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Research paper

Effect of Some Chemical Additives in Increasing the Electrical Conductivity of the Liquid Fuel Dimethyl Aminoethyl Azide (DMAZ)

Shahram G. Pakdehi*, Hamidreza Taffazolinia,
Sajjad Rezaei, Manoochehr Fathollahi

*Faculty of Chemistry and Chemical Engineering, Malek Ashtar
University of Technology, P.O. Box 16765/3454, Tehran, Iran
E-mail: sh_ghanbari73@yahoo.com

Abstract: The novel liquid fuel 2-dimethylaminoethyl azide (DMAZ) is a good candidate for the replacement of the hydrazine family in space programs. However, it suffers from low electrical conductivity, which is dangerous during the production process and its transportation. A simple and effective way to increase its electrical conductivity is the use of antistatic additives. In this article, the electrical conductivity of DMAZ was initially predicted *via* an artificial neural network. Then, the increase in electrical conductivity of DMAZ was investigated using some chemical surfactants (such as hexylamine, octylamine and tributylamine), acetonitrile as a polar aprotic substance and a DMAZ salt (DMAZ·HCl) as an ionic liquid. Of these additives, acetonitrile had the greatest effect on the electrical conductivity of the fuel, exhibiting an increase in conductivity of about 45%. Adding a two-component mixture of acetonitrile and the DMAZ salt, further increased the electrical conductivity of the fuel by about 55%, relative to pure DMAZ. Tributylamine did not have a significant effect on the electrical conductivity of the fuel.

Keywords: DMAZ, electrical conductivity, artificial neural network, chemical additives, acetonitrile

Nomenclature

b_j	Bias of the neuron j of the hidden layer
f	Activation or transfer function of the neuron
j	Hidden neuron
MSE	Mean square error
N	Total number of data values used
n_j	Output of a neuron
R	Determination coefficient
w_{jr}	Weight of the connection among the input neurons with the hidden layer
X_i	Real value
X_{max}	Maximum actual values
X_{min}	Minimum actual values
X_n	Normalized value
x_r	Input neuron
y	Experimental value with network
y_f	Calculated value with network

1 Introduction

2-Dimethylaminoethyl azide (DMAZ, $\text{Me}_2\text{NCH}_2\text{CH}_2\text{N}_3$) is a highly energetic liquid fuel and a good candidate for use in the aerospace industry. It is an alternative fuel for the carcinogenic hydrazine family [1-4]. However, in contrast to the hydrazine family, DMAZ suffers from low electrical conductivity problems [5, 6]. During work on DMAZ production on an industrial scale, it was observed that this fuel has a relatively high sensitivity to electrostatic discharge. In addition, high chargeability was observed during the transfer process of this fuel, which leads to fire and explosion. Thus, to prevent these hazards, the conductivity of the fuel should be increased.

Unfortunately, there is very little information on the electrical properties of liquid fuel DMAZ. However, different methods have been employed to remove or control static electricity in flammable fuels. One of the simplest ways to discharge an electric charge is grounding. Other methods used for this purpose include increasing the humidity, use of neutral gases, ventilation, *etc* [7, 8]. However, none of these methods are fully effective and do not completely discharge the electrical charge accumulated in the fuel. One of the most effective ways to solve this problem is to reduce the electrical resistance of the fuel *via* antistatic additives [9]. Antistatic additives have the advantage of requiring the addition of only a few parts per million (ppm) to increase the conductivity

of an insulating liquid by several orders of magnitude without affecting its other thermophysical or performance properties. However, they do not prevent the creation of static electricity. Rather, they only contribute to the relaxation of electrical charges in hazardous sites by increasing the electrical conductivity of the fuel, which, in return, may contribute to an increase in the rate of fuel transfer [8]. Several groups of the additives exist, such as surfactants, ionic liquids and polar aprotic materials [9-15].

Various methods have been reported for the prediction of the electrical conductivity in liquids, such as quantum calculations, molecular dynamics (MD) simulation, some Monte Carlo simulations, hole theory, the computer-aided reverse design method, the QSPR method, the back-propagation artificial neural network (BP ANN) method, group contribution-based prediction method, structure-based method, *etc.* These methods were completely discussed by Cao *et al.* [16]. They stated that the disadvantages of these methods included weaknesses in predicting the conductivity of new liquids and needed complex parameters. However, one conventional predictive method for nonionic high energetic liquids is the artificial neural network or ANN method. DMAZ is a nonionic liquid. Therefore, in this article, the electrical conductivity of the liquid fuel DMAZ will be initially predicted by an artificial neural network and then validated experimentally. For new liquid fuels, this is a novel idea. Afterwards, the effects of the chemical additives on increasing the electrical conductivity of the fuel will be studied. It is hoped that the results of this research will be helpful in increasing safety in the production and transportation of the liquid fuel DMAZ in the aerospace industry.

2 Artificial Neural Network Theory

2.1 Network design

Artificial neural networks (ANNs) are nonlinear learning mathematical models that are designed as simulation of human brain procedures and have been used in many scientific disciplines [17-19]. A neural network consists of a number of simple processing elements, called the neurons. Each neuron of the neural network is connected to others by means of direct communication links, each with an associated weight, which represents information being used by the network to solve the problem. The output of a neuron is computed from the following Equation 1 [19]:

$$n_j = f\left(\sum_{i=1}^N w_{jr} x_r + b_j\right) \quad (1)$$

where w_{jr} is the weight of the connection among the input neurons with the hidden layer, j is the hidden neuron, x_r is the input, the term b_j corresponds to the bias of the neuron j of the hidden layer, and f is the activation or transfer function of the neuron.

Different types of transfer functions have been proposed for ANNs, such as linear, logarithmic sigmoid, hyperbolic tangent sigmoid, and radial basis transfer functions [20]. In the present study, the hyperbolic tangent sigmoid (Equation 2) and the linear (purelin) function (Equation 3) were utilized as the transfer functions of the input and output layer.

$$f(x) = \frac{e^x - e^{-x}}{e^x + e^{-x}} \quad (2)$$

$$f(x) = x \quad (3)$$

2.2 Normalization

Data should be normalized because raw data entry reduces the network speed and accuracy. Since each parameter has its own classifications, in order to equalize the range of their changes data normalization occurs to prevent over-weighting of the network weights. In this article, Equation 4 was applied for data normalization [21].

$$X_n = \frac{X_i - X_{\min}}{X_{\max} - X_{\min}} \quad (4)$$

where X_n , X_i , X_{\max} and X_{\min} are normalized, real, maximum actual and minimum actual values respectively.

2.3 Network evaluation factors

The basis for deciding which network is best for each application run is the determination coefficient (R) and mean square error (MSE). They are given as follows [22]:

$$R = \sqrt{\frac{\sum_{i=0}^N (y - y_f)}{\sum_{i=0}^N y^2 - \frac{\sum_{i=0}^N y_f^2}{N}}} \quad (5)$$

$$MSE = \frac{\sum_{i=0}^N y_f^2}{N} \quad (6)$$

where y , y_f and N are experimental and calculated values with the network and the total number of data values used, respectively. The best model performance based on the R criterion is one and zero for the other criteria [23].

2.4 Predicting the electrical conductivity of liquids *via* an artificial neural network

Multi-layer perceptron (MLP) and radial basis function (RBF) neural networks, two of the most well-known neural networks, were used to predict the electrical conductivity. For this purpose, the input data for the MLP and RBF neural networks were: dipole moment, dielectric constant, boiling point, freezing point, molar volume, highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energies. The values of the HOMO and LUMO energies may be calculated *via* Gaussian 09 software. The optimized structures of DMAZ were drawn with Gauss view 5.0 and then the best angles and bond lengths were calculated *via* Gaussian 09 software using the B3LYP method. A schematic diagram of the neural network for the electrical conductivity calculation for liquids is given in Figure 1.

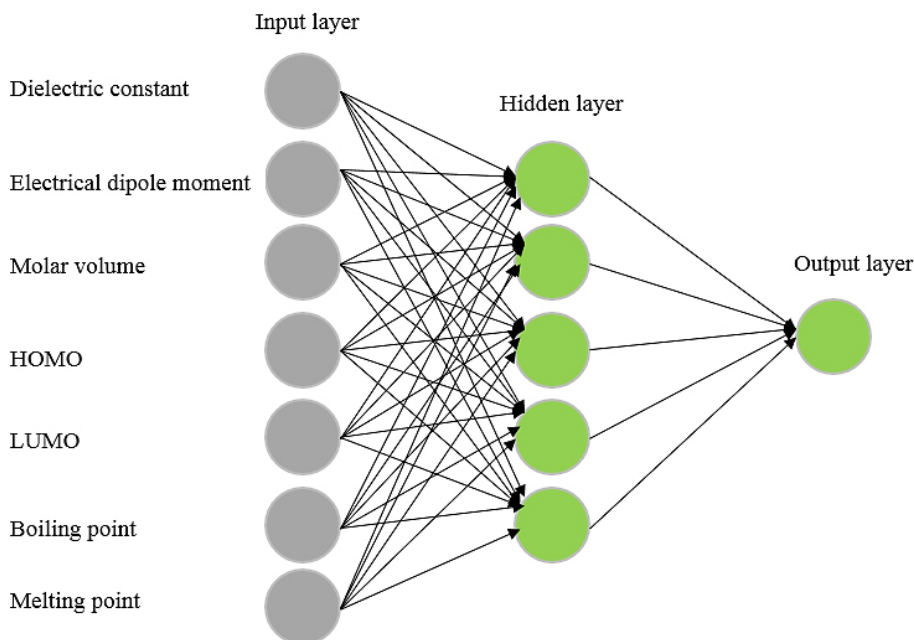


Figure 1. Schematic diagram of the neural network used

3 Experimental

3.1 Materials and equipment

The materials used to increase the electrical conductivity of liquid fuel DMAZ were surfactants [such as hexylamine ($C_6H_{15}N$), octylamine ($C_8H_{19}N$), tributylamine ($C_{12}H_{27}N$)], polar aprotic materials (for example acetonitrile (CH_3CN)) and an ionic liquid (for example a DMAZ·HCl salt ($C_4H_{10}N_4 \cdot HCl$)). The surfactants and acetonitrile were analytical grade and purchased from Merck Co. (Germany).

DMAZ·HCl salt ($C_4H_{10}N_4 \cdot HCl$) was synthesized by the reaction between an equimolar molar ratio of pure liquid DMAZ (purity >99.9 wt.%, purchased from 3M Co., USA) and aqueous hydrochloric acid solution. The final white solid was dried overnight at 70 °C.

The electrical conductivity was measured using a calibrated electrical conductometer (Model 856, Metrohm Co., Swiss). The accuracy of the device was $\pm 1 \mu S/m$.

3.2 Experimental procedure

The electrical conductivity of pure liquid fuel DMAZ was measured as $10 \mu\text{S/m}$. Specified amounts of the additives were added to the fuel and stirred well with a magnetic stirrer. The conductivity of each solution was measured using the conductometer at $25 \text{ }^\circ\text{C}$.

4 Results and Discussion

4.1 Optimal structure selection for artificial neural network

The neural network structure is optimized by varying the number of hidden layers and the number of neurons within each hidden layer. According to the Cybenko theorem [24], the multi-layer perceptron artificial neural network and a hidden layer can predict any kind of nonlinear problem. The neural network structure used in this article has a hidden layer for predicting the electrical conductivity of liquids. The number of neurons in the hidden layer was determined through an optimization process to reduce the error. The number of appropriate neurons in the hidden layer depends on three factors:

- (i) the complexity of the relationship between the input and output data,
- (ii) the number of training and testing data,
- (iii) noise intensity applied by the dataset.

Too many neurons in the neural network may not be able to achieve the desired error, while they may cause overfitting. In this research, the number of neurons in the hidden layer was optimized by Equations 5 and 6. Of the neurons used, the best results were obtained when 5 neurons were used in the hidden layer, with a least squares error of 0.04119 and a regression coefficient of 0.94073. Therefore, from the optimal neural network structure used by seven neurons for data entry, five neurons in the hidden layer and one output neuron were applied because there was only one output (electrical conductivity) (Figure 1).

In this research, 73 data values were used for the neural network: 51 data values for network training, 11 data values for network validation and 11 data values for network testing. The data used for various chemicals [25, 26] were calculated after normalization according to Equation 4. The MLP neural network regression results are shown in Figure 2. As can be seen from Figure 2, the results were more than 85% convergent. Figure 3 shows the mean square error of this artificial network, which is approximately 0.007.

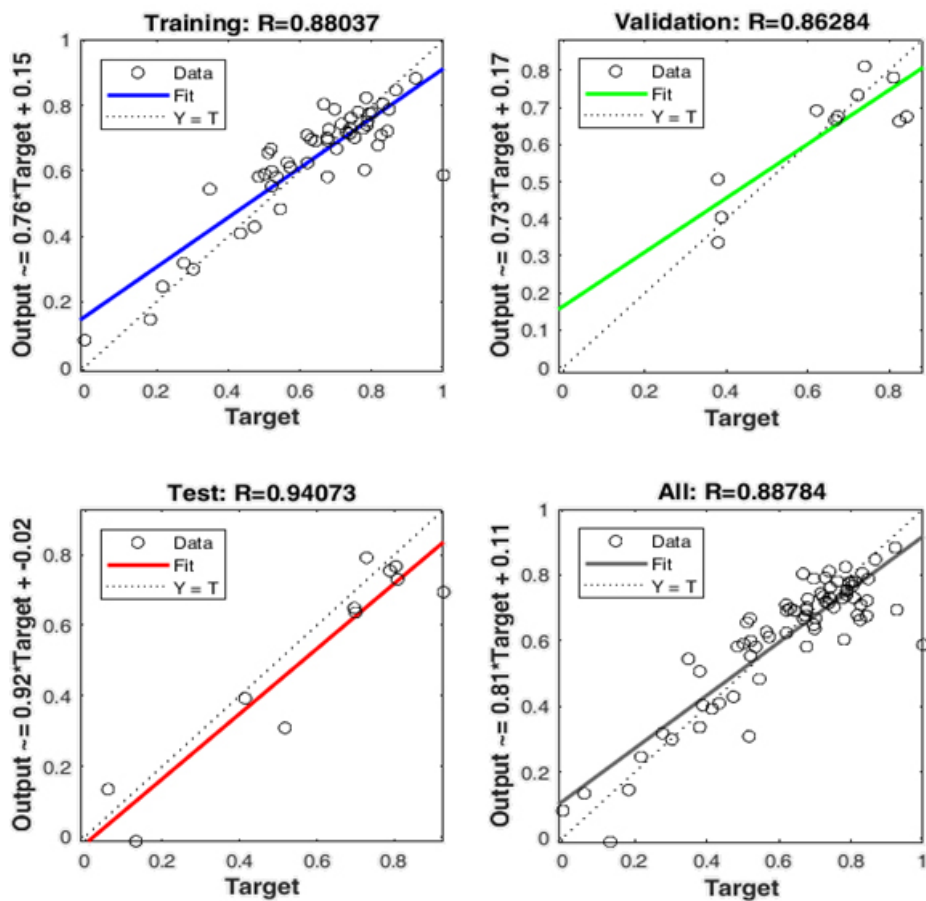


Figure 2. Regression values for the MLP artificial neural network

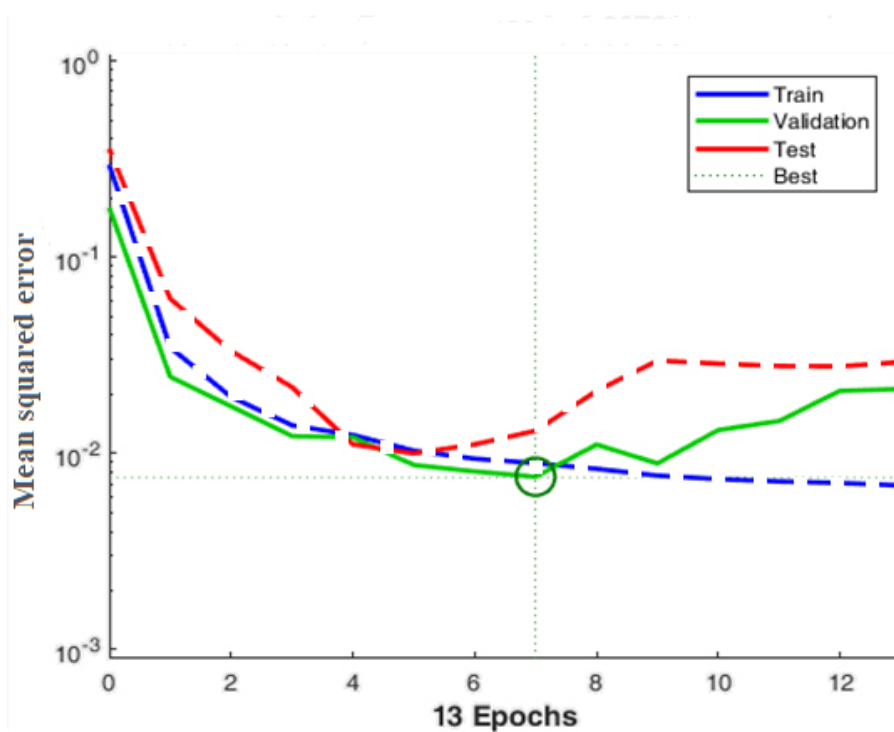


Figure 3. Mean squared error values (best validation performance is 0.0076034 at epoch 7)

The RBF neural network, like MLP, was designed to make a comparison, but the RBF transmission function is Gaussian. Figure 4 shows the output values of the RBF network regression against the target values. Also, Table 1 shows the mean squared error values for the MLP and RBF neural networks.

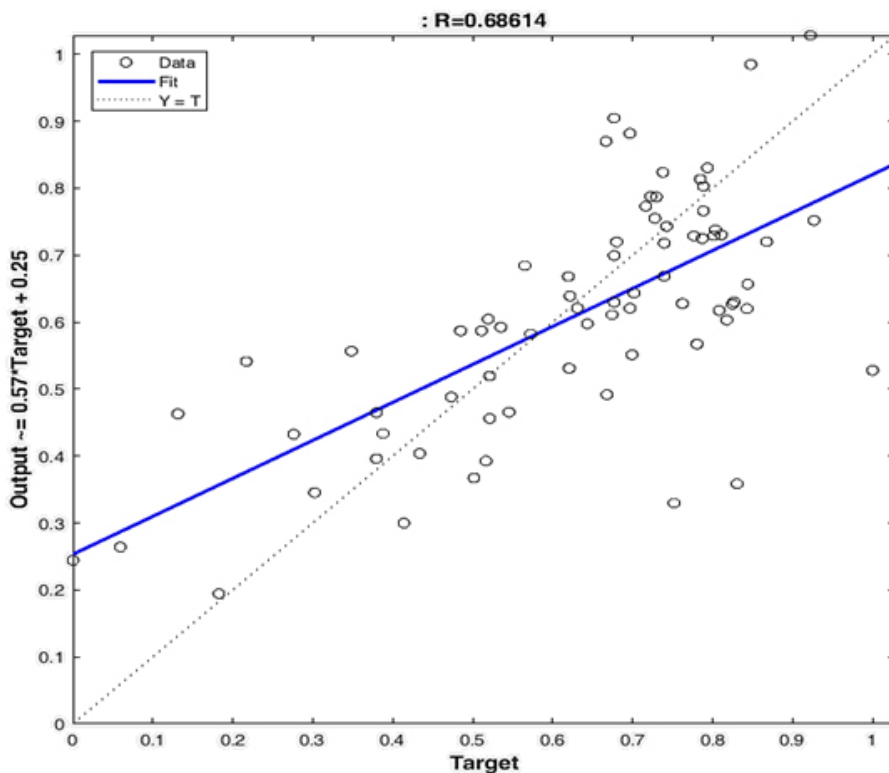


Figure 4. Regression values for the RBF neural network

Table 1. Mean squared error values for the MLP and RBF networks

Neural network	Mean squared error value
MLP	0.0070
RBF	0.0202

Taking these results into account, it is clear that the MLP neural network gives a better prediction than the RBF network. The electrical conductivity of some low-conductivity pure fuels and organic liquids was investigated by the MLP neural network method, and is presented in Table 2. As shown, the values predicted by the neural network are close to the measured values. Using this method, the electrical conductivity value of DMAZ was predicted as 9.1 and 8.8 $\mu\text{S}/\text{m}$ by the MLP and RBF methods, respectively. These are very close to the measured value of 10 $\mu\text{S}/\text{m}$ (as stated later). It should be noted that in some cases the error is high. This may be for two reasons:

- low number of data values used,
- use of more descriptors to better predict the electrical conductivity.

It is hoped that with the development of this method, the conductivity of the fuel mixture can be predicted.

Table 2. The results of the neural network prediction for some organic liquids

Material	Electrical conductivity [$\mu\text{S}/\text{m}$] [25]	Predicted electrical conductivity <i>via</i> neural network [$\mu\text{S}/\text{m}$]	Error [%]
Acetone	$6 \cdot 10^{-6}$	$6.09 \cdot 10^{-6}$	1.50
Ethanol	0.135	0.123	8.89
Toluene	$8 \cdot 10^{-7}$	$7.1 \cdot 10^{-7}$	11.25
Methanol	$40 \cdot 10^{-6}$	$44 \cdot 10^{-6}$	10.00
Diethyl ether	$30 \cdot 10^{-6}$	$30.4 \cdot 10^{-6}$	1.34
Water	5.1	5.32	4.31
Acetonitrile	$92 \cdot 10^{-3}$	$105 \cdot 10^{-3}$	12.38

4.2 Experimental results

4.2.1 Effect of amine surfactant additives

As was mentioned, the amine additives were investigated due to their compatibility with DMAZ. They serve as surfactant. Some surfactants are capable of increasing electrical conductivity due to the production of micelles or reverse micelles. Surfactants were thought to increase the electrical conductivity of liquid fuel DMAZ. For this reason, materials such as hexylamine, octylamine and tributyl amine, all of which have a polar head and a nonpolar head, were used. Hexylamine and octylamine were used to compare the chain length [27]. They were added to the fuel at various concentrations. Preliminary experiments showed the effective concentrations of the additives used were 3000, 4000 and 5000 ppm. At concentrations lower than 3000 ppm, no change in the electrical conductivity of the fuel was observed.

The electrical conductivity test data (in $\mu\text{S}/\text{m}$) are presented in Figure 5. As is shown in this figure, octylamine and hexylamine increased the electrical conductivity to a small extent, but tributylamine did not change the electrical conductivity of the test fuel.

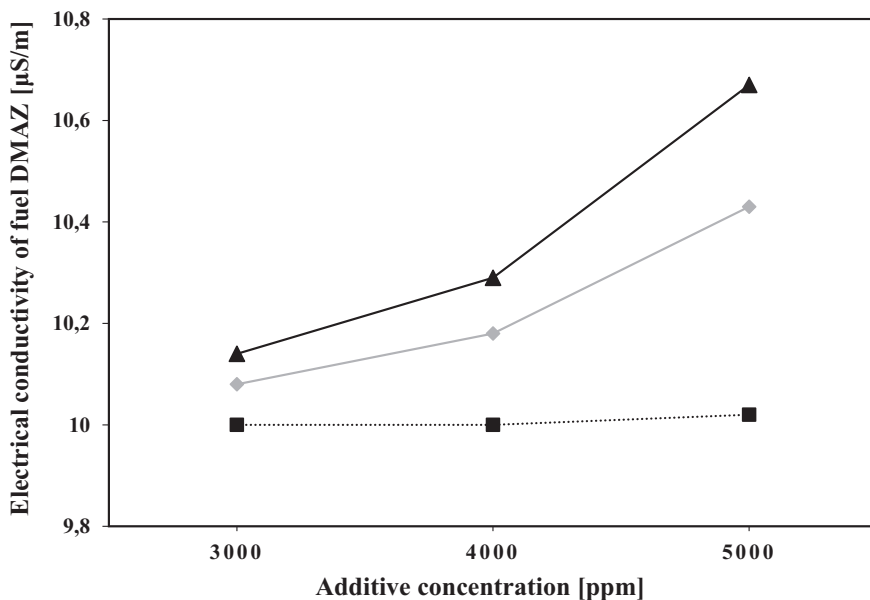


Figure 5. Effect of different concentrations of hexylamine, octylamine and tributylamine on the electrical conductivity of liquid fuel DMAZ (-▲- Octylamine, -◆- Hexylamine, ···■··· Tributyl amine)

There is the possibility of micelle formation in the fuel due to the addition of a surfactant. Therefore, one of the causes could be micelle formation and consequently increased electrical conductivity. By forming micelles and collisions between them, charged micelles could be formed which affect the electrical conductivity. As a consequence, the presence of micelles in the solution and their collision with each other transfers energy between them and may cause an electric charge to affect the electrical conductivity [28].

According to Figure 5, it is apparent that octylamine increased the electrical conductivity more than hexylamine. As the hydrocarbon chain is increased (octylamine *versus* hexylamine), the critical micelle concentration is decreased. Therefore, micelles are formed faster and are more effective in the transfer of electrical current. As a result, the mechanism on adding octylamine is similar to that of hexylamine, except that the critical micelle concentration for octylamine is lower [29].

There was no change in electrical conductivity on addition of tributylamine. This amine is unable to produce micelles because its hydrocarbon chain is small. As a result, it failed to increase the electrical conductivity.

4.2.2 Effect of aprotic and ionic liquid additives

The electrical conductivity of the test fuel increased on addition of acetonitrile (as a low price aprotic material) and DMAZ·HCl salt (Figure 6). Octylamine, the surfactant which had the highest effect on DMAZ's electrical conductivity, is shown for comparison.

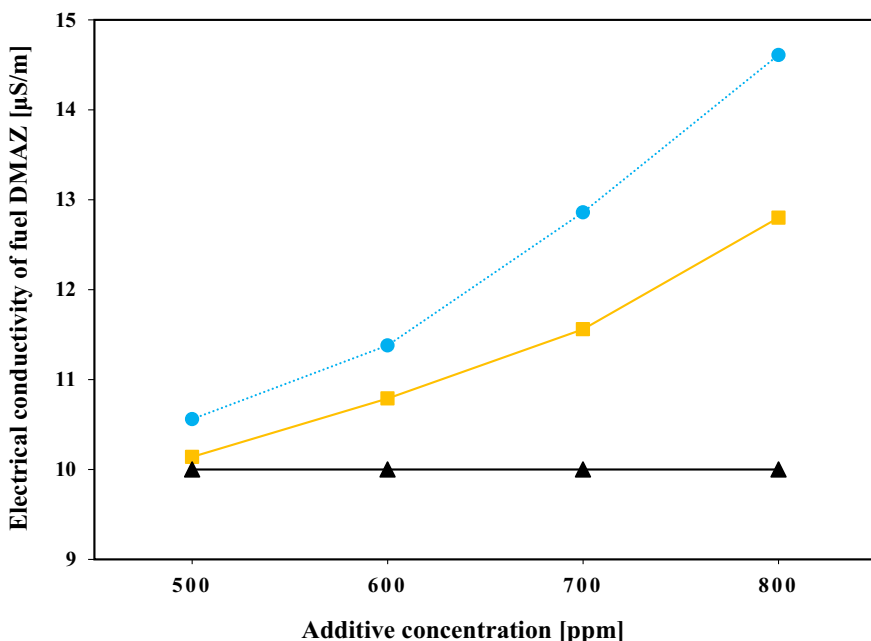


Figure 6. Changes in electrical conductivity of liquid fuel DMAZ on adding acetonitrile and DMAZ·HCl salt (···●·· acetonitrile, -■- DMAZ·HCl salt, -▲- octylamine)

Acetonitrile has a high dielectric constant (about 39) [26]. When the acetonitrile concentration is increased, the dielectric constant is increased [30], enhancing molecular dissociation of acetonitrile. Thereby, ion production will be increased, and hence the electrical conductivity of the fuel solution will be increased.

Of the ionic liquids, DMAZ·HCl salt has the least unwanted side-effects on the fuel properties. Therefore, this was selected as the ionic liquid additive. The addition of DMAZ·HCl salt increased the electrical conductivity of the fuel. One explanation for this effect is that by adding the ionic liquid, ions are produced which carry the charges in the fuel. Higher salt concentrations lead to greater production of ions and hence higher electrical conductivity of the fuel.

The average value of the dielectric constant for the majority of ionic liquids is 15.5 [30]. Acetonitrile has a greater dielectric constant than DMAZ·HCl salt (39 *versus* 15.5). Therefore, the electrical conductivity with acetonitrile is greater than with DMAZ·HCl salt. Figure 6 confirms this deduction.

By adding octylamine to the fuel, there was no change in electrical conductivity in the concentration range 500-800 ppm. So, as was stated, the concentration of octylamine would have to be increased to 3000-5000 ppm in order to show a significant increase in the electrical conductivity of the fuel.

4.2.3 Effect of a mixture of acetonitrile and DMAZ·HCl salt on the electrical conductivity of liquid fuel DMAZ

As was stated in the previous subsection, acetonitrile and DMAZ·HCl salt had the greatest effect on the electrical conductivity of the fuel. It may therefore be advantageous to use a mixture of these two compounds. The effect of adding the mixture (50/50 by weight) on the electrical conductivity of the fuel is demonstrated in Figure 7. The electrical conductivity with each individual material is also shown for comparison.

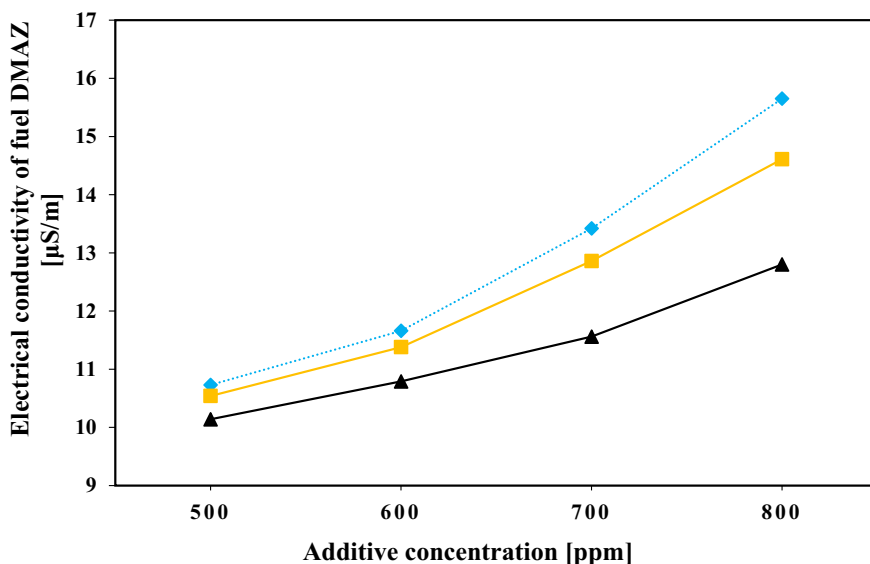


Figure 7. Electrical conductivity with a two-component mixture of acetonitrile and DMAZ·HCl salt compared with the conductivity with each individual component (··◆·· acetonitrile + DMAZ·HCl salt, -■- acetonitrile, -▲- DMAZ·HCl salt)

It is clear from Figure 7 that adding the two-component mixture had a greater effect on the electrical conductivity than when the two components were added separately. In other words, acetonitrile and DMAZ·HCl salt synergistically increase the electrical conductivity.

5 Conclusions

- ◆ The RBF and MLP artificial neural networks were used to predict the electrical conductivity of a novel liquid fuel 2-dimethylaminoethyl azide (DMAZ). The MLP neural network gave a better prediction than RBF.
- ◆ The electrical conductivity of some low-conductivity pure fuels and organic liquids was investigated by the MLP neural network method, and gave results close to the measured values. The MLP method predicted the electrical conductivity of DMAZ as 9.1 $\mu\text{S/m}$, compared to the experimental result of 10 $\mu\text{S/m}$. The increase in the electrical conductivity of DMAZ was then investigated after adding some chemical surfactants (such as hexylamine, octylamine, and tributylamine), acetonitrile as a polar aprotic substance and DMAZ·HCl salt as an ionic liquid. High concentrations of octylamine and hexylamine increased the electrical conductivity due to the formation of micelles. Tributylamine did not have a significant effect on the electrical conductivity of the fuel. Of the other additives, acetonitrile had the greatest effect on the electrical conductivity of the fuel, increasing the conductivity by about 45%.
- ◆ Adding a two-component mixture of acetonitrile and DMAZ·HCl salt, further increased the electrical conductivity of the fuel and enhanced DMAZ conductivity by about 55%, relative to pure DMAZ, compared to when each compound was added separately.

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