

Study of Surface Tension of Binary Mixtures of Poly (Ethylene Glycol) in Water and Poly (Propylene Glycol) in Ethanol and its Modeling Using Neural Network

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Abstract

In this work, the surface tension (σ) of aqueous solutions of PEGs (poly ethylene glycol) with molecular weights of 200, 300 and 6000 in water, and poly (propylene glycol) (PPG) with a molecular weight of 2000 in ethanol, were measured in the (293.2-338.2) K temperature range and atmospheric pressure. The results of the measurements were modeled by an Artificial Neural Network(ANN) with input of mass fraction of polymer and solution temperature. The predictions of the artificial neural network model fit the experimental data perfectly.

Keywords: *Surface Tension, Polymer Solutions, Polyethylene Glycol, Propylene Glycol Neural Network*

1. Introduction

Thermophysical properties are required for both engineering and product design. The surface tension of a liquid mixture is an important property which plays a vital role in the mass and heat transfer, like in liquid-liquid extractions, gas adsorption, distillation and condensation. In the chemical industries, it determines the quality of the products (i.e. coatings, paints, detergents, cosmetics and agrochemicals).

Aqueous polymer solutions, especially poly (ethylene glycol) + water systems, have found widespread applications, mostly because of their use in two-phase aqueous

systems for separation of binary mixtures [1-3]. Despite the success of the aqueous two-phase separation technique, data on the properties of interfaces that are necessary for the design of extraction processes and also for the development of models that predict phase partitioning are few [4]. Although reviewed surface tensions of pure liquid polymers and the interface properties of polymer-polymer systems are available [5-12], there is no available data for polymer-non polymer systems i.e., PPG-ethanol or PEG-water.

Neural networks have recently been widely used in many fields of engineering and

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science [13-20]. The important reason for this rapid development and various applications of neural networks is their suitability to evaluate nearly any function in a stable and efficient way. Therefore, they make a platform on which different models can be developed [14-16]. One significant problem in estimating the surface tension of pure liquids and their mixtures is an accurate theory that is computationally infeasible due to the use of many adjustable parameters that sometimes require a sufficient amount of data for calibration and validation purposes that make them computationally inefficient, hence only approximate estimations could be made. To date, there are no publications concerning the use of ANNs in surface tension on the polymer field. It is expected that a neural network approach would offer a helpful and beneficial new solution to solve this particular problem.

In this work, for the first time, the surface tensions of aqueous solutions of various PEGs (poly ethylene glycol) with molecular weights of 200, 300 and 6000 in water and of poly (propylene glycol) (PPG) with a molecular weight of 2000 in ethanol were measured in the (293.15-338.15) K temperature range and atmospheric pressure. The results of measurements were modeled by neural networks with polymer mass fraction and solution temperature as input.

2. Materials and methods

Absolute ethanol GR (>99.8%), poly (ethylene glycol) with average number molecular weights of 200 and 300 (stated purity $\leq 99.0\%$ (mass)) were purchased from Merck. Also, poly (ethylene glycol) with MW=6000 (purity $\leq 99.0\%$ (mass)) and

propylene glycol 2000 (purity $\leq 99.0\%$ (mass)) were from Fluka and Riedel-deHaen, respectively. The polymer's polydispersity was equal to 1 and double-distilled water was used in making the solutions.

The solutions were prepared in mass, using an analytical balance with ± 0.1 mg accuracy. The surface tension measurements were carried out using a thermostated tensiometer (model K9 Kruss Germany) with an accuracy of ± 0.1 mN/m, at temperatures of (298.2, 308.2, 313.2, 318.2, 328.2, and 338.2) K and atmospheric pressure. The tensiometer, which is equipped with a Du Noüy platinum-iridium ring (6 mm circumference), was calibrated with distilled water ($\sigma / \text{mN/m} = 72.8 @ T=25^\circ \text{C}$).

A constant temperature water bath was used to control the temperature of the solutions with an accuracy of ± 0.1 K. Measurements for each solution were repeated five times. It should be noted that density is needed to calculate its surface tension. The density measurements were carried out using a 10 cm³ glass pycnometer. The volume of the pycnometer was calibrated as a function of temperature using double-distilled water. The accuracy of the density measurements was 0.0002 g.cm³.

3. Artificial neural network

Artificial neural networks (ANNs) are highly flexible mathematical constructs that have been inspired by the workings of the biological nervous system. ANNs have a natural tendency for storing experiential knowledge and making it available for use [21]. ANNs can simply be viewed as general nonlinear models which have the ability to encapsulate the underlying relationship that

exists between a series of inputs and outputs of a system.

There are many different ANN structures like MLP (multi layer perception), RNN (recurrent neural network) and RBF (radial basis function). Each of these structures has been used for the modeling of different case studies [21].

Feedforward neural networks (FFNN) are undoubtedly the most common neural network structures used in engineering applications. It has been shown that a three-layer (input-hidden-output) FFNN can represent any function provided that a sufficient number of neurons are present [22]. The Feedforward neural networks that have been used in this examination are presented in Fig. 1.

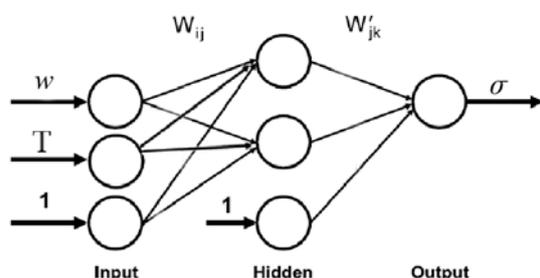


Figure 1. Feedforward neural networks used to represent temperature-dependent isotherm data sets.

The input layer receives the process inputs and fans out this information to all functional neurons of the hidden layer. Each neuron of the hidden layer essentially accomplishes two tasks: (1) a weighted summation of all process inputs; and (2) a non-linear transformation, via a neuron transfer function of the weighted summation to produce the output of each neuron of the hidden layer which then serves as input to the neurons of

the output layer. The output layer performs the same task as the neurons of the second layer to produce the final output of the FFNN. Typical transfer functions that are used in the hidden and output layers are linear, sigmoid or hyperbolic tangent.

4. Results and discussion

The measured surface tensions of different polymer solutions are reported in Tables 1-4. The results show that when the concentration of PEGs increases, surface tension decreases, but when the concentration of PPG2000 increases, surface tension increases. Also, in our selected systems, surface tension decreases with increasing temperature for any given concentration of alcohols or water, because as temperature rises, so does the kinetic energy of the molecules, and thus intermolecular attraction decreases. This increase in temperature represents the work necessary to pull the new molecules into the surface against the attractive forces of the interior. Thus, enrichment of the surface occurs, and the force of surface tension decreases. Several authors have correlated linear and nonlinear behavior of surface tensions in the binary mixtures with temperature [24-25].

In addition, many published investigations [26-29] have focused on the molecular dynamics simulation of PEG/water solutions to interpret the solubility of PEGs in water in different conditions. Due to flexibility of the PEG molecule, the conformation of PEG in aqueous solutions ranges from a random coil to a helical structure that appears under certain conditions of temperature and concentration. The helical structure in low temperature creates a segment of the PEG

chain which has a rather large dipole moment and could be expected to interact favorably with water, whereas the random coil structure in high temperature will have smaller or no dipole moments and can be

assumed to interact less favorably with water. This is another major reason why PEG decreases the surface tension of water with increasing of temperatures.

Table 1. Measured surface tension of PEG 200 + Water solutions at various temperatures/K and concentrations.

$T / K = 303.15$		$T / K = 308.15$	
W	$\sigma_{exp.} / (mN / m)$	W	$\sigma_{exp.} / (mN / m)$
0.1493	59.85	0.1493	58.81
0.222	57.88	0.222	56.85
0.2329	57.68	0.2329	56.64
0.3828	54.63	0.3828	53.58
0.4481	52.79	0.4481	51.75
0.467	52.05	0.467	51.08
0.5197	51.49	0.5197	50.46
0.5586	50.63	0.5586	49.62
0.6411	49.39	0.6411	48.28
0.712	47.58	0.712	46.54
0.7328	47.26	0.7328	46.22
$T / K = 313.15$		$T / K = 318.15$	
W	$\sigma_{exp.} / (mN / m)$	W	$\sigma_{exp.} / (mN / m)$
0.1493	57.87	0.1493	56.83
0.222	55.81	0.222	54.99
0.2329	55.59	0.2329	54.55
0.3828	52.65	0.3828	51.71
0.4481	50.82	0.4481	49.81
0.467	50.19	0.467	49.39
0.5197	49.45	0.5197	48.55
0.5586	48.72	0.5586	48.12
0.6411	47.12	0.6411	46.17
0.712	45.51	0.712	44.69
0.7328	45.19	0.7328	44.28
$T / K = 323.15$		$T / K = 328.15$	
W	$\sigma_{exp.} / (mN / m)$	W	$\sigma_{exp.} / (mN / m)$
0.1493	55.77	0.1493	54.66
0.222	54.07	0.222	53.3
0.2329	53.5	0.2329	52.56
0.3828	50.99	0.3828	50.36
0.4481	49.21	0.4481	48.19
0.467	48.49	0.467	47.43
0.5197	47.52	0.5197	46.44
0.5586	46.84	0.5586	45.49
0.6411	45.25	0.6411	44.23
0.712	43.98	0.712	43.17
0.7328	43.57	0.7328	43.06
$T / K = 333.15$		$T / K = 338.15$	
W	$\sigma_{exp.} / (mN / m)$	W	$\sigma_{exp.} / (mN / m)$
0.1493	53.66	0.1493	52.73
0.222	52.57	0.222	51.63
0.2329	51.42	0.2329	50.49
0.3828	49.25	0.3828	48.23
0.4481	46.93	0.4481	45.99
0.467	46.26	0.467	45.34
0.5197	45.41	0.5197	44.39
0.5586	44.48	0.5586	43.25
0.6411	43.09	0.6411	42.51
0.712	42.35	0.712	41.61
0.7328	42.65	0.7328	41.93

Table 2. Measured surface tension of PEG 300 + Water solutions at various temperatures/K and concentrations.

$T / K = 298.15$		$T / K = 303.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.108	60.03	0.108	59.09
0.1998	57.69	0.1998	56.66
0.2987	53.6	0.2987	52.67
0.3926	51.98	0.3926	50.92
$T / K = 308.15$		$T / K = 313.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.108	58.16	0.108	57.44
0.1998	55.71	0.1998	54.58
0.2987	52.27	0.2987	51.54
0.3926	49.92	0.3926	48.41
$T / K = 318.15$		$T / K = 323.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.108	56.73	0.108	55.46
0.1998	53.3	0.1998	52.42
0.2987	50.52	0.2987	49.51
0.3926	47.89	0.3926	46.81
$T / K = 328.15$		$T / K = 333.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.108	54.73	0.108	53.58
0.1998	51.44	0.1998	50.37
0.2987	48.38	0.2987	47.21
0.3926	46.29	0.3926	45.27
$T / K = 338.15$			
w	$\sigma_{\text{exp.}} / (mN / m)$		
0.108	52.58		
0.1998	48.57		
0.2987	46.35		
0.3926	44.19		

Table 3. Measured surface tension of PEG 6000 + Water solutions at various temperatures/K and concentrations.

$T / K = 298.15$		$T / K = 303.15$	
w	$\sigma_{exp.} / (mN / m)$	w	$\sigma_{exp.} / (mN / m)$
0.0399	59.87	0.0399	58.84
0.0601	59.02	0.0601	58.4
0.0982	58.26	0.0982	57.23
0.1974	57.13	0.1974	56.11
$T / K = 308.15$		$T / K = 313.15$	
w	$\sigma_{exp.} / (mN / m)$	w	$\sigma_{exp.} / (mN / m)$
0.0399	57.98	0.0399	57.25
0.0601	57.4	0.0601	56.37
0.0982	56.71	0.0982	55.79
0.1974	54.84	0.1974	53.69
$T / K = 318.15$		$T / K = 323.15$	
w	$\sigma_{exp.} / (mN / m)$	w	$\sigma_{exp.} / (mN / m)$
0.0399	55.74	0.0399	54.69
0.0601	55.35	0.0601	54.29
0.0982	54.73	0.0982	53.37
0.1974	52.65	0.1974	51.62
$T / K = 328.15$		$T / K = 333.15$	
w	$\sigma_{exp.} / (mN / m)$	w	$\sigma_{exp.} / (mN / m)$
0.0399	53.64	0.0399	52.61
0.0601	53.83	0.0601	52.22
0.0982	52.23	0.0982	51.19
0.1974	50.59	0.1974	49.47
$T / K = 338.15$			
w	$\sigma_{exp.} / (mN / m)$		
0.0399	51.27		
0.0601	51.42		
0.0982	50.17		
0.1974	48.28		

Table 4. Measured surface tension of PPG 2000 + Ethanol solutions at various temperatures/K and concentrations.

$T / K = 298.15$		$T / K = 303.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.1	22.14	0.1	21.54
0.2	22.44	0.2	21.84
0.3	22.64	0.3	22.14
0.4	22.94	0.4	22.44
$T / K = 308.15$		$T / K = 313.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.1	21.24	0.1	20.54
0.2	21.34	0.2	20.84
0.3	21.64	0.3	21.34
0.4	22.24	0.4	21.94
$T / K = 318.15$		$T / K = 323.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.1	20.34	0.1	19.84
0.2	20.44	0.2	20.13
0.3	20.84	0.3	20.44
0.4	21.44	0.4	21.24
$T / K = 328.15$		$T / K = 333.15$	
w	$\sigma_{\text{exp.}} / (mN / m)$	w	$\sigma_{\text{exp.}} / (mN / m)$
0.1	19.44	0.1	19.26
0.2	19.63	0.2	19.32
0.3	20.34	0.3	20.03
0.4	21.54	0.4	20.68
$T / K = 338.15$			
w	$\sigma_{\text{exp.}} / (mN / m)$		
0.1	18.78		
0.2	18.92		
0.3	19.42		
0.4	20.29		

Table 5. Weight matrices and bias for the systems used in this study.

System	Weight Matrix for Hidden Layer	Bias Matrix for Hidden Layer	Weight Matrix for Output Layer	Bias Matrix for Output Layer
PEG 200 + Water	3.7813 0.7806 0.0585 0.0311	-6.9526 -0.5766	8.0177 -16.1449	0.6021
PEG 300 + Water	0.3693 -15.6317 -12.0188	-0.1736 3.4368	-1.4571 -0.0194	0.7716
PEG 6000 + Water	-0.2070 -0.6522 57.1624 -3.1968	0.5810 -6.2828	1.1851 -0.0196	0.3482
PPG 2000 + Ethanol	2.6071 0.0285 -0.1391	-5.6801 -1.1840	0.1393 20.7577	18.1325

It may be suggested that the presence of extensive intermolecular H-bonding between water and PEG could be responsible for decreasing surface tension with increasing concentration of PEGs. The H-bonding between the $-O-$ of water and the termini $-H$ of PEGs and that between the $-H$ of water and the $-O-$ of the termini $-OH$ of PEGs could be assigned as the major contributor here. However, relatively weaker H-bonding between the $-H$ of water and the $-O-$ of the $-H_2C-O-CH_2-$ of PEGs may also have lower contribution. While for the mixture of PPG and ethanol, with increasing the concentration of PPG, the H-bonding of ethanol-ethanol and PPG-PPG and also PPG-ethanol is decreased in comparison to the interaction of $-H_2C$ with $-CH_2-$ of PPGs. Therefore, the surface tension of the mixture (PPG-ethanol) is increased with increasing concentration of PPGs.

Pure ethanol has lower surface tension (22.8 mN/m) than pure PPG (27.2 mN/m). In the primary solution of PPG and ethanol (0.1 mass polymer fraction), the ethanol concentration is higher than PPG in the surface, and therefore the surface has a lower surface tension (according to Eberhart [30] the surface layer of the liquid is enriched in the component of lower surface tensions). But an increase in the PPG concentration of the mixture leads to an increase in the intermolecular lipophilic interaction (methylene units) of ethanol and PPG and to a movement of the ethanol molecules to the bulk of the mixture; while the PPG molecules are moved in the opposite direction and are accumulated in the surface of the mixture. Thus, there is an increase in surface tension of the mixture. These results are indicated in

Table 4. Pure water has higher surface tension than pure PEG. In the primary solution of PEG and water, the water concentration is higher than PEG, and therefore has a high surface tension in the surface of the mixture. But an increase in PEG concentration of the mixture leads to a decrease in the surface tension according to the Eberhart [30] expression. Table 1-3 shows a decrease in surface tension of the mixture with an increase in PEG concentration.

The results were modeled by a neural network with three hidden neurons, including the bias. Using MATLAB Neural Network Toolbox, a feed-forward ANN model was designed using a back-propagation training algorithm. In this modeling, mass fraction of polymer (W) and the solution temperature (T) are represented as input variables and surface tension as the output variable. Series of data of PEG 200 with temperatures of 298.15, 303.15, 308.15, 313.15, 318.15, 323.15, 333.15 and 338.18 K, series of data of PEG 300, PEG 6000 and PPG 2000, with temperatures of 298.15, 303.15, 308.15, 313.15, 318.15, 333.15 and 338.18 K were used as the training data set to fit the mass fraction of the polymer-dependent neural network, and the remaining series for PEG 200 (323.15K), PEG 300, PEG 6000 and PPG 2000(323 K) were used as the validation data set to assess the performance of the neural network fractions for the data that were not used during the fitting process. Results obtained with many neural network structures with different transfer functions were very similar. Based on this, a decision was made to use a conventional structure with sigmoid transfer functions for the

hidden layer and a linear transfer function for the output layer. The input and output to the FFNN are usually scaled between 0.1 and 0.9. The learning process consists of determining the weight matrices W_{ij} and W'_{jk} that produce the best fit of the predicted outputs over the entire data set. Table 5 shows weight matrices and bias for the mentioned systems. In this investigation, the quasi-Newton optimization algorithm was used to fit the data. A neural network with a hidden layer having three neurons, including the bias neuron, was used to represent very accurately the surface tension data in all cases. As shown in Figs. 2-5, the neural network model is able to predict very well the observed data points for both the training and validation data sets.

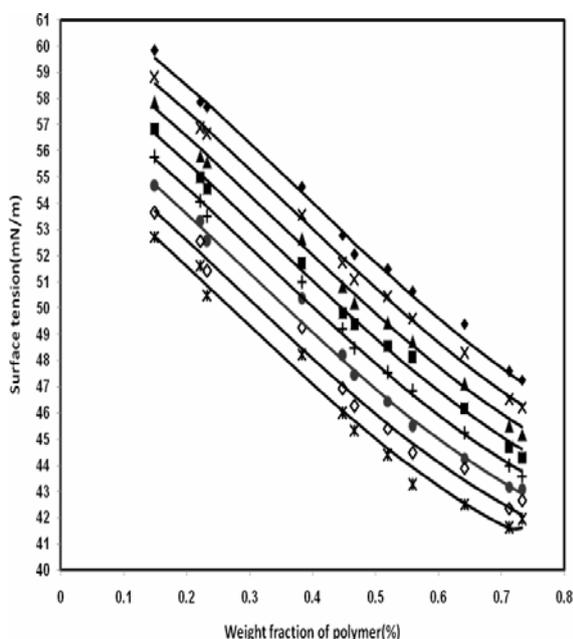


Figure 2. Surface tension of PEG 200 in water at different temperatures and concentrations : ,■, Δ,T=298.15 K; ◆,T=303.15 K; ×,T=308.15 K; ▲,T=313.15 K; ■,T=318.15 K; +,T=323.15 K; ●,T=328.15 K; ◇,T=333.15K; *,T=338.15; - Neural network.

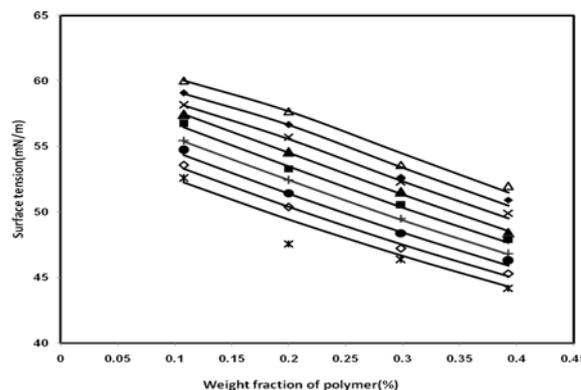


Figure 3. Surface tension of PEG 300 in water at different temperatures and concentrations■, Δ,T=298.15 K; ◆,T=303.15 K; ×,T=308.15 K; ▲,T=313.15 K; ■,T=318.15 K; +,T=323.15 K; ●,T=328.15 K; ◇,T=333.15K; *,T=338.15; - Neural network.

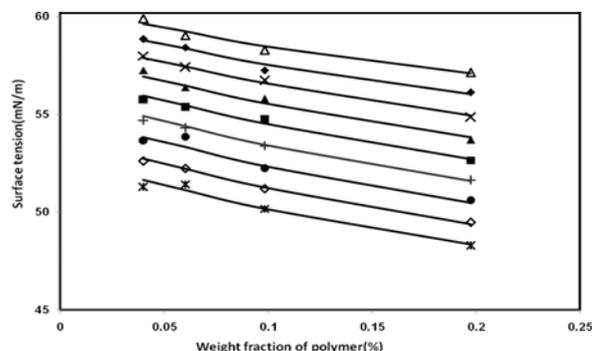


Figure 4. Surface tension of PEG 6000 in water at different temperatures and concentrations: ■, Δ,T=298.15 K; ◆,T=303.15 K; ×,T=308.15 K; ▲,T=313.15 K; ■,T=318.15 K; +,T=323.15 K; ●,T=328.15 K; ◇,T=333.15K; *,T=338.15; - Neural network.

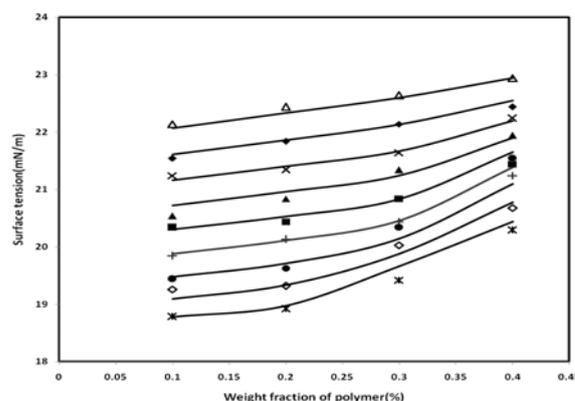


Figure 5. Surface tension of PPG 2000 in ethanol at different temperatures and concentrations: ■, Δ,T=298.15 K; ◆,T=303.15 K; ×,T=308.15 K; ▲,T=313.15 K; ■,T=318.15 K; +,T=323.15 K; ●,T=328.15 K; ◇,T=333.15K; *,T=338.15; - Neural network.

5. Conclusions

The surface tension (σ) of aqueous solutions of PEGs (poly ethylene glycol) with molecular weights of 200 at range 0.15 to 0.74 polymer mass fraction, 300 at range 0.1 to 0.4 polymer mass fraction and 6000 at range 0.04 to 0.2 polymer mass fraction in water at range 0.15 to 0.74 polymer mass fraction and poly propylene glycol (PPG) with a molecular weight of 2000 in ethanol at range 0.1 to 0.4 polymer mass fraction were measured in the (293.2-338.2)/K temperature range. The results of measurements were modeled by a neural network. In this modeling, the mass fraction of polymer and the solution temperature are represented as input variables and surface tension as output variable. The predictions of the artificial neural network model fit the experimental data perfectly.

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