

Determination of Surface Tension and Viscosity of Liquids by the Aid of the Capillary Rise Procedure Using Artificial Neural Network (ANN)

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ABSTRACT: *The present investigation entails a procedure by which the surface tension and viscosity of liquids could be predicted. To this end, capillary experiments were performed for porous media by utilizing fifteen different liquids and powders. The time of capillary rise to a certain known height of each liquid in a particular powder was recorded. Two artificial neural networks (ANNs) were designed and used to separately predict the surface tension and the viscosity of each liquid respectively. The surface tension predictor network had six inputs, namely: particle size, bulk density, packing density and surface free energy of the powders as well as the density of the probe liquids together with the capillary rise time of the liquids in the corresponding powders. The viscosity predictor network had surface tension as an extra input. In order to correlate the surface tension and viscosity as predicted by the corresponding artificial neural network to their experimentally determined equivalents, two different statistical parameters namely the product moment correlation coefficient (r^2) and the performance factor (PF/3) were used. It must be noted that for a perfect correlation $r^2 = 1$ and PF/3 = 0. The results of the present work clearly showed that the artificial neural network approach is able to predict the surface tension (i.e. $r^2 = 0.95$, PF/3 = 16) and viscosity (i.e. $r^2 = 0.998$, PF/3 = 13) of the probe liquids with unsurpassed accuracy.*

KEY WORDS: *Surface tension, Viscosity, Capillary rise method, Artificial neural network (ANN).*

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INTRODUCTION

Thermodynamic and transport properties of liquids are fundamental in processes involving liquid flow and heat and mass transfer. Two most important of these properties are surface tension and viscosity. Surface tension of a liquid characterizes the free energy per unit area required for the formation of a liquid-air interface at constant temperature, pressure, and composition. Viscosity of a liquid, on the other hand, characterizes its resistance to flow. Due to the vast number of chemicals in active use, and the lack of experimentally determined data for surface tension and viscosity there is an intensive and growing need for reliable and easily carried out procedures to provide such required data. The present study is an attempt to provide one such procedure.

Many approaches for determining the surface tension and the viscosity of liquids have been proposed [1-6]. One method, is the capillary rise procedure. The theoretical background of this procedure is as follows:

The first approach to analyse imbibition was reported by *Bell et al.* [7] who found a square root dependency of time on the height of imbibition. In 1921, *Washburn* [8] derived the time dependency of the height rise of a liquid into a single, straight line capillary of radius r . The derivation of this equation was based on Eq. (1). The capillary pressure causing the wetting phenomenon may be related to the liquid surface tension (γ_{lv}) and the contact angle (θ) by what is known as the *Laplace's* equation :

$$\Delta P_c = \frac{2\gamma_{lv} \cos\theta}{r} \quad (1)$$

where ΔP_c is the capillary pressure and r is the capillary radius. The *Washburn's* equation (Eq. (2)) was then derived by equating the capillary pressure to the pressure loss due to internal friction (*Hagan-Poiseuille's* equation [9]):

$$h^2 = \frac{r\gamma_{lv} \cos\theta}{2\eta} t \quad (2)$$

where h is the height to which the liquid has risen, η is the liquid viscosity and t is the time of capillary rise. In 1918, *Lucas* [10] independently derived the same equation.

Measurement of the viscosity of liquids by the

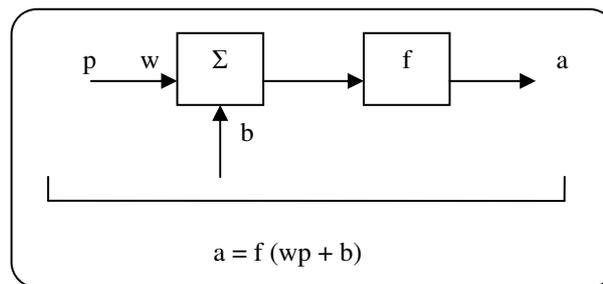


Fig. 1: The neuron model.

capillary rise procedure, was first proposed by *Levitt* [11]. *Levitt* determined the viscosity of liquids using the half-time of rise in a fine vertical capillary. Using this method viscosity of water, methanol, ethanol, benzene, acetone, and chloroform were measured at room temperature using marine barometer tubing. In 1998, *Rye et al.* [12] suggested an open capillary technique for simple measurement of the surface tension and the viscosity individually, as well as a ratio of the two. By this method viscosity and surface tension of 1,4-butanediol, cyclohexanol, 1-butanol, 2-octanol, diethylene glycol, 1-heptanol were calculated and compared with the literature values. In the present work, the performance of designed ANNs for the prediction of the surface tension and the viscosity of liquids using time of capillary rise to a certain height in porous media were put to test.

An artificial neural network (ANN) can be considered as a black box consisting of a series of complicated equations for the calculation of outputs based on a given series of input values. Therefore, the task of the network is to perform a set of mappings between its inputs and outputs. Neural networks consist of collections of connected processing elements or neurons. As shown in Fig. 1, the neuron consists of an input p , which is multiplied by a weight w , and then is summed by a bias b . The product is computed by a mathematical function f which determines a neuron output a . This mathematical function is called the activation or the transfer function.

Neural networks are currently being extensively applied in many fields of science and engineering [13-20]. The major reason for this rapid growth and diverse application of neural networks is their ability to estimate virtually any function in a stable and efficient way. Hence, they create a platform on which different models can be constructed. One main problem in predicting surface tension and viscosity of liquids is the

Table 1: Some physical properties and chemical nomenclature of the probe powders.

Powders	Active component	Particle size (μm)	Bulk density (kg/m^3)	Supplier
Aerosil 200	Silica	0.012	50	Degussa
Printex V	Carbon black	0.025	190	Degussa
Bayferrox 130	Iron oxide	0.17	1050	Bayer
Kemira 650	Titanium dioxide	0.22	900	Kemira
Ti-Pure R-900	Titanium dioxide	0.41	820	DuPont
Penta Calcium Carbonate	Calcium carbonate	1.9	700	Omya Pars
Super Talk WT-2500	Magnesium silicate	2.4	500	Sanaye Poodre Sangsar
Baryte Ba-2500	Barium sulfate	2.4	1060	Sanaye Poodre Sangsar
Kaoline KSP	Aluminium silicate	2.4	440	Khorasan Kaoline
Kaoline KCC	Aluminium silicate	3.8	610	Khorasan Kaoline
Syloid ED 30	Silica	5.5	130	Boysan
Super Talk WT-1500	Magnesium silicate	8.3	630	Sanaye Poodre Sangsar
Super Talk WT-800	Magnesium silicate	15	790	Sanaye Poodre Sangsar
Milky Calcium Carbonate-800	Calcium carbonate	15	820	Micronized Powder
Zinc Oxide Z-1	Zinc oxide	45	620	Farayand Powder

fact that the application of an exact theory is computationally impracticable, hence only approximate estimations could be made. It is expected that a neural network approach would offer a helpful and beneficial new solution to solve this particular problem.

MATERIALS AND METHODS

Materials

Imbibition experiments were carried out with porous columns made from fifteen packed powders. Physical and chemical properties of which are given in Ref. [21] and are summarized in table 1. The suppliers of these powders are also mentioned in table 1. The powders provide a wide range of particle size (i.e. 0.012-45 μm) and bulk density (i.e. 50-1600 kg/m^3). Some properties of the probe liquids used are shown in table 2. Formamide was provided by the Roth Company and all the other liquids except distilled water were provided by the Merck Company. The properties of these liquids were obtained from Ref. [22]. The liquids provide a wide range of surface tension (i.e. 15.45-71.99 mJ/m^2), and viscosity (i.e. 0.25-21 $\text{mPa}\cdot\text{s}$) and were used as received without any further purification.

Methods

Capillary rise experiments

Two different approaches for capillary rise experiments have been broadly employed: In the first approach, the weight increase caused as a consequence of the capillary rise in the porous media is evaluated; whilst in the second approach, evaluation is made of the length advanced by the liquid through the porous media. However, *Labajos-Broncano et al.* [23] proved that the measurement of mass or length in the experimental procedure give equivalent results and have no effect on the final outcome. In this work, the distance-time imbibition was employed.

The apparatus, depicted in Fig. 2, was used for the capillary rise experiments and consisted of a reservoir of a certain liquid and a graduated vertical cylindrical tube supported by a clamp. The cylindrical tube was in fact a 10-ml volumetric pipette with the bottom tip cut off. Glass wool was packed into the bottom of the tube to act as a support bed for each powder. The tube was then filled with a known mass of a given powder. For each powder, the weight and height were always kept constant by vibrating the tube to a certain volume fraction, in order

Table 2: Some properties of the probe liquids.

Liquids	Density (kg/m ³)	Surface tension (mN/m)	Viscosity (mPa.s)
Ethane-1,2-diol	1110	47.99	21
2,6,10,15,19,23- Hexamethyltetracosane-2,6,10,14,18,22-hexaene	860	31.85	15
Hexadecane	770	27.05	3.59
Formamide	1130	57.03	3.31
Diiodomethane	3320	66.18	2.8
Propan-2-ol	800	20.93	2.2
Nitrobenzene	1200	43.45	1.86
Distillated water	1000	71.99	1
Decane	730	23.54	0.81
Chlorobenzene	1110	32.99	0.8
Methanol	790	22.07	0.60
Ethyl acetate	900	23.39	0.44
Butanal	800	24.36	0.41
Acetone	790	23.46	0.32
Pentane	630	15.45	0.25
Glycerine ^a	1230	62.82	1412

a) Glycerine was only used to determine the surface free energy of Printex V (i.e. Carbon black powder) since the times of capillary rise for the other liquids were too small.

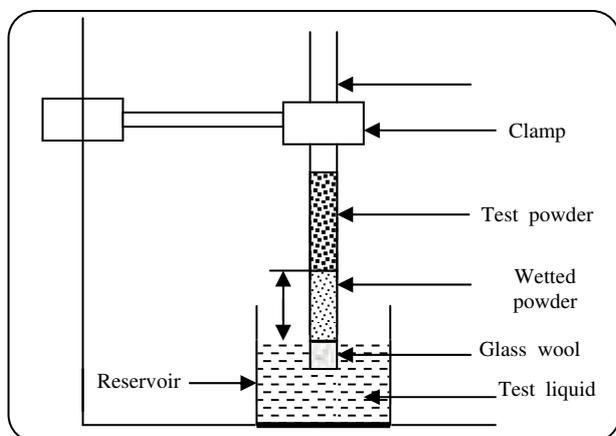


Fig. 2: Schematic representation of the apparatus for the capillary rise experiments.

to ensure a constant packing density. Then, the tube was lowered into the reservoir and a time of zero was recorded when the liquid first touched the powder. For each liquid, the penetration time to a known given height of powder column was then recorded. Before using the tube for the next experiment, the tube was cleaned

carefully with cleansing solution followed by repeated rinsings with distilled water and finally with acetone. The tube was then thoroughly dried in an oven. When the tube was not in use, it was covered at both ends to prevent the smallest traces of dust from entering. All column wicking experiments were performed at the temperature of 25 °C. The temperature of the system was controlled by a Multi Temp III thermostat with a precision of ± 0.1 °C. Each reported value is the average of three independent measurements for liquids for which small standard deviations were observed. In the case of liquids where large standard deviations were obtained, the reported value is the average of five to six measurements.

In any case, it was ensured with a 95 % confidence limit that the standard deviation was always less than 8 % of the measured values.

Determination of surface free energy of the powders

Since surface free energy of the powders (γ_{sv}) plays a critical role in analysing the diffusion of liquids in porous media and its value is usually not provided by the

supplier, then it had to be determined separately. This was carried out with reference to Eq. (2). The radius r in this equation was replaced by K , and this substitution was assumed to account for all porous media nonidealities such as variations in the porous media porosity and the tortuous nature of the flow path. With such a change, the modified Lucas-Washburn equation becomes:

$$h^2 = \frac{K\gamma_{lv} \cos\theta}{2\eta} t \quad (3)$$

It is evident from Eq. (3) that $2\eta h^2/t$ equals $K\gamma_{lv} \cos\theta$, therefore a plot of $2\eta h^2/t$ versus the liquid surface tension (γ_{lv}) of the test liquids used, would give valuable information regarding γ_{sv} . In fact this plot will show a maximum which is analogous to the γ_{sv} of the investigated material [24-27]. Indeed, it is equated with the largest value of a liquid surface tension that yields $\cos\theta=1$. It must be noted that the penetration time for each liquid to reach a certain height for each powder was considered. The estimated surface free energies are also summarized in table 3.

Prediction of surface tension and viscosity of the liquids using artificial neural networks

The Neural Network Toolbox of MATLAB 7.0 [28] was used for the prediction of surface tension and viscosity of the liquids. Two artificial neural networks (ANNs) were used separately to predict surface tension and viscosity of the liquids. The network used for surface tension determination contained six inputs corresponding to the density as well as the time of the capillary rise to a certain height, in the corresponding powders of known particle size, bulk density, packing density, and surface free energy. The network employed for the viscosities included an extra input, namely, the surface tension.

Two initial preexamination steps were undertaken prior to the prediction by ANNs.

As a first step, the Curve Fitting Toolbox of MATLAB 7.0 was used to correlate each input variable of the network to its equivalent output. The results showed that use of only one input variable did not correlate well with the outputs of the networks.

In the second step, the principal components analysis (PCA) of the Statistics Toolbox of MATLAB 7.0, was used in order to determine the minimum number of input variables required to give the maximal accurate

Table 3: Estimated surface free energy of the probe powders.

Powders	Surface free energy (mJ/m ²)
Kemira 650	35.6
Super Talc WT-2500	28.1
Baryte Ba-2500	27.9
Penta Calcium Carbonate	28.2
Aerosil 200	58.3
Printex V	63.9
Syloid ED 30	27.8
Ti-Pure R-900	35.4
Bayferrox 130	26.0
Zinc Oxide Z-1	25.5
Kaoline KSP	27.6
Super Talc WT-800	27.9
Milky Calcium Carbonate-800	28.3
Kaoline KCC	29.2
Super Talc WT-1500	28.1

prediction. All the possible number of variables from two to a maximum of all inputs for each network were used. In this investigation, the results showed that compression of data was not possible and only when all variables were used, the errors were minimized.

Since the number of inputs for the networks were determined by statistical means, the next step of the work was to determine the most appropriate architecture for the networks. For each network, several networks were created, trained and tested. The number of layers, the optimum number of neurons per layer and the transfer function(s) in the hidden layer(s) for each network, were obtained by trial and error. Care was taken to avoid overtraining. If overtrained, in a sense, the ANN adapts to the training data too well, so that further improvements of the ANN based on the training data will no longer provide further improvement in the prediction of the testing data. Therefore it was ensured not to include more weights and biases in both networks than the number of data in the training set. The 15 liquids with 154 data

points were divided in two groups: a training set (11 liquids with 138 data points) and a testing set (4 liquids with 16 data points). Networks were trained using the Levenberg-Marquardt back propagation algorithm. A linear activation function was used in the output layer of the networks. All the input and output data were normalized to the interval [-1 to 1] before training and testing. The performance factor (PF/3) and the product moment correlation coefficient (r^2) were used in order to assess the accuracy of the experimental data in comparison with the data predicted by the trained networks. The performance factor (PF/3) was derived by Guan *et al.* [29] and in fact combines three different statistical measures of fit (suitably weighted) into a single numerical value which is given in Eq. (4). This facilitates ease of comparison by declaring the errors involved in the predictions, as follows:

$$PF/3 = \frac{100 \times \left[\gamma + V_{AB} + \frac{CV}{100} - 1 \right]}{3} \quad (4)$$

where CV is the coefficient of variation, γ was proposed by Coates *et al.* [30], and V_{AB} was derived by Schultz [31], and each is respectively given in Eqns. (5) to (7):

$$CV = \frac{\sqrt{\frac{1}{N} \sum (X_i - fY_i)^2}}{\bar{X}} \times 100 \quad (5)$$

$$\text{where } f = \frac{\sum X_i Y_i}{\sum Y_i^2}$$

$$\log(\gamma) = \sqrt{\frac{1}{N} \sum \left(\log\left(\frac{X_i}{Y_i}\right) - \log\left(\frac{X_i}{Y_i}\right) \right)^2} \quad (6)$$

$$V_{AB} = \sqrt{\frac{1}{N} \sum \frac{[X_i - (FX_i)]^2}{X_i F Y_i}} \quad (7)$$

$$\text{where } F = \sqrt{\frac{\sum \frac{X_i}{Y_i}}{\sum \frac{Y_i}{X_i}}}$$

where N is the number of data set, and X_i and Y_i are values for element i of data set X and Y, respectively.

It must be noted that for a perfect correlation $r^2 = 1$ and PF/3=0. The number of epochs (i.e. the number of

presentation of a set of training (input and/or target) to a network for the calculation of new weights and biases) were fixed at 2000. At the same time, the initial values of the weights and biases were always initiated from random values, therefore, each run might have produced different output values. Therefore, each artificial neural network was made to run five times and the average value for r^2 and PF/3 were reported. Results of this investigation depicted that both ANNs having a hidden layer of 12 neurons had the maximum r^2 and the minimum PF/3. Also, in both cases, the tan-sigmoid transfer function applied to calculate the output value of the neurons of the hidden layer gave the best result. Therefore these architectures were selected for the rest of the work. Figs. 3 and 4 show the learning progress of the selected ANNs with time.

RESULTS AND DISCUSSION

Figs. 5 and 6 show the experimentally determined surface tensions and viscosities, respectively, at 25 °C of the liquids versus the predicted values using the corresponding neural network. The present approach of artificial neural networks were able to predict with high accuracy the surface tension (i.e. $r^2 = 0.95$, PF/3 = 16) as well as the viscosity (i.e. $r^2 = 0.998$, PF/3 = 13) of the probe liquids. These results indicate that neural networks can be considered as efficient tools for calculating surface tension and viscosity within the working range. Furthermore, prediction of surface tension and viscosity of liquids using ANNs has the following further advantages:

1- Common methods for surface tension measurement of liquids include:

(i) Force methods that derive the surface tension from the weight of the entrained meniscus at the perimeter of a device, such as a Wilhelmy plate or Du Noüy ring, impinging upon the interface.

(ii) Pressure methods that determine the surface tension from the maximum pressure required to blow a bubble.

(iii) Shape methods that analyse the distortion of a droplet surface as a function of surface tension (e.g., spinning drop, pendant drop and sessile drop methods).

A drawback of these methods is that the macroscopic sizes of these interfaces mentioned above require relatively large amounts of materials for the preparation of films and can also require long monolayer equilibrium

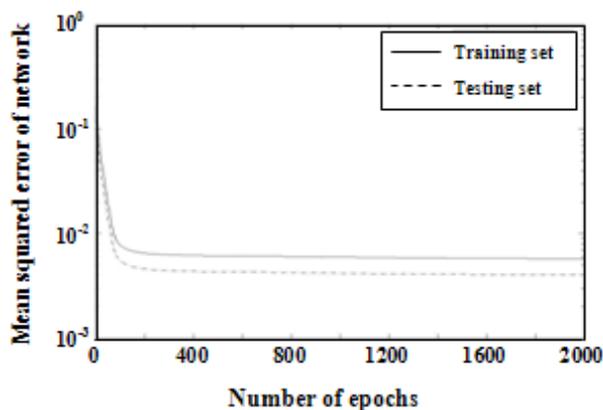


Fig. 3: Plot of MSE versus number of epochs for both training and testing data of the first designed ANN.

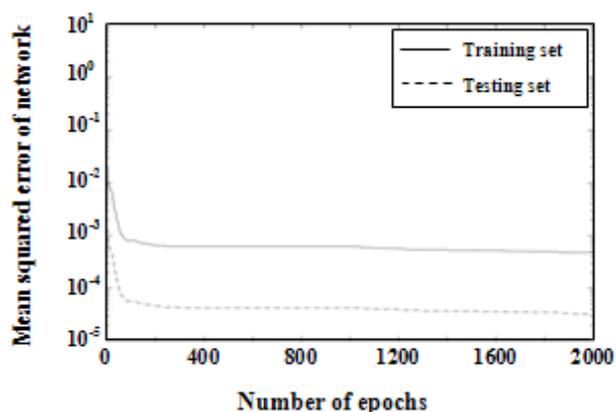


Fig. 4: Plot of MSE versus number of epochs for both training and testing data of the second designed ANN.

times. While the present approach requires relatively little amounts of materials and times of experimental procedure are relatively small.

2- In the past few years, researchers have developed experimental procedures for the determination of surface tension and viscosity of liquids mixture [32-35]. Since, ANNs have the ability to generalize beyond the training data, designed and trained ANNs are able to predict the surface tension and the viscosity of liquid mixtures at 25 °C and ambient pressure.

3- A further advantage of ANN is its inherent fault tolerance, in other words, the overall performance is not affected significantly even if a few data are abnormal because of experimental errors.

4 - Application of this procedure is easy and simple and requires only a representative database of properties for the powders and the liquids. Determination of the

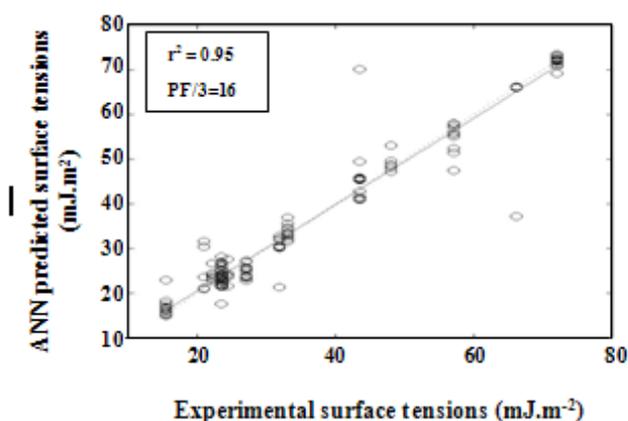


Fig. 5: Plot of surface tensions of the liquids predicted by ANN versus their experimental values.

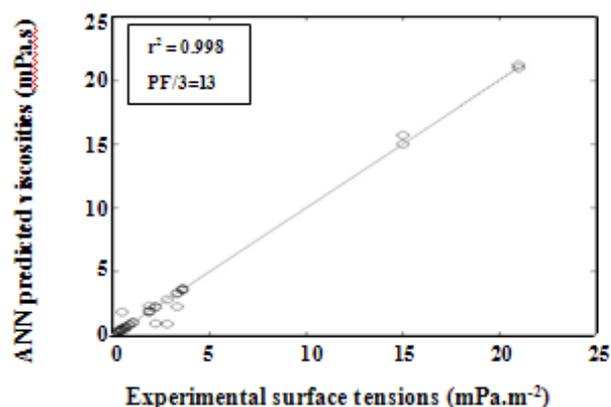


Fig. 6: Plot of viscosities of the liquids predicted by ANN versus their experimental values.

time of rise of liquids to a certain height of powder columns, and the software to create and train adequate networks able to learn the relationship between the liquid properties and network's inputs is easily attainable.

It should be noted that in order to estimate the value of viscosity of a liquid by this approach, the value of its surface tension must be known. If both viscosity and surface tension of a liquid were unknown, this method would still be beneficial to calculate these quantities. In this case, the value of surface tension is initially determined by the aid of an initial first neural network. Then, the value of viscosity of liquid would be achieved using a final second neural network.

CONCLUSIONS

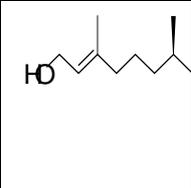
An approach has been devised and used to determine the surface tension and the viscosity of liquids. In this

approach surface tension and viscosity of liquids were predicted using artificial neural network (ANN) by the aid of the capillary rise procedure. The results showed that the ANN was able to predict with high accuracy the surface tension (i.e. $r^2 = 0.95$, PF/3 = 16) as well as the viscosity (i.e. $r^2 = 0.998$, PF/3 = 13) of liquids.

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