Indian Journal of Fibre & Textile Research
Vol. 41, March 2016, pp. 67-72

Predicting hydrophobicity of silica sol-gel coated dyed cotton fabric by artificial neural network and regression

Samander Ali Malik[a], Sidra Saleemi[b] & Naveed Mengal

Department of Textile Engineering, Mehran University of Engineering & Technology, Jamshoro 76062, Sindh, Pakistan

Received 13 August 2014; accepted 19 September 2014

Artificial neural network (ANN) and multiple linear regression (MLR) have been used to predict the hydrophobicity of silica sol-gel coated dyed cotton fabric using different nanoparticle concentrations, dye concentrations, dye types and cross linker types as predictors. A total of 32 samples have been dyed with reactive and direct dyes using two dye concentrations at HT dyeing machine. To develop nano roughness on dyed fabric, with an aim to create super hydrophobic dyed cotton, different concentrations of silica nanoparticles with a combination of silane hydrophobes (alkyltrialkoxysilanes), and silane cross-linkers, i.e. tetraethoxysilane (TEOS) and teramethoxysilane (TMOS) are applied by sol-gel technique using dip-dry-cure process. The hydrophobicity is measured by AATCC spray rating technique. The coefficient of determination (R^2) indicates that there is a strong correlation between the measured and the predicted values with a trivial mean absolute error; ANN is found to be more powerful predicting method than MLR. The most influencing variables revealed through correlation coefficient and *P-values* of regression model are silica nanoparticle and dye concentration. Empirical and statistical models have been proposed to predict dyed cotton fabric hydrophobicity without any prior trials, which reduces cost and time.

Keywords: Cotton, Hydrophobicity, Neural network method, Regression method, Silica nanoparticle, Sol-gel coated fabric

1 Introduction

Hydrophobic textiles are becoming today's need, especially when exposed to rainy and snowing environment. Different finishing agents have been developed to meet the required hydrophobicity in the finished textile goods, such as silicones, fluoro chemicals, PVA (polyvinylalchohol) and waxes. These fluoro base chemicals are expensive and environmentally hazardous, because during curing process due to the emission of non-volatile fluorinated compounds, it can yield the risk for health and skin diseases; whereas, other treatments are non-durable[1, 2].

Treatment with nanosol solutions is not only suitable to maintain the existing properties of textile materials but it also enhances other properties such as repellency, soil release, biological and UV-resistance[3]. Durability towards repellency of textile coated material was improved using silica nanopaticles and silane additives through sol-gel route on desized, scoured and bleached cotton samples[4]. Furthermore, when silica nanoparticles are applied on dyed cellulosic substrate by sol-gel cross-linking process, it forms thin smooth film on fabric surface with high degree of homogeneity but it can alter color of dyed substrate[5].

Previously, research has been conducted to make the cellulosic fabrics hydrophobic using nanosols considering contact angle measurement for bleached and dyed fabrics[3-6]. Spray rating method is very appropriate, because this resembles to the actual environment for which hydrophobic/water repellent fabrics are designed, simulating to rainy and snowing environment. Therefore, in the present domain of research, a combination of silica nanoparticles, silane hydrophobes (alkyltrialkoxysilanes), and silane cross-linkers, i.e. tetraethoxysilane (TEOS) and tetramethoxysilane (TMOS) were applied on 100% cotton reactive and direct dyed (0.5% and 3% o.w.f) fabric. The hydrophobicity of coated samples was evaluated through AATCC spray rating test. The detailed experimental outcomes have already been published[7, 8], while this study only deals with the development of empirical and statistical models for prediction of hydrophobicity.

As silica sol-gel coating recipe consists of different constituents, the interaction between these variables and fabric hydrophobicity is complex. So, it is

[a]Corresponding author.
E-mail: samander.malik@faculty.muet.edu.pk
[b]Present address: Department of Textile Engineering and Technology, University of the Punjab, Lahore.

difficult to determine the relation analytically. Therefore, neural network and regression techniques are used to predict hydrophobicity of silica nanoparticle coated dyed cotton fabric. ANN is an adaptive system which simulates the biological nerve cell system. It understands the underlying complex interactions between input and output variables through supervised learning process by adjusting its synaptic weights, otherwise it is not possible by any analytical method. Neural network has already been used in textile related applications. It is used to predict H_2O_2/UV and textile dye solution de-colorization processes[9,10], dyeing process optimization, and fabric quality characteristics prediction[11] and computer match prediction of fluorescent dyes[12]. It is important to judge the efficiency of ANN models, and for this purpose several researchers[13-15] have compared ANN with MLR technique.

From the literature survey, it is evident that ANN and MRR techniques have been widely used to predict different properties of textiles. In the present study, it is explored that these empirical and statistical models will help the manufacturers of water repellent goods to predict dyed cotton fabric hydrophobicity without any prior trials, which also reduces cost and time.

2 Materials and Methods

2.1 Materials

A commercially available ready-to-dye cotton plain woven fabric (136 g/m^2) was used as sample substrate and commercial reactive dye (Drimarene Reactive Red 5B) and direct dye (Indosol Red-BA) were used as colorants. The nanosol solution was prepared from silica nanoparticals (Aerosil® 200), silane hydrophobe (*n*-octyltriethoxysilane) with tetraethoxysilane (TEOS) and/or tetramethoxysilane (TMOS) as cross-linker, while ethanol (96%) and 0.01 N hydrochloric acid were used as solvent and *p*H controller respectively.

2.2 Dyeing of Samples

Bleached cotton fabric samples (7 g) were dyed (0.5% and 3% o.w.f) with reactive dyes (Drimarene Reactive Red-5B) and direct dye (Indosol Red-BA) on Rapid HT dyeing machine by exhaust method. Dyeing was done at liquor-to-goods ratio of 20:1, while addition of other dyeing auxiliaries and chemicals, washing, drying and curing were followed according to guidelines provided by manufacturer (Clariant Pakistan Pvt. Limited).

2.3 Sol Preparation

The silica sols were prepared according to the method as mentioned earlier[4, 6] using Ultrasonic cleaner (FRT-200B). The sols were made using 0- 0.2% (on weight of bath) silica nanoparticles, 4 mL silane hydrophobe, 20 mL silane cross-linker, 96 mL reagent grade ethanol (96%) as solvent and 24 mL hydrochloric acid (0.01 N). Except silane hydrophobe, all ingredients were mixed in a beaker. Two hours later, silane hydrophobe was added to the solution. To make the sol-gel ready for application, the solution was subjected to ultrasonic treatment for 30 min in chilled or ice bath using an ultrasonic probe.

By varying the concentration of silica nanoparticles (0, 0.02, 0.1 and 0.2 % on weight of bath), a series of sols were prepared and applied on samples by dipping the fabric in solution evenly, dried at room temperature and cured at 100 $^{\circ}$C for 1 h in laboratory oven[3]. The treated samples were then stored in air tight polyethylene bags till they were tested.

2.4 Feed Forward Backpropagation Artificial Neural Network

ANNs are adaptive models that are able to learn by examples, the underlying complex interaction between input variable (s) and output variable (s), otherwise not have any analytical solution. As compared to other methods, ANN has advantage over its theoretical and statistical counterparts, because of its example based iterative learning. Feedforward backpropagation artificial neural network is widely used in textile field to solve function approximation, process optimization and prediction problems.

Feedforward backpropagation is so called because during network training, the information flows in forward direction while error propagated back to hidden layers, this iterative process will continue till any specified condition is reached.

2.4.1 Network Training

For training neural network model, multilayer feedforward neural network architecture was designed into ANN toolbox function ‘trainlm’ integrated in MATLAB. It is an incorporation of the Levenberg-Marquardt (LM) algorithm into backpropagation to train the neural networks. Backpropagation is the steepest descent method, the biases and network weights move in the opposite direction to the error gradient. Backpropagation have drawback of slow convergence rate and local minima in error surface[16]. Therefore, to speed up the backpropagation learning, LM technique is commonly used. In LM technique, to change weights, the following updated rule is used:

$$\Delta W = (J^T J + \mu I)^{-1} J^T e \qquad \ldots (1)$$

where J is the Jacobian matrix of derivatives of each error to each weight; μ, the scalar; e, the error vector; I, the identity unit matrix; and superscript T, the matrix transposition. Further detailed description is available in published research [17, 18].

Multilayer feedforward ANN with two hidden layers was trained in Matlab toolbox function 'trainlm'. As there is no certain rule for selection of network structure, number of layers, neurons in layers, learning rate and momentum rate were decided on hit and trial basis after training several networks. The architecture and parameters of developed model are given in Fig.1 and Table 1. Sigmoid (logsigmoid) and linear transfer functions were used in the hidden and output layers respectively, while mean square error (MSE) was used as performance function. To analyze the error in real values it was recorded in the form of mean absolute error (MAE). As there were two nominal input variables, for network training they were systematically coded into 0/1 direct coding system, although for ANN training other coding system (1/2 or 1-4) can be used but that would not be suitable for development of regression equations. In this coding system, 1 indicates presence of variable's category and 0 otherwise; further detail is given in Section 2.6. For better generalization, the experimental data were pre-processed to 0 and 1, then after training it was post processed to original form using suitable matlab codes. The experimental data were randomly divided into two sets, i.e. training and testing.

Table 1—Network parameters

Network	:	$4\text{-}[\text{-}3\text{-}2]_2\text{-}1$
Transfer functions	:	{logsig,logsig,purelin}
Inputs	:	4
Output	:	1
Learning rate	:	0.3
Mu	:	0.5
Epochs	:	2000
Performance goal	:	0.001
Training function	:	'trainlm'

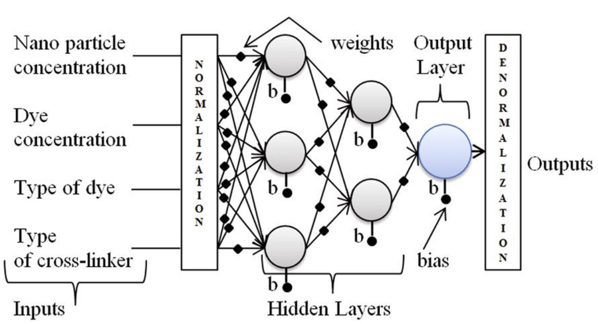

Fig. 1—Architecture of ANN model [b – bias term and ( ) – synaptic weight]

2.5 Multiple Linear Regression

Multiple linear regression (MLR) is commonly recognized statistical prediction technique in textile technology. The advantage of this technique is to give a quantitative equation of developed model and also reflects the significance of model and individual input variables. Therefore, in present study, MLR has been used to compare the outcome with ANN model and to establish a quantitative relationship between input variables to the hydrophobicity of dyed cotton treated by silica nanoparticle through sol-gel route.

2.6 Introducing Categorical Dummy Variables

Modeling, especially multiple regression requires regressors as well as dependent variables in the numeric form. As the dye type and cross linker are the nominal variables, to develop models, numerical variables must be introduced in their place by using suitable coding methodology. As they are categorical variables of type dichotomous (can take only two values, i.e. 0 or 1), the values 0 and 1 are used as dummy variables to develop ANN and regression model. The general model will take the form as stated in following equation:

$$\text{Fabric hydrophobicity [AATCC spray rating]} = \beta_0 + \beta_1 \times (\text{Silica conc.}) + \beta_2 \times (\text{Dye conc.}) + \beta_3 \times D_1 + \beta_4 \times D_2 \qquad \ldots (2)$$

where β_0 is the intercept; β_1-β_4, the coefficients of corresponding variable; D_1, the dummy variable for dye type (1 is indicative of reactive dye and 0 represent direct dye); and D_2, the dummy variable for cross linker type (1 represents TEOS and 0 is for TMOS).

2.7 Measurement of Water Repellency

Water repellency of 100 % cotton, dyed and sol coated, samples was assessed by spray rating test according to standard test method as recommended by AATCC (AATCC 22 - 2010). Water repellency AATCC rating scale ranging from AATCC 50 (highly absorbent) to AATCC 100 (highly hydrophobic) is given in Table 2.

3 Results and Discussion

3.1 Fabric Hydrophobicity Modeling with Artificial Neural Network

The influencing input variables and their values are given in Table 3. For neural network prediction, the data was divided into training and test sets. From 32 data points, 26 data pairs were selected for network training and remaining six pairs were used to test the developed model. The performance of trained model was judged on unseen data by analyzing coefficient of determination (R^2) and mean absolute error (MAE). The developed model and its performance on novel data is presented in Figs 2(a) & (b) and Table 4. The results reveal that there is a very strong correlation between the experimental values and the neural network responses. The trend lines of predicted and experimental spray rating on training and novel data show excellent correlation and reveal no over-fitting situation, which reflects the legitimacy of ANN.

Furthermore, Table 4 expresses the coefficient of determination (R^2), percentage mean absolute error and absolute error (in terms of AATCC spray rating) of training and test sets. The coefficient of determination (R^2) shows that 99% of the variability in the predicted variable in the training and test set is explained by the model predictors. This shows the significance of predicting variables used in the present model to predict dyed fabric hydrophobicity. The reported mean absolute errors 0.42 and 0.49, expressed in terms of AATCC spray rating on training and testing respectively, indicate the prediction capability and good precision of the ANN. As the difference between training and test results is insignificant, it is inferred that the network is generalized very well without any over fitting (Fig. 2), this also expresses the actual and predicted values of hydrophobicity. The developed ANN model demonstrates that it is possible to predict the hydrophobicity of dyed cotton fabric by using concentration of silica nanoparticles, dye type, dye concentration and type of cross-linkers. As cotton fibre is naturally water loving, this tendency is due to the presence of hydroxyl groups which attracts water molecule and make fabric wet. By increasing silica nanoparticle concentration, fabric surface roughness increases, furthermore, the reactive dyes form

Table 2—Spray rating

Ratings	Explanation
100	No wetting and adherence of small drops to sprayed surface
90	No wetting, but adherence of small drops to sprayed surface
80	No wetting, but adherence of drops to sprayed surface
70	Wetting of half of sprayed surface
50	Wetting of whole sprayed surface

Table 3—Selected parameters for prediction

Influencing variables	Value
Dye type	Reactive (1) & Direct (0)
Dye concentration, %	(0.5) & (3.0)
Cross-linker type	TEOS (1) & TMOS (0)
Silica concentration, %	0, 0.02, 0.1 & 0.2
Total samples	32

Table 4—Performance of ANN and MLR models

Parameter	Hydrophobicity		
	Artificial neural network		Multiple linear regression
	Training	Testing	
R^2	0.99	0.99	0.73
R^2 (Adj)	-	-	0.69
MAE, % / MS	2.09	2.46	11.2
MAE	0.42	0.49	2.45

MME = Mean absolute error, in terms of AATCC spray rating.
MS = Mean square of regression model.

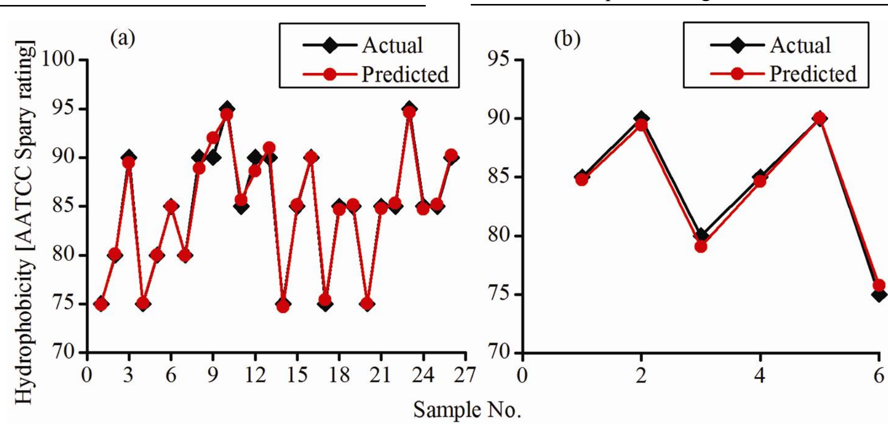

Fig. 2—Performance of ANN on hydrophobicity prediction (a) training and (b) testing

covalent bonds with cotton fabric and hence with higher the concentration of dye more hydroxyl groups of fabric will be occupied, thus increasing the hydrophobicity. Additionally, silane oligomers form hydrogen bonds with the hydroxyl groups of cotton but after curing, hydrogen bonds will be converted to covalent bonds. This helps to increase hydrophobicity and longevity of the hydrophobic finish when applied through sol-gel route in their presence. Therefore, it is inferred that all selected parameters have influence on fabric hydrophobicity which helps the successful training of ANN.

3.2 Fabric Hydrophobicity Modeling with Multiple Linear Regression

Multiple linear regression models were developed from the experimental data of the dyed fabric hydrophobicity. The general regression model of dyed cotton fabric hydrophobicity in terms of AATCC spray rating is given in following equation:

$$\text{Hydrophobicity [AATCC spray rating]} = 76.746 + 60.988 \times (X_1) + 1.125 \times (X_2) + 0.938 \times (D_1) + 0.938 \times (D_2) \quad \ldots (3)$$

$$D_1 \begin{cases} 1\,\text{Reactive dye} \\ 0\,\text{Direct dye} \end{cases}, \quad D_2 \begin{cases} 1 = \text{TEOS} \\ 0 = \text{TMOS} \end{cases}$$

Where X_1 is the concentration of silica nanoparticle (%); X_2, the dye concentration (%); D_1, the dummy variable for dye type; and D_2, the dummy variable for cross-linker type.

Above equation states that all predictors have positive effect on fabric hydrophobicity. This effect is more prominent with concentration of silica nanoparticle and dye concentration, as is evident from the p-value of both predictors. The reasons are already explained in Section 3.1. Effect of reactive dye and TEOS is 0.938 units more than that of direct dye and TMOS. This is due to the dye chemistry. The reactive dyes react with hydroxyl (-OH) groups of fabric by covalent bonding leaving behind lesser free hydroxyl groups to react with water molecule, whereas direct dyes attach with cellulose via weak hydrogen bonding. It may be longer carbon chain structure of TEOS which makes it 0.938 units more effective than TMOS. From *p-value* of model, it has been observed that, model is statistically significant with silica nanoparticle and dye concentration as most significant variables. Table 4 states that there is good correlation between experimental and predicted fabric hydrophobicity with a coefficient of determination (R^2) = 0.73 and an acceptable MAE =2.45.

Table 5—Correlation between input and output variables

Input parameters	Correlation coefficient (r)
Dye type	0.079
Dye concentration, %	0.238
Cross linker type	0.079
Silica concentration, %	0.812

3.3 Comparison of ANN and MLR Models

From results as expressed in above section and in Fig. 2 and Table 4, it is evident that the coefficient of determination of ANN model is much higher than that of MLR model; it states that in ANN model, input variables have explained 99% variance in the predicted variable as compared to that in MLR model (73% variance). On other hand; other key aspect of prediction is absolute error, which is the difference between actual and predicted outputs. The lesser the error the better is the prediction. MAE in terms of AATCC spray rating of ANN is 0.42 and 0.49 on training and testing respectively as compared to 2.45 of MLR model. Therefore, it can be concluded that neural network has better prediction capabilities as compared to regression model for prediction of dyed fabric hydrophobicity.

3.4 Determination of Correlation Coefficient

Correlation coefficients (r) were measured to analyze the relationship between input and output variables (Table 5). It is observed that there is a strong correlation between silica nanoparticle concentration and hydrophobicity, dye concentration shows around 24% relationship while dye and cross-linker type reflects only 8% correlation with fabric hydrophobicity. This analysis also supports the theoretical understanding about the effect of used regressorson fabric hydrophobicity.

4 Conclusion

In this study, empirical and statistical models are proposed to predict dyed cotton fabric hydrophobicity without any prior trials, which reduces cost and time. Hydrophobicity of dyed 100% cotton plain woven fabric is predicted successfully from dye type, dye concentration, cross-linker type and concentration of silica nanoparticles by using multilayer feedforward backpropagation artificial neural network and multiple linear regression and results are compared.

Neural network has predicted fabric hydrophobicity with excellent precision and trivial mean absolute error of 0.42 and 0.49 in term of AATCC spray rating scale on training and testing respectively. Reported coefficient of determination states that 99% variance in dependent variable is explained by the model. As the difference between training and test results is insignificant, the network is generalized very well without any over fitting. MLR model has explained only 73% variance in the predicted variable with a mean absolute error of 2.45 in terms of AATCC spray rating scale. Therefore, it can be concluded that ANN is more powerful prediction tool as compared to MLR for prediction of sol coated dyed fabric hydrophobicity.

The advantage of using MLR is to reveal the significance of model and individual input values through P-values. From *P-value* of MLR model, it has been observed that the model is statistically significant with silica nanoparticle and dye concentration as most significant variables, as is also evident from correlation coefficients of input variables. These models will benefit in designing and developing hydrophobic cotton surfaces for water repellent and rain wear applications.

References

1 Heywood D, *Textile Finishing* (Society of Dyers and Colorists, Bradford), 2003, 187.
2 Lewandowski E, Grzegorz M & Eugeniusz M, *J Hazard Mater*, 136 (2006) 385.
3 Mahltig B, Helfried H & Horst B, *J Mater Chem,* 15(2005) 4385.
4 Roe B & Zhang X, *Text Res J,* 79(2009) 1115.
5 Hou A, Yu Y & Chen H, *J Carbohyd Polym,* 79 (2010) 578.
6 Mahltig B & Böttcher H, *J Sol-Gel Sci Technol*, 27 (2003) 43.
7 Saleemi S, Ejaz A, Almas R & Tanwari A, *Sci Int*, 25 (2013) 823.
8 Saleemi S, Malik S A, Syed U & Tanwari A, *J Eng Fiber Fabric,* 9 (4) (2014) 16.
9 Slokar Y M, Zupan J & MarechalAM, *Dyes Pigm,* 42 (1999) 123.
10 Daneshvar N, Khataee A R & Djafarzadeh N, *J Hazard Mater*, 137(2006) 1788.
11 Kuo CF J, Chang C D, Su T L & Fu C T, *Polymer Plast Technol Eng*, 47(2008) 678.
12 Bezerra C M &Hawkyard CJ, *Color Technol*, 116 (2000) 163.
13 Baldua R K, Rengasamy RS & Kothari V K, *Indian J Fibre Text Res*, 39(2014) 157.
14 Ureyen M E & Gurkan P, *Fiber Polym*, 9 (2008) 87.
15 Hussain T, Malik Z A & Tanwari A, *Indian J Fibre Text*, 35 (2010) 243.
16 Rumelhart D E , Hinton G E & Wiliams R J, *Nature*, 323(1986) 533.
17 Wilamowski B M, Iplikci S, Kaynak O & Efe M Ö, *Proceedings. IJCNN '01, International Joint Conference on Neural Networks* (IEEE, Washington DC), 2001, 1778.
18 Fine T L, *Feedforward Neural Network Methodology* (Springer Verlag, New York), 1999.