Comparison between Neural Network Technique and Mathematical Modelling of Steam Extraction of Essential Oil

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***Abstract*—**A Neural Networks (NN) feed-forward multi-layer with Levenberg-Marquardt training algorithm was developed to predict the yield for steam extraction of eucalyptus essential oil. The developed techniques are based on four independent variables including temperature, residence time, particle size and mass flux per unit mas of substratum that affected on the yield of leached oil. Different networks were trained and tested with different network parameters for training and testing data sets. To confirm the network generation, an independent data set was used and the predictability of the network was statistically assessed. The regression coefficient (R^2) of 0.9737 implies that the predicted values have an excellent agreement with the experimental date. A mass transfer based mathematical model was developed for constant rate period and diffusion-controlled regime of steam extraction. The proposed model was numerically solved using the Fick's second law in steady-state for one dimensional diffusion in rectangle coordinates. The predicted results have shown that the applied technique of the neural network has better adjusted the experimental data when compared with the mathematical model.

***Keywords*—**Essential oil, mathematical model, neural network, steam extraction.

I. Introduction

Steam extraction is the traditional method used in aromatic industry to obtain essential oils from leaves. This extraction method is less expensive when it is compared with technologically advanced methods such as supercritical fluid extraction [1]. Mathematical models are used to simulate and optimize the process without having to perform experiments in order to know its extraction process behavior, aiming the reduction of experimental procedures in process operation and design. For this purpose, different approaches have been proposed in the literature. Reference [2] applied steam extraction for essential oil of coriander fruits. The authors modelled the steam extraction as an irreversible desorption of essential oil in water, with first-order rate and flashing due to the formation of phase equilibrium. Reference [3] developed a mathematical model for hydro distillation and steam extraction where two types of particles were taken into consideration. The first are leaves where a part of the solute is deposited on the surface in fragile glandular trichomes, and the second are the ground particles with initially homogeneous solute distribution. Reference [1] used a diffusion model to model the extraction of cymbopogon winterianus essential oil at laboratorial scale thus allowing a good prevision on industrial scale. Reference [4] modelled the steam extraction process on semi continuous mode considering three stages in the process of obtaining oil. First a thermal oil exudation from the glandular trichomes occurred, secondly vapor-liquid equilibrium at the interface is assumed, and finally the mass transfer of the oil in vapor-phase. Reference [5] used the same model to simulate the steam extraction process data for rosemary, basil, and lavender essential oils. Reference [6], [7] investigated the effect of temperature and extraction time on the extraction process with oil vapor and established a mathematical model based on Fick's first law applied to material balance for the pilot plant. Reference [8] used steam extraction for Rosemary (R. Officinallis) to study a theoretical-experimental dynamic process of oil extraction. The mathematical modelling is composed of a differential equation of balance of matter in fluid phase that considers the accumulation in the fluid and the solid material. Reference [9] developed a mathematical model based on the assumption of intact particle streams. In the first of which the oil extraction is governed by diffusion processes, and in the second state of equilibrium between the solid and liquid phase is established. Reference [10], investigated the steam extraction of the essential oils for Lavandin Super Oil according to the model Dunkhorst-Houghton, which considers the effect of axial dispersion, and the use of optimization procedures to establish the model parameters. Experimental data for lavadin super oil were used to correlate model parameters. Reference [11] developed a model based on diffusion of essential oil from eucalyptus leaves to optimize the oil recovery. Using the numerical method, the best diffusion coefficient was established for different operating conditions by comparing the model concentration of oil remaining in the leaves with the experimental amount of oil recovered. In most of the mentioned steam extraction modelling approaches, essential oil was considered as a single compound. However, essential oil is not a single compound and its composition varies during the extraction process therefore these methods results in inaccurate diffusivities. Diffusivity of the solute in the solid matrix is another important parameter which should be determined

Manuscript received September 4, 2017.

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https://doi.org/10.17758/EARES.EAP1117055

experimentally or optimized based on experimental yield data. As a result of these constraints, almost in all of steam extraction modelling, the researchers consider some hypothesis in their case to simplify their model and decrease the model parameters, then they tune their model with one or two of the mentioned parameters to fit the experimental data. It is clear that the process of steam extraction of essential oil is complex because the solid matrix containing the essential oil has different structure and that the essential oil is a mixture of many components formed basically by terpenes. In some cases oil may be characterized based on a single component if this is in high concentration relative to the other. On the other hand, the mathematical model that simulate the dynamic process of extraction of essential oil by steam extraction introduces uncertain parameters value such as diffusion coefficient in the particle and mass transfer coefficient that are dependent on the geometry and nature of the plant. Due to complexity of the process, it is difficult to be modelled and simulated using conventional mathematical model. In recent years, considerable advancement in artificial intelligent techniques has been taken place to predict the response in complex and difficult situations. Such techniques can enhance predicting capability of the model when mathematical or statistical methods are difficult to formulate and fails to predict with desired accuracy [12]. NN, artificial intelligent technique is considered as promising tool because of their simplicity toward simulation, prediction and modelling [13], [14]. In this study, a NN technique is developed for simulating steam extraction of essential oil of eucalyptus leaves based on mass transfer differential equations for constant rate and diffusion-controlled regime of extraction. In order to compare predictability of NN model and mathematical technique, the results of each are compared with experimental data at the same conditions.

II. Procedure for Paper Submission

A. Neural Network

Neural network is known for their ability of learning, simulation, and prediction of data. The inspiration of NN came from studies on the structure and function of the brain and nerve systems as well as the mechanism of learning and responding [13], [15]. The network consists of numerous individual processing units (neurons) and commonly interconnected in a variety of structures. The strength of these interconnections is determined by the weight associated with neurons. The multilayer perceptron (MLP) and back propagation (BP) are the most common and successful NN architecture with feed-forward network topologies in the modelling applications and supervised learning technique, respectively [15]. BP is the process by which derivatives of network error, with respect to the networks, is feedback to the network. This algorithm is used to adjust the weights so that the error decreases and the neural model get closer to producing the desired outputs. In this way, BP offers a method of minimizing errors between obtained outputs and desired target values [16]. Preprocess the data, create the network object, train the network and simulate the network response to new inputs are the four steps to develop a BPNN for modelling [16], [17]. In this study, firstly the inputs and targets data were scale within the range 0 and 1. Secondly, the data set was divided into training set for computing the gradient and updating the network weights; validation set for improving generalization and testing set for validating the network performance. The data in each subset were selected randomly to create a network. A total of ten training algorithms were conducted to simulate the test data. The performances of the network in each training process and the best network with the highest prediction performances were recorded.

B. Development of a Neural Network Model

Neural network Toolbox 5.1 in MATLAB 7.4 (R 2017a) mathematical software was used to predict the leaching yield of essential oil. According to [18] stating that in a multi-layer neural network with one hidden layer will never require neurons more than twice as many as inputs, a 10-neuron hidden layer was initially used in the network.

Since the main goal was to find the network having the best performance on new data, seven networks involving 4-10 hidden neurons were trained and tested while ten BP training algorithm and transfer functions were applied to each. The approach to the comparison of the networks was to evaluate an error function using data which were independent on those used for training.

Each network was separately trained and tested by minimization of an appropriate error function defined with respect to data set. This means that during training and testing process, the error decreased. However, when the network began to overfit the data, the error on the testing set began to rise. When the testing error increased for a specified number of iterations, the training was stopped, and the weights and biases at the minimum of the testing error were returned. To assess the network a third independent data set called validating set was used and an in biased estimate of the validating error, which is called generalization error, was obtained and evaluated. Each selected network was run on the validating data set of nine different extraction conditions. The network received inputs including residence time, temperature, particle size and mass flux and predicted yield of essential oil as output. In order to evaluate the results, obtained regression coefficient and mean square error, predicted values were statistically compared with experimental data. The configuration of the BPNN giving the smallest MSE was LMA (4-10-1) with a tangent sigmoid transfer function (tansig) at hidden layer with 10 neurons and a linear transfer function (purelin) at out layer.

To improve the generalization of the selected network, the network with the above topology was run on experimental data and regression coefficient as well as regression line between predicted values from the neural network and the experimental data were obtained to assess the network predictability.

C. Mathematical Modelling

The mathematical model of steam extraction of essential oil was formulated using a model based on Fick's second laws in steady-state for one-dimensional diffusion in rectangle

coordinates on mass balance of the extracted oil as discussed by [1]. The mathematical model is based on the following assumptions:

1. The essential oil extracted is composed of several chemical components, but it was considered as being represented by one pseudo component.
2. Steam is uniformly distributed in the extractor.
3. The mass transfer per unit volume extraction bed between solid and fluid phases are governed by a linear driving force.

Diffusion occurs normal to a surface with an area A and through a volume element AΔx. A material balance on component i entering at x and leaving at x+Δx yield.

The mathematical formulation is expressed as:

$$\frac{\partial C_i}{\partial t} A\Delta x = R_i A\Delta x \tag{3}$$

For the case of diffusion

$$\frac{\partial C_i}{\partial t} = D_{ij} \frac{\partial^2 C_i}{\partial x^2} + R_i \tag{4}$$

where D is the diffusion coefficient and C_i is the initial concentration.

When no chemical reactions occur, $R_i = 0$,

$$\frac{\partial C_i}{\partial t} = D_{ij} \frac{\partial^2 C_i}{\partial x^2} \tag{5}$$

$$\frac{\partial^2 C_A}{\partial x^2} = \frac{1}{D} \frac{\partial C_A}{\partial t} \qquad \text{in } 0 \le x \le L \tag{6}$$

The initial and boundary conditions are:

$$C_A = C_{AO} \qquad \text{in t = 0} \tag{7}$$

$$C_A = 0 \qquad \text{in x = 0} \tag{8}$$

$$C_A = 0 \qquad \text{in x = L} \tag{9}$$

where L is the thickness of the leave and C_{A0} is the initial concentration at time, t = 0.

Applying these equations to the boundary conditions,

$$C_A(x,t)\big|_{x=L} = 0 \quad x > 0 \tag{10}$$

$$\left.\frac{\partial C_A}{\partial t}\right|_{x=0} \qquad t > 0 \tag{11}$$

The system of partial differential equation is solved using a separation of variables techniques. $C_A(x,t)$ is assumed to be separable into independent functions of position and transformed time of the form.

$$C_A(x,t) = \Psi(x)\Gamma(t) \tag{12}$$

Equation (6) becomes:

$$\frac{1}{\Psi(x)} \frac{d^2\Psi}{dx^2} = \frac{1}{\Gamma(t)} \frac{d\Gamma}{dt} \equiv -\beta^2 \tag{13}$$

where β^2 is the arbitrary constant.

$$\Gamma(t) = \exp(-\beta^2 t) \tag{14}$$

$$\frac{d^2\Psi}{dx^2} = -\beta^2 \Psi(x) \tag{15}$$

With the boundary condition of:

$$\Psi(x) = 0 \qquad \text{x = L} \tag{16}$$

$$\frac{d\Psi}{dx} = 0 \qquad \text{x = 0} \tag{17}$$

The solution of (15) has the general form of

$$\Psi(x) = A\sin(\beta x) + B\cos(\beta x) \tag{18}$$

From (18), we can see that A = 0 and B may be arbitrarily chosen. Without loss of generality, set β = 1.

Thus the solution to the spatial problem is:

$$\Psi_n(x) = \cos(\beta_n x) \tag{19}$$

where β_n: n = 1, 2, 3… are given by the positive roots defined by the boundary condition in (19), that is,

$$\cos(\beta_n L) = 0 \tag{20}$$

$$\beta_n = \frac{\pi}{L}\left(n - \frac{1}{2}\right) \tag{21}$$

Initial conditions, the complete solution for the system involves Φ(x, t) being constructed by a linear supposition of the solutions

$$C_A(x,t) = \sum_{n=1}^{\infty} \delta_n \Psi(\beta_n, x)\Gamma(\beta_n, t) \tag{22}$$

(14) and (19) in (22)

$$C_A(x,t) = \sum_{n=1}^{\infty} \delta_n \cos(\beta_n x)\exp(-\beta_n{}^2 t) \tag{23}$$

Where δ_n are constant coefficients used to satisfy the initial boundary conditions. The values of δ_n can be explicitly determined by solving (23) at t = 0.

$$\Phi(x,t)\big|_{t=0} = \sum_{n=1}^{\infty} \delta_n \cos(\beta_n x) = C_{A0}(x) \tag{24}$$

And using the property that the cosine functions are orthogonal

$$\int_0^L \cos(\beta_m x)\cos(\beta_n x)dx = 0 \text{ for } m \ne n \tag{25}$$

$$\int_0^L \cos(\beta_m x)\cos(\beta_n x)dx = \frac{L}{2} \text{ for m = n} \tag{26}$$

Operating both sides of (25) and (26) with the operator $\int_0^L \cos(\beta_n x)dx$ for an arbitrary r, these can obtain:

$$\sum_{n=1}^{\infty} \delta_n \int_0^L \cos(\beta_r x)\cos(\beta_n x)dx = \int_0^L \cos(\beta_r x) C_{A0}(x)dx \tag{27}$$

$$\delta_n = \frac{2}{L}\int_0^L \cos(\beta_n Z) C_{A0}(x)dx \tag{28}$$

Substituting in these coefficients, the generalized solution becomes

$$\partial C_A(x,t) = \frac{2}{L} C_{A0} \sum_{n=1}^{\infty} \exp(-\beta_n{}^2 t)\frac{(-1)}{\beta_n}\cos(\beta_n x) \tag{29}$$

where $\beta_n = \frac{\pi}{L}\left(n-\frac{1}{2}\right)$ n = 1, 2, 3,… (30)

The mass flow as function time was obtained from the mass flux at the boundary multiplied by normal surface area A resulting.

$$w_A(t) = \frac{4C_{A0}DA}{L}\sum_{n=1}^{\infty} e^{\frac{D_m^2\pi^2 t}{L}} \quad (31)$$

Thus the extracted mass of solute constituent is

$$w_A(t) = \frac{8m_{AO}}{\pi^2}\sum_{m=0}^{\infty}\frac{\left(1-e^{\frac{(2m+1)^2 n^2 Dt}{L^2}}\right)}{(2m+1)^2} \quad (32)$$

and the degree extraction is defined by

$$e(t) = \frac{w_A(t)}{w_A(\infty)} = \frac{\sum_{m=0}^{\infty}\frac{\left(1-e^{\frac{(2m+1)^2\pi^2 Dt}{L^2}}\right)}{(2m+1)^2}}{\sum_{m=0}^{\infty}\frac{1}{(2m+1)^2}} \quad (33)$$

III. RESULTS AND DISCUSSION

In the diffusion model based on material balance in the two phases (solid-liquid) present in the extraction process, the parameter D was directly adjusted using (33) with one term in the series. The parameter D was estimated by minimization of the sum of square of error between the experimental data and the prediction model. Analyzing the Fig. 1, it can be seen that diffusion model based on mass transfer fitted very well the experimental data. The diffusion model is based in material balance across internal surface of particle assuming that the components to be extracted are uniformly distributed inside the particle and the surface resistance is negligible. Fig. 1 illustrates the total extraction yield curve (total yield versus extraction time) of eucalyptus leaves resulted from mathematical model and corresponding experimental data at the temperature of 97°C, residence time of 0.2 minutes, leave particle size of 0.00005 m and mass flux of 0.01 kg/m^{2}s.

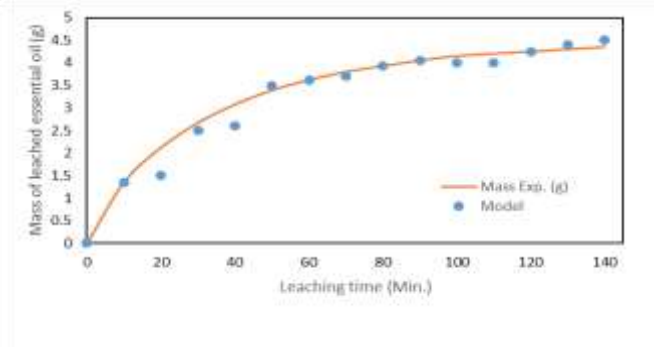

Fig. 1. Predicted results of mathematical model in comparison with experimental data

A schematic NN structure is shown in Fig. 2: a three-layer NN, with tangent sigmoid transfer function (*tansig*) at hidden layer with 10 neurons and a linear transfer function (*purelin*) at output layer. A multi-layer perceptron (MLP) was selected as the relevant network type for training with experimental data. Selected input variables were: temperature, residence time, mass flux and the particle size with yield of leached oil as the output variable. In order to estimate the behavior of the steam extraction, the values for input variables were randomly collected from the experimental dataset and divided into three partitions. The first set (70% of data) was used as training data, second set (20%) for validation and evaluation of network quality during the training phase. The third set (10%) for testing network performance. MLP network was studied by different number of neurons in the hidden layer and with different transfer function. The final best model was selected based on the minimum root-mean-square error and the maximum coefficient of determination.

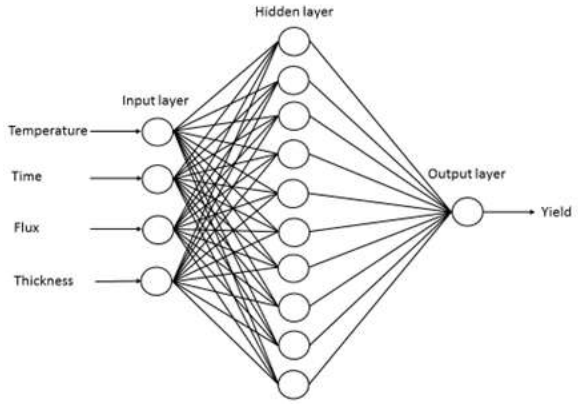

Fig. 2. A schematic diagram of the neural network structure

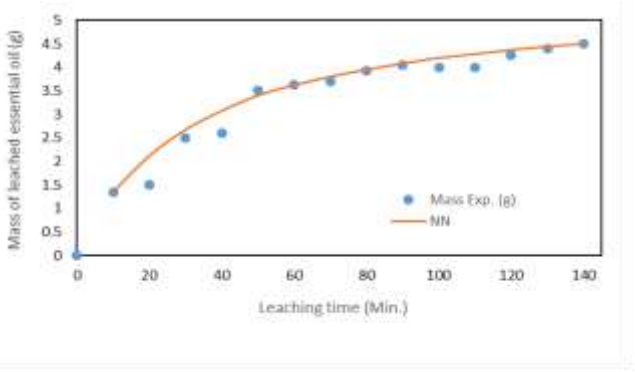

Fig. 3. A schematic diagram of the neural network structure

Fig. 3 shows a comparison of predicted results of NN technique with experimental data at temperature of 90°C, mass flux 0.10 kg/m^{2}s, time of 0.2 min and particle size of 0.00005 m. Mathematical model, neural network and experimental data were juxtaposed in Fig. 4. As can be seen, neural network can predict at various operational conditions much better than mathematical model.

https://doi.org/10.17758/EARES.EAP1117055

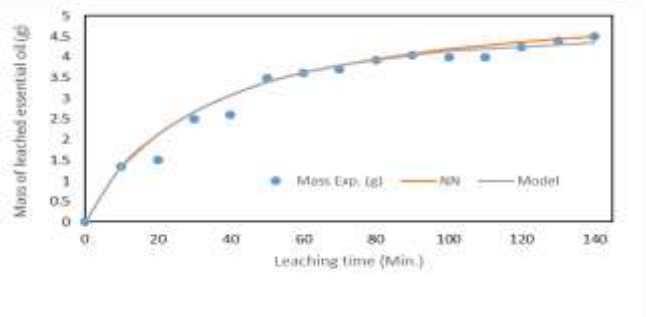

Fig. 4. Comparison between Mathematical model, neural network and Experimental data

As it is shown in Fig. 4, the predictions result of the neural network model have a very good agreement with the experimental data compared to the mathematical model results. It is noticeable that during the training process the entire network parameters weights are optimized simultaneously to minimize mean square error between predictions and training data set that involves the experimental data.

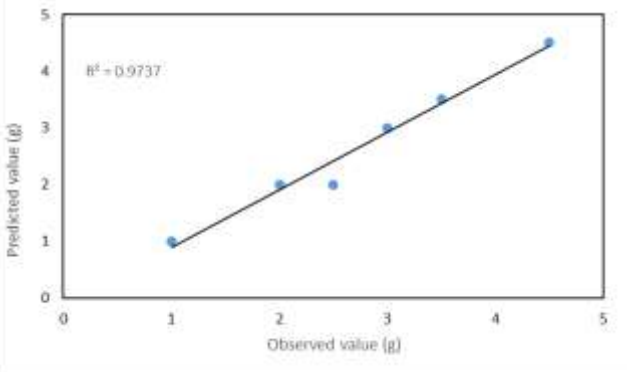

Fig. 5. Correlation between measured and predicted values of total extraction yield

A regression analysis of the network response between NN outputs and the corresponding targets was performed. The graphical output of the network outputs plotted versus the targets is illustrated in Fig. 5. Taking into account the non-linear dependence of the data, linear regression shows an excellent agreement between NN outputs (predicted data) and the corresponding targets (experimental data) with the best linear equation y = 0.9472 + 0.2066 and regression coefficient (R^2) of 0.9725.

TABLE I: PREDICTED AND EXPERIMENTAL VALUES AT DIFFERENT OPERATIONAL CONDITIONS

Series number	Time (min)	Temperature (^oC)	Particle size (m)	Mass flux ($kg/m^2.s$)
1	0.20	95	0.00005	0.12
2	0.25	95	0.00005	0.11
3	0.22	95	0.00005	0.10
4	0.20	97	0.00005	0.10
5	0.30	97	0.00005	0.11
6	0.32	99	0.00005	0.12
7	0.18	99	0.00005	0.10
8	0.20	99	0.00005	0.10
9	0.20	99	0.00005	0.09

TABLE II: PREDICTED AND EXPERIMENTAL VALUES FOR THE YIELDS (KG OIL/KG EUCALYPTUS LEAVES)

Series number	Experimental values	Predicted values	Deviation (%)
1	0.0165	0.0150	9.09
2	0.0170	0.0175	9.71
3	0.0160	0.0160	0.00
4	0.0171	0.0150	8.77
5	0.0203	0.0160	7.98
6	0.0057	0.0060	9.50
7	0.0051	0.0040	7.84
8	0.0027	0.0026	9.60
9	0.0067	0.0055	8.21

Generalization of the network was evaluated by comparing predicted values from the neural network model with experimental data that reported by [6] at the same operational conditions. The Table I presents a series of the operational conditions and experimental data of yields from the literature [6] as well as predicted values from the neural network model at the same conditions. While the Table II shows the deviation between experimental and predicted values of total yields for each operational condition. The average deviation of 7% implies that the predictability of the neural network model is satisfactory. In order to compare the mathematical model results with the neural network results, the mathematical model and the neural network model were run on the same experimental conditions and predicted results of the mathematical model were compared with experimental data. Better performance of NN techniques was confirmed by comparing mean squared error (MSE) and root mean square error (RMSE) with the mathematical model as illustrated in Table III.

TABLE III
MEAN SQUARED ERROR AND ROOT MEAN SQUARE ERROR FOR NN AND MATHEMATICAL MODEL

Error	NN	MM
MSE	0.928	0.746
RMSE	0.950	0.636

IV. CONCLUSION

Since mathematical modelling is based on some assumptions made to derive and solve the model. The model results may have deviations from the reality. The importance of this kind of deviations becomes more crucial when the mathematical model is used for simulating and optimizing a very costly process, such as steam extraction of eucalyptus leaves essential oil. Neural networks technique with a tangent sigmoid transfer function (*tansig*) at hidden layer and a linear transfer function (*purelin*) at output layer were proposed. The proposed NN model showed a good, precise and an effective prediction of the experimental data with a satisfactory result with of R^2 = 0.9737 for four independent variables that affected the yield of leached oil. The results showed an excellent agreement between NN results and experimental data. The predictability of the neural network model was better than that of the mathematical model.

https://doi.org/10.17758/EARES.EAP1117055

ACKNOWLEDGMENT

The Author wishes to thank the Vaal University of technology for providing all necessary materials and funding.

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