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Artificial Neural Network Estimation of Lignocellulosic Material Acidity

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The present study describes a simple and efficient artificial neural network (ANN) modelling to predict the hot water and total acidities of lignocellulosic materials including wood and agricultural residues from the hot water and alkali solubilities and pH values. The performance of the proposed model trained by Levenberg-Marquardt algorithm was evaluated by analysis of the predicted as well as the experimental data. The prediction error of 1.31 % and the correlation R^2 values varying between 0.9983 and 0.9940 confirmed that three layered ANN model with 3 hidden neurons produced more accurate results.

Key Words: Wood, Agricultural residue, Acidity, Artificial neural network.

INTRODUCTION

Artificial neural networks (ANNs) inspired by structure and function of biological neurons in the human brain are computer-based trainable techniques, which have been frequently applied in the field of chemistry[1-3]. An ANN consists of three or more layers *i.e.*, an input layer, one or more hidden layers and an output layer. These layers are made up of a number of interconnected neurons which have activation function. The strength of connections between the neurons are quantified with connection weight which can be modified during the training of ANN. The activation function and back propagation algorithm are applied for weights optimization of ANN. The ANNs with different number of hidden neuron are trained by repeatedly presenting a series of input-output sets to network. The ANN gradually learns the input-output variable's relationship of interest by adjusting the connection weights to minimize the error between the actual output values and their estimated values of the training set. The trained network is usually checked through a separate test set to monitor performance and validity for prediction of external data sets[4-8].

The wood acidity is important, since it influences fixation of wood preservatives[9], hardening of synthetic resins used to bind particles in fibre board and particle board[10],

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corrosion of metals in contact with wood[11-13], wood-cement compatibility[14], discolouration of wood, pulping and plastification of wood[15].

Different wood species have different acidities. This difference of acidity between species depends upon free and bound organic acids of extractives and the phenolic compounds[11,16] and also noncellulosic polysaccharides[9] in wood. These components in wood differ both in amount and kind from species to species[15].

For investigation of acids present in wood, the method has been provided by Subramanian *et al.*[17] to determine total and water soluble acids and pH values. When determining the acidity, titration procedure is dependent on stability of the pH value. After each addition of alkali, some time is needed for the solution to reach a constant pH value and during the procedure only one sample can be analyzed under specific conditions[18]. Balaban and Ucar[19] found that the correlation presents quadratic equations between acidities (hot water and total) and solubilities (hot water and NaOH) in hardwoods and cubic relations in softwoods, respectively.

Besides wood, some lignocellulosic agricultural residues, *e.g.* grapevine (*Vitis vinifera*) canes and poppy (*Papaver somniferum*) stalks were interested as raw material for the wood industry[20,21]. Presumably, the acidity can also effect on production processes and products which are obtained from these agricultural materials.

Artificial neural networks (ANNs) are used for complex linear or non-linear relationships[22,23]. Between acidities (hot water and total) and solubilities (hot water and NaOH) of different woods, the relationships were found to be non-linear[19] and titration procedure for determination of wood acidity is laborious and time consuming[18]. Therefore, we aimed rapid and easily estimation of the acidities from solubilities and pH values of lignocellulosic materials using ANN modelling.

EXPERIMENTAL

Nine hardwood and 8 softwood species (Softwoods: *Abies bornmülleriana*, *A. equi-trojani*, *Cupressus sempervirens*, *Juniperus excelsa*, *Picea orientalis*, *Pinus brutia*, *P. nigra* and *P. sylvestris*; hardwoods: *Carpinus orientalis*, *Castenea sativa*, *Eucalyptus camaldulensis*, *Fagus orientalis*, *Quercus frainetto*, *Q. petraea*, *Q. vulcanica*, *Populus tremula* and *Tilia platyphyllos*) were used as test woods. Wood materials were prepared according to Balaban and Ucar[19]. Trees were felled. Three discs taken from each stem at the bottom, middle and top of the tree were used. The wood discs of each tree were debarked and chipped. After air-drying, the wood chips were ground to 1 mm. Heartwood and sapwood of *Castenea sativa*, *Quercus frainetto*, *Q. petraea* and *Q. vulcanica* were treated separately.

Stalks of poppy (*Papaver somniferum*) and canes of grapevine (*Vitis vinifera*) cultivars including cv. *Italia*, cv. *Cardinal*, cv. *Alphonse lavallee*, cv. *Yuvarlak cekirdeksiz*, cv. *Baris*, cv. *Hafizali*, cv. *Razaki*, cv. *Tekirdag cekirdeksiz*, cv. *Cavus*, cv. *Kozak beyazi* and cv. *Trakya ilkeren* were used as lignocellulosic agricultural residues. The stalks and canes were cut into 2-3 cm length and air-dried. The air-dried material was also ground to 1 mm.

Method: Hot water acidity was determined according to the procedure of Balaban and Ucar[19] and expressed in meq mol/100 g oven dried material. Total acidity was estimated upon the method of Subramanian *et al.*[17] and expressed also in meq mol/100 g oven dried material. Hot water solubility and 1 % NaOH solubility were determined by T 207 om-93 and T 212 om-93.

ANN Modelling: For ANN modelling, MATLAB software was used. Back propagation with Levenberg-Marquardt algorithm were employed as efficient method for non-linear weight optimization of ANN. To construct the neural network models, representative data sets were prepared based on the available experimental results and the data were randomly mixed within each set. 22 data pairs were used for training, 8 data pairs for testing and 3 data pairs for validation. A neural network, having an input layer of 4 neurons which are hot water pH, hot water solubility, sodium acetate pH and 1 % NaOH solubility and an output layer of 2 neurons which are the hot water and total acidities, was structured as shown in Fig. 1 and the topology of used network is indicated in Fig. 2.

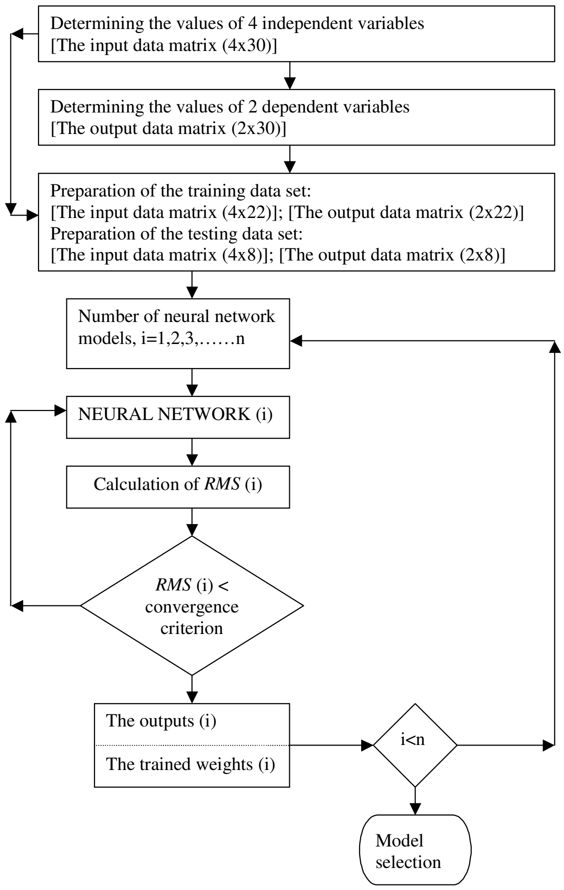


Fig. 1. Operational sequence of the simulation method of ANN

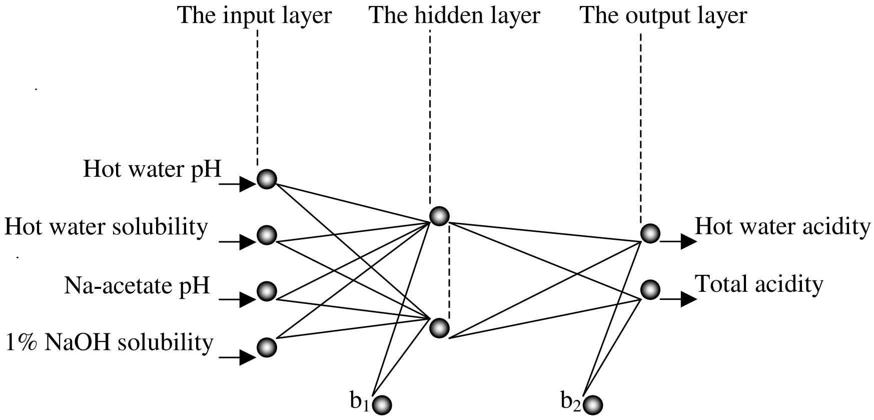


Fig. 2. Network architecture used for estimation of the acidities of lignocellulosic materials

Because the sigmoidal logistic function was used as the activation function, before presenting to the network, all data were normalized into a range 0.1-0.9 using following equation:

$$X_N = 0.1 + \frac{0.8(X - X_{min})}{(X_{max} - X_{min})} \quad (1)$$

where X_N is normalized value of the data (hot water pH, hot water solubility, sodium acetate pH, 1 % NaOH solubility, hot water acidity and total acidity), X is original value of the data and X_{max} and X_{min} are the maximum and the minimum original values of the data.

Root mean square (RMS) error was used as a criterion of the fitting quality of networks, which is defined as

$$RMS = \sqrt{N^{-1}\sum_{i=1}^{N}(X'_i - X_i)^2} \quad (2)$$

where N is the number of data, X'_i is the target value and X_i is the output value predicted by the network.

RESULTS AND DISCUSSION

The experimental results indicated that hardwoods were more soluble and acidic than softwoods. The hot water acidities of hardwood species were found between 1.29 and 25.09 meq mol/100 g, total acidities between 2.87 and 29.75 meq mol/100 g, hot water solubilities between 2.02 and 11.98 %, 1 % NaOH solubilities between 15.57 and 27.61 %, hot water pH values between 3.39 and 5.47 and sodium acetate pH values between 5.82 and 6.68, whereas, the hot water acidities of softwood species were determined between 0.47 and 6.93 meq mol/100 g, total acidities between 3.13 and 10.88 meq mol/100 g, hot water solubilities between 1.42 and 5.13 %, 1 % NaOH solubilities between 10.37 and 16.68 %, hot water pH values between 4.14 and 5.57 and sodium acetate pH values between 5.87 and 6.71, respectively. The analytical results of some wood species were in agreement with finding from this previous study[19]. The results indicated that canes of the grapevine cultivars and stalks of the poppy stand mainly in the acidic case and they were fairly soluble in hot water and 1 % NaOH. The hot water acidities of these agricultural residues varied between 6.48 and 10.41 meq mol/100 g, total acidities between 7.9 and 13.31 meq mol/100 g, hot water solubilities between 6.46 and 11.08 %, 1 % NaOH solubilities between 18.88 and 22.04 %, hot water pH values between 4.14 and 4.47 and sodium acetate pH values between 5.96 and 6.20, respectively (Figs. 3-5).

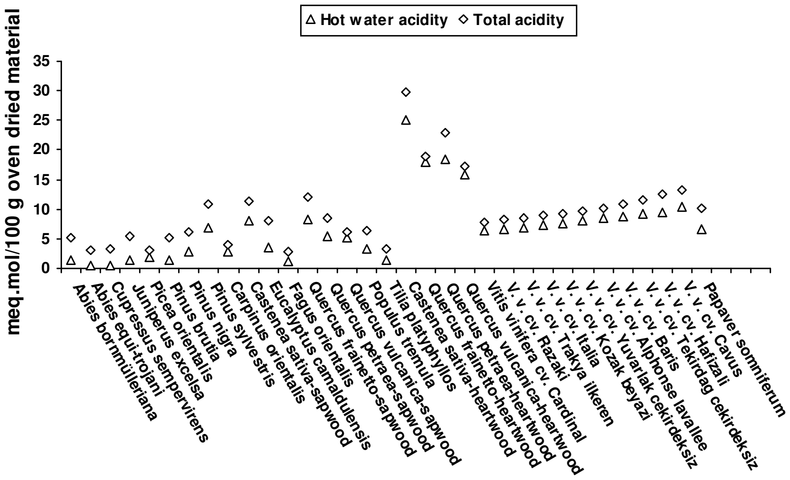
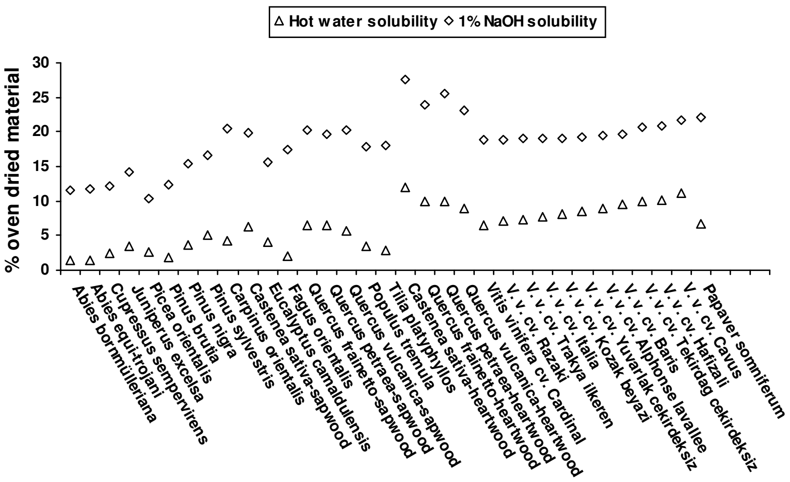


Fig. 3. Hot water and total acidities of different softwoods, hardwoods and agricultural residues



Fi. 4. Hot water and 1 % NaOH solubilities of different softwoods, hardwoods and agricultural residues

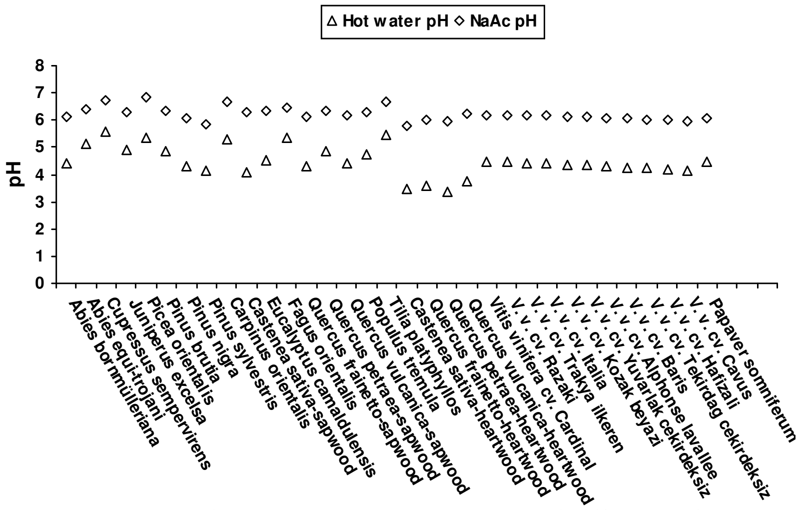


Fig. 5. Hot water and sodium acetate (NaAc) pH values of different softwoods, hardwoods and agricultural residues

Training and testing experiments were conducted to find a network topology with greatest accuracy in predicting of validation data set. The performances of different network models are shown in Table-1. Fitness of the models was compared using the root mean square (RMS) error. The NN3 model with 3 hidden neurons and RMS error of 0.023266 and 0.010584 in the testing could be chosen as the best fit model to predict the hot water and the total acidities of lignocellulosic materials.

TABLE-1
ACCURACY OF THE NEURAL NETWORK MODELS

Model	RMS error			
	Hot water acidity		Total acidity	
	Training	Testing	Training	Testing
NN1 4-1-2	0.027791	0.024477	0.027108	0.039202
NN2 4-2-2	0.022575	0.034285	0.018873	0.047544
NN3 4-3-2	0.007877	0.023266	0.009672	0.010584
NN4 4-4-2	0.006150	0.023700	0.006367	0.036426
NN5 4-5-2	0.005219	0.038009	0.003295	0.069858
NN6 4-6-2	0.005429	0.039424	0.004388	0.048720
NN7 4-7-2	0.002128	0.033445	0.004269	0.036301
NN8 4-8-2	0.002133	0.063504	0.002160	0.125349

When the NN3 model was allowed to estimate the hot water and the total acidities, linear regressions of the training and the testing data sets indicated that agreement between the predicted and the actual acidities was generally good. The correlation R^2 values were 0.9983, 0.9973, 0.9940 and 0.9946, respectively (Figs. 6 and 7).

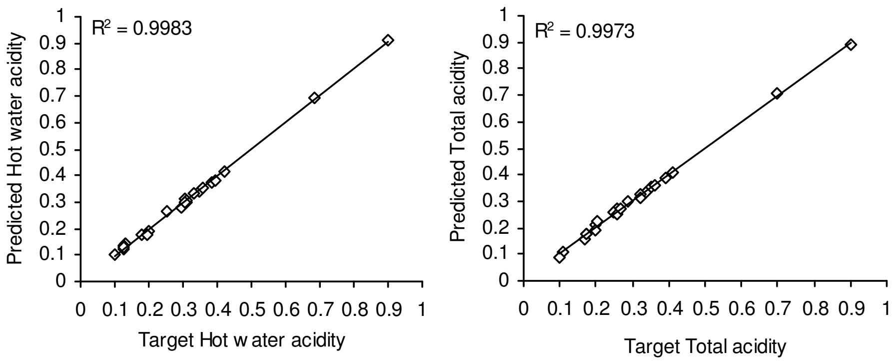


Fig. 6. Relationship between the predicted values by NN3 model and the actual values of hot water and total acidities for training data set

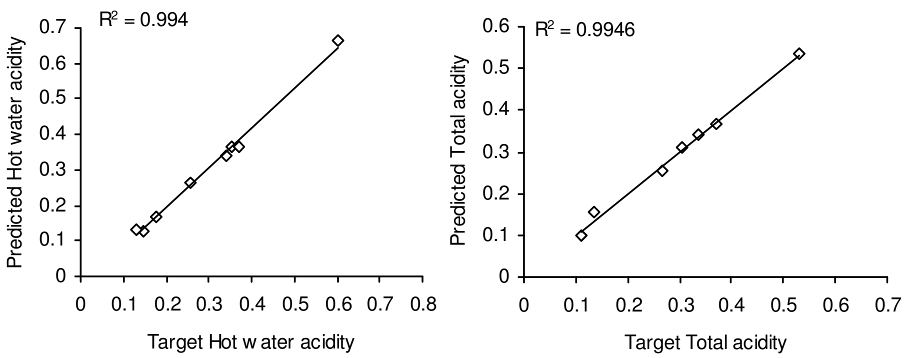


Fig. 7. Relationship between the predicted values by NN3 model and the actual values of hot water and total acidities for testing data set

The data pairs of *Cupressus sempervirens*, *Vitis vinifera* cv. *Italia* and *Quercus frainetto*-heartwood were used for validation step of the model NN3 which clearly highlights the superior predictive capability in the training and testing. The results of validation are listed in Table-2. Average per cent relative standard error between the predicted and the actual values closed to 1.31 % indicate good model performance. Therefore, ANN is considered excellent to estimate the hot water and the total acidity values of lignocellulosic materials.

TABLE-2
PERFORMANCE OF THE NN3 MODEL IN THE VALIDATION

Species	Hot water acidity (meq mol/100 g)			Total acidity (meq mol/100 g)		
	Actual	Predicted	RE %	Actual	Predicted	RE %
Cupressus sempervirens	0.52	0.51	–1.92	3.24	3.23	–0.31
Vitis vinifera cv. Italia	7.27	7.09	–2.48	8.87	8.72	–1.69
Q. frainetto-heartwood	17.88	18.07	1.06	19.01	18.93	–0.42

Conclusion

Using Levenberg-Marquardt algorithm for building and optimizing calibration model, three-layered ANN model was proposed to estimate the lignocellulosic material hot water and total acidities which were the output variables. In this model which was very simple and rapid, the hot water pH, Na-acetate pH, the solubility in hot water and the solubility in 1 % NaOH were used as the input variables. Furthermore, the network had 3 hidden neurons. The performance of the proposed model was validated by application of the model to analyze the predicted and experimental data. It was obtained that the proposed model could predicted the hot water and the total acidity values of lignocellulosic materials with very low prediction error of 1.31 % and high correlation R^2 values varied between 0.9983 and 0.9940.

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