INFERENCE OF OIL CONTENT IN PETROLEUM WAXES BY ARTIFICIAL NEURAL NETWORKS

Lima, Anie D. M.[1,2]; Silva, Danilo do C.S. [1]; Silva[1], Valéria S., De Souza Jr., Maurício B.[2]

[1]*Research And Development Center (CENPES), Lubricants and Special Products, PETROBRAS, Rio de Janeiro, Brazil*
[2]*School of Chemistry, Federal University of Rio de Janeiro (UFRJ), Rio de Janeiro, Brazil*

Abstract: The reduction of the time required to determine oil content is important in the production of petroleum waxes. Here, it is aimed to generate a model whose output (the inferred oil content) is obtained from inputs given by other characterization parameters (needle penetration, viscosity, density and refractive index) that are obtained from simpler experiments. Laboratory experiment data together with industrial data were employed in the modeling. These 'real' data were compared with predictions made by the linear models and artificial neural networks. The networks outperform the linear models, as they generate smaller residuals in the whole operational range considered.

Keywords: Nonlinear analysis system, Artificial intelligence, Process parameter estimation, Product quality, Neural-network models, Backpropagation algorithms

1. INTRODUCTION

Oil content in petroleum waxes is presently measured by standard experiments recommended by ASTM: ASTM D 721 (to oil content lower than 15% m/m) and D 3235 (to oil content bigger than 15% m/m). The experiments are done with complex glass apparatus and demand a lot of time. It is possible to develop correlations between physic properties and oil content to help works that need the result of this property in a short time. To do this, one of the methods is the use of artificial neural networks.

Artificial neural networks are computational technics that present a mathematic model inspired in neuron structures of intelligent organisms and that acquire knowledge from experience (Haykin, 1999). The use of neural networks depends on the ability to adapt it to the problem under consideration, by changing the synaptic weights (in the 'learning' phase) to increase efficiency.

Neural networks have been extensively used to represent non linear input-output dependencies, as it has been proved that they can approximate arbitrary well any continuous function (Funahashi; Hecht-Nielsen, 1989; Hornik, 1989).

This work comprises two kinds of investigation: experimental and modeling. In the first approach, values of needle penetration, viscosity, density and refractive index from samples of one kind of wax with different oil contents were acquired. These properties depend on composition and crystallization of wax. In the second one, those data were processed to develop linear models and neural networks in order to predict this characteristic. This technique allows the development of a calculation program to be used works in a refinery environment, so that, based on it, the operator can decide about variables of the process. It is also possible to design a control procedure that acts on the process based on inference of the model

2. SCIENTIFIC METHODOLOGY

2.1 Production and analysis of petroleum waxes

One of the processes of production of waxes is deoiling, that is, extraction of oil in waxes. The process consists in cooling slack wax until a temperature in which only waxes get solid, allowing their separation by filtration. The kind of crystallization determines if the wax will get more oil content during this process, determining the solvent consumption. Hydrocarbon waxes constituted mostly by n-alkanes (macrocrystallines), with crystals like 'dishes', have a structure easier to remove oil. Branches Waxes (microcrystallines), with crystals like 'needles', present more difficulty to remove oil in deoiling (Speight, 2001).

The excess of oil in a wax reduces hardness, and this is inconvenient to store the final product. Oil is also responsible for the appearance of spots, which is a bad characteristic when the end use product is candle. Hardness is measured by needle penetration (ASTM D1321, 2004).

Oil in a wax means that the product has some structures that have more affinity with oil than wax. These can be identified by the following experiments: viscosity (ASTM D445, 2004), density (ASTM D4052, 1996) and refractive index (ASTM, D1218, 2002) in waxes and that is why these parameters and needle penetration are important to predict oil content (Lima et. al, 2005).

2.2 Experimental methodology

Oil Content (ASTM D 721, 1985)

The sample is dissolved in methyl ethyl ketone, afterwards the solution is cooled to –32°C (–25°F) to precipitate the wax, and filtered. Evaporating the methyl ethyl ketone and weighing the residue determine the oil content of the filtrate.

Viscosity (ASTM D445)

This method specifies a procedure for the determination of the kinematic viscosity by measuring the time for a volume of liquid to flow under gravity through a calibrated glass capillary viscometer. The dynamic viscosity can be obtained by multiplying the kinematic viscosity by the density of the liquid.

Density (ASTM D4052)

This experiment covers the determination of the density or relative density of petroleum distillates and viscous oils. A small volume (approximately 0.7 mL) of liquid sample is introduced into an oscillating sample tube and the change in oscillating frequency caused by the change in the mass of the tube is used in conjunction with calibration data to determine the density of the sample.

Needle Penetration (ASTM D1321)

The depth penetrated (0.1 mm) in a cylinder of wax by a standard needle, with a load of 100g, in a specific temperature, during 5 seconds, corresponds to the 'needle penetration' measurement.

Refractive Index (ASTM D1218)

The refractive index is measured using a high-resolution refractometer of an optical-mechanical or automatic digital type with the prism temperature accurately controlled. The instrument principle is based on the critical angle concept.

2.3 Basic concepts about neural networks

A common network has multilayer configuration with parallel processing. The most used is MLP (multilayer perceptron), with an input layer, a hidden layer and an output layer (Fig. 3.).

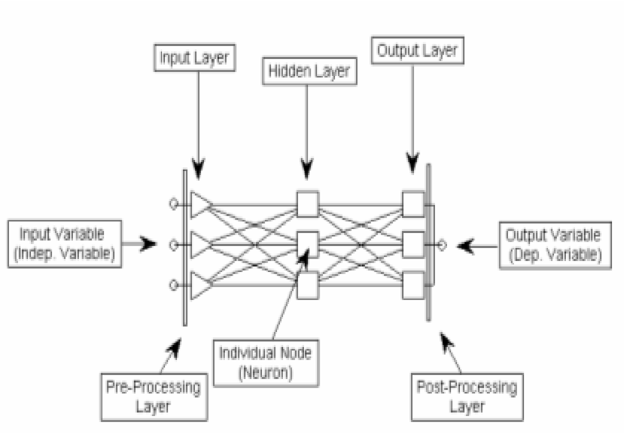

Fig. 3. Example of MLP network (De Souza Jr.,1993).

Data are fed in the input layer, which has a neuron per each input variable. Each one of the neurons in input layer is connected to each neuron of the hidden layer. Seemingly, each hidden neuron is connected to each unit of the output layer. The number of neurons in the output layer is the same number of output variables. Signals arriving on a neuron go to cell body, where they are added to others that come from other neurons of the previous layer. The 'j' neuron (Fig. 4.) from the layer (k+1) receives a set of inputs $s_{pi,k}$ (i = 1, ..., nk) corresponding to the outputs of nk neurons from previous layer. These outputs were influenced by wjik weights that correspond to each connection. The neuron sums inputs and the resultant value is added to a bias (an inner limit of activation) represented by $\theta_{j,k+1}$. The response $s_{pj,k+1}$ is produced by 'j' neuron to this signal, according to an activation function f( ) called transfer function (De Souza Jr., 1993).

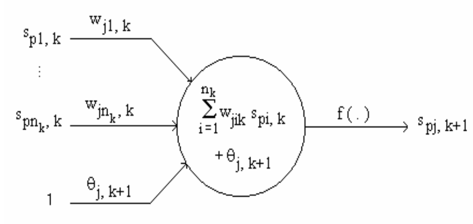

Fig. 4. The jth Neuron (De Souza Jr., 1993).

Common transfer functions are:

- Linear function:

$$f\left(\lambda_{pj,k+1}\right) = \lambda_{pj,k+1} \tag{1}$$

- Sigmoidal function:

$$f\left(\lambda_{pj,k+1}\right) = \left[1 + \exp\left(-\lambda_{pj,k+1}\right)\right]^{-1} \tag{2}$$

- Hyperbolic function:

$$f\left(\lambda_{pj,k+1}\right) = \tanh\left(\lambda_{pj,k+1}\right) \tag{3}$$

The training phase of a neural network consists in giving a set of data, with inputs and outputs known, so weights and biases for each neuron of network are adjusted with training algorithms using prediction errors, until network results in correct predictions of outputs. The procedure is iterative and continues until minimization of the global error function is reached. A second subset of data is randomly chosen for selection section or validation. These data are not used in the adjustment of weights and biases during training section, but performance of network is checked during the training with them. If error of selection data is not decreasing or begins to increase for a specified number of iterations, training is stopped. If training does not have restrictions, neural networks can describe training data very well, but usually describe new data poorly. Because of this, a third subset of data is randomly chosen as an additional check of the capacity of generalization of the neural network (De Souza Jr., 1993).

2.4 *Experimental Procedure*

In this section a sample of macrocrystalline wax 150/155 produced from heavy oil was utilized. The experiment of oil content was performed according to ASTM D 721 and resulted 0.69% m/m. Different fractions of heavy base oil were added to 15 portions of original sample to obtain new samples with oil content ranging from 1 to 15% m/m. The experiments of needle penetration 25ºC, refractive index 70ºC, viscosity 80ºC and density 70ºC were performed three times for each sample.
In addiction to the laboratory data, results of experiments performed on final products of a refinery like 120/125, 130/135, 140/145 and 150/155 waxes, produced by the same petroleum, shown in Table 1 were considered in the study. The complete set of results (industrial plus laboratory) is presented on Figures 5,6,7 and 8.

Table 1. Results from experiments with different kinds of final waxes from the same petroleum.

Oil Content (%m/m)	Needle Penetration (1/10 mm)	Viscosity 80ºC (cSt)	Density a 70ºC	Refractive Index
0.97	21.0	9.228	0.7895	1.4410
0.98	21.2	9.285	0.7892	1.4410
0.99	21.8	9.069	0.789	1.4405
1.47	22.5	8.149	0.7871	1.4401
1.06	25.2	8.417	0.7877	1.4402
1.06	22.0	8.369	0.7876	1.4402
1.01	22.0	8.671	0.7885	1.4404
1.07	24.0	8.348	0.7876	1.4402
0.99	20.0	9.018	0.7889	1.4408
1.09	16.0	5.254	0.7757	1.4355
0.99	16.0	5.183	0.8097	1.4352
0.91	25.0	5.322	0.7752	1.4353
0.44	15.2	5.19	0.7748	1.4339
0.54	25.0	5.449	0.7752	1.4369
0.94	43.0	5.343	0.7759	1.4344
0.54	15.0	5.141	0.7747	1.4352
0.82	20.0	5.235	0.7751	1.4341
3.01	31.6	4.655	0.7733	1.4335
2.96	43.6	4.217	0.7712	1.4320

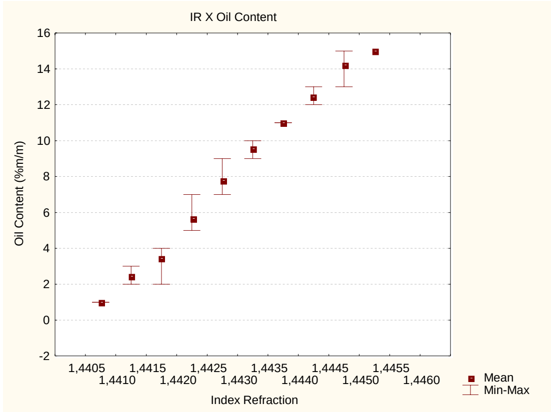

Fig. 5. Results of Index Refraction vs Oil Content

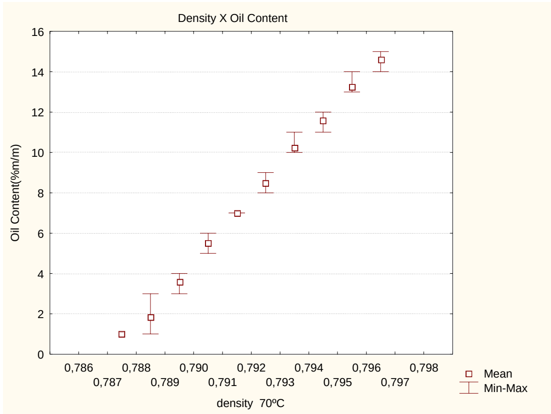

Fig. 6. Results of Density vs Oil Content

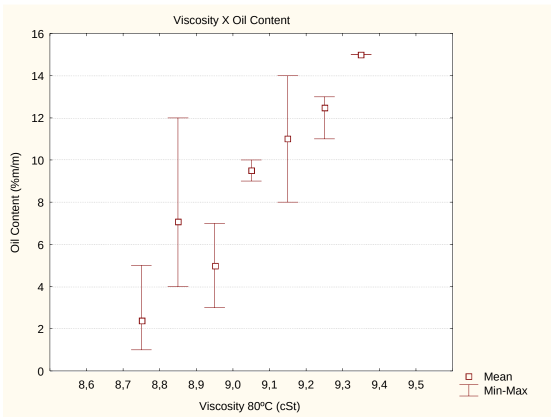

Fig. 7. Results of Viscosity vs Oil Content

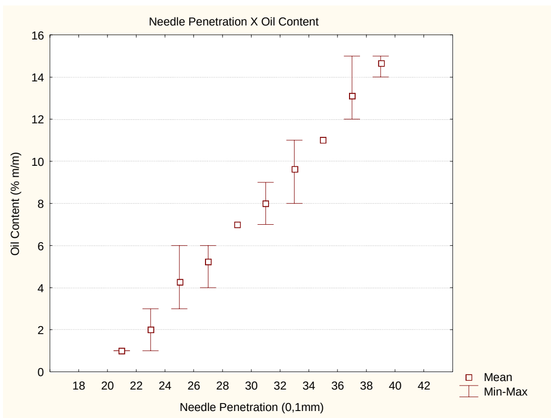

Fig. 8. Results of Needle Penetration vs Oil Content

2.5 *Simple Linear Regression*

Prior to linear regression, the data were analysed for autlier detection (value outside the range between + or – $2,5\sigma/n^{1/2}$) and no outliers where found. The observation of Figures 5 to 8 shows that the viscosity measurement has a large variability. Additionally, it is noticed that for medium and high oil content values an approximate linear dependence is observed between this characteristic and the other ones studied. So, simple linear regression models were tested first.
The simple linear regression between each variable and the oil content is presented in Figures 9 to 12.

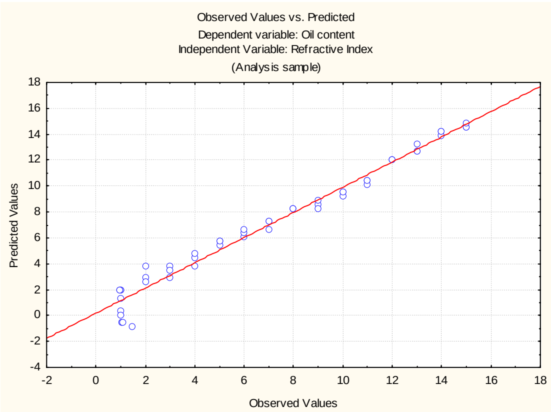

Fig. 9. Linear Regression – Refractive Index X Oil Content

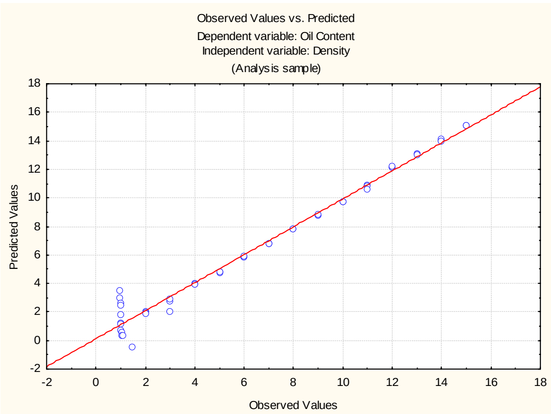

Fig. 10. Linear Regression – Density vs Oil Content

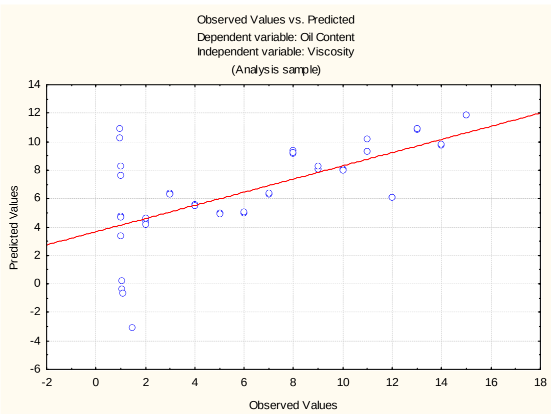

Fig. 11. Linear Regression – Viscosity vs Oil Content

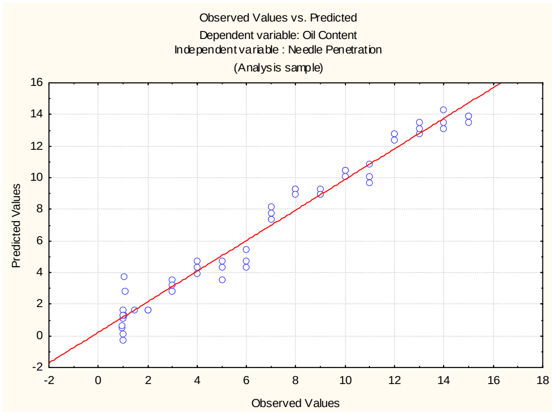

Fig. 12. Linear Regression – Needle Penetration vs Oil Content

The R-square (also known as determination coefficient) values of the linear regressions are in Table 2. This parameter indicates the percentage of the data variation that is explained by the linear model.

Table 2. R-square of linear regressions

Variables X Oil Content	
	R-square
Penetration	0.9564
Viscosity	0.6908
Density	0.9823
Refractive Index	0.9725

Viscosity has the worst result for linear analysis, due to its higher variation. For values between 0 and 6% m/m of oil content, linear predictions show the higher deviations. However, this range is the most interesting one, taking into consideration the final products. This motivated the use of non-linear models in this work. Neural networks were assumed, as they do not demand that the specific non-linear dependence is explicitly described.

2.6 *Building neural network*

The complete set of data was randomly separated in three subsets (in proportion 2:1:1) for training, selection (validation) and test (a second independent validation).

The software S TATISTICA© 6.0 was used to select the best network, based on data training and validation, through statistical analysis of the residuals of the predictions.

3. DISCUSSIONS AND RESULTS

A descriptive statistic analysis can be observed on Table 3. SD ratio is the ratio between standard deviation of difference between predict values and outputs, and Standard deviation between outputs and its average, that is, ratio between prediction deviation and deviation of real data from its average.

The best regression model is the one with lower SD ratio and networks with the best performances has this value closer to 0. An MLP with performance ranked as 'Excellent' was obtained, that uses backpropagation method of calculation, with an input layer operating with 4 neurons and linear functions, 9 neurons in hidden layer with hyperbolic functions and 1 linear neuron in output layer. The performance of non-linear network was compared with the best results obtained from the best linear network, with 4 input neurons and one output, as shown in Table 3.

An observation of SD ratio indicates that the networks can be used to predict the property proposed and MLP network is better than linear, as the MLP network has a SD ration five times lower than the linear model.

Table 3. Results from Statistica© of statistics analysis of networks

Descriptive Statistics	Linear	MLP
Average Error	0.2472	-0.04306
SD Error	1.465	0.3129
Absolute Average Error	0.9363	0.2544
SD Ratio	0.3053	0.06517
Correlation	0.9525	0.9979

In Table 4, it is possible to see the performance parameters for training, selection, and test that represent SD ratios for those sections. The MLP network has lower values to SD ratios and errors, proving itself as more adequate to be used for prediction.

Table 4. Results from Statistica© for training , selection and test of networks.

Training Summary- Performances						
Parameter	Train	Selection	Test	Error Train	Error Selection	Error Test
MLP	0.06162	0.05458	0.09213	0.02151	0.02449	0.02541
Linear	0.1840	0.4667	0.2866	0.06054	0.1610	0.09181

In Figure 12 it is possible to see the results of predicted versus observed values for MLP and Linear networks. The straight line to MLP shows that predictions are satisfactory. The linear model presents very high deviations at low oil content values, which makes it unadvisable to use in that range.

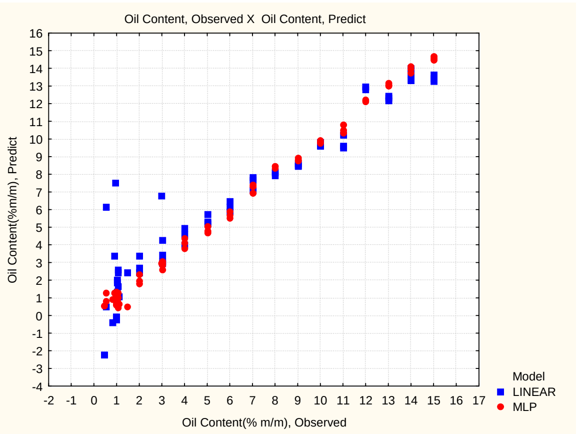

Fig.12. Predict oil contents X observed oil contents

The sensitivity analysis of inputs shows the relative contribution of each variable. Each variable is treated as if it were unavailable for analysis, being substituted by its average value. Global error of network when the variable is not available is divided by global error when it is available, resulting a ratio that should be bigger than 1.0, if variable contributes to the solution of the problem. The results are in Table 5. All variables have influence on oil content, receiving rank 1st the most influent variable.

Table 5. Results of sensitivity analysis from Statistica© for networks

Sensibility Analysis				
	Needle Penetration	Viscosity	Density	Refractive Index
MLP Ratio	4.287	11.51	7.855	9.258
MLP Rank	4 th	1st	3rd	2nd
Linear Ratio	1.779	3.070	1.034	4.895
Linear Rank	3rd	2nd	4th	1st

4. CONCLUSIONS

It is possible to see that the trained neural network makes good predictions to the set of data obtained from the proposed experiments and to the set of results obtained from final products of a refinery. The model proposed of a multilayer network as a MLP (4-9-1), with hyperbolic functions in the hided layer, presented correlation 0.9979 against 0.9525 of the linear model, beyond of best train, selection and test performances.

Additionally, as neural networks may outfit the data, special care was taken here on order to avoid this risk by using two validation (selection and test) data sets

The linear models can be considered as satisfactory if the range of oil content is above 6 % m/m. However for values between 0 to 6 %, linear models produce very large errors (even negative values may be obtained).
As this low range is important for the final product, the ANNs reveal itself as the best option to infer oil content from characteristics that may be obtained through more rapid experiments.

The neural network can be converted in a program written in C++, and interfaced, using a man machine interface, to the operator, so that he or she can use to get the result of oil content from the inputs considered. Additionally, a controller can be implemented to act on process by changing variables based on that inference.

The characteristics of petroleum processed to production of waxes have influence on the physical properties used to the prediction context proposed in this paper. This study was performed with a product of one kind of petroleum. To another kind of petroleum, it would be necessary to retrain the neural network including the new data.

REFERENCES

Baughman, D. R. and Y. A. Liu (1995); *Neural Networks In Bioprocessing and Chemical Engineering,* Academic Press, San Diego.

De Cerqueira, E. O, J. C. De Andrade and R. J. Poppi (2001); *Redes Neurais e suas Aplicações em Calibração Multivariada*, Quim. Nova, **Vol. 24**, **No. 6**, 864-873, 2001.

De Souza JR., M.B (1993); *Redes Multicamadas Aplicadas a Modelagem e Controle de Processos,* D.Sc. Dissertaion, PEQ/COPPE/UFRJ (in portuguese).

Funahashi, K.; *On the Approximate Realization of Continuous Mappings by Neural Networks*, Neural Networks, **2**, 183-192.

Haykin, S. (1999). *Neural networks – A comprehensive foundation*, Prentice Hall, Upper Saddle River, New Jersey.

Hecht-Nielsen, R.; *Theory of the Backpropagation Neural Network*, IEEE Int. Conf. on Neural Networks, **Vol. I**, 593-605, San Diego, 1989.

Hornik, K.; M. Stinchcombe & White; *Multilayer Feedforward Networks are Universal Approximators*, Neural Networks, **2**, 359-366, 1989.

Lima, A. D. M. & N. F. Pereira (2005); *Manual de Parafinas - Qualidade e Toxicologia*; Workshop de Avaliação dos Objetivos da Produção, Petrobras, CENPES/PDAB/LPE, 2005.

Speight, James G. (2001); Handbook of Petroleum Analysis, John Wiley & Sons.

Stipanovic, A. J.; G. P. Firmstone and M. P. Smith (1997); *Having Fun With Base Oils: Predicting Properties Using Neural Networks; Symposium on Worldwide Perspectives on the Manufacture*, Characterization and Application of Lubricant Base Oils, American Chemical Society, San Francisco, CA, April-1997