.15144
123	423.15	0.1122	0.8878		922.3	922.21163
124	423.15	0.5412	0.4588		919.5	919.54473
125	423.15	0.7264	0.2736		914.1	914.13966
126	423.15	1	0		906.4	906.37070
Validation set						
127	298.15	0.1643	0.8357		1016.3	1016.24908
128	298.15	0.7264	0.2736		1020	1019.97633
129	303.15	0.2278	0.7722		1018.2	1018.26485
130	303.15	0.7264	0.2736		1016.2	1016.21613
131	308.15	0.3067	0.6933		1018.2	1018.15288
132	313.15	0.1122	0.8878		1003.5	1003.67682
133	313.15	0.1643	0.8357		1008.3	1008.22279
134	313.15	0.5412	0.4588		1013.5	1013.53844
135	318.15	0	1		990.2	990.19181
136	323.15	0.1643	0.8357		1002.3	1002.32716
137	323.15	0.7264	0.2736		1000.6	1000.55254
138	338.15	0.3067	0.6933		997.1	997.05200
139	343.15	1	0		975.8	975.72152
140	348.15	0.1122	0.8878		983	982.89602
141	353.15	0.1122	0.8878		979.4	979.54005
142	353.15	0.5412	0.4588		982.1	982.17075
143	358.15	0	1		968.6	968.66456
144	373.15	0.3067	0.6933		969.4	969.45973
145	383.15	0.1643	0.8357		959.1	959.26831
146	403.15	0	1		935.1	935.04195
147	403.15	0.1122	0.8878		940.6	940.57638
148	403.15	0.4077	0.5923		941.9	941.89977
149	413.15	0.7264	0.2736		923.6	923.66691
150	423.15	0.1643	0.8357		923.3	923.43058
151	423.15	0.2278	0.7722		924.3	924.23176
152	423.15	0.3067	0.6933		924.1	924.33899
153	423.15	0.4077	0.5923		922.8	922.92461
Testing set						
154	298.15	0.3067	0.6933		1024.8	1024.66160
155	303.15	0.5412	0.4588		1021	1021.07644
156	308.15	0	1		994	994.00377
157	313.15	0.3067	0.6933		1014.7	1014.77733
158	313.15	1	0		1000	1000.05633
159	318.15	0.7264	0.2736		1004.5	1004.51245
160	323.15	1	0		992	991.96308
161	328.15	0.7264	0.2736		996.7	996.59193
162	328.15	1	0		988	987.89476
163	333.15	0	1		983.2	983.14499
164	333.15	0.1122	0.8878		992.3	992.41228
165	338.15	0.1643	0.8357		992.7	992.76996
166	338.15	0.7264	0.2736		988.7	988.65045
167	343.15	0	1		977.7	977.71704
168	353.15	0	1		971.8	971.79896
169	353.15	0.2278	0.7722		984.5	984.50538
170	358.15	0.2278	0.7722		980.8	980.70320
171	363.15	0.5412	0.4588		973.9	973.81634
172	363.15	0.7264	0.2736		968.1	968.08259
173	373.15	0.4077	0.5923		968.5	968.51752
174	383.15	0.2278	0.7722		960.6	960.48864
175	383.15	1	0		942.3	942.15172
176	393.15	0.1643	0.8357		950.7	950.82941
177	393.15	0.2278	0.7722		952	951.87621
178	393.15	0.7264	0.2736		941.7	941.80301
179	403.15	0.3067	0.6933		943.1	943.22401
180	413.15	0.4077	0.5923		932.6	932.60333

Where x_i, y_i, $\bar{x}$, $\bar{y}$ and n the experimental data, predicted data, mean experimental data, mean predicted data and the number of data points respectively. The acquired results of above mentioned statistical quantities (R^2, *RMSE*, *ARD*, *SD* and *AAD*) for density of the predicted model focused around ANN for every three sets (training, validation and testing) as well as the total data set was arranged in Table 2. From table 2, it can be presumed that the three chosen input parameter (temperature, mole fraction of MEA and water) were suitable for predicting the density and the predicted results by the model are dependable on the grounds that their correlation and error values were in a satisfactory range. Furthermore, R^2 values of the predicted model were very near to unity and their relating errors are very small (or negligible).

The plot of density data versus operational temperature for the literature data and ANN predicted data is shown in Fig. 6. It can been seen that predicted data showed excellent harmony with the literature data, demonstrating the applicability of ANN model to predict the density of aqueous MEA.

A cross plot of density data of whole literature and predicted data is shown in Fig. 7. It is obvious from Fig. 7 that the predicted data was in very good agreement with the experimental data, confirming the approach of ANN modeling in predicting the density successfully.

Table 2: Statistical parameters for Training, Validation and Testing data sets.

Parameter	Training	Validation	Testing	All
R^2	0.99999	0.99999	0.99999	0.99999
RMSE	0.07553	0.09316	0.08415	0.07973
ARD	0.00006	0.00008	0.00007	0.00006
SD	0.07583	0.09493	0.08575	0.07995
AAD	0.05852	0.07493	0.06977	0.06267

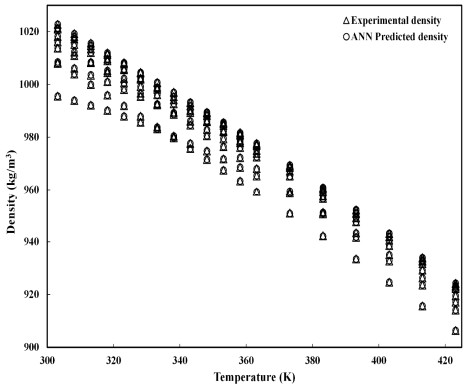

Fig. 6: Density as a function of temperature for experimental data and ANN predicted data.

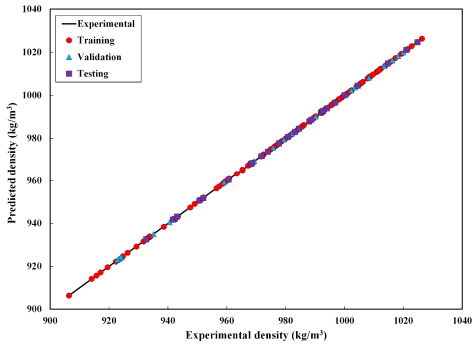

Fig. 7: Comparison of Experimental data with ANN predicted data in different data sets.

Conclusion:

In the present work, we have developed a model using the nonlinear artificial neural network (ANN) technique to predict the experimental density data of aqueous MEA based on temperature and mole fraction of MEA and water. The predicted results disclose that the chosen parameters i.e. inputs, were extremely suitable for the estimation of density of aqueous MEA. In addition, the statistical quality represented by different parameters and the low anticipated error of the developed model show that it can precisely anticipate the density of the aqueous MEA solution.

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