Black Box Modelling of Gasoil Hydrotreating by Artificial Neural Networks

Ghazi Otmanine*, Kahina Bedda and Noureddine Bentahar

Univ M'hamed Bougara, Dep. of Chemical Process Engineering, Faculty of Hydrocarbons and Chemistry - Technology Laboratory Hydrocarbons Avenue de l'Indépendance, 35000 Boumerdes, Algeria

Abstract: Hydrotreating of gasoil is one of the most important processes in petroleum refining; it helps to improve the characteristics of diesel fuel to make it meet the required specifications and pollution standards. Modelling of this process would, among other things, allow to predict the product quality as a function of different process variables (temperature, pressure, space velocity). There are several modelling techniques, among them the Black Box type modelling by Artificial Neural Networks (ANNs), which is used to model the hydrotreating process at pilot scale. The approach used in this work is an artificial intelligence approach. The model developed is a neural network Multi Layer Perceptron (MLP) type. Network learning (NL) is carried out according to the Backpropagation Gradient algorithm with momentum; The Early Stopping (ES) technique has been used to prevent the effect of overfitting and thereby ensure a good generalization of the model. Experimental and predicted results show good agreement with an error not exceeding 4%.

Keywords: Hydrotreating, gas oil, modelling, artificial neural networks.

INTRODUCTION

Gas oil hydrotreating process is a catalytic process, in which gas oil obtained from either primary distillation of crude oil or conversion processes (visbreaking, coking and catalytic cracking), is treated under hydrogen pressure [1-3]. This process helps obtain a quality diesel fuel with low contents of sulphur, nitrogen, aromatics and olefins together with a better stability and a high cetane number (CN) [1].

The process of gas oil hydrotreating has known in recent years considerable development, as a result of changing pollution standards which are becoming increasingly more stringent particularly in Europe and the USA (Table **1**) [2,4]. Great efforts are expanded by the largest refining companies and reputable research laboratories, to improve and develop technologies and process catalysts, reflecting the importance of such a process in refining.

Sulphur more specifically sulphur dioxide SO_2 is a major among those particular products that contribute to pollution. Its concentration in gas oil is the object of strict regulations which seek to limit gas emissions of SO_2 of car exhausts [5]. When transformed first into sulphur trioxide SO_3 then mixed with exhaust water vapour, it produces sulphuric acid [5]. This acid pollution can then be transported by wind over several hundred kilometres and made to fall as acid rain. Sulphur dioxide causes respiratory diseases and can worsen cardio vascular ones [6]. On the other hand, sulphur has a negative effect on the proper operation of the catalytic converter itself whose main function is to reduce pollutant gas emissions in to the atmosphere. Furthermore, the presence of appreciable quantities of SO_2 in the fuel easily oxidable into SO_3 can poison the catalysts of the catalytic converter [6,7]. Sulphur of straight run (SR) fractions derived directly from atmospheric distillation is strongly depending on that of crude oil. However, it has been noticed that gas oil contains about twice less sulphur than the crude from which it is derived [8]. Up until 1994 gas oil specifications allowed a sulphur content of up to about 0.2-0.3 %. Current legislation requires a content of no more than 0.05 %, which entails new process design and a modification of the operation of existing industrial units [9]. Table **2** shows the progressive development of specifications on sulphur levels in gasoline and diesel in the European Union [10].

Although reaching such a low level of sulphur content requires setting up hydro desulphurization units operating under relatively more stringent conditions. Until recently, reduction of sulphur content in gas oil was beneficial to the engine against risks of corrosion wear. However, marketing of highly desulphurized gas oil has led to other corrosion effects affecting the injection system and no longer the engine. These effects will appear in the form of sticking of injection pumps caused by wear effects and taking place after several thousands km of run [11.12]. Therefore hydrotreating by the desulphurization process of gas oil

*Address correspondence to this author at the Univ M'hamed Bougara, Dep. of Chemical Process Engineering, Faculty of Hydrocarbons and Chemistry - Technology Laboratory Hydrocarbons Avenue de l'Indépendance, 35000 Boumerdes, Algeria; Tel: +213 24 79 51 70; Mob: +213 77 33 88 929;
E-mail: oth_gha@yahoo.fr, oth_gha@umbb.dz

E-ISSN: 2409-787X/14

Table 1: Changing Diesel Fuel Specifications in Europe and the U.S. (a) [2], (b) [4]

Gas oil characteristics	Europe (a)			USA (b)	
	1996	2000	2005	1993	2006
Cetane index	49	51	51 - 58	40	40
Aromatics (% vol)	-	-	-	35	35
Polyaromatics (% poids)	Unspecified	11	11 - 2	-	-
Sulphur (ppm)	500	350	50 -10	500	15

Table 2: Progressive Development of Specifications on Sulphur Levels in Gasoline and Gas Oil Fuel in the European Union

	1994	1996	2000	2005	2009
gasoline (ppm)	1000	500	150	50	10
Diesel fuel (ppm)	2000	500	350	50	10

is highly desirable even necessary to reduce particulate emissions and risk of corrosive wear of diesel engines & injection systhems.

The hydrotreating catalysts are composed of an oxide support and an active phase in the form of molybdenum sulphide or tungsten enhanced by cobalt or nickel. Commonly used formulas are the associations CoMo, NiMo and NiW for the active phase and alumina γ with large specific surface area for the support [13,14].

In industry, the most widely used catalyst for the hydrotreatment of gas oil fuel is the CoMo / Alumina, but now with the new specifications endorses, this catalyst performance is no longer satisfactory. Therefore its doping with phosphorous is necessary. Indeed a new catalyst of the formula $CoMoP/Al_2O_3$ and referenced C-606a [15] has been marketed in Japan. It is characterized by high activity and great stability; it also allows the production of a fuel with sulphur content below 10 ppm.

Another method has been developed for improving hydrotreating catalysts performance by their doping with noble metals such as Platinum (Pt) and Rutheium (Ru). The adding effects of transition metals such as nickel, ruthenium and tungsten $CoMo/\gamma\text{-}Al_2O_3$ [16] to a commercial catalyst has been studied in terms of activity in sulphur removal. Improved catalytic performance has been achieved; and several catalysts have been developed, such as the following catalysts: $PtMo/Al_2O_3$ and $RuMo/Al_2O_3$, $RuCoMo/\ Al_2O_3$ but are still not yet used in industry [17-19].

On the other hand, a two-stage hydrotreatment using the same catalyst for each stage $CoMo/Al_2O_3$ allows to obtain a good quality gas oil fuel. Excellent results have been achieved particularly for the hydrodesulphurization; gas oil fuel with sulphur content below 10 ppm have been produced [20].

Given its importance in refining, several models of gas oil hydrotreating process have been developed [21-24]. These models are based on the laws of chemical kinetics and the equations of mass transfer between the phases in the hydrotreating reactor. They allow to predict product characteristics (content in sulphur, aromatics, and olefins) as a function of the different operating variables such as temperature, space velocity and hydrogen partial pressure. These models are often used for simulation of industrial hydrotreating units.

Nevertheless, these latter present a great disadvantage in that each model is specific to the gas oil for which it has been developed. This is due essentially to the assumptions made in order to simplify mathematical equations governing the process. For e.g. in the model presented by Murali *et al.* [24] all sulphur compounds present in the gas oil fuel (mercaptans, thiophenes, benzothiophene, dibenzo-thiophenes.) are considered as one pseudo-component. During a change in the load composition, the model provides meaningless results for the pseudo-component chosen is no longer representing the new gas oil sulphur compounds.

2. HYDROTREATING PROCESS MODELLING

It would be very interesting to develop a more general model that could predict the product characteristics for all hydrotreated gas oil fuels. Such an approach is impractical using the fundamental laws that govern process engineering. To achieve this objective, a *black box* type model based only on experimental data is therefore proposed. This technique is increasingly used in process engineering [25, 26]. It is used in modelling fluid catalytic cracking FCC process for obtaining gasoline [27], process water treatment [28] and modelling apparatus such as heat exchangers [29]. The most suited technique for developing this type of modelling is the Black Box modelling by Artificial Neural Networks (ANNs).

An *ANNs* is a computational model whose design is basically inspired from the operation of real neurons [30]. Neural network modelling has found extensive applications in the field of process engineering in recent years greatly encouraged by results achieved through this approach [31]. This technique is very powerful in that only a small identification effort is enough to construct non-linear multivariable models that can yield excellent agreement between experimental and predicted values.

There are two families of neural networks: feedforward neural networks and feedback neural networks. An feedforward neural network is presented graphically by a set of interconnected neurons. The information flowing from inputs to outputs without backtracking; if the neural network is represented by a graph whose nodes are neurons and the edges are the connections between nodes, the graph of a network is not completed acyclic; moving through the network, from any one neuron and following the connections, we can not return to the starting neuron [30,31]. The great majority of applications of neural networks use networks with layers as depicted in the example in Figure **1**.

This feedforward neural network with n_i inputs, n_h hidden neurons and n_o output neurons, implement nonlinear functions of its n input variables (are often designated by weight $x_1, x_2, \ldots x_n$) through composition of n_{al} algebraic functions performed by its hidden neurons [30].

It should be emphasized that time plays no functional role in an open loop neural network if the inputs as well as the outputs are constant. The time required for the computation of the function performed by each neuron is negligible and, functionally, this computation can be considered as instantaneous. For this reason feedforward networks are often called "static networks", as opposed to feedback neural networks which are called "dynamic networks" [30,31].

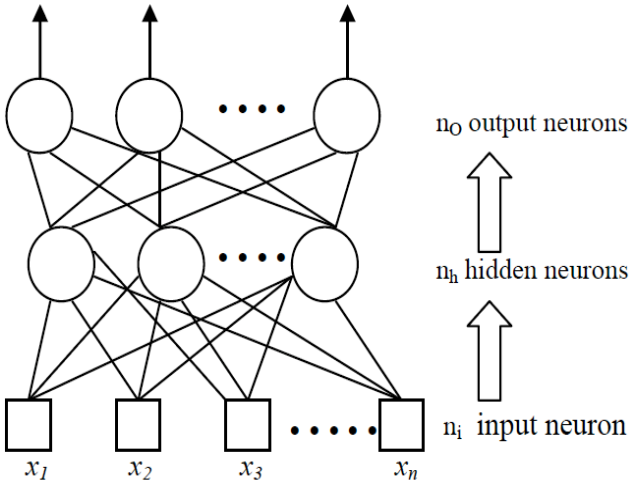

Figure 1: Representation of an feedforward neural network.

For feedback neural networks, the graph architecture is different from that of feedforward neural networks, where the graph is cyclic: as one moves in the network by following the direction of connections, it is possible to find at least one path that returns to its starting point (such a path is referred to as a cycle). The exit of a neuron on the network can be a function of itself, it is clearly conceivable that if the concept of time is explicitly considered [30,31]. In these conditions, any graph cycle of the feedback neural networks must include at least one connection of non-zero delay.

Figure **2** shows an example of a feedback neural network. The numbers displayed in the squares represent the delay linked with each connection, expressed as a multiple of the unit of time (or sampling period) T. This network includes a cycle which begins from the neuron 3, goes to the neuron 4, and back to neuron 3, the connection of 4 to 3 is linked to a delay of one time unit (q-1) [30,31]

All feedback neural networks whatever its complexity, can be put into canonical form comprising a feedforward neural network, in which some outputs (state variables) are returned to the inputs by loopbacks of a unit delay [32]. This canonical form is consequently made up of an acyclical connection graph of one unit delay connecting certain outputs of this graph to the inputs of the network. For example, the

feedback neural network shown in Figure **2** can be put into the canonical form shown in Figure **3** [30]. This network has a single state variable x, it is therefore, a first order. The gray part of the canonical form is a feedforward neural network.

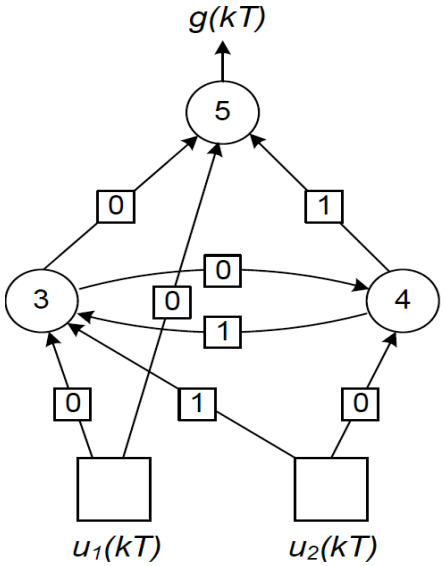

Figure 2: Representation of a feedback neural network.

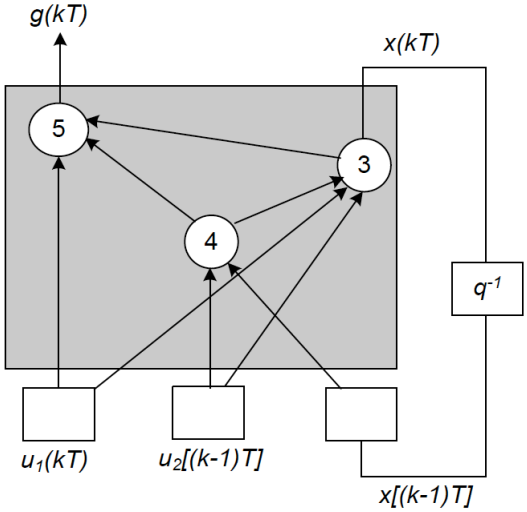

Figure 3: Form canonical on the feedback neural network.

Thus, any neural network can be put into a canonical form consisting of a static neural network, consequently, the general properties of either feedback or feedforward neural networks, therefore depend on the properties of feedback neural networks.

Feedforward neural networks with layers whose hidden neurons have a function of sigmoid activation are often called Multilayer Perceptron (MLP) [30]. Example applications of MLP in process engineering are the modelling of the fluid catalytic cracking process and crude oil distillation process [33]. MLP can also estimate the characteristics of products obtained by different methods such as biodiesel fuels density [34] and cetane index [35].

Note that artificial neural networks of the MLP type were utilized to model the two-stage process of hydrotreating of diesel fuel on a pilot scale. The developed model links the sulphur content of hydrotreated diesel with the following operating variables: temperature (t), pressure (p), hourly space velocity (HSV) and the sulphur content of the load & number of hydrotreating stages.

3. DATA AND RESULTS OF EXPERIMENTS

Five light gas oil fuels have been provided by a refineries in Algeria with varying sulphur content were obtained by atmospheric distillation and have been tested under variable operating conditions of temperature, pressure & HSV. The hydrogen/load ratio was maintained at 500ml. ml^{-1} for all experiments. Experiments were performed both on one (1) single stage as well as two (2) stages. The characteristics of these gas oil fuels are shown in Table **3**. The amount of catalyst used for diesel hydrotreating is 50 c

The catalyst is placed in the pilot reactor between two layers of 25g of silica (silica allows a better distribution of liquid and gas phases on the catalyst bed). The catalyst utilized was the CoMo/alumina type

Table 3: Loads Characteristics

Gasoil fuel	A	B	C	D	E
Density	0.864	0.846	0.,838	0.835	0.827
Sulphur content (ppm)	12000	10000	7500	1870	990
Distillation ASTM (°C)					
10% vol	259	255	251	245	241
50% vol	292	289	286	282	279
90% vol	341	337	333	330	328
FP	357	354	351	349	346

whose characteristics are shown in Table **4**. The sulphur content of loads and products were determined by X fluorescence.

Table 4: Catalysts Properties of a Sulphurized Commercial Catalyst of the Type CoMo /Alumina

Chemical composition (wt %)	
Co	4.2
Mo	16.7
S	4.6
Co/Mo atomic ratio	0.4
Characteristics:	
Density (g cm^{-3})	0.87
Specific surface area (m^2 g^{-1})	130.00
Porosity (cm^3 g^{-1})	0.19
Pores average diameter (nm)	4.5

A total of twenty five (25) experiments were conducted to study the variation of sulphur content of hydrotreated diesel fuel as a function of process operating conditions, sulphur content of the load and number of hydrotreating stages. Table **5** depicts the results obtained for each individual experiment.

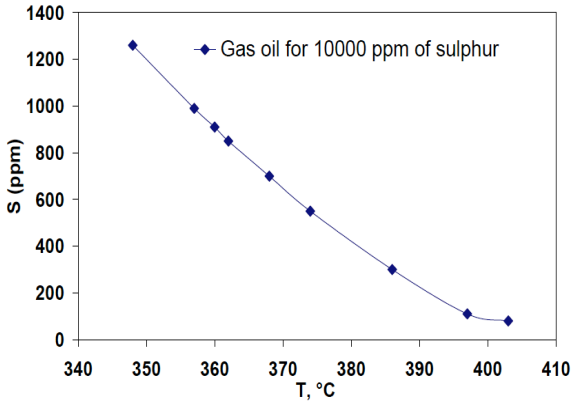

Figure 4: Effect of hydrotreating temperature on the product sulphur content (P=800 psi, HSV=2.6 h^{-1}).

Figures **4**, **5** and **6** indicate that increasing the temperature or pressure will result in a reduction in the

Table 5: Characteristics and Results of Experiments of Different Hydrotreating Gasoil from Atmospheric Distillation

n°exp	T (°C)	p (psi)	HSV (h^{-1})	Sulphur content in load (ppm)	N° of stages	Sulphur content in product (ppm)
1	348	800	2.6	10000	1	1200
2	357	800	2.6	10000	1	990
3	362	800	2.6	10000	1	850
4	368	800	2.6	10000	1	700
5	374	800	2.6	10000	1	550
6	386	800	2.6	10000	1	300
7	397	800	2.6	10000	1	110
8	403	800	2.6	10000	1	80
9	360	800	2.8	10000	1	910
10	360	1200	2.8	10000	1	780
11	360	1600	2.8	10000	1	720
12	360	2000	2.8	10000	1	700
13	360	800	0.4	12000	1	350
14	360	800	0.47	12000	1	390
15	360	800	0.67	12000	1	470
16	360	800	1.14	12000	1	640
17	360	800	2.37	12000	1	890
18	360	800	2.6	12000	1	920
19	360	800	6.0	7500	1	1870
20	360	800	3.0	7500	1	1250
21	360	800	6.0	1870	2	1140
22	360	800	1.5	1870	2	350
23	360	800	2.1	990	2	170
24	360	800	1.0	990	2	50
25	360	800	0.4	990	2	10

sulphur content of hydrotreated diesel fuel; whereas an increase in space velocity will result in an increase in sulphur content.

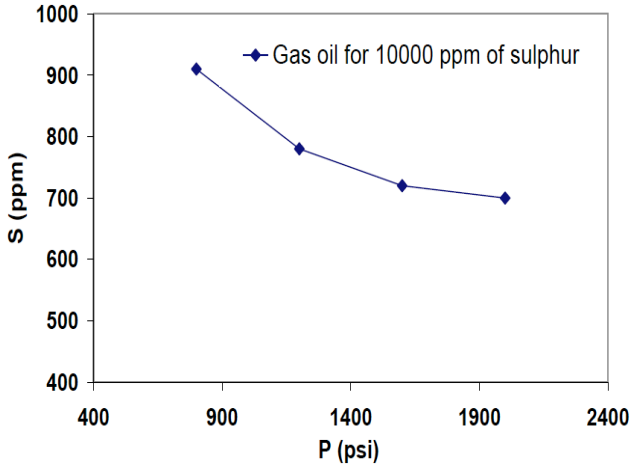

Figure 5: Effect of the hydrotreating pressure on the product sulphur content (T=360°C HSV=2.8 h^{-1}).

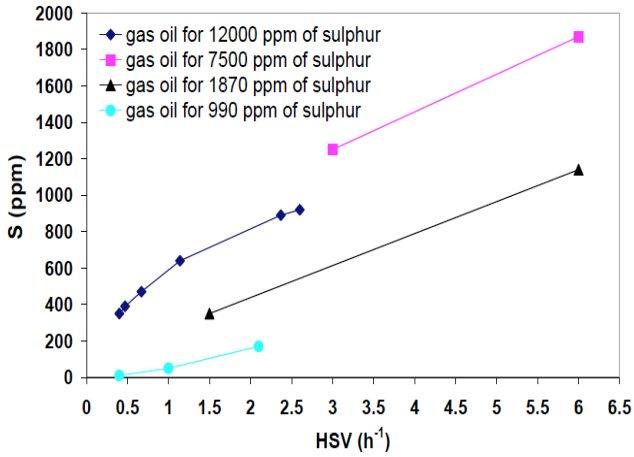

Figure 6: Effect of the hydrotreating space velocity on product sulphur content (T=360°C, P=800 psi).

According to the graphical in Figure **4** representation of experiments N° 1 to 8 the sulphur content of products decreases with increasing temperature. Experiments N° 9-12 shown in Figure **5** indicate that the increase in pressure reduces the sulphur content of products in particular when it varies between 800 and 1200 psi; between 1200 and 2000 psi, the decrease in sulphur content is less important. Figure **6** representing the experiments N° 13 to 18, N° 19 and 20, N° 21 and 22, N° 23 to 25 indicates that the increase of the hourly space velocity increases the sulphur content of gas oil hydrotreating. The hydrotreated diesel of experiment N°2 is used as feed for the tests N° 23, 24 and 25, while the diesel obtained from the experiment N°19 is used as feed for the tests N° 21 and 22. A second stage of hydrotreating allows obtaining results much more satisfactory, for example the single stage experiment N°2 provids a diesel with a sulphur content of 990 ppm, by substituting a second stage (experiment N° 25) the sulphur content decreases until 10 ppm.

4. METHODOLOGY AND APPLICATIONS

The Multi Layer Perceptron MLP neural networks are best suited for modelling processes such as hydrotreating of diesel. The practical advantage of these networks compared to classical techniques of nonlinear modelling lies in their ability to produce models with comparable accuracy with less experimental data (or in their capacity to build more accurate models from the same amount of data). This is the property of universal approximation [30].

It goes without saying that the universal approximation property is not specific to neural networks: polynomials, Fourier series, spline functions possess the same particularity. Therefore, what distinguishes neural networks from other common universal approximators is their parsimony [36]

To obtain an approximation of a given accuracy, the neural networks use fewer parameters than the usual approximators.

In practice, neural networks are advantageous compared to conventional approximators, when attempting to solve a problem for more than one or two variables [31]. This advantage is particularly interesting in terms of computation time, especially in terms of the amount of information necessary for the calculation of coefficients

The implementation of this model has been carried out using the MATLAB software (version 7.3) [37] which provides a special interface for artificial neural networks, no programming was necessary. The process variables: temperature, pressure, space velocity, load sulphur content, and the number of hydrotreating stages are the inputs to the network and the sulphur content of the hydrotreated gas oil is its output.

The Multi Layer Perceptron (MLP) will thus comprise three layers as shown in Figure **7**.

- The first layer consists of five input variables: temperature, pressure, sulphur content of the load, number of stages.

- The second layer will contain a given number of hidden neurons: x hidden neurons.
- The third layer consists of a single output neuron.

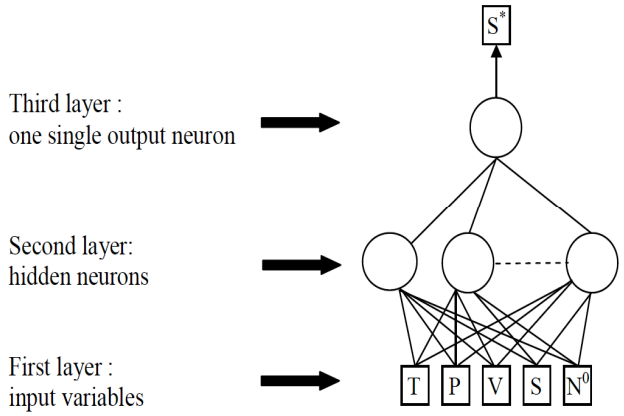

Figure 7: Network architecture used.

To ensure the universal approximation property of the network, the activation function of hidden neurons was chosen to be a sigmoid function. The function mostly used in practice is the logistic function [37] given by:

$$f(v) = \frac{1}{1+\exp(-v)} \quad (1)$$

Where: (ν) is the potential given by:

$$v = w_0 + \sum_{i=1}^{n-1} w_i x_i \quad (2)$$

Where:

x_i: Inputs / Starters or network variables. w_i: Parameters or weight of the network. w_o: Bias.

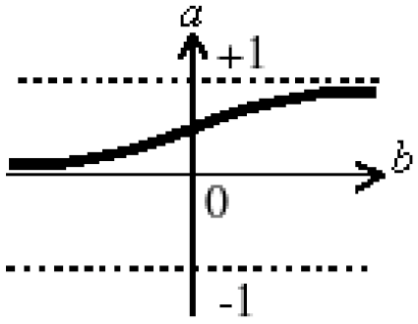

Figure 8: a = logsig (b).

This function allows limiting the amplitude of the outputs between 0 and 1 as shown in Figure **8** a; it is called under MATLAB environment MATLAB l*ogsig*.

According to the Universal Approximation Theorem, the activation function of the output neuron must be linear: *f(v)* = *v* and it is known under MATLAB as *Purelin* (b) as shown in Figure **9** a.

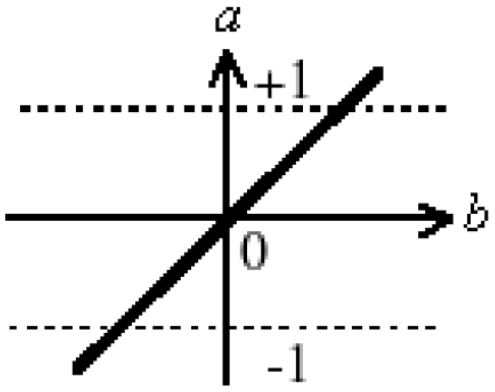

Figure 9: a = Purelin (b).

In practice, the model input and output data are normalised over the interval [0.1, 0.9], which will allow a better network convergence. The function used for normalizing the different variables is given by the following equation [37]

$$y = (y_{\max} - y_{\min}) * (x - x_{\min}) / (x_{\max} - x_{\min}) + y_{\min} \quad (3)$$

Where:

x: is an element of the (input or output) vector to normalize.

x_{max}: is the value of the largest vector element to normalize.

x_{min}: is the value of the smallest vector element to normalize. *y*: is the normalized value of x.

y_{max}: is the maximum value of the interval of normalization, i.e 0.9.

y_{min}: is the minimum value of the interval of normalization i.e 0.1.

The MATLAB function that performs this operation is called "*mapminmax*". Normalized data are compiled in Table **6**. Learning in the present case is a supervised non-adaptive supervised learning. It consists of determining from cases submitted to the network all the parameters *(w)* which minimizes the cost function of the least squares *J(w)* [37] given by:

$$J(w) = \frac{1}{2} \sum_{k=1}^{N} \left[y_p\left(x^k\right) - g\left(x^k, w\right) \right]^2 \quad (4)$$

Table 6: Normalized Data of the Process (Learning, Validation, Test)

N°exp	T (°C)	P (psi)	HSV (h^{-1})	Sulphur content in load (ppm)	N° of stages	Sulphur content in product (ppm)
1	0.1000	0.1000	0.4143	0.7547	0.1000	0.6376
2	0.2309	0.1000	0.4143	0.7547	0.1000	0.5215
3	0.3036	0.1000	0.4143	0.7547	0.1000	0.4613
4	0.3909	0.1000	0.4143	0.7547	0.1000	0.3968
5	0.4782	0.1000	0.4143	0.7547	0.1000	0.3323
6	0.6727	0.1000	0.4143	0.7547	0.1000	0.2247
7	0.8127	0.1000	0.4143	0.7547	0.1000	0.1430
8	0.9000	0.1000	0.4143	0.7547	0.1000	0.1301
9	0.2745	0.1000	0.4429	0.7547	0.1000	0.4871
10	0.2745	0.3667	0.4429	0.7547	0.1000	0.4312
11	0.2745	0.6333	0.4429	0.7547	0.1000	0.4054
12	0.2745	0.9000	0.4429	0.7547	0.1000	0.3968
13	0.2745	0.1000	0.1000	0.9000	0.1000	0.2462
14	0.2745	0.1000	0.1000	0.9000	0.1000	0.2634
15	0.2745	0.1000	0.1386	0.9000	0.1000	0.2978
16	0.2745	0.1000	0.2057	0.9000	0.1000	0.3710
17	0.2745	0.1000	0.3814	0.9000	0.1000	0.4785
18	0.2745	0.1000	0.4143	0.9000	0.1000	0.4914
19	0.2745	0.1000	0.9000	0.5730	0.1000	0.9000
20	0.2745	0.1000	0.4714	0.5730	0.1000	0.6333
21	0.2745	0.1000	0.9000	0.1639	0.9000	0.5860
22	0.2745	0.1000	0.2571	0.1639	0.9000	0.2462
23	0.2745	0.1000	0.3429	0.1000	0.9000	0.1688
24	0.2745	0.1000	0.1857	0.1000	0.9000	0.1172
25	0.2745	0.1000	0.1000	0.1000	0.9000	0.1000

Where: g(x,w): Approximation of the regression function.

Learning of the Multilayer Perceptron (MLP) is accomplished using the Gradient Backpropagation Algorithm with Momentum. Modifying the network weights was based upon second order_Levenberg-Marquardt optimization technique. The Early Stopping (ES) technique was utilized to avoid the effect of model overfitting [37].

MODEL STRUCTURE

The approach consists of varying the number of neurons of the hidden layer. Several networks with different structures will be tested and the network yielding the smallest error will be chosen.

The initialization of weights and biases is carried out randomly between -1 and +1. For a given number of hidden neurons, several parameter initialization attempts were undertaken in order to maximise chances to arrive at the optimal model.

The determination of the optimal structure of the model is achieved iteratively.

Stages for the choice of the model structure is depicted in Figure **8**. Data is randomly divided into three groups:

- A learning set consisting of **10** test examples, used to compute the gradient and update the values of network parameters.
- A validation set consisting of **2** test examples, the error is being tracked throughout the learning process. As the validation error increases for a specified number of iterations (i.e.6), learning is stopped, that is to say, before complete

convergence of the algorithm, and the overfitting is kept within limits. The weights and biases were those that correspond to the minimum of the validation error.

- A test set containing **3** examples, not used during learning, it helps to determine the optimal number of hidden neurons that allows the best approximation of hydrotreated gas oil sulphur content while ensuring adequate generalization.

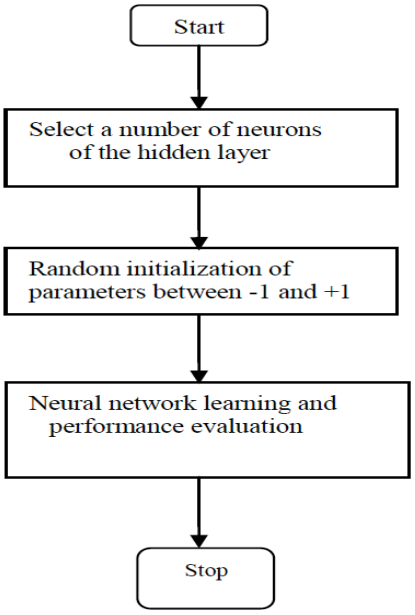

Figure 10: Applied algorithm of modeling.

RESULTS AND DISCUSSION

The number of neurons in the hidden layer have been varied between 1 and 10. The criterion for choosing between these different structures is the mean quadratic error of the test (TMQE). The network with seven (7) hidden neurons has the smallest TMQE (2.9462e-004) with very good mean quadratic errors on the training sets (1.6952e-005) and validation set (8.7257e-005). This model is the optimal model for determining the best approximation of the sulphur content of gas oil hydrotreating. The Mean Quadratic Errors across the model: Learning LMQE, test TMQE and validation VMQE are computed using the following equations $LMQE = \sqrt{\frac{1}{N_L}\sum_{k=1}^{N_A}\left[y_p^k - g\left(x^k, w\right)\right]}^2$

(5)

$$TMQE = \sqrt{\frac{1}{N_T}\sum_{k=1}^{N_T}\left[y_p^k - g\left(x^k, w\right)\right]}^2 \quad (6)$$

$$VMQE = \sqrt{\frac{1}{N_V}\sum_{k=1}^{N_V}\left[y_p^k - g\left(x^k, w\right)\right]}^2 \quad (7)$$

Where:

N_L, N_T, N_V: are respectively the numbers of examples of the learning set, validation and testing. g(x,y): Approximation of the regression function Variations of LMQE, TMQE and VMQE parameters during the learning process of the optimal network are shown in Figure **11**.

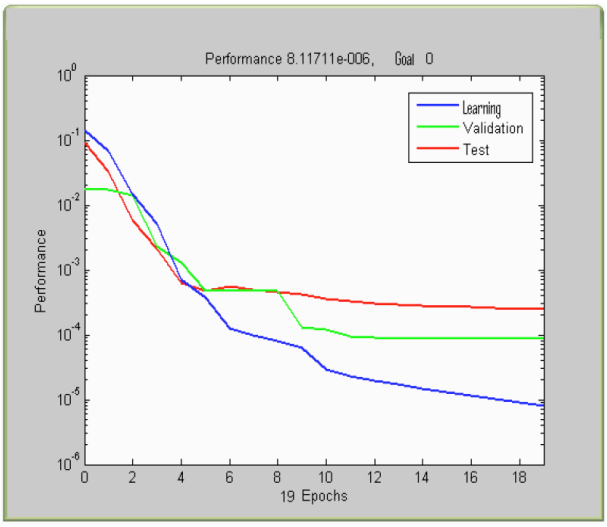

Figure 11: Variations of LMQE, TMQE and VMQE during the learning process of seven the hidden neurons network.

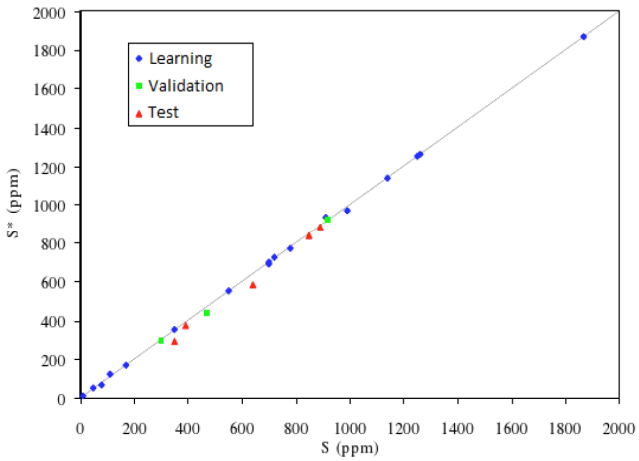

Figure 12: Simulated sulphur content (S*) versus measured sulfur content (S) of hydrotreated gas oils.

In order to make a comparison between the measured values (S) and the predicted ones (S*) of hydrotreated diesel sulphur content, normalization of the output values of the network was accomplished with the MATLAB function "mapminmax", 'reverse'. The points on Figure **12** represent predicted (S*) versus

measured values of sulphur content (S). It should be noted that these points are distributed around the axis "y = x", indicating a good agreement between predicted and measured values. The mean relative error regarding learning was 2.31 percent, validation set error was 3.62 percent and the entire test error was 6.74 percent. In summary, the network has achieved good learning as well as satisfactory generalization of the model.

CONCLUSION

Gas oil hydrotreating process has been modelled using artificial neurons of the Multi Layer Perceptron (MLP) network type. The determination of the number of neurons in the hidden layer and the choice of initial parameters of the network are made by trial and error. The results obtained are very good, the test error being less than 4%. The technique of artificial neural networks is very well suited for modelling this type of process.

Furthermore, it would be very interesting to enhance this type of model by including additional input variables, for example the load characteristics such as aromatics content, olefins content, and cetane index, or the process characteristics such as the catalyst type and the quantity of hydrogen. The developed model of gas oil hydrotreating process in two stages has provided very good results; this proves the ability of neural networks to solve this type problem.

The use of such a model on an industrial scale could make enormous services; it would focus the attention of operators when there are failures in the working of the process, for example during the aging of catalyst. It is important to note that the implementation of such models in a refinery is much simpler than the pilot scale because of the availability of large quantities of data. In fact, the monitoring of the process is carried out in a rigorous manner and this several times a day. The application of such an approach for other refining processes and petrochemicals would be very beneficial.

NOMENCLATURE

CO = carbon monoxide

g(x,w) = Approximation of the regression function

J(w) = Cost function of the least squares

HSV = hourly space velocity

N° = number of stages

NL = number of examples of the learning

NT = number of examples of the testing

NV = number of examples of the validation

n_i = inputs

n_h = hidden neurons

n_o = output neurons

n_{no} = nonlinear functions

n_m = nanometre

P(psi) = pressure (pound square inch)

PPM = parts per million

S = sulphur

(S) = sulphur measured

(S*) = sulphur predicted

SO_2 = sulphur dioxide

SO_3 = sulphur trioxide

T (°C) = temperature (Celsius degree)

V = volume

x = input or output vector. *x1, x2*, … *xn* weight input vector

x_i = inputs / starters or network variables

x_{max} = the largest vector element

x_{min} = the smallest vector element

y = normalized value of x

y_{max} = maximum value of the interval 0.9

y_{min} = minimum value of the interval 0.1

w_i = weight of the network

w_o = Bias

Abbreviations

ANNs = Artificial Neural Networks

MLP = Multi Layer Perceptron

NL = Network learning

ES = Early Stopping

FCC = Fluid Catalytic Cracking

HC = Hydrocarbons

HDS = Hydrodesulphurization

HDT = Hydrotreatment

LCO = Light Cycle Oil

LMQE = Learning Mean Quadratic Error

PAHs = Polycyclic Aromatic Hydrocarbons

SR = Straight Run

TMQE = Test Mean Quadratic Error

VMQE = Validation Mean Quadratic Error

VB = Visbreaking

REFERENCES

[1] Leprince P. Le Raffinage du Pétrole: Procédés de transformation. Editions Technip, Paris 1998; Vol. 3.

[2] Courty P, Gruson JF. Refining clean fuel for the future. Oil Gas Sci Technol – Rev IFP 2001; 56: 551-524.

[3] Kaminskii EF, Khavkin VA, Kurganov MV, *et al.* Production of ecological clean diesel fuels. Chem Technol Fuels Oils 1996; 32(2): 68-70

[4] Bensaïd B, Saint-Antonin V. Le Diesel aux Etats-Unis, Panorama. Institut Français du Pétrole, Paris 2004.

[5] Truex TJ, Pierson WR, McKee DE. Sulphate in Diesel Exhaust Environ Sci Technol 1980; 14(9): 1118-1121. http://dx.doi.org/10.1021/es60169a013

[6] Guibet JC. in: Techniques de l'ingénieur, Carburants liquides, BE 8545 1998.

[7] Guibet JC. Carburants et Moteurs. Editions Technip, Paris 1997; Vol. 2: pp. 505-506.

[8] Girard C, Guibet JC, Billon A, *et al.* Technico-economic aspects and environmental impact of gas-oil desulphurization. International Congress: State of the Art. Potential. SIA, Lyon, France 1993.

[9] Heinrich G, Kasztelan S, Kerdraon L. Diesel fuel upgrading: hydroprocessing for deep desulphurization and/or aromatics saturation, IFP Publication, Industrial Direction, Paris 1994.

[10] Monot F, Warzywoda M. Désulfuration microbiologique des produits pétroliers. In: Vandecasteele J-P (ed) Microbiologie pétrolière, Concepts Implications Environnementales, Editions Technip, Paris 2005; Vol. I: pp. 737-755.

[11] Lacey PJ, Lestz SJ. Effects of low lubricity fuels on diesel injection pumps. Part 1: field performance, part 2: laboratory evaluation. SAE 920823 and 920824 1992.

[12] Korres DM, Anastopoulos GE, Lois A, *et al.* A neural network approach to the prediction of diesel fuel lubricity. Fuel 2002; 81: 1243-1250. http://dx.doi.org/10.1016/S0016-2361(02)00020-0

[13] Knudsen GK, Cooper HB, Topsoe H. Catalyst and process technologies for ultra low sulphur diesel. Appl Catal A 1999; 189: 205-215. http://dx.doi.org/10.1016/S0926-860X(99)00277-X

[14] Eijsbouts S. Life cycle of hydroprocessing catalysts and total catalyst management. Stud Surf Sci Catal 1999; 127: 21-36. http://dx.doi.org/10.1016/S0167-2991(99)80391-7

[15] Fujikaa T, Kimura H, Kiriyama K, *et al.* Development of ultra-deep HDS catalysts for production of clean diesel fuels. Catal Today 2006; 111: 188-193. http://dx.doi.org/10.1016/j.cattod.2005.10.024

[16] Deuk KL, In Chul L, Seong IW. Effects of transition metal addition to Co/Mo γ-Al_2O_3 catalyst on the hydrotreating reactions of atmospheric residual oil. Appl Catal A 1994; 109(2): 195-210.

[17] Grange P, Vanhaeren X. Hydrotreating catalysts, and old story with new challenges. Catal Today 1997; 36: 375-391. http://dx.doi.org/10.1016/S0920-5861(96)00232-5

[18] Pinzon HM, Merino LI, Centeno A, *et al.* Performance of noble metal-Mo/γAl_2O_3 catalysts: Effect of preparation parameters. Stud Surf Sci Catal 1999; 127: 97-104. http://dx.doi.org/10.1016/S0167-2991(99)80397-8

[19] Pessayre S, Geantet C, Bacaud R, *et al.* Platinum doped hydrotreating catalysts for deep hydrodesulfurization of diesel fuels. I & EC Research 2007; 46: 3877-3883.

[20] Ramirez S, Cabreara C, Aguilar C, *et al.* Two stages light gasoil hydrotreating for low sulphur diesel production. Catalysis Today 2004; 98: 323-332. http://dx.doi.org/10.1016/j.cattod.2004.07.045

[21] Vradman L, Landau MV, Herskowitz M. Deep desulphurization of diesel fuels: kinetic modeling of model compounds in trickle-bed. Catal Today 1999; 48: 41-48. http://dx.doi.org/10.1016/S0920-5861(98)00356-3

[22] Cotta RM, Maciel MRW, Filho RM. A cape of HDT industrial reactor for middle distillates. Comput Chem Eng 2000; 24: 1731-1735. http://dx.doi.org/10.1016/S0098-1354(00)00451-8

[23] Froment GF. Modeling in the development of hydrotreatment processes. Catal Today 2004; 98: 43-54. http://dx.doi.org/10.1016/j.cattod.2004.07.052

[24] Murali C, Voolapalli RK, Ravichander N, *et al.* Trickle bed reactor model to simulate the performance of commercial diesel hydrotreating unit. Fuel 2006; doi:10. 1016/j.fuel. 09 019.

[25] Boumahrat M, Gourdin A. Méthodes numériques appliquées. Office des Publications Universitaires, Algérie 1993.

[26] William L, Luyben. Process modelling, simulation and control for chemical engineers. Mc Graw-Hill, International Edition, USA 1990.

[27] Wang Z, Yang B, Chen CJ. Modelling and optimization for the secondary reaction of FCC gasoline based on the fuzzy neural network and genetic algorithm. Chem Eng Process 2007; 46: 175-180. http://dx.doi.org/10.1016/j.cep.2006.05.011

[28] Farouk S, Mjalli S, A-Ashed HE, *et al.* Use of artificial neural network black-box modelling for the prediction of waste water treatment plants performance. Environ Manage 2007; 83: 329-338. http://dx.doi.org/10.1016/j.jenvman.2006.03.004

[29] Reza M, Nasr J, Givi MM. Modeling of crude oil fouling in preheat exchangers of refinery distillation units. Appli Therm Eng 2006; 26: 1572-1527. http://dx.doi.org/10.1016/j.applthermaleng.2005.12.001

[30] Dreyfus G, Martinez JM, Samuelides M, *et al.* Réseaux de neurones: Méthodologie et applications, Editions Eyrolles, Paris 2004.

[31] Corriou JP. Les réseaux de neurones pour la modélisation et la conduite des procédés, Lavoisier Technique et Documentation, Paris 1995.

[32] Nerrand O, Roussel-Ragot P, Personnaz L, Dreyfus G, Marcos S. Neural networks and nonlinear adaptive filtering: unifying concept and new algorithms. Neural Comput 1993; 5: 165-197. http://dx.doi.org/10.1162/neco.1993.5.2.165

[33] Weimin L, Wenkai L, Hui C-W. Integrating neural network models for refinery planning. Comput Aided Chem Eng 2003; 15: 1304-1309. 8th International Symposium on Process Systems Engineering doi: 10.1016/S1570-7946

[34] Baroutian S, Aroua MK, Raman AA, Sulaiman NM. Estimation of vegetable oil-based ethyl esters biodiesel densities using artificial neural networks. J Appl Sci 2008; 8(17): 3005-3011.

[35] Ramadhas AS, Jayray S, Muraleedhanan C Padmakumari KJ. Artificial Neural Networks used for the prediction of cetane number of Biodiesel. J Renewable Energy 2006; 31: 2524-2533. http://dx.doi.org/10.1016/j.renene.2006.01.009

[36] Barron AR. Universal Approximation bounds for superposition of a sigmoidal function. IEEE T Inform Theory 1993; IT-39: 930-945. http://dx.doi.org/10.1109/18.256500

[37] Demuth H, Beale M, Hagan M. Neural Network Toolbox 5: User's Guide, Guide d'utilisation des réseaux de neurones sur le logiciel MATLAB 2007.

Received on 20-08-2014 Accepted on 14-09-2014 Published on 17-11-2014

DOI: http://dx.doi.org/10.15377/2409-787X.2014.01.01.4