MODELING AND OPTIMIZATION OF THE EXTRACTION OF LIGNOSULFONATE FROM BARLEY STRAW BY USING ARTIFICIAL NEURAL NETWORKS

Maria Guadalupe Serna-Diaz[a], Joselito Medina-Marin[b], Ainhoa Arana-Cuena[c], Juan Carlos Seck-Tuoh-Mora [d], Alejandro Tellez-Jurado[e], Yuridia Mercado-Flores [f], Angelica Jimenez-Gonzalez[g], Norberto Hernández-Romero [h]

[a](c) (e) (f) (g) Polytechnic University of Pachuca, Pachuca de Soto, Hidalgo, México
[b] (d) (h) Autonomous University of Hidalgo State, Advanced Research Centre in Industrial Engineering, Pachuca de Soto, Hidalgo, México

[a]sedg0210@yahoo.com.mx, [b]jmedina@uaeh.edu.mx, [c]ainhoa@upp.edu.mx, [d]jseck@uaeh.edu.mx, [e]alito@upp.edu.mx, [f]yuridiamercado@upp.edu.mx, [g]ajimenez@upp.edu.mx, [e]nhromero@uaeh.edu.mx

ABSTRACT
Artificial Neural Networks (ANNs) have been applied to model dynamic systems from a wide range of areas. In particular, we are interested in the application of ANN in fitting data sets to model the extraction process of lignosulfonates from barley straw. In order to know real data about the percentage of delignification taking into account three variables from the experimental process: cooking time of the barley straw, percentage of sulfite concentration applied to the barley straw, and barley size. With the data gathered from 75 experiments performed three times each one, we created an ANN optimal with a minimum mse, and we calculate an optimal value from this ANN model.

Keywords: Artificial neural network, Optimization, Lignosulfonate extraction, Data fitting.

1. INTRODUCTION

Lignocellulosic biomass is considered as a key element to avoid the fossil fuel dependence and encourage the use of raw material friendlier with the environment (Zhu, Sun, Su, Zhao, Ma, Zhu, Shi, and Gu, 2013).

The lignocellulosic biomass decomposition is based on three main compounds: cellulose, hemicellulose and lignin, which are joined in microfibrillar grouped in a chain of crystalline cellulose, covered with structures of hemicellulose and holocellulose protected by lignin (Abud, Costa, Wanderley, Souza, and Sant'Anna 2013). Nevertheless, these biopolymers are better utilized individually (Kahar, Taku, and Tanaka 2013).

Lignocellulosic material has a broad range of applications, but the use of each compound depends on the treatments utilized to extract them and how easy and cheap is the separation of the three compounds. These treatments can be physical (Subhedar and Gogate 2014; Iskalieva, Yimmou, Gogate, Horvath, Horvath, and Csoka 2012), chemical (Yang, Kuittinen, Zhang, Keinänen, and Pappinen, 2013; Duque, Manzanares, Ballesteros, Negro, Oliva, Saez, Ballesteros, 2013) or biological (El-Zawawy, Ibrahim, Abdel-Fattah, Solimanb, Mahmoud, 2011; Wang, Manley, and Feldmant, 2011).

Nowadays, the utilization of biomass extracted from agricultural residuals is often applied to obtain polymers. In the case of Barley (*Hordeum vulgare*), one kilogram of barley corresponds to 0.750 of straw (Carrera and Mateo 2005). For this reason, barley straw is a good option for extracting Lignocellulosic material (Singh, Shukla, Tiwari, and Srivastava, 2014). Moreover, lignin has many applications in industry, such as dispersant agent for mixture of cement and gypsum (Stráněl and Sebök, 1997), as emulsifier (Weis and Bird, 2001), in polymers production (Effendi, Gerhauser, and Bridgwater, 2008), and a new trend is the use of lignin in agricultural industry as soil conditioner (Deng et al., 2011) and vegetal growth promoter (Ertani et al., 2011).

The applications mentioned above utilize the lignin in the form of lignosulfonate (LS), which is the sulfonate lignin obtained from the separation with sulfite (Chakrabarty, Saha, and Ghoshal 2009). This process requires high temperature and pressure in order to depressurize the fibers of cellulosic material and reach its disaggregation. Furthermore, it is important to consider the size of the biomass (pieces of barley straw) and the concentration of the sulfite to find the maximum performance of delignification.

In this research work we took into account three independent variables: barley straw size, sulfite concentration, and cooking time. And as dependent variable we considered the performance percentage of delignification.

Experimental results were fitted through an Artificial Neural Network model in order to find an optimal combination of values that produce the maximum delignification performance.

2. MATERIALS AND METHODS

The lignocellulosic material of barley straw utilized in this work was taken from crops located in Zempoala, Hidalgo, Mexico. The separation was carried out in a semi-industrial autoclave AV-3580 Prendo®, which was modified to be able to reach 3 atm of pressure.

2.1. Variables

The variables for this experiment were: size of straw with a sieve number 8 (2.0 mm), 10 (1.68 mm) and 20 (0.84 mm); cooking time was 15, 30, 45, 60, and 90 minutes; finally, sodium sulfite concentrations with values of 1%, 3%, 5%, 10%, and 15%. The temperature was kept at 137 ºC, the pressure at 3 atm, and the relation between straw and sulfite was 1 part of straw for 25 of liquor. Then, a total of seventy five lab experiments were done ($3 \times 5 \times 5 = 75$). Table 1 shows the combination values for each experiment.

Table 1: Combination of values for each experiment

Experiment number	Sieve number	Cooking time	Sulfite concentra-tion
1	8	15	1
2	8	15	3
…	…	…	…
6	8	30	1
7	8	30	3
…	…	…	…
26	12	15	1
27	12	15	3
…	…	…	…
31	12	30	1
32	12	30	3
…	…	…	…
51	20	15	1
52	20	15	3
…	…	…	…
56	20	30	1
57	20	30	3
…	…	…	…
74	20	90	10
75	20	90	15

2.2. Lab experimental method

The straw was ground to separate the fibers. It was winnowed with chain mails of 8 (2.0 mm), 10 (1.68 mm) and 20 (0.84 mm). Then, 2g of straw for each size were mixed with 100 mL of sodium sulfite, in different concentrations (1%, 3%, 5%, 10%, and 15%). Every experiment was done in triplicate.

The digestive process was carried out in a closed system inside glass bottles and the cooking process was done in an autoclave with temperature of 137 ºC and pressure of 3 atm, during 15, 30, 45, 60, and 90 minutes. From this process, the cellulose pulp and the "liquor" were produced. Lignosulfonates contained in the liquor were separated by means of vacuum filtration, in filter paper with pore of 1.6 μm.

To quantify the solubilized material, the material retained by the filter paper was burned at 550 ºC during 6 hours. Then, the counting was done by weight difference, with a previous correction of the humidity and solubilized material in a sample without sulfite only with water (Northey, 1992; Ekeberg, Gretland, Gustafsson, Braten, and Fredheim, 2006).

The performance of delignification material is calculated with Equation 1.

$$PSM = \frac{DWs - (DWf - A - W)}{DWs} \times 100 \quad (1)$$

Where:
PSM: is the percentage of solubilized material.
DWs: is the initial dry weight.
DWf: is the final dry weight.
A: are the residual ashes.
W: is the sample without sulfite (only water).

2.3. Data fitting method

The data fitting was done through an Artificial Neural Networks (ANNs), which was trained, validated and tested with the 75 experimental results from the combination of 3 sizes of barley straw, 5 cooking times, and 5 sulfite concentrations.

2.3.1. Artificial Neural Network

An ANN is computational technique that performs a multifactorial analysis. It is inspired in biological neural networks, and is composed of nodes distributed in layers, and interconnected through lines with assigned weights.

Nodes represent artificial neurons, which are processing units performing a no-linear sum (Dayhoff, and Deleo, 2001). Connections weights are adjusted according to a training process of the ANN. An adequate training produces an ANN that helps in values forecasting, objects classification, function approximation, pattern recognition with multifactorial data, and modeling and optimization of data fit for different phenomena (Sinhaa, Chowdhurya, Sahaa, and Dab, 2013; Aghav, Kumar, and Mukherjee, 2011; Duque, Manzanares, Ballesteros, Negro, Oliva, Saez, and Ballesteros, 2013; Maache-Rezzoug, Pierre, Nouviaire, Maugard, and Rezzoug, 2011), among other applications.

In this work, we applied the Matlab® Neural Network Toolbox to perform the data fitting. The ANN model was obtained through an iterative process. In it we were changing the number of neurons, and the ANN model was trained ten times, saving the ANN with the less Mean Square Error (mse). The method used for training was the Levenberg-Marquardt method, and the data division for training, validating, and testing was done randomly.

3. RESULTS

3.1. Lab experimental results

Results obtained from straw delignification with different values for sulfite concentration, cooking time, and size of barley straw are showed in figure 1.

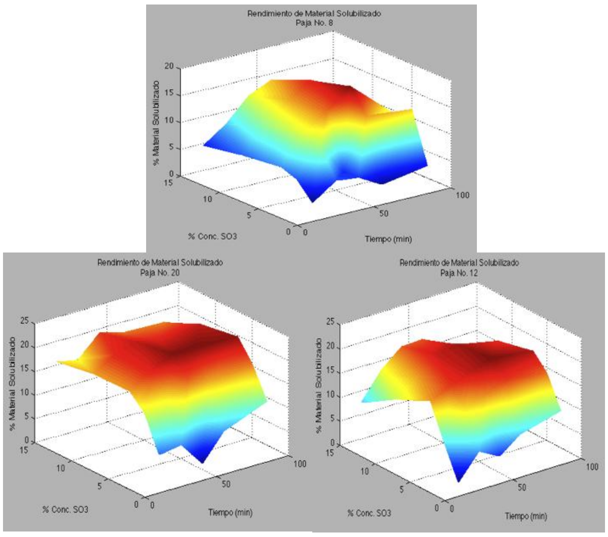

Figure 1: Percentage of solubilized material from straw barley of size 8(a), 12(b) and 20(c).

The lower percentages of delignification are represented by dark blue color and the higher percentages are represented by dark red color.

Figure 1 shows that the maximum value of percentage of delignification is located in the same value for cooking time (60 minutes) and sulfite concentration (5%) for each value of barley straw size. For the barley straw size 8, the maximum value is 18.7 ± 0.53, for size 12 the maximum value is 20.4 ± 0.47, and for size 20 the maximum value is 22.5 ± 1.3.

3.2. ANN model results

The data fitting was performed through an ANN model, which was created with the Neural Network Toolbox included in Matlab®.

Furthermore, a Matlab® script was implemented to find an ANN model with a minimum mse. This script creates ANN models starting from a model with 3 layers; the first one considers the three input variables (barley straw size, cooking time, and sulfite concentration); the second one is the hidden layer where the connections weights are set from the training process; and the last layer contains only one neuron with the value of the percentage of solubilized material.

70% of the experimental data (56 samples) were utilized in the training process, and they were chosen randomly. 15% of the data (11 samples) were utilized to validate the model, and the other 15% (11 samples) of data were utilized to test the ANN model.

The ANNs created in the scritp were trained with the Levenberg-Marquardt backpropagation algorithm, and we iterate from an ANN with 3 neurons until an ANN with 50 neurons. Each one was trained ten times in order to find a model with the minimum mse.

Taking into account this criterion, we found an ANN model with 20 neurons and the less mse of 1.2214 (Figure 2).

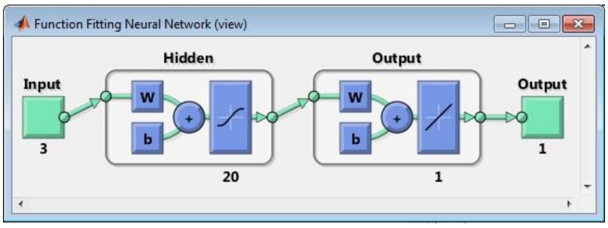

Figure 2: ANN model with minimum mse.

Figure 3 contains the graphic for lab experimental data and the data obtained from the ANN model. The seventy five values obtained from lab experiments are denoted as small red circles, and the dashed line denotes the data forecasted from the ANN model, for the same seventy five combinations. The axis of abscissas represents the experiment number, where each experiment was performed in a combination of values for sieve number, cooking time, and sulfite concentration. The axis of ordinates indicates the percentage of solubilized material obtained from each combination.

The model showed in figure 2 was utilized to estimate the parameters for the optimal value of percentage of delignification. The input value for barley straw size was iterated from 8 to 20, the input value for cooking time was iterated from 15 to 90 minutes, and the input value for sulfite concentration was iterated from 1 to 15. Each parameter was incremented in steps of one, and we obtained a total of 14 820 combinations in order to evaluate the ANN model for each one.

The highest value of solubilized material calculated with this ANN model was 24.6432%, with barley straw size of 16, a cooking time of 42 minutes, and a sulfite concentration of 15%.

Figure 4 shows the graphic generated with all the data forecasted with the ANN model. As in figure 3, the small red circles denotes the seventy five lab experimental data, and the green graphic denotes the 14 820 forecasted data from the ANN model. Similar to figure 3, the axis of abscissas represents the experiment number, and the axis of ordinates indicates the percentage of solubilized material.

The circle filled in red indicates the optimal value of solubilized material.

4. CONCLUSIONS

Lignocellulosic material has been applied in different areas, and it has been gotten from different sources. Currently, agricultural residuals are used to extract lignin with good results.

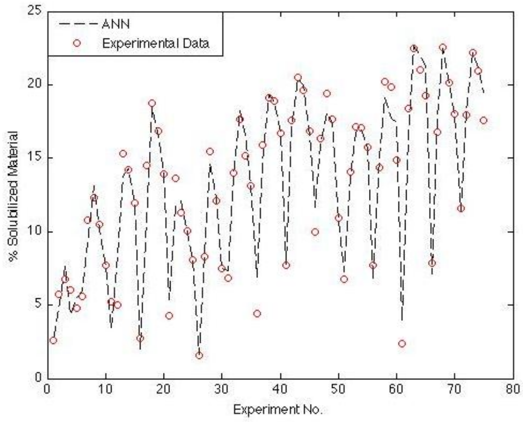

Figure 3: Comparative graphic between experimental data and data predicted with the ANN model.

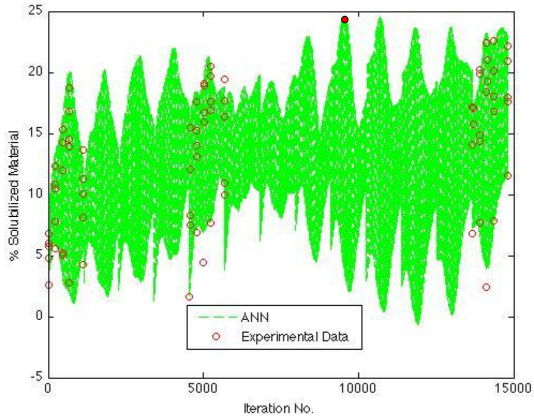

Figure 4: optimal value found in all the data forecasted with the ANN model.

In this work, we used barley straw to get lignin from biomass, and to analyze the effect of factors as barley straw size, cooking time, and sulfite concentration, a total of 75 experiments were carried out. The three barley straw sizes considered were 8, 12, and 20; the five cooking times utilized were 15, 30, 45, 60, and 90 minutes; and the five sulfite concentrations were 1%, 3%, 5%, 10%, and 15%.

Experimental data were used to create ANNs with different number of neurons and trained ten times. The ANN with the lower mse (1.2214) was taken to search the optimal value for percentage of delignification. The maximum value found for the solubilized material with the ANN model was 24.6432%, for a barley straw size of 16, cooking time required of 42 minutes, and sulfite concentration of 15%.

As future work, we are going to take into account some economic considerations, in order to estimate the cost for producing lignosulfonates from barley straw, and to determine if the optimal value found by the ANN model can be obtained with a low cost.

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AUTHORS BIOGRAPHY

Maria Guadalupe Serna-Diaz. She received the M.S. degree in Biotechnology from the Polytechnic University of Pachuca, in Pachuca de Soto, Hidalgo, Mexico, in 2011. Currently, she is a PhD student of the Polytechnic University of Pachuca, Pachuca de Soto, Hidalgo, Mexico.

Joselito Medina-Marin. He received the M.S. and Ph.D. degrees in electrical engineering from the Research and Advanced Studies Centre of the National Polytechnic Institute in Mexico, in 2002 and 2005, respectively. Currently, he is a Professor of the Advanced Research in Industrial Engineering Centre at the Autonomous University of Hidalgo State in Pachuca, Hidalgo, México. His current research interests include Petri net theory and its applications, active databases, simulation, and programming languages.

Ainhoa Arana-Cuena. She received the Ph.D. degree in Molecular Biology from the Autonomous University of Madrid in Spain, in 2002, She is working as a full time professor in the Polytechnique University of Pachuca, in Hidalgo State, México. Her research interest is the applications of agricultural residuals.

Juan Carlos Seck-Tuoh-Mora. He received the M.S. and Ph.D. degrees in electrical engineering (option: Computing) from the Research and Advanced Studies Centre of the National Polytechnic Institute in Mexico, in 1999 and 2002, respectively. Currently, he is a Professor of the Advanced Research in Industrial Engineering Centre at the Autonomous University of Hidalgo State in Pachuca, Hidalgo, México. His current research interests include cellular automata theory and its applications, evolutionary computing and simulation.

Alejandro Jurado-Tellez. He received the M.S. degree in Biotechnology from the Autonomous Metropolitan University, in México City, México in 1997, and the Ph.D. degree in Molecular Microbiology from the University of Alcalá in Spain, in 2002. He is working as a full time professor in the Polytechnique University of Pachuca, in Hidalgo State, México. His research interest is the applications of agricultural residuals.

Yuridia Mercado-Flores. She received the M.S. and Ph.D. degrees in Biotechnology from the National Polytechnic Institute in México, City, México, in 2002 and 2005, respectively. She is working as a full time professor in the Polytechnique University of Pachuca, in Hidalgo State, México. Her research interest is the applications of agricultural residuals.

Angélica Jiménez-González. She received the M.S. and Ph.D. degrees in Biotechnology from the Autonomous Metropolitan University, in México City, México, in 2003 and 2008, respectively. She is working as a full time professor in the Polytechnique University of Pachuca, in Hidalgo State, México. Her research interest is the applications of agricultural residuals.

Norberto Hernandez-Romero. He received the M.S. degree from Department of Electrical Engineering, Laguna Technological Institute at México, in 2001 and the Ph. D. from Autonomous University of Hidalgo State at México in 2009. Currently, he is a professor of the Advanced Research in Industrial Engineering Centre at the Autonomous University of Hidalgo State at Pachuca, Hidalgo, México. His current research interests include system identification, feedback control design, genetic algorithms, fuzzy logic, neural network and its applications