Forecast of Polyethylene Properties in the Gas Phase Polymerization Aided by Neural Network

Nasrin Bakhshizadeh, Ashkan Forootan

Abstract—A major problem that affects the quality control of polymer in the industrial polymerization is the lack of suitable on-line measurement tools to evaluate the properties of the polymer such as melt and density indices. Controlling the polymerization in ordinary method is performed manually by taking samples, measuring the quality of polymer in the lab and registry of results. This method is highly time consuming and leads to producing large number of incompatible products. An online application for estimating melt index and density proposed in this study is a neural network based on the input-output data of the polyethylene production plant. Temperature, the level of reactors' bed, the intensity of ethylene mass flow, hydrogen and butene-1, the molar concentration of ethylene, hydrogen and butene-1 are used for the process to establish the neural model. The neural network is taught based on the actual operational data and back-propagation and Levenberg-Marquart techniques. The simulated results indicate that the neural network process model established with three layers (one hidden layer) for forecasting the density and the four layers for the melt index is able to successfully predict those quality properties.

Keywords—Polyethylene, polymerization, density, melt index, neural network.

I. INTRODUCTION

THE melting and density indices are two important control parameters in the process of polyolefin production that determine the quality of the products. Accurate control of these two indices significantly reduces the amount of producing incompatible products. Analysis of density properties and melting index in the first and second gas phase reactor lasts 1 to 4 hours in laboratory. This time latency could cause instability in the quality of the product and difficulty in its control. So far, some researches have been carried out to forecast those two parameters based on other operational parameters [1], [2].

The mechanistic and black box models are among the models that are used with the aim of controlling the process and system. The mechanistic models are extracted from initial scientific principles and by using the physics and chemistry laws that govern the process while Black box models are obtained from operational data and by employing neural networks and describe the relationship between input and output data of the system [1]-[3].

McGregor et al. presented a theoretical model based on the

Nasrin Bakhshizadeh is Polymer plant Process Engineer, Jam Petrochemical Complex, Assaluyeh, Iran (corresponding author, e-mail: n_bakhshizadeh@jpcomplex.com).
Ashkan Forootan is Head of Chemical plants' Process Engineering, Jam Petrochemical Complex, Assaluyeh, Iran (e-mail: a_forootan@jpcomplex.com).

initial regulations that could on-line predict the specifications of melt and density indices of low density polyethylene (LDPE) in the fluid bed reactors (UNIPOL). [4]

Ohschima et al presented an on-line model for predicting the melt index in which, the reaction time was taken much shorter than the time of remaining in the reactor and the structure of each polymer particle stays similar to the other polymers [5]. Their model is a linear formula between reactants' concentration, catalyst's concentration logarithm term and temperature in the following order:

$$log(MI_i) = \beta + \alpha_1 log \frac{[H_2]}{[C_2]} + \alpha_2 log \frac{[C_3]}{[C_2]} + \alpha_3 log \frac{[C_4]}{[C_2]} + \alpha_4 log[R] + \alpha_5 [T] \quad (1)$$

$$\frac{dlog(MI_c(t))}{dt} = \frac{1}{\tau(t)} log(MI_i(t)) - \frac{1}{\tau(t)} log(MI_c(t)) \quad (2)$$

Recently, the neural networks are employed to model the non-linear processes and their control. Among the activities that have taken place on modeling by neural network, one may note the models presented by Thitiyasook et al. [6]

The simulation results showed that the neural network models presented with two hidden layers are fairly able to predict the melt and density indices and therefore, they could be used for predicting those two basic variables. In the present project, an on-line applicable plan has been proposed to estimate the melt and density indices, which is a neural network based on the input-output data of the polyethylene production industrial unit in linear style. Temperature, the level of reactor bed, the intensity of mass flow of ethylene, hydrogen and butene-1, the polyethylene concentration, hydrogen and butene-1 related to the process are used to model the network and the neural network is trained based on actual operational data and the Back-propagation and Levenberg-Marquart techniques.

II. DESCRIPTION OF THE PROCESS

The LLDPE (Linear Low Density Poly Ethyelene) production unit of Jam Petrochemical Company which is based on spiral process uses Zigler-Nata catalysts and gas reactors for producing various LLDPE grades. After preparation and mixing with the co-catalysts and passing through initial stages of the reaction, the catalysts and propylene are injected to pre-polymerization. The pre-polymer, the non-reacted propylene and the propane flow into the first gas phase reactor from the pre-polymerization section. The second gas phase reactor receives the formed polymer particles. The monomer which is fed to the first and second

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

reactors depends on the ratios and the concentrations demanded for each type of grade (with respect to the melt index and its density). The analyzers on each reactor measure the amount of monomer, propane and hydrogen. The polymer powder produced in the reactor will flow in steamer after being separated from the gas in filter. The moist polymer powder is transferred to the dryer by the weight and low pressure difference between the steamer and dryer. The polymer powder with the required mixed additives enters the extruder hopper and then it is injected into the extruder through the hopper. Pellets that have concerned sizes are transferred by gravity to the silos. Then, they are packed in big bags and become ready to be marketed.

III. NEURAL NETWORKS

Artificial neural network is a model for information processing which is a copy of biological neural networks such as human brain. The artificial neural networks transmit the knowledge or hidden law beyond data to the network structure by process on empirical data, an act which is called learning. Essentially, the ability to learn is the most important characteristic of an intelligent system. A system that can learn is more flexible and could be programmed easier; therefore, it can be more responsive to new problems and relations posed to it. As it could be seen in Fig. 1, the network becomes compatible based on conformity and co-assessment between the input and target than the network output and our concerned output (known as "target"). Generally, a large number of those input and output couples are used so the network is taught in this so-called supervised learning process.
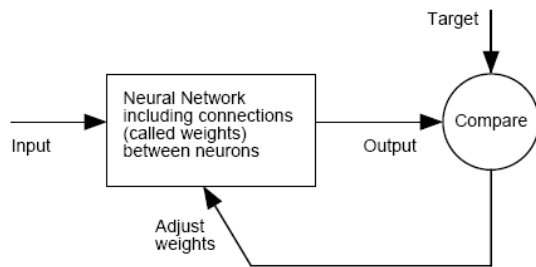

Fig. 1 Neural network

The connections between neurons determine the network function. The main goal is to determine the method of establishing suitable connections for solving specific problems. Fig. 2 (a) shows the model of a neuron with an input. This simple neuron is made of the two key elements: Weight (w) and transfer function (f).

The input P is applied to the neuron and by multiplication in the weight (w), it becomes weighed and applied to the transfer function (f) as an input to acquire the final output. By adding bias to the structure, the biased input is created in form of neuron (Fig. 2 (a)). The bias input is fixed (I) value. The bias value sums up with the w.p. product and in fact, shifts the function leftward. By f, it means the transfer function. This function is usually a step function (I) and/or a Sigmoid function which has received argoman (n) and produces output

(a).

$$a=f(wp+b) \qquad (3)$$

The b and w are two adjustable parameters in the neurons. One or more neurons make a layer of network. A network could consist of one or more such layers. Fig. 3 shows a single layer network with R input and S neuron.
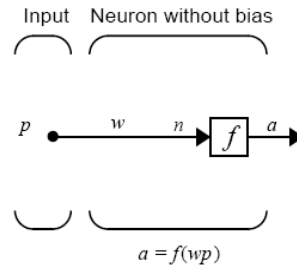

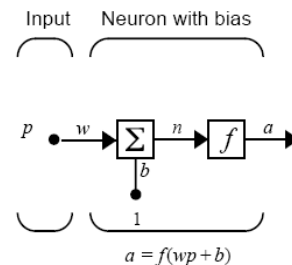Fig. 2 (a) The model of a neuron with an input
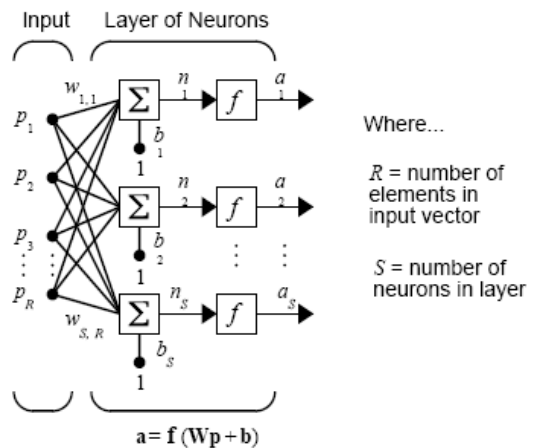

Fig. 2 (b) The model of a neuron with an input

Fig. 3 A single layer network

In this network, the members of input P vector are applied to all neurons and after multiplication in the weights vectors and sum of the bias, they are applied in transfer function and the output is obtained. Two or more neurons could combine together in a form of a layer. A specific network could be made of several layers. Each layer in the network has its own weights matrix, bias vector and its own output. A network could contain different number of neurons in its different layers. The outputs of each mid layer are used as the input of the next layer.

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

The layers of several layered networks have different duties, a layer that creates the network output (the last layer) is known as output layer and other layers are labeled as hidden layers.

The multi-layer networks are very powerful. For example, a two-layer network with a sigmoid first layer and a two linear necessary layer could estimate any given functions with limited numbers of disconnected points. This type of two-layer network is extensively applicable in the post-emission networks.

IV. Procedures of Work

The first step in establishing network is to define the problem. In the supervised networks such as post-emission networks, the question definition is done through defining the target input vectors and target vectors (desirable outputs). In the course of MLP network education by using BP learning algorithm, first, calculations are done from the input of the network to the network output and then, the values of the calculated errors are emitted to the previous layers. At first, the output calculation is made layer to layer and the output of each layer will be the input of the next layer. In post-emission state, first, the output layers are adjusted; as for each one of the neurons of output layer, there is a desirable value and the

weights could be adjusted by them and the up-dating principle. At first data collection of density properties and melt index has been done. It is the main target to predict them for desirable grades. 495 samples of the density measurement results have been taken from the first gas phase rector, 495 samples of density measurement results from the second gas phase reactor, 495 samples of the results from melt index measurement of the first gas phase reactor and 495 samples of the results of melt index measurement in the second gas phase reactor of the mentioned four (4) grades are collected. Those results are related to the times that the plant is in normal operation service; so the results could be attributable. It should be mentioned that the tests have been performed in the central laboratory of Jam Petrochemical Company and by using the test methods listed in Table I.

TABLE I
THE LIST OF TEST METHODS OF MELT INDEX AND DENSITY TESTS

Properties	METHOD
Density	ASTM D1505
Melt index	ASTM D1238

The place for taking samples from the polymer powder of first and second phase gas reactors are shown in Fig. 4.
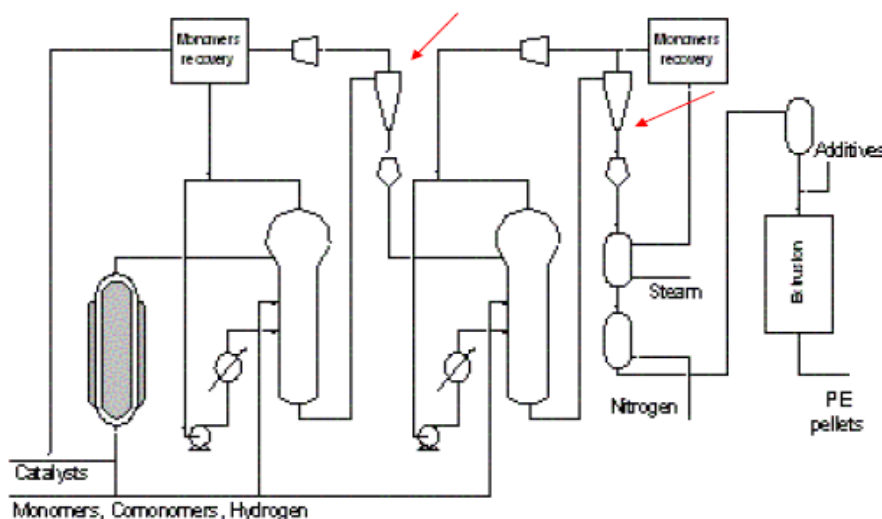

Fig. 4 The sampling place in the gas phase reactors of Jam Petrochemical Company

Selecting the variables of a process with highest relations and effects on the output data is the second step in creating the network. The following parameters are the input data considered in this research for forecasting the density and predicting melt index in the first reactor of gas phase:

1- The mass flow intensity of ethylene feed that is input to the first reactor
2- The mass flow intensity of hydrogen feed that is input to the first reactor
3- The mass flow intensity of butene feed input to the first reactor
4- The ethylene concentration in the first gas phase reactor
5- The hydrogen concentration in the first reactor of gas phase

6- The butene-1 concentration in the first reactor of gas phase
7- The temperature of the first reactor of gas phase
8- The level of the bed of first reactor of gas phase

Input data for predicting the density and melt index related to the second reactor are the entire parameters listed above, as well as, the entire data related to its prior reactor (first gas phase reactor). Then, the educational and testing values of the network should be selected. In the present project, of the 495 sets of existing data, 450 data are considered for education and 45 remaining data are for test.

Ordinarily, the input vector and target is divided at random into three sets of education 60%, evaluation, 20% and trial 20%. The evaluation complex is used in line with maintaining

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

the generality of the network. The education procedure continues to the time when the network error shows reduction in evaluation network; in this way, the over fitting network on education complex is prevented.

In this project, approximately 90% of data is used for education and 10% for evaluation.

The education process needs a series of examples for the behavior which is expected from the network that includes the network input and target. In the process of education, the weights and biases are adjusted to minimize the efficiency functions.

In the present project, the Back-Propagation method which is briefly called BP is used to learn the weights of a multi-layer network and by using gradient descent, it is tried to minimize the square errors between networks outputs and the target function. After emission of a multi-layer network with non-linear transfer function and learning principle is Widro-Hoff.

The neuro networks could be divided into feed forward networks and Recurrent networks (in which, output feedback is used) based on their information process methods. The most customary network which is used for post-emission principle is a multi-layer feed forward network. The multi-layer networks could be used for learning non-linear problems as well as problems with several decision making (processes). For example, for the first reactor, by recalling new function in MATLAB environment, it is possible to create a three-layer network with 8 neurons in the input layer, 16 neurons in the hidden layer and a neuron as the desirable data of the problem; and for the second reactor; too, a three-layer network with 16 neurons in the input layer, 16 neurons in the hidden layers and a neuron as the desirable data of the problem. In first and second layer, the TANSIG function and in the last layer, the Purlin function has been used. The reason is that if the last layer contains the linear neurons, the output could take any values.

The desirable characteristics of the output signal determine the type of output layer transfer function. One must note that in establishing a neural network, finding the suitable size of the network and the optimized limits of the number of the neurons of the hidden layer are the most important part of the work. The size of network is not depended only on the number of the network layer, but also it is related to the number of nodes in each layer and the number of connections between those nodes. One must note that although increase in the number of neurons of the hidden layer of the network- due to putting more effective parameters in access- increases the flexibility of the network in optimization line, if the size of hidden layers is taken too large, the procedure of education encounters problem and while loss of time and space, no suitable results are obtained and perhaps, using large networks for the set of data without high numbers might direct network towards creating unsatisfactory results. It is not possible to determine the number of neurons of hidden layers by paying attention to the parameters of the problem. This is a great problem and an issue in many scientific researches. The final selection of the applied functions, as well as the concerned

network structure and the number of mid-layers along with the number of neurons of each layer is an issue that reaches to answers by trial and error. In fact, with respect to the extension of parameters of artificial neural networks, in order to achieve a desirable prediction, long periods of time must be spent for calibration of those parameters that ultimately, in this project, the best result was obtained through creating three-layer neural network for predicting density and the four-layer network for predicting melt index.

Recently, researches have been carried out in establishing a suitable optimizing tool for determining the optimized combination of parameters effective in the calibration of ANN coefficient and by using them, it becomes possible to determine the hidden layers and the number of effective neurons in each one of the layers to yield the best state in the predicted values of the flow [5].

A point must be considered is that the neural networks are dynamic complexes that have the ability of learning through seeing the educational information and consequently, as the amount of education information which is introduced to the network becomes higher, the network will show better output. One of the cases that somehow cause inaccurate network design is that the design starts designing the network when having little education information. In this case, after viewing the information for 10 to 15 times (10 to 15 epoch), the network reveals a very great performance (the assumption was that the education information was 10 to 15 items). In this condition, the network is over-trained; that is, it cannot make decisions for the inputs rather than the educational data; thus, it is suggested to increase the number of educational data as much as possible.

Regarding the number of layers, one may say that most networks which are used are two and three layers; however, networks with 4 or more layers are also used. In the present research, the number of neurons of the hidden layers is chosen as twice the neurons of first layer (input) For the number of neurons and the layers, one must point out that those values are determined by error and trial; however, there are two ways for determining them:

- Taking the network small, followed by increase in the layers and neurons (construction technique)
- Taking the network large and then, lowering the layers and neurons (pruning).

The first method which is also used in this report has two advantages:

- Increase in calculations is done gradually.
- The optimized size of the network is obtained by this.

Prior to the education of a feed forward network, first, the initial values of weights and biases must be determined. The NEWFF function automatically gives value to the weights; however, if we plan to do so, we might use the init command. In addition, it should be noted that in the present research the Levenberg-Marquart algorithm is used for education. The efficiency index in the multi-layer networks is the mean square error. If the probability of occurrence of each one of the target vectors is considered, the average mean square will be compatible with sum of errors squares in the educational

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

complex.

Usually, BP algorithm, before its end, is repeated by using the same education data for thousands of times. Various conditions could be used to end the algorithm.

- Stop after repetition in certain frequencies
- Stop when the error becomes less than a certain amount. Of course, it could lead to over fitting.
- Stop when the error in the examples of the confirmation complex follows a certain rule.

If the recurrence frequencies are little, we will have error and if they are too many, the over-fitting will occur. After the education of network, it could be used for simulation of new data. In this line, the SIM function is used. In the present project, due to the possibility of making comparison between the simulated results and the results obtained from lab

analysis, the test data are used for simulation as concerned data.

V. RESULTS

With respect to the configuration of the neural network defined in this project, in accordance with Table II, after defining the problem, specifying the input and output data and ultimately, network education, the results are as follows:

TABLE II
CONFIGURATION OF NEURAL NETWORK FOR PREDICTING POLYMER PROPERTIES

Predicted polymer quality variables	Obtained NN configuration
Density of first and second reactor	Model #1=8-16-1 NN
MFR of first and second reactor	Model #2=8-16-16-1 NN

A. The Predicted Densities for the First and Second Reactors
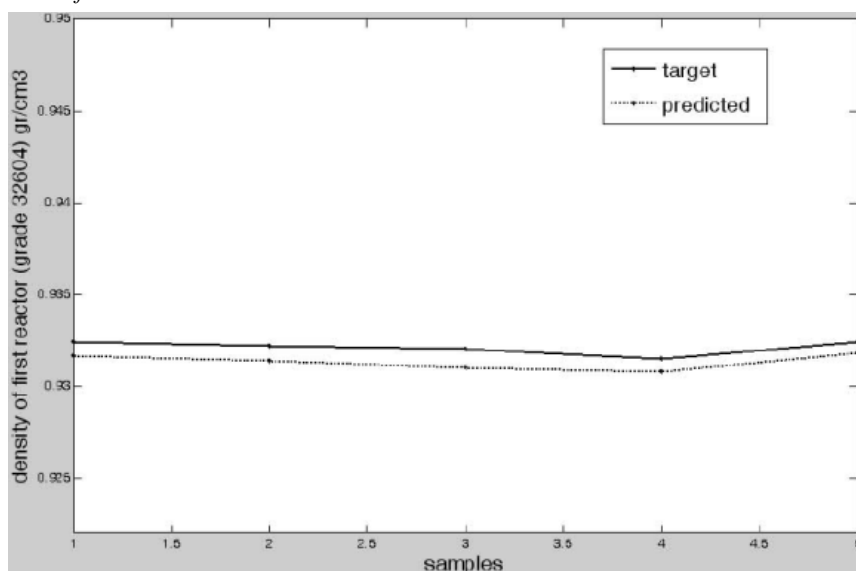
Fig. 1 (a) Prediction of density in the first reactor for grade (32604)

Fig. 1 (b) Prediction of density in the first reactor for grade (60507)

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019
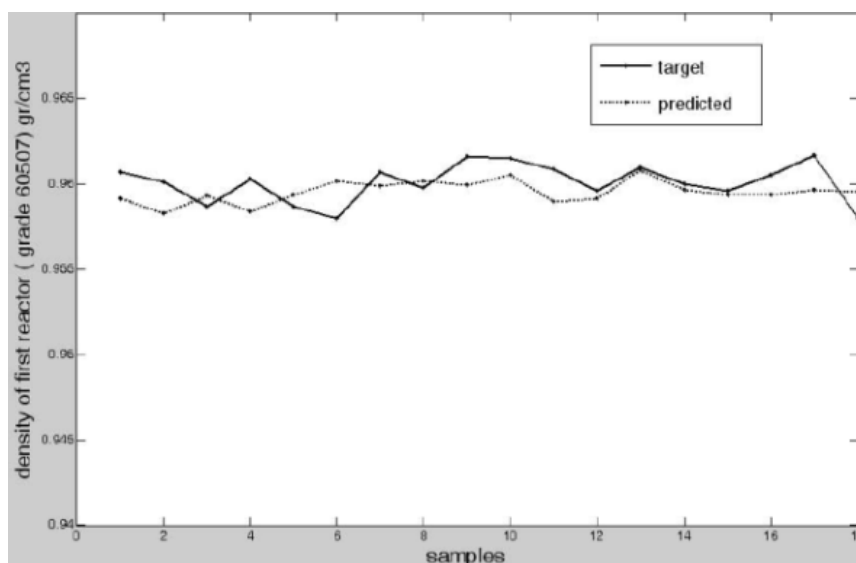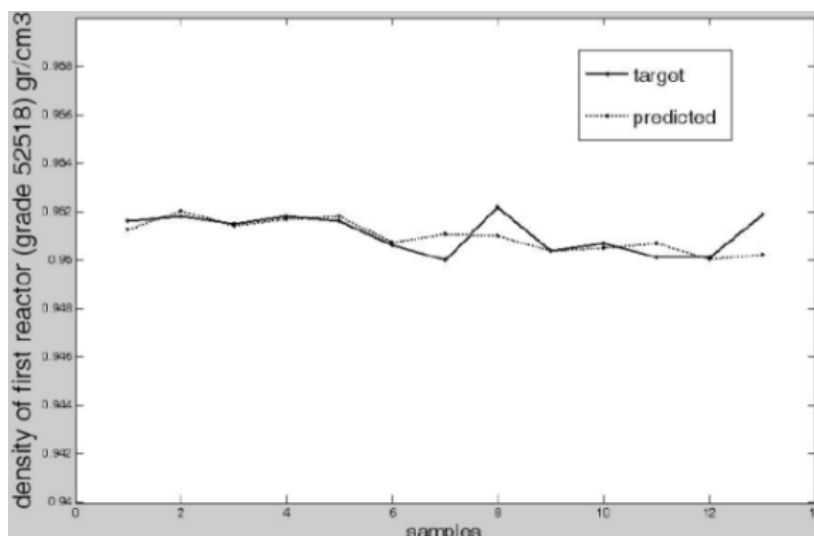
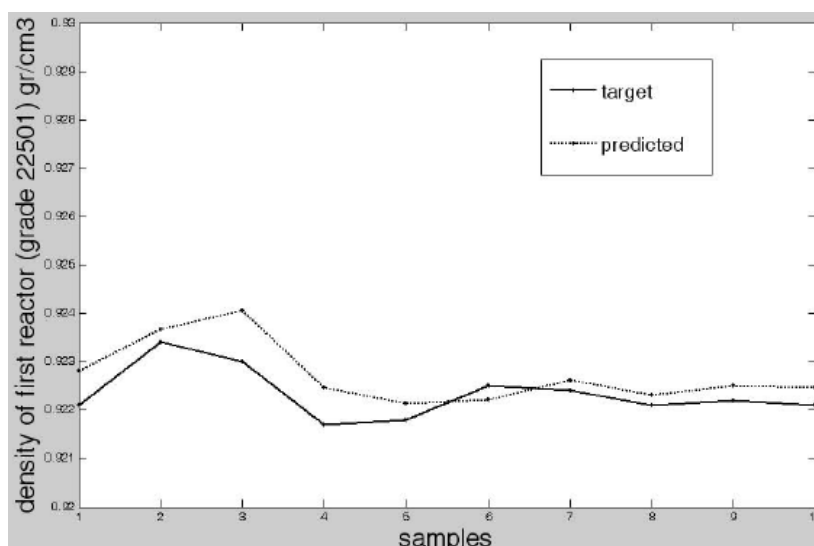Fig. 2 (a) Prediction of density in the first reactor for grade (52518)

Fig. 2 (b) Prediction of density in the first reactor for grade (22501)
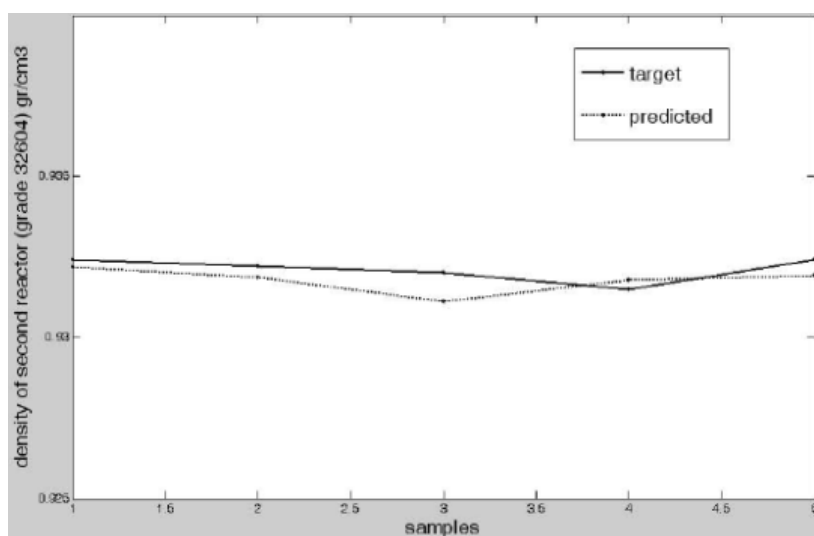

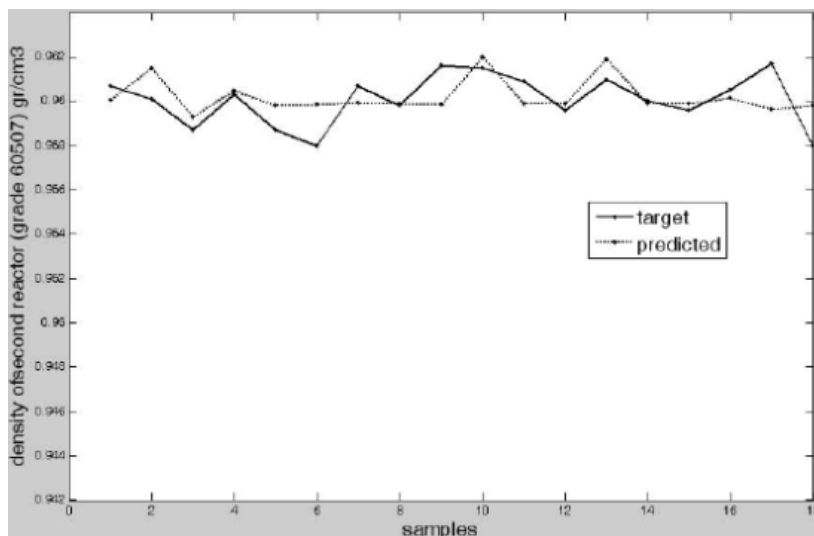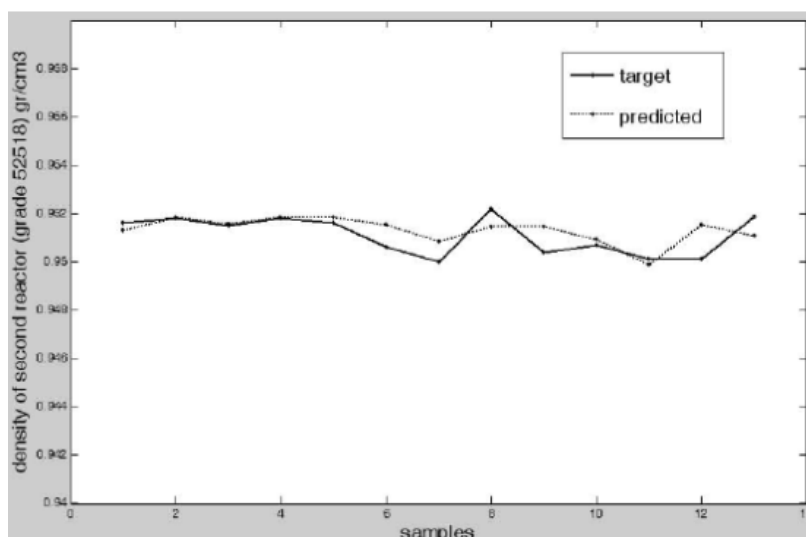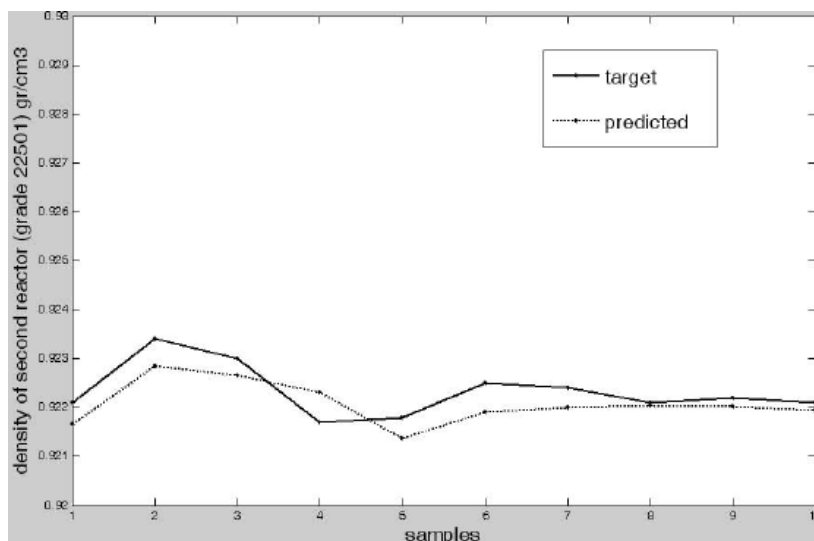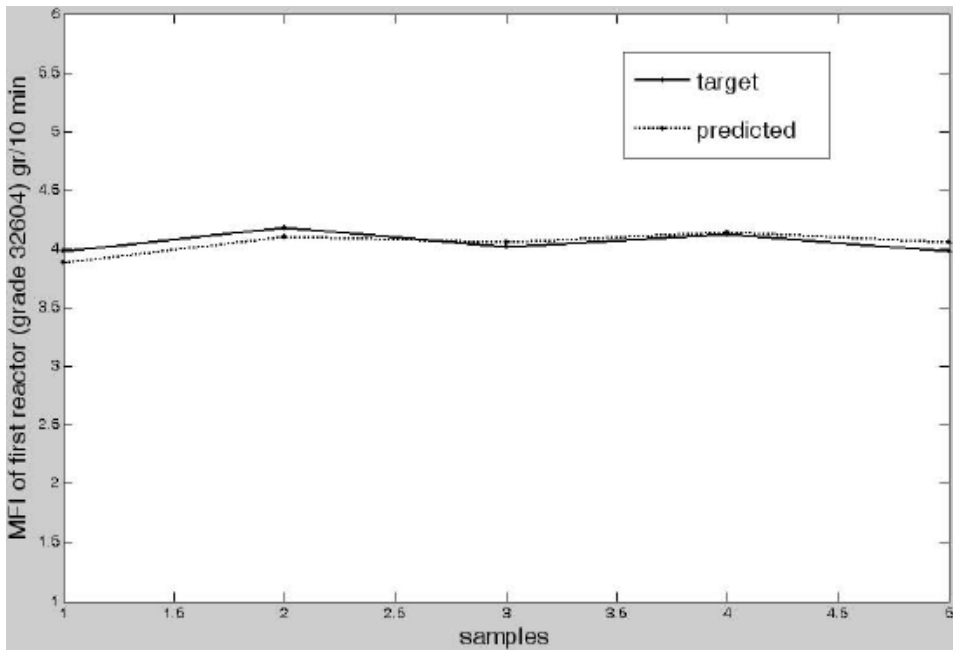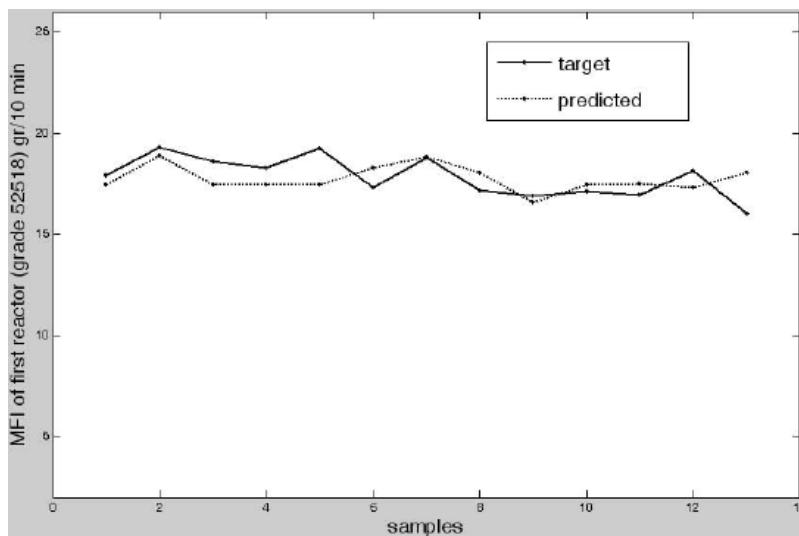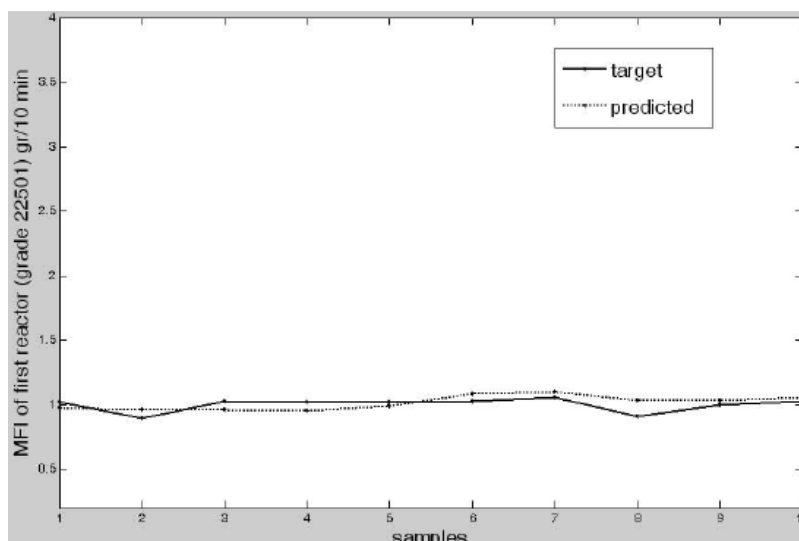Fig. 3 (a) Prediction of density in the second reactor for grade (32604)

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

Fig. 3 (b) Prediction of density in the second reactor for grade (60507)

Fig. 4 (a) Prediction of density in the second reactor for grade (52518)

Fig. 4 (b) Prediction of density in the second reactor for grade (22501)

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

B. The Predicted Melt Index for Reactors First and Second

Fig. 5 (a) Prediction of melt index in the first reactor for grade (32604)

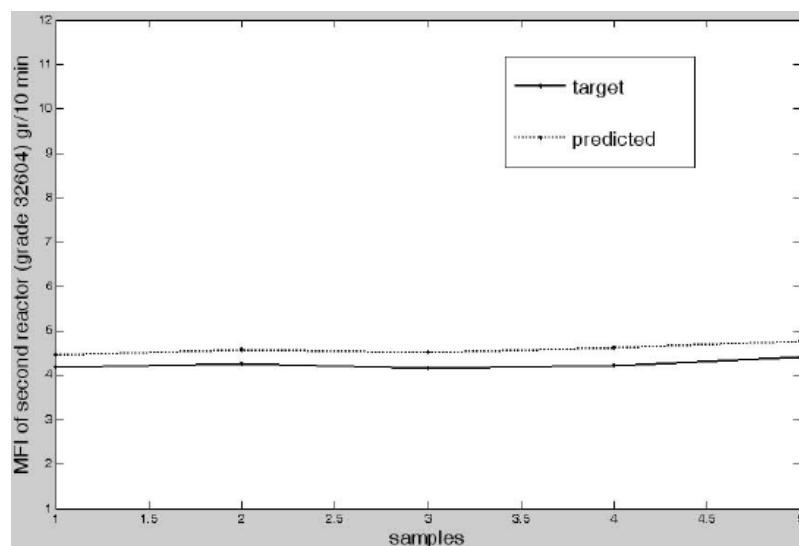Fig. 5 (b) Prediction of melt index in the first reactor for grade (60507)

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019
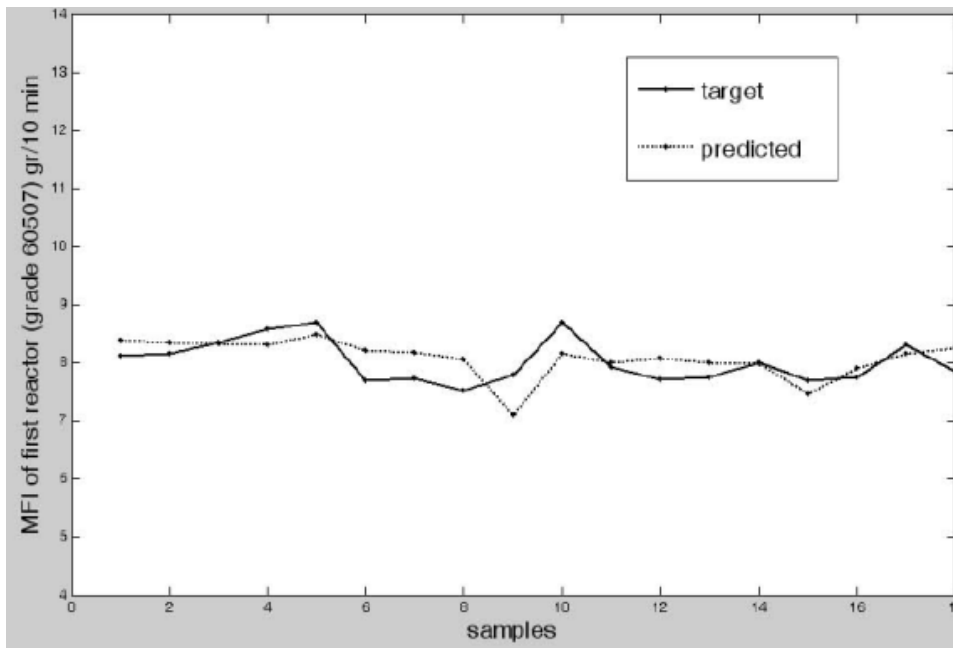
Fig. 6 (a) Prediction of melt index in the first reactor for grade (52518)

Fig. 6 (b) Prediction of melt index in the first reactor for grade (22501)

Fig. 7 (a) Prediction of melt index in the second reactor for grade (32604)
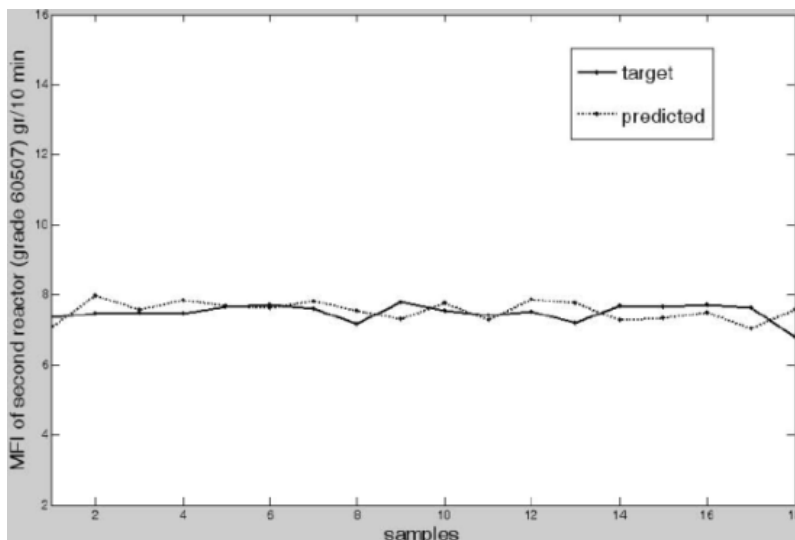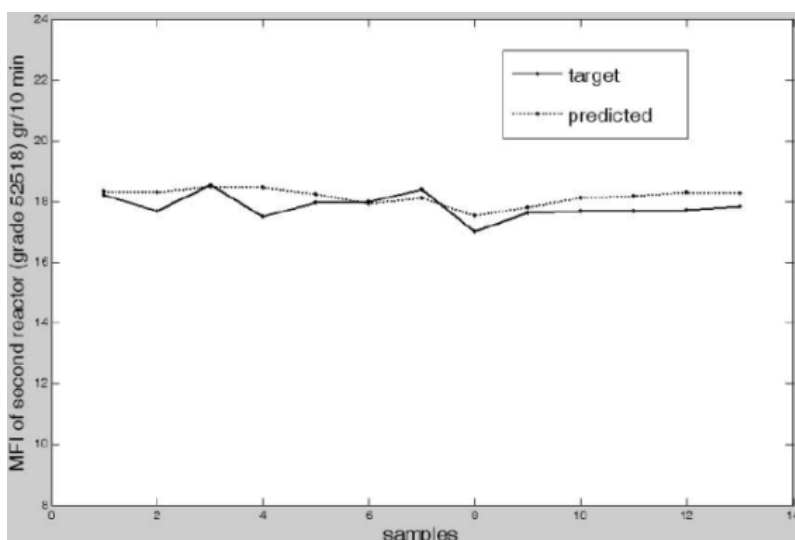
World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

Fig. 7 (b) Prediction of melt index in the second reactor for grade (60507)

Fig. 8 (a) Prediction of melt index in the second reactor for grade (52518)
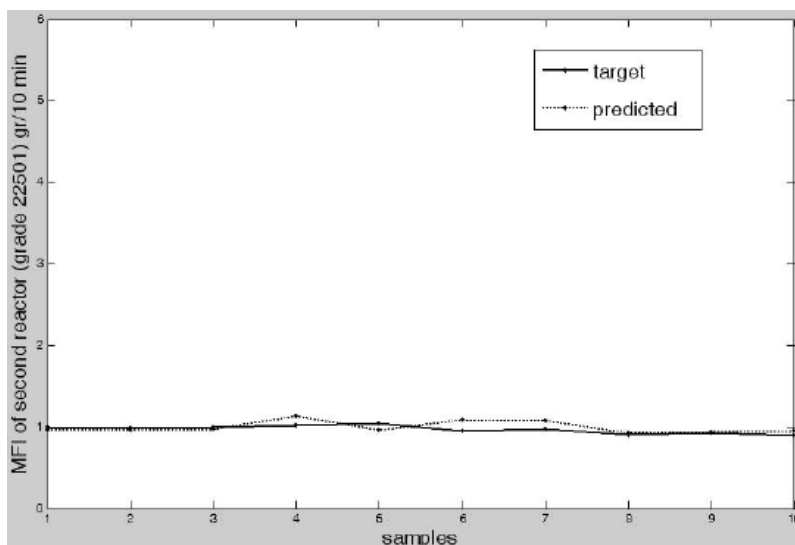

Fig. 8 (b) Prediction of melt index in the second reactor for grade (22501)

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:13, No:8, 2019

The scheme of the education process of the neural network and the learning curve in MATLAB environment is shown in Figs. 9 and 10.

As it could be seen, the simulation results indicate that the created neural network model could fairly well predict the melt index and density. To predict the melt index, the network needed a more complicated structure than the density and as it has been mentioned before, to forecast the density, a three-layer with one hidden layer, and for predicting the melt index, the four-layer network with two hidden layers are used.

In this research, the results of analysis of four (4) grades with melt index range of 0.85 to 20 gr/10 min and density in 0.920 to 0.962 gr/cm^2 are used for network education. As a result, this network could be used for predicting the properties of grades which the melt index and density place in this range; too. However, expansion of neural network to other operational scope could not be guaranteed. In another word, in a range in which the lab data are present, a function which is obtained from neural network has a profile similar to other models while beyond that scope, each model shows different profiles. Ohshima has noted this in his paper by showing Figs. 11-15 [7]-[9].
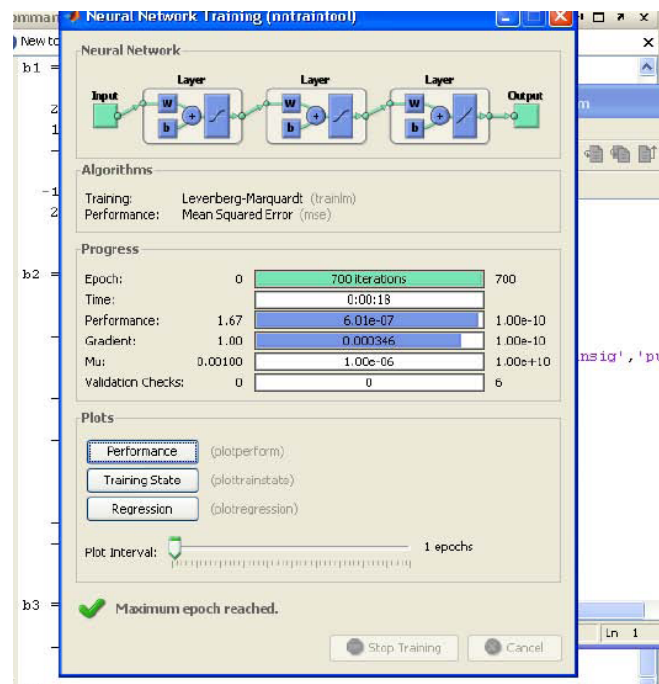

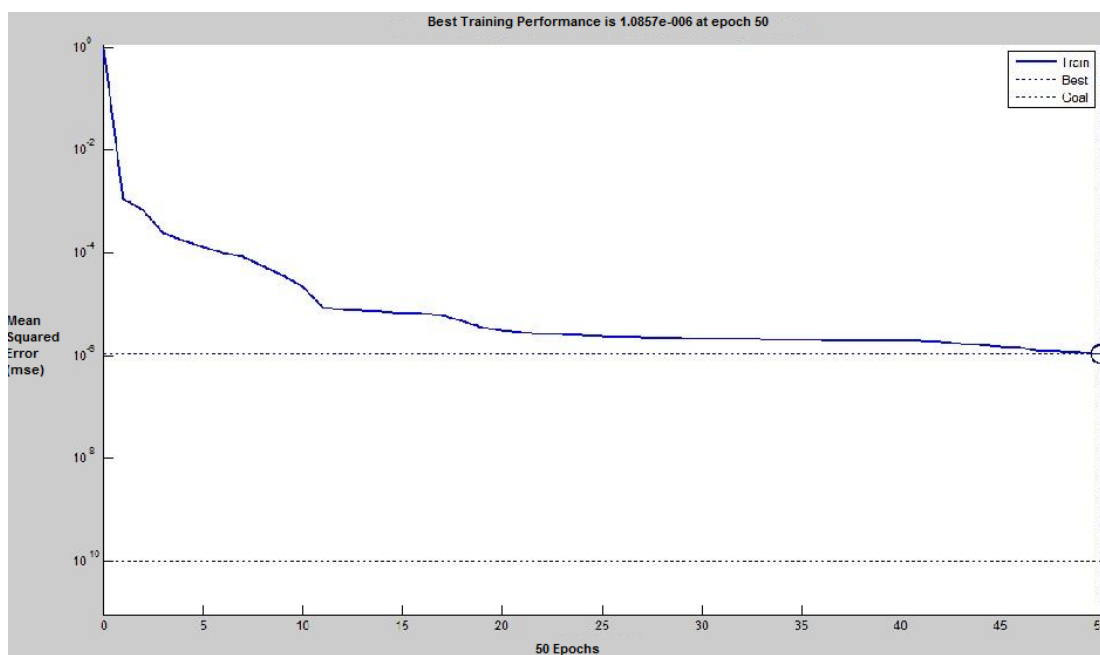Fig 9 Education process of neural network in MATLAB Environment

Fig. 10 The learning curve of network in MATLAB Environment

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
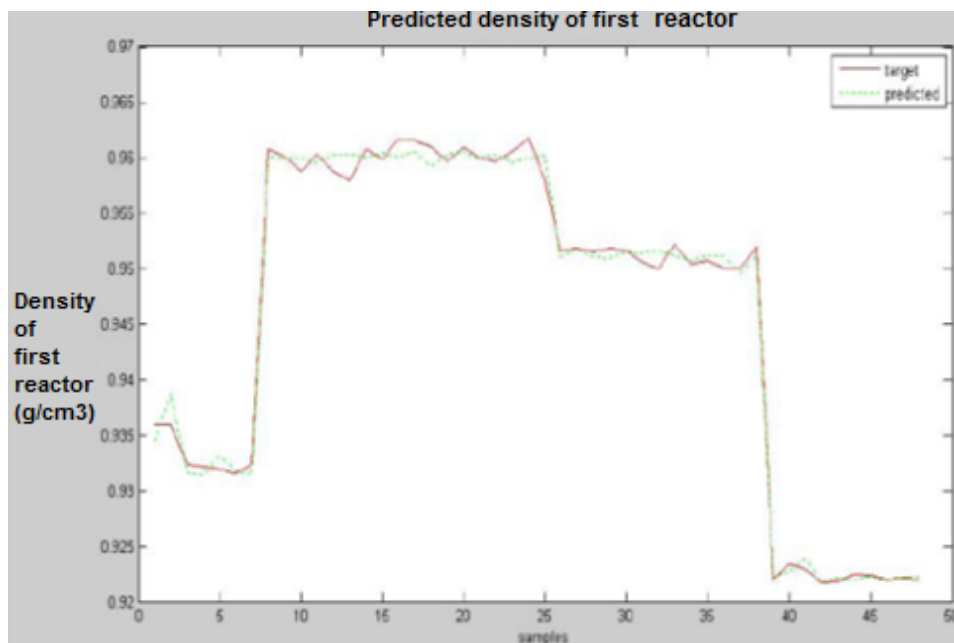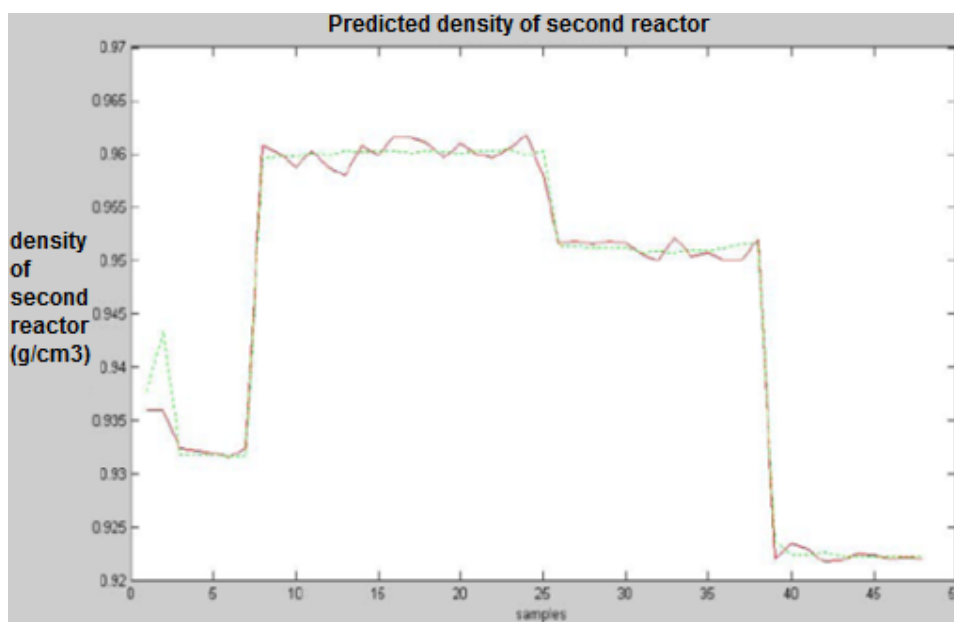Vol:13, No:8, 2019

Fig. 11 Prediction of density in the first reactor for grade 326040 and grade 60507, 22501 and 52518

Fig. 12 Prediction of density in the second reactor for grade 32604 and grade 60507, 52518 and 22501

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
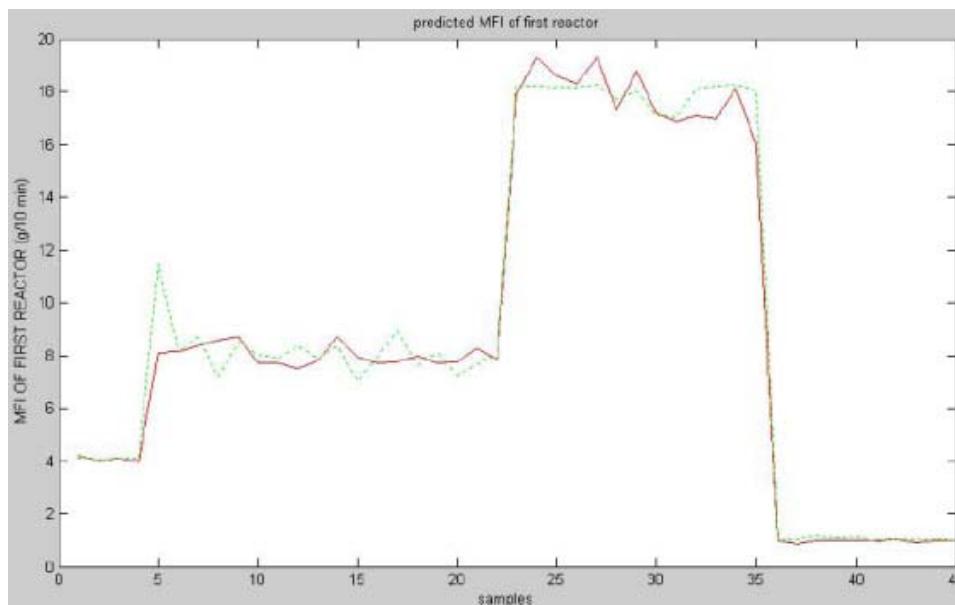Vol:13, No:8, 2019

Fig. 13 Prediction of melt index in the first reactor for grade 32604 and grade 60507, 52518 and 22501
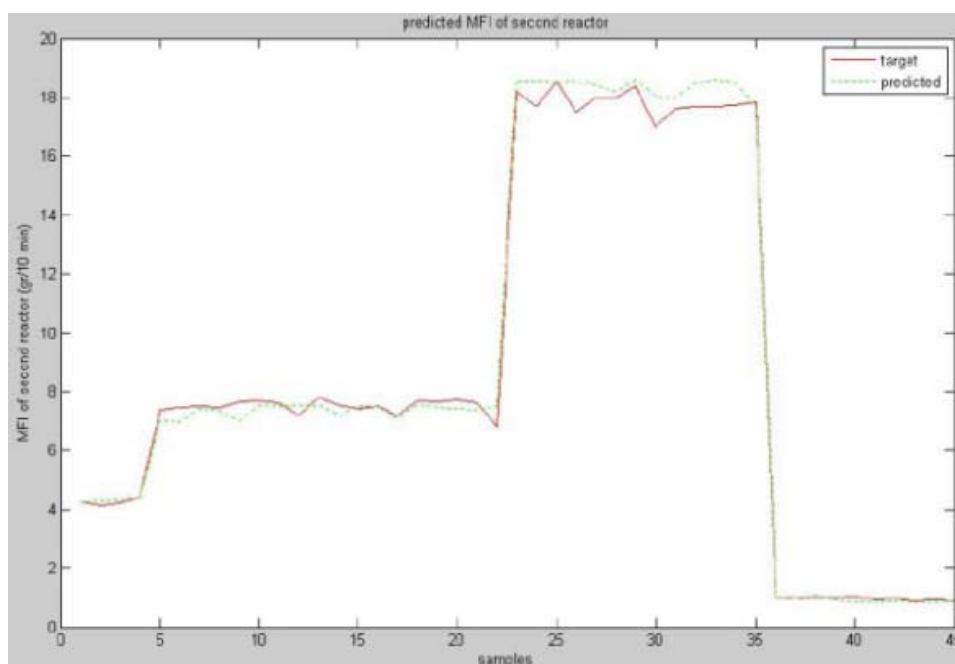

Fig. 14 Prediction of melt index in the second reactor for grade 32604 and grade 60507, 52518 and 22501
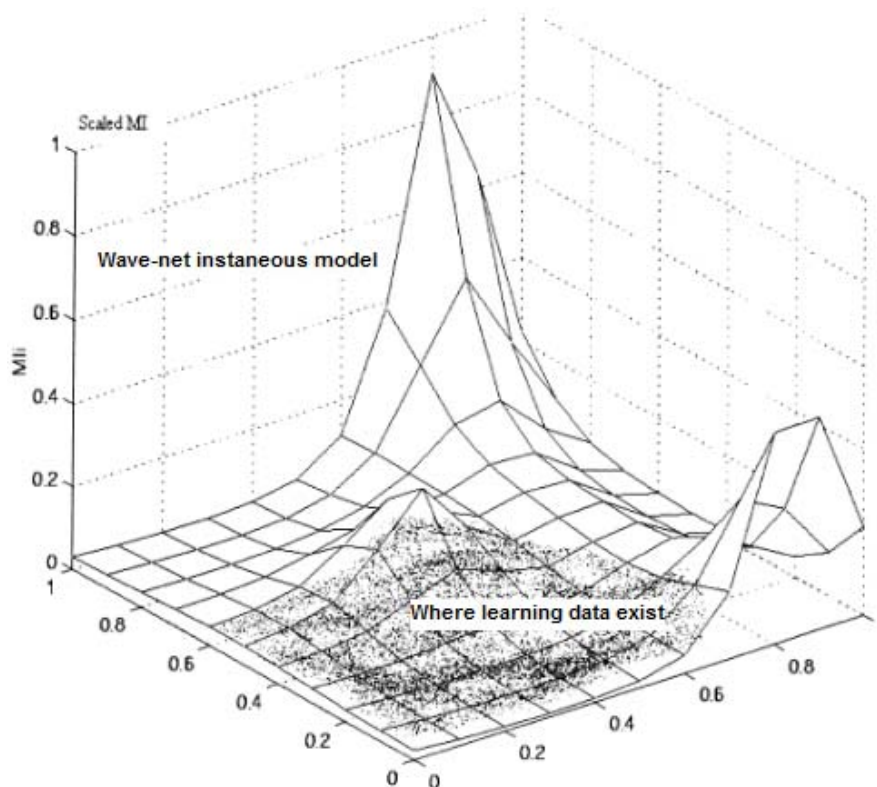
Fig. 15 Using neural network beyond its education scope

REFERENCES

[1] Abedi, S. Vahdadi Asl, Fall 2007, publisher: Petrochemical Research & Technology Co. 1st Ed. Polyolefin.

[2] F. Rodriguez, "principles of polymer systems", Taylor & Francis, 1996, p2.

[3] Resin plastics (LDPE,HDPE,PP,PS,PVC,ABS,PMMA). Chem. Rep.2000, 120, 37 .

[4] K.B. MCAuley, J.F. MacGregor, "on-line inference of polymer properties in an industrial polyethylene reactor", ALCHE J. 37 (6) (1991) 825-835

[5] M. Ogawa, M. Ohshima, K. Morinaga and F. Watanabe; "Quality inferential control of an industrial High Density Polyethylene Process," J. Process Control, 9, 51-59 (1999)

[6] P. Thitiyasook, P. Kittisupakorn, S. Niamsuwan, K. Konakom; "on-line neural network estimator of polymerization plant".17th European Symposium on Computer Aided Process Engineering – ESCAPE17 (2007)

[7] M. Ohshima, M.Tanigaki, "Quality control of polymer production processes", J. of process control, 2000, pp.135-145

[8] M.Karthikeyan, J. Chem. Inf. Model. , 45, 581 (2005)

[9] A. Afantitis, G Melagraki, K. Makridima, A. Alexandridis , H.Sarimveis and O. Iglessi- Markopoulou, J. Molecular Structure, 716 , 193(2005)