Petroleum and Coal

Article **Open Access**

Modeling of Zeoforming Process Variables Using GMDH Neural Network on Pilot Scale Experiments

Saeed Soltanali, Reza Seif Mohaddecy, Maryam Mashayekhi, Mehdi Rashidzadeh

Catalysis Technologies Development Division, Research Institute of Petroleum Industry (RIPI), Tehran, Iran

Received June 28, 2020; Accepted October 15, 2020

Abstract

In this paper, the group method of data handling (GMDH) networks are applied for modeling the momentous process variables of a bench scale zeoforming. The proposed model can predict the product research octane number (RON), benzene content in the product, liquid recovery (%), coke deposition (%) on the catalyst and sulfur removal conversion (%) by using a grand polynomial correlation which is a function of weight hourly space velocity (WHSV), reactor inlet temperature and reactor pressure. To do such a task, twelve experiments were performed in the bench scale pilot for 1,400 hours. Then, modeling were done by GMDH software.

The results showed that this model can precisely estimate the process variables and product properties. Moreover, it is confirmed that the proposed model is capable of predicting of the product research octane number (RON), benzene content in product, liquid recovery (%), coke deposition(%) on the catalyst and sulfur removal conversion (%) with the average absolute deviation (AAD%) of 0.29% , 0.33%, 0.41%, 0.28% and 3.2% respectively. Moreover, the root means square error (RMSE %) of the mentioned parameters are 0.34%, 0.41%, 0.64%, 0.41% and 3.4%, respectively.

Keywords: *Modeling; GMDH; Neural network; Zeoforming; RON; Liquid recovery; Sulfur removal conversion.*

1. Introduction

Like typical catalytic reforming, the Zeoforming process converts low-octane hydrocarbons into high-octane gasoline components. *n*-Alkanes are transformed mainly into aromatic hydrocarbons via recombination of olefins formed as a intermediate phase whereas the conversion of *iso*-alkanes and naphthenes takes place on a limited scale [1-2]. In this way the formation of aromatic rings is possible from n-pentane and other light n-alkanes as well as from n-hexane or higher. The Zeoforming process runs with an endothermic heat effect in the presence of catalyst consisting of hydrogen form of zeolite ZSM-5 and a binder Al_2O_3. Contrary to typical reforming [3-8], where the dehydrogenation of naphtenes and dehydrocyclization of n-paraffins are the dominating reactions, practically no hydrogen is produced and light hydrocarbons are main by-products only. The Zeoforming process was first developed in the Institute of Catalysis Zeosit in Novosybirsk, Russia and it was investigated on a pilot plant in 1987–1992 [1-2].

On the other hand, developing a black box model, which is exclusively obtained from experimental data, can provide other practical methods in the field of process modeling. These models provide a dynamic relationship between input and output variables and bypass underlying complexity inside the system. Most of these common approaches rely on linear system identification models. The major processes found in chemical engineering are unfortunately nonlinear processes, and previously mentioned approaches fail to respond regarding process nonlinearity. As an alternative to fundamental models, artificial neural networks (ANNs) are a valuable estimate tool, and up to now, numerous applications of ANN models in the engineering area have been reported [9]. ANN can perform better than regression models, and is tolerant to noise in data [10-13]. The increased importance of ANNs arises from their possibility to parallel process of data despite their components are independent of each other [14]. On the

other hand, straightforward theories do not offer adequate precision for estimation of experimental data.

However, ANN's structure contains a massive complicated of equations within its nodes and layers. Furthermore, the arrangement of network is chosen manually or randomly which does not assure the best possible network. As a better alternative, the group method of data handling (GMDH) provides a self-organizing neural network to express the genome of system as well as using the most suitable configuration by means of minimization process. In the other word, the GMDH utilizes feed-forward network whose coefficients are determined using regression together with imitation of self-organizing activity [15]. The algorithm chooses the most suitable polynomial expressions built by combination of two independent variables at a time.

Some artificial neural network models have been developed in the literatures to predict and control parameters in industrial processes such as catalytic reforming unit [16-17]. But based on our literature review, there is no study on using GMDH to model the Zeoforming process. Therefore, the present study is devoted to model the RON of the product, benzene content in product, liquid recovery (%), coke deposition(%) on the catalyst and sulfur removal conversion using GMDH for a pilot scale test of Zeoforming process. To validate the proposed model, several tests were performed with different operating conditions in the Zeoforming pilot plant (about 1440 hrs).

2. Materials and methods

2.1 Process description of the industrial scale Zeoforming unit

A block flow diagram of the industrial scale zeoforming unit is presented in Figure 1.

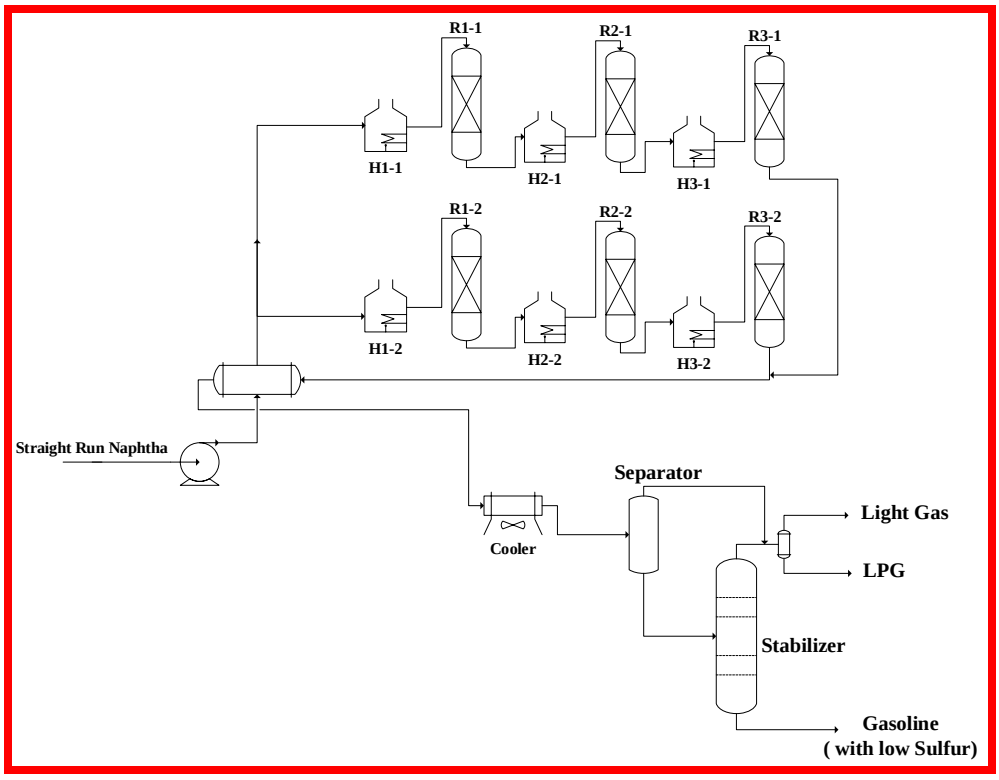

Figure 1. Block flow diagram of the industrial scale Zeoforming unit

The Zeoforming plant has been producing unleaded gasoline component during the last few years. In the conventional Zeoforming plant there are two lines of reactors work alternatively. Both lines consist of three reactors. Each line of reactors works during 7-10 days and then the catalyst is regenerated. The total time of the catalyst work is about one and a half year.

2.2. Experiments in the pilot scale system

The experiments were carried out in a pilot test system, which is licensed by the Research Institute of Petroleum Industry (RIPI). This device can tolerate temperatures and pressures up to 500°C and 70 bar, respectively. The simplified diagram of pilot scale is presented in Figure 2. As can be seen, the temperature along the reactor bed is controlled by use of three thermocouples (TIC 1-3).

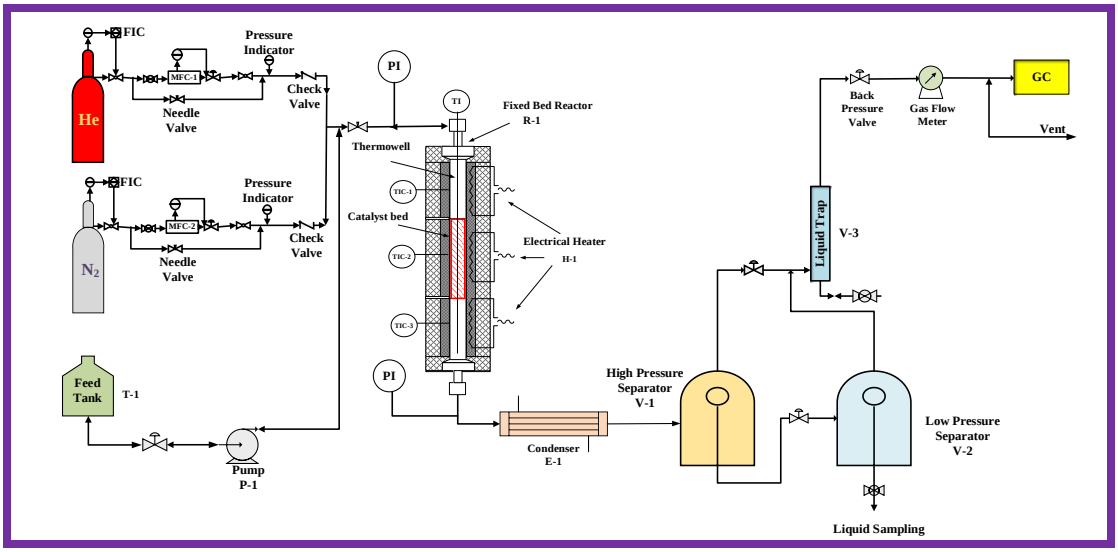

Fig. 2. Schematic diagram of the experimental Zeoforming unit

As can be seen, the feedstock is pumped from the reservoir (T-1) using a piston pump (P-1) to the top of the reactor. Helium and nitrogen gas for purging are sent from their cylinders to the top of the reactor, the flow rate being controlled by mass flow controllers (MFC-1). The unit consists of a stainless steel reactor (internal diameter of 2.2 cm and length of 51 cm), kept at isothermal condition using a three-zone electric furnace. In the reactor inlet, the temperature of inlet feedstock increases to the reaction temperature using the preheating zone of the reactor (zone-1). Reactions are carried out at the following conditions: Temperature of 300-440°C; Pressure of 5-20 bar and WHSV of 1-4. As mentioned, through the bed, there are three thermocouples to control the bed temperature. At the reactor outlet, the reaction effluent is cooled using a water-cooled heat exchanger (E-1). Then, the vapor and liquid phases are separated in a high and low pressure separators (V-1, V-2). After reducing the pressure, the liquid is allowed to flow from the separator to the other flash drum, maintained at atmospheric pressure. The liquid sample is discharged discontinuously into sampling bottles. Finally, the output of gas flow rate was metered by gas-meter and composition of the this stream is determined using an online gas chromatograph (Agilent 8790-A).

The operation conditions for the catalytic activity testing in the pilot and products specifications are shown in Tables 1 and 2.

Table 1. Operation conditions of pilot testing

No.	WHSV h^{-1}	T Units °C	P bar	No.	WHSV h^{-1}	T Units °C	P bar
1	1.22	358	5	7	3.61	361	15
2	1.02	400	10	8	3.5	402	5
3	1.2	440	15	9	3.52	440	10
4	2.35	360	10	10	4.02	314	20
5	2.33	400	15	11	3.9	340	17.7
6	2.31	434	5	12	3.01	355	12.5

Table 2. Pilot test products specification

No.	Liquid recovery	Octane number	Coke Deposition	S Conversion	Benzene content
	Units				
	vol /vol	-	%wt	%	%wt
1	55	83.3	1.5	93.3	3.4
2	56	82.7	21.0	39.5	3.9
3	39	67.9	38.0	16.7	8.1
4	71	82.5	9.9	66.7	2.3
5	70	80.4	22.5	39.5	2.0
6	52	85.7	35.0	23.3	4.5
7	82	80.8	2.1	64.3	1.4
8	79	81.9	12.2	69.5	2.0
9	46	84.1	31.8	7.1	4.7
10	84	78.9	8.2	95.7	1.0
11	93	80.7	6.0	48.2	0.8
12	80	80.6	4.2	71.7	2.5

3. GMDH modeling approach for the Zeoforming pilot scale experiments

The basic structure of the brain has been widely employed for various fields such as modeling, control, and pattern recognition. The GMDH, introduced by Ivakhnenko [18], is a hierarchical and learning network structure that provides an effective approach to identify higher order non-linear systems. Its main purpose is the identification of relations in large complex non-linear multidimensional systems as well as their approximation and prediction. In the GMDH network, the part which corresponds to the neuron of a neural network is called the "N-Adaline", and is generally expressed by a polynomial. The N-Adaline is composed of two inputs and one output, and the latter is generated by combinations of two inputs [19]. Inputs, x_i and x_j, are then combined to produce a partial descriptor based on the simple quadratic transfer function as the following:

$$\hat{y}_n = a_0 + a_1 x_{i_n} + a_2 x_{j_n} + a_3 x_{i_n} x_{j_n} + a_4 x_{i_n}^2 + a_5 x_{j_n}^2 \quad (1)$$

where $\hat{y}_n$ is determined using the least squares method, and coefficients i.e. a_0 to a_5 are determined statistically, and are unique for each transfer function. These coefficients can be thought as analogous to weights found in other types of neural networks. The GMDH topology is usually determined using a layer by layer pruning process based on a pre-selected criterion of what constitutes the best nodes at each level. The traditional GMDH method is based on an underlying assumption that data can be modeled by using an approximation of the Volterra series or Kolmorgorov-Gabor polynomial as follows:

$$y = a_0 + \sum_{i=1}^{M} a_i x_i + \sum_{i=1}^{M}\sum_{j=1}^{M} a_{ij} x_i x_j + \sum_{i=1}^{M}\sum_{j=1}^{M}\sum_{k=1}^{M} a_{ijk} x_i x_j x_k \ldots \quad (2)$$

where X (x_1, x_2 ,…., x_M) is the vector of input variables, and A (a_1 , a_2 ,…., a_M) is the vector of summand coefficients [19].

During constructing GMDH, all combinations of inputs are generated, and sent into the first layer of the network. Outputs from this layer are then classified and selected as input for the next layer with all combinations of the selected outputs, sent into the layer 2. This process is continued as long as each subsequent layer (n+1) produces a better result than layer (n). When layer (n+1) is found to not be as good as layer (n), the process will be stopped. Now, each layer consists of nodes that a pair of inputs is its source.

In GMDH topology, each node produces a set of coefficients (a_i & $i \in \{1,2,3,..,5\}$) that are estimated by using training data. Then, the fitness is tested by evaluating the mean square error of the modeled ($\hat{y}$) and actual (pilot data) (y) values as follows:

$$Error = \sum_{n=1}^{N} (\hat{y}_n - y_n)^2 \quad (3)$$

To identify the coefficients with the best fit, the partial derivatives of Eq.(3) are calculated with respect to each constant value a_i, and set it equal to zero as follows:

$$\frac{\partial Error}{\partial a_i} = 0 \quad (4)$$

Finally, to compare the modeled and actual values, average absolute deviations (AAD%) and root mean squared error (RMSE) are calculated as follows:

$$ADD\% = \frac{\sum_{i=1}^{N_t=50}\sum\left|\left(\frac{Y_i^{Actual}-Y_i^{modeled}}{Y_i^{Actual}}\right)\right|}{N_t} \quad (5)$$

$$RMSE\% = \sqrt{\frac{\sum_{i=1}^{N_t=50}\sum\left(\frac{Y_i^{Actual}-Y_i^{modeled}}{Y_i^{Actual^2}}\right)}{N_t}} \quad (6)$$

where N_t, Y_i^{actual}, $Y_i^{modeled}$ are the number of test runs, actual variables, and the modeled values.

4. Results and discussion

To build up the GMDH model for the Zeoforming pilot plant, as previous mentioned fourteen test runs with specific operating condition about 1400 hours have been done. These points were included of product research octane number (RON), benzene content in product, liquid recovery (%), coke deposition (%) on the catalyst, sulfur removal conversion (%), temperatures of reactor, Weight hourly space velocity (WHSV), reactor pressure. The corresponding polynomial equations of the proposed models for the growth period are presented in Table 3 to 7. Figures 3 to 7 show the comparison between the measured output variables (pilot data) and the modeled ones using the GMDH network. From these figures, a reasonable agreement can be observed.

Table 3. Nodal expressions for GMDH neural network of product RON

RON = 49.025 + A^2*B*5.9819e-05

A =-1412.42+T*6.63879–T*D^2*0.0011457-T^2*0.0155909+T^2*D*0.000207261+ ^2*0.25228

B = 70.918 - T*4.01667 + T*C*0.0978574 - T*C^2*0.000591001

C = 88.868 - T^2*P*4.82775e-06

D =199.971-WHSV*181.677 + WHSV *T*0.476092 + WHSV ^2*30.7452 - WHSV ^2*T*0.0806093 - T^2*0.000805396

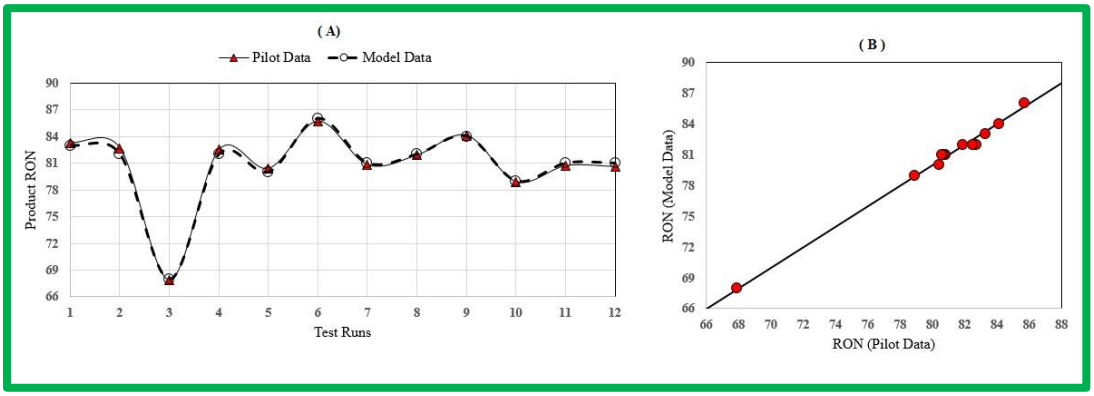

Figure 3. Comparison plots: A) pilot product RON vs. modeled and B) pilot RON product vs. modeled

Table 4. Nodal expressions for GMDH neural network of benzene content in product

Benzene content= 2.44239 + A*1.4666 - A*B^2*0.0383337 + A^2*B*0.00157376 - B*2.64507 + B^2*0.535629

A = 15.2436 - WHSV*3.56789 + WHSV ^2*C*0.248011 - C*4.63411 + C^2*0.555185

B = 4.36356 - P*D*0.175582 + P*D^2*0.0362233

C = -7.74906 + WHSV*3.7904 - WHSV*T*0.0119091 + T^2*8.51803e-05

D= 4.56679 - WHSV *P*0.121769 + P^2*0.015789

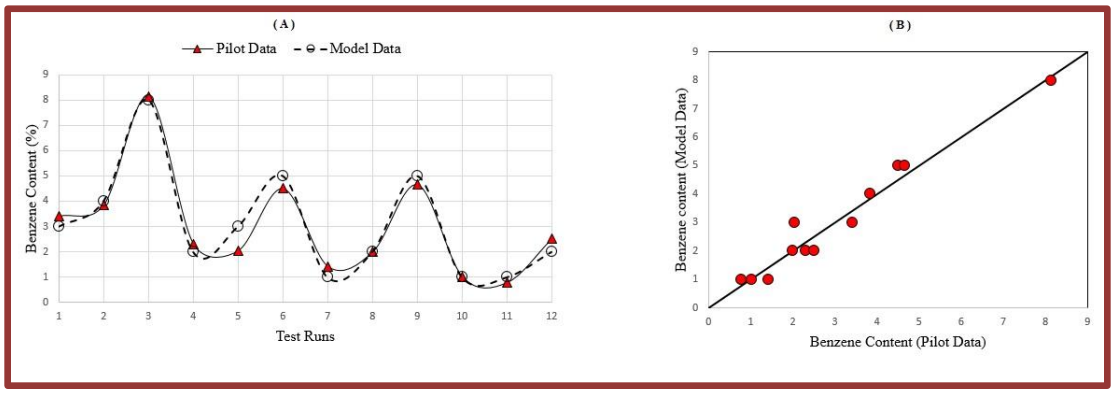

Figure 4. Comparison plots: A) Pilot benzene content in product vs. model data and B) Pilot benzene content in product vs. modeled

Table 5. Nodal expressions for GMDH neural network of coke deposition % on the catalyst

Coke Deposition = -3.32288e-13 + A*1
A= -1.66955 -WHSV *B^2*0.00282549 + B*1.27244
B = 360.994 - T*2.10261 + T ^2*0.00310333

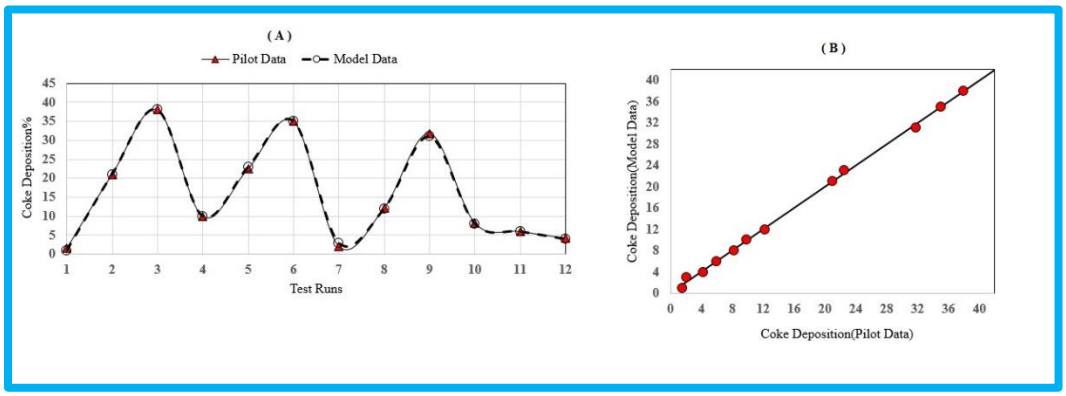

Figure 5. Comparison plots: A) Pilot coke deposition content on the catalyst vs. model data and B) Pilot coke deposition content on the catalyst vs. modeled.

Table 6. Nodal expressions for GMDH neural network of coke deposition % on the catalyst

Liquid recovery %=-76.1252+B*A^2*0.000764728-B^2*A*0.000422058+A*4.90851- A^2*0.0645144
A = -26.7678 + WHSV *B*0.159235 - WHSV ^2*1.68632 + B*1.94443 - B^2*0.011157
B = -843.718 + WHSV*35.3068 - WHSV*T ^2*0.000166972 + T*4.47515 - T^2*0.00561868

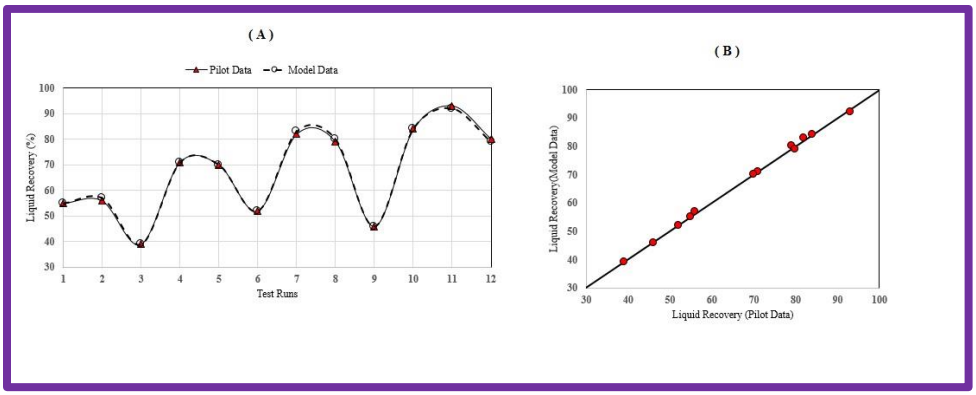

Figure 6. Comparison plots: A) Pilot liquid recovery vs. model data and B) Pilot liquid recovery vs. modeled

Table 7. Nodal expressions for GMDH neural network of removal sulfur conversion %

S Conversion = -414.602 - B*A*0.375169 + B*A^2*0.00137611 + A*26.6726
A = 17.1374 + B^2*0.0104304
B = 328.735 - T *0.679671 - T*P^2*0.000254402

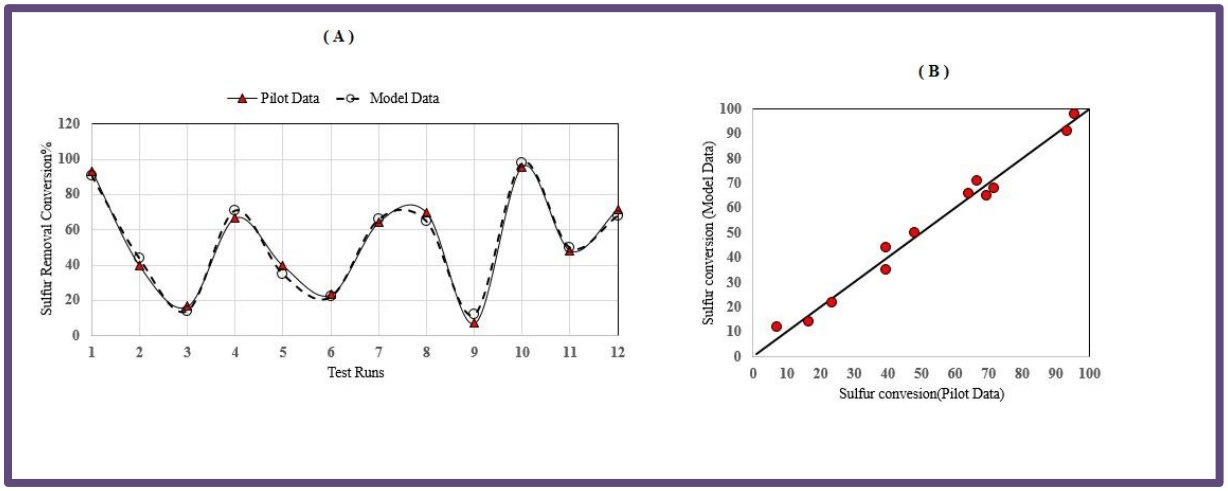

Figure 7. Comparison plots: A) Pilot vs. model data and B) sulfur conversion Pilot vs. modeled

The results show that this model can predict well the RON, benzene content in product, coke deposition, liquid recovery, sulfur conversion with the AAD % of 0.29%, 0.33%, 0.28%, 0.41 and 3.2 %, respectively. Moreover, the RMSE % of the mentioned variables are 0.34%, 0.41%, 0.41%, 0.64% and 3.4%, respectively.

It is supposed that the main deviation can be related to some factors including power fluctuation of instruments, calibration of analysis devices, human errors, and signal transmission that cannot be excluded from the collected data. However, from the presented results, it can be concluded that the proposed approach is reliable enough to be utilized for predicting the behavior of the heavy naphtha catalytic reforming unit.

5. Conclusion

In this work, a group method of data handling (GMDH) modeling approach was applied to predict and model significant output variables of a zeoforming unit on bench scale experiments. These process output variables were RON, benzene content in product, coke deposition, liquid recovery and sulfur conversion. Then, by using the proposed model, the influence of operating conditions variables were studied.

Twelve test runs were used to construct and train the GMDH network for the target zeoforming plant. It was concluded that a GMDH network with three neurons in the intermediate layer was satisfying to simulate output variables of zeoforming i.e. RON, benzene content in product, coke deposition, liquid recovery and sulfur conversion with small value of the AAD% and RSME%. Consequently, the GMDH can be a reliable and accurate tool to model zeoforming plant for sensitivity analysis, optimization and troubleshooting purposes without tackling the complexity of fundamental methods.

References

[1] Stepanov VG, lone KG, and Snytnikova GP. Zeolite catalysts in the upgrading of low-octane hydrocarbon feedstocks to unleaded gasolines. Catalysts in Petroleum Refining and Petrochemical Industries, 1995; 477-482.

[2] Kornblit L, Marchut EA, Jęczmionek L, Bozek S, Lenartowicz L, Mikrut G. Petroleum & coal, 2004; 46(3): 63-68.

[3] Anunziata OA, and Pierella LB. Selective conversion of light gasoline into aromatic hydrocarbons over shape selective zeolites, I. Catalytic activity of various zeolites for aromatization of light gasoline, heavy gasoline and coker naphtha. React. Kinet. Catal. Lett., 1993; 49(2): 311-317.

[4] Samborskaya MA, Mashina VV, Cherednichenko OA, Makarovskikh AV. Modeling of reactor of straight–run gasoline fractions refining on zeolite catalysts. Procedia Chemistry, 2015; 15: 237-244.
[5] Hodala JL, Halgeri AB, Shanbhag GV, Reddy RS. Aromatization of C5-rich Light Naphtha Feedstock over Tailored Zeolite Catalysts: Comparison with Model Compounds (n-C5-n-C7). Chemistry Select, 2016; 1: 2515-2521.
[6] Ostrovskii NM, Rovenskaja SA, and Echevski GV. Aromatization of Light Naphtha Fractions on Zeolites. 2. Model of Catalyst Deactivation. Chem Ind.,2004; 58(3): 104-108.
[7] Rovenskaja SA, Ostrovskii NM. Aromatization of Light Naphtha Fractions on Zeolites. 1. Kinetic Model. Chem Ind., 2003; 57(9): 399-403.
[8] Stepanov VG, and Ione KG. Petrol Production from Virgin Fraction of oil and Gas Condensates with the use of Zeoforming Process. Chemistry for Sustainable Development, 2005; 13: 805-818.
[9] Golmohammdi H, Rashidi A, Safdari SJ. Prediction of ferric iron precipitation in bioleaching process using partial least squares and artificial neural network. Chem. Ind. Chem. Eng. Q., 2013; 19: 321-331.
[10] Elkamel A, Al-Ajmi A, Fahim M. Modeling the Hydrocracking Process Using Artificial Neural Networks. Pet. Sci. Technol., 1999; 17: 931-954.
[11] Eslamloueyan R, Setoodeh P. Optimization of Fed-Batch Recombinant Yeast Fermentation for Ethanol Production Using A Reduced Dynamic Flux Balance Model Based on Artificial Neural Networks. Chem. Eng. Commun.,2011; 198(11): 1309-1338.
[12] Zahedi G, Fgaier H, Jahanmiri A, Al-Enezi G. Artificial Neural Network Identification and Evaluation of Hydrotreater Plant. Pet. Sci. Technol. 2006; 24(12): 1447–1456.
[13] Joseph B, Wang FH, Shieh PS. Exploratory data analysis: A comparison of statistical methods with artificial neural networks. Comput. Chem. Eng., 1992; 16(4), 413-423.
[14] Milic P, Rajkovic KM, Milicevic PM, Milic SM, Brdaric TP, Pavelkic VM. Comparison, Artificial Neural Network Modeling and Genetic Algorithm Optimization of The Resinoid and Potassium Yields from White Lady's Bedstraw (*Galium mollugo* L.) By Conventional, Reflux and Ultrasound-Assisted Aqueous-Ethanolic Extraction. Chem. Ind. Chem. Eng. Q. 2013; 19(1), 141-152.
[15] Atashrouz S, Amini E, Pazuki G. Ionics, Modeling of surface tension for ionic liquids using group method of data handling. 2015; 21(6): 1595-1609.
[16] Manamalli D, Kanagasabapathy P, Dhivya K. Expert Optimal Control of Catalytic Reformer Using ANN. Chem. Eng. Commun., 2006; 193:729-742.
[17] Sadighi S, Mohaddecy SR. Int. J. Technol., Predictive modeling for an industrial naphtha reforming plant using artificial neural network with recurrent layers. 2013; 2: 1-11.
[18] Ivakhnenko AG. Polynomial Theory of Complex Systems. IEEE Transactions on Systems, Man, and Cybernetics, 1971; 364-378.
[19] Fujii K, Yamamoto T. Development of a Nonlinear Soft Sensor Using a GMDH Network for a Refinery. Crude Distillation TowerElectr. Eng. Jpn.,2014; 188(6): 31-38.

To whom correspondence should be addressed: Reza Seif Mohaddecy, Catalysis Technologies Development Division, Research Institute of Petroleum Industry (RIPI), P.O. Box 14665-137, Tehran, Iran,
E-mail: *seifsr@ripi.ir*