Regular Article

A Neuro-Fuzzy Algorithm for Modeling of Fischer-Tropsch Synthesis over a Bimetallic Co/Ni/Al$_2$O$_3$ Catalyst

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An alumina supported Co/Ni catalyst was prepared by sol-gel procedure to study the catalytic behavior during Fischer-Tropsch synthesis in a fixed-bed reactor. The effect of CO conversion (10-50%) on hydrocarbon product distribution (CH$_4$, C$_{5+}$ and C$_2$-C$_4$ olefin selectivities) was studied. Selectivity for CH$_4$ decreased, while those of C$_{5+}$ and olefin selectivities increased with increasing CO conversion. The catalysts properties were characterized at different stages using powder X-Ray Diffraction (XRD), Brunauer-Emmett-Teller (BET) surface area measurements, and Scanning electron microscopy (SEM). A neuro-fuzzy model called locally liner model tree (LoLiMoT) was applied to predict the catalytic behavior during Fischer-Tropsch reaction over the Co/Ni/Al$_2$O$_3$ catalyst. The predicting system was established on CO conversion values as a target based on three variables, including partial pressure of CO and H$_2$, and H$_2$/CO feed ratios as the input. To evaluate the generalization performance of the system, the k-fold cross validation was applied so that an excellent prediction was observed with mean square error (*MSE*) which equals 7.4211e-004. Finally, the extrapolation ability of LoLiMoT was perused (beyond the training range). The obtained data from LoLiMoT were compared with the experimental data, and the results indicated that LoLiMoT is a worthy system modeling with high capability for data prediction, both within and beyond the training range.

Keywords: locally linear model tree, Cross-validation, Extrapolation, CO conversion, Operational condition, Training range

INTRODUCTION

Regarding the growth in the rate of energy consumption, there is a worry about encountering the shortage of energy sources in the future. Then, one of the most challenging problems of modern world is to find a suitable substitute for oil sources. In this field, Fischer-Tropsch (F-T) synthesis is an effective pathway to produce the fuel using coal and natural gas [1]. The critical determinants of product distribution are the process variables such as temperature, pressure, gas flow rate, and H$_2$/CO feed ratio [2]. Unfortunately, despite the F-T synthesis benefits in fuel producing, the process is very costly. Therefore, seeking to find advantages methods to decline experimental cost will improve the economic strategies. Combination of industry with computer sciences could be a beneficial way to reduce

time and finance cost due to experiments [3]. In order to get useful information from the experimental data and convert this to valuable knowledge, an effective data mining technique such as artificial neural networks (ANN), or neuro-fuzzy (NF) model should be employed. Several successful applications of neural network modeling have been reported for the data sets obtained by conventional or high throughput experimentations for various catalytic systems [4-9]. One of the most significant discussions on predicting the result for real reactions is data prediction beyond training range (extrapolation). Unfortunately, because of the standardization of inputs and outputs that is required to run ANN, a problem arises in extrapolation: if the training data set does not contain the maximum possible output value, an unmodified network will be unable to synthesis this peak value. Therefore, the confidence in the ANN model can be greatly enhanced if some methodology could be found for prediction beyond the training range of

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the calibration and verification procedure in addition to the available measured data sets. The present study focuses on developing a model for data prediction in F-T process beyond the training range.

The k-fold cross-validation is one of the most common performance estimation methods [10] that the system is trained multiple times in order to measure the performance of each parameter combination. In a k-fold cross-validation, the training data are randomly split into k non-overlapping segments (the folds) of approximately equal sizes: k-1 samples are used as training sets and the remaining sample is used as validation set [11].

In this research, a $Co/Ni/Al_2O_3$ catalyst was prepared by sol-gel procedure for synthesis of light olefins in F-T synthesis in a micro fixed-bed reactor. Afterward, an algorithm was developed based on the so-called NF model of LoLiMoT to predict of the catalyst activity in F-T synthesis over $Co/Ni/Al_2O_3$ catalyst. The LoLiMoT method was investigated as a predictor that can predict data in the range of the experimented conditions (*i.e.* interpolation). Then, the LoLiMoT extrapolation ability (data prediction beyond training range) was studied for data prediction during F-T process.

EXPERIMENT AND METHODS

Catalyst Preparation

An $80\%Co/20\%Ni/15\%Al_2O_3$ catalyst was prepared by conventional sol-gel method. Aluminum triethylate $Al(OC_2H_5)_3$ (1.23 g) dissolved in 2 ml ethanol with vigorously stirring at 60 °C. $Co(NO_3)_2.6H_2O$ (17.31 g) and $Ni(NO_3)_2.6H_2O$ (4.29 g) were dissolved in 5 ml ethanol and added into the solution with vigorously stirring at 60 °C. After stirring for 30 min at 60 °C, 2 ml nitric acid 65% was added drop wise to the mixture. Finally, the solution was hydrolyzed by adding a H_2O/C_2H_5OH (15/10 ml/ml) mixture. The solution was stirred to gel over approximately 3 h. Then, the sample was stirred at 120 °C for 16 h to give transparent monolithic gel. The catalyst was calcined at 600 °C for 6 h.

The Methodology of Catalyst Testing

A schematic representation of the experiment setup is shown in Fig. 1. The experiments were carried out in a fixed bed tubular stainless steel micro reactor. In every experiment one gram of catalyst is put in the reactor. Before starting every experiment, the catalyst is reduced by the use
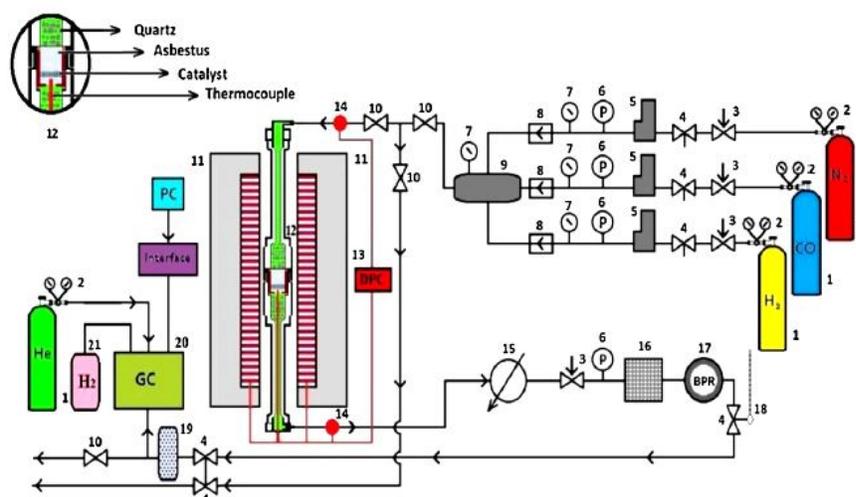
Fig. 1. Schematic representation of the reactor in a flow diagram used. 1-Gas cylinders, 2 Pressure regulators, 3-Needle valves, 4-Valves, 5-Mass FlowControllers (MFC), 6-Digital pressure controllers, 7-Pressure gauges, 8-Non return valves, 9-Ball valves, 10-Tubular furnace, 11-Temperature indicators, 12-Tubular reactor and catalyst bed, 13-Condenser, 14-Trap, 15-Air pump, 16-Silica gel column, 17-Gas Chromatograph (GC), 18-Mixing chamber, 19-BPR: Back Pressure Regulator (Electronically type), 20-CP (Control panel).

of N_2 and H_2 gases. After reduction is done, the temperature of the oven is set to the experiment and when the temperature reaches the desired point for the experiment the CO valve is opened and the flow ratio of feed gases is set by the control panel. After 8 h, the gas chromatograph (GC) is turned on and the gas products are measured to obtain the percent of CO conversion. The catalysts were extremely fine particles so intraparticle diffusion could be neglected. The gas hourly space velocity (GHSV) increased to the value in which the CO conversion was almost the same for a variety of catalyst weight indicating that external diffusion can be neglected above this GHSV. Hence, the kinetic experiments were conducted free from internal and external mass transfer limitations.

Locally Linear Model Tree (LoLiMoT)

The local linear neuro-fuzzy (LLNF) network is depicted in Fig. 2. It is based on a dived-and-conquer strategy. LoLiMoT algorithm provides a simple, fast, and deterministic model which has low number of trial-and-error steps for system identification. It has been introduced as a local linear model (LLM) algorithm [12,13] and can be regarded as a radial basis function that the output layer weights are replaced with a linear function of the network inputs (Fig. 2). Each neuron realizes an LLM and an associated validity function, and determines the region of validity of the LLM [12]. It is an incremental tree-construction algorithm that partitions the input space by axis-orthogonal splits. In iteration, a new rule or LLM is added to the model. Thus, LoLiMoT belongs to the class of incremental or growing algorithms [14]. Also, there is the validity loop (upper levels) during iteration that determines the parameters for nonlinear partitioning of the input space (structure) and the inner loop (lower levels) that estimates the parameters of those LLMs [15]. The validity functions, which are similar to basis functions in RBF and could be Gaussians, are normalized such that for any input u, $\sum_{i=1}^{m} \phi_i(\underline{u}) = 1$, and the output of this model is computed as:

$$\hat{y} = \sum_{i=1}^{m} (w_{i0} + w_{i1}u_1 + w_{i2}u_2 + w_{i3}u_3 + \ldots + w_{ip}u_p)\phi_i(\underline{u})$$

(1)

where the LLMs and the validity functions depend on $\underline{u} = [u_1 \quad u_2 \quad \ldots \quad u_p]^T$ as input of the model and p is number of the dimension of the model input.
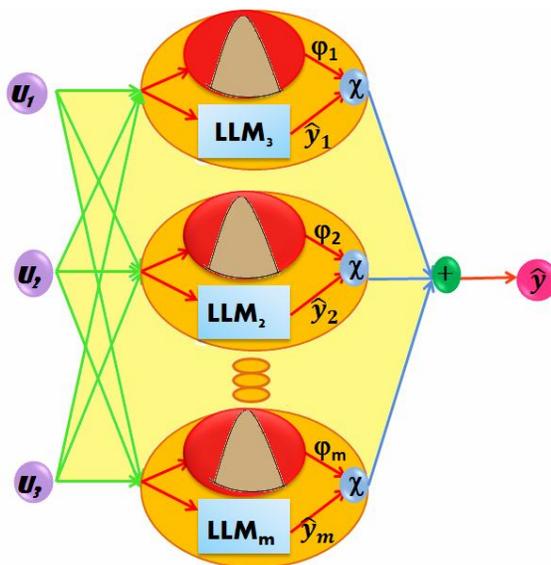
Fig. 2. local linear Neuro-fuzzy model architectedfor Co-Ni/Al$_2$O$_3$ catalyst.

This network simply interpolates linear hyper-planes, which are used to approximate the functions locally, by nonlinear neurons called validity function. A choice for validity function is normalized Gaussians. If these Gaussians are furthermore axis-orthogonal the validity functions are

$$\phi_i(\underline{u}) = \frac{\mu_i(\underline{u})}{\sum_{j=1}^{m} \mu_j(\underline{u})} \tag{2}$$

$$\mu_i(\underline{u}) = \exp\left(\frac{(u_1 - c_{i1})^2}{-2\sigma_{i1}^2}\right) \times \ldots \times \exp\left(\frac{(u_p - c_{ip})^2}{-2\sigma_{ip}^2}\right) \tag{3}$$

The coordinates of the center c_{ij} and standard deviations σ_{ij} as the hidden layer parameters of the ANN are nonlinear network parameters and each weight w_{ij} as the *j-th* local weight of the linear system i is a linear parameter. Each Gaussian function in Eq. (4) performs as a membership function with input vector $\underline{u}$ for the locally linear model. The global parameter vector contains *m(p+1)* elements:

$$w = [w_{10} \quad w_{11} \quad \ldots \quad w_{1p} \quad w_{20} \quad w_{11} \quad \ldots \quad w_{m0} \quad w_{mp}] \tag{4}$$

And the associated regression matrix *X* for *N* measured data samples are:

$$X = [x_1 \quad x_2 \quad \ldots \quad x_m] \tag{5}$$

$$x_i = \begin{bmatrix} \phi_i(\underline{u}(1)) & u_1(1)\phi_i(\underline{u}(1)) & \ldots & u_p(1)\phi_i(\underline{u}(1)) \\ \phi_i(\underline{u}(2)) & u_1(2)\phi_i(\underline{u}(2)) & \ldots & u_p(2)\phi_i(\underline{u}(2)) \\ \vdots & \vdots & \vdots & \vdots \\ \phi_i(\underline{u}(N)) & u_1(N)\phi_i(\underline{u}(N)) & \ldots & u_p(N)\phi_i(\underline{u}(N)) \end{bmatrix} \tag{6}$$

Therefore:

$$\hat{y} = X\hat{w} \quad , \quad \hat{w} = (X^T X)^{-1} X^T y \tag{7}$$

The input space is decomposed to axis orthogonal style yielding hyper-rectangles which centers of Gaussian membership functions $\mu_i(u)$ are placed. The standard division of these Gaussians is set to 0.157 of the length of their rectangles in each dimension.

$$\sigma_{ij} = k_\sigma \, \Delta_{ij} \tag{8}$$

$$k_\sigma = 0.157 \tag{9}$$

where Δ_{ij} denotes the extension of the hyper rectangle of local model i in dimension u_j. k_σ has been found by trial and error method that 0.157 has the best accuracy from other values. The LoLiMoT algorithm is classified as follow: (1) start with an initial model: start with a single neuron, which is a global linear model over the whole input space with $\phi_1(u) = 1$ and set $m = 1$. If there is a priori input space partitioning, it could be used as the initial structure; (2) find the worst LLMs: calculate a local loss function *e.g.* sum square error (SSE) for each of the $i = 1,...,m$ LLMs, and find the worst performing neuron; (3) check all divisions: the worst LLM is considered for further refinement. The hyper rectangle of this LLM is split into two halves with an axis orthogonal split. Divisions in all dimensions are tried, and for each of the p divisions the following steps are carried out: (a) construction of the multi-dimensional validity functions for both generated hyper rectangles; (b) construction of all validity functions; (c) estimation of the rule consequent parameters for newly generated LLMs; (d) calculations of the loss function for the current overall model; (4) find the best division: the best of the *p* alternatives, checked in step 3, are selected and the related validity functions and LLMs are constructed. The number of LLM neurons is incremented (*m* = *m* + 1); (5) test for convergence: if the termination condition is met, then stop, else go to step 2.

Statistical Parameters

Three statistical parameters, R^2, *MSE* and standard deviation, were used to interpret the accuracy of the model. The coefficient correlation (R^2) is calculated from following equations:

$$\rho = \frac{1}{N} \sum_{i=1}^{N} X_{CO,i}^{exp} \tag{10}$$

$$R^2 = 1 - \left(\frac{\sum_{i=1}^{N_p} \left(X_{CO,i}^{exp} - X_{CO,i}^{cal}\right)^2}{\sum_{i=1}^{N} \left(X_{CO,i}^{exp} - \rho\right)^2}\right)^2 \tag{11}$$

The second one is mean square error (*MSE*) which is defined by Eq. (12)

$$MSE = \frac{1}{N} \sum_{i=1}^{N} (X_{CO,i}^{exp} - X_{CO,i}^{cal})^2$$

(12)

Also, the Standard Deviation is measured as:

$$\text{Standard Deviation} = \frac{1}{N} \sqrt{\sum_{i=1}^{N} (X_{CO,i}^{exp} - X_{CO,i}^{cal})^2}$$

(13)

$X_{CO,i}^{exp}$ and X_{COi}^{cal} indicate the experimental (target) and calculated (output) CO conversion values, respectively, and *N* clarifies the number of input patterns.

RESULTS AND DISCUSSION

Catalyst Characterization

The XRD technique performed for calcined (before the test) is shown in Fig. 3. Scans were taken with a 2θ range from 0 to 70° to identify the actual phases of 80%Co/20%Ni/15wt%Al$_2$O$_3$ catalyst .The actual phases identified for calcined catalyst were NiCo$_2$O$_4$ (fcc), NiO (rhombohedral) and Co$_3$O$_4$ (fcc) which were crystallized with diffraction peaks at 18.5, 23.5, 31.3, 36.9, 44.8, and 59.4°. Here to be noted that, oxidic phases are highly selective for the preparation of olefins.

The SEM observations have shown differences in morphology of precursor and optimal calcined catalysts (before and after the reaction). The electron micrograph obtained from catalyst precursor depicts several larger agglomerations of particles (Fig. 4a) indicating that this material has a less dense and homogeneous morphology. After the calcination at 600 °C for 6 h, the morphological features are different from the precursor sample and show that the agglomerate size is greatly reduced compared to the precursor (Fig. 4b). It may be due to cover the calcined catalyst surface with small crystallite of cobalt and nickel oxide. However, the size of these grains grew larger by
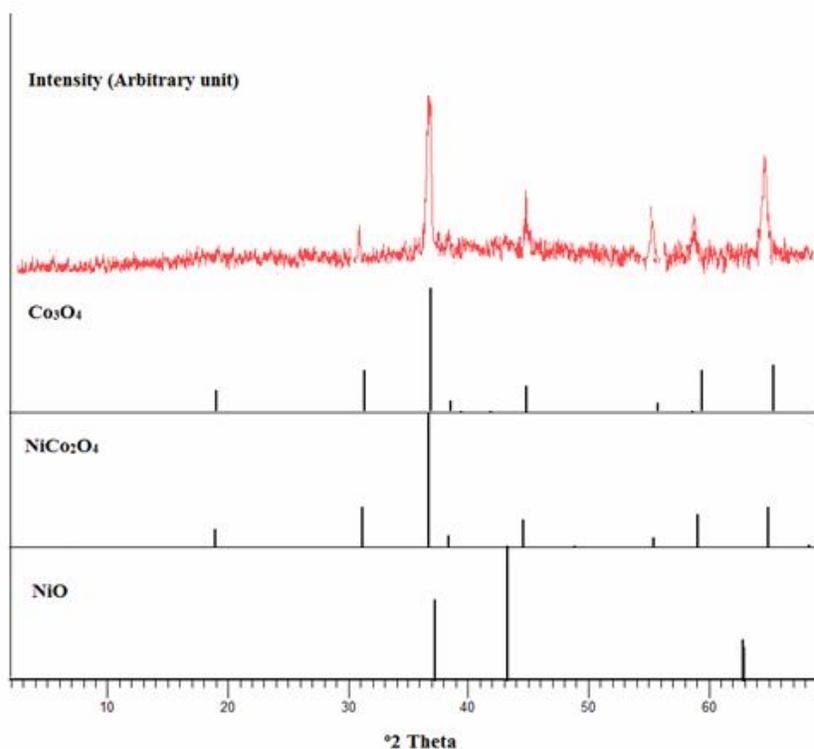
Fig. 3. XRD pattern of calcined sample for the 80%Co-20%Ni/15wt% Al$_2$O$_3$ catalyst.
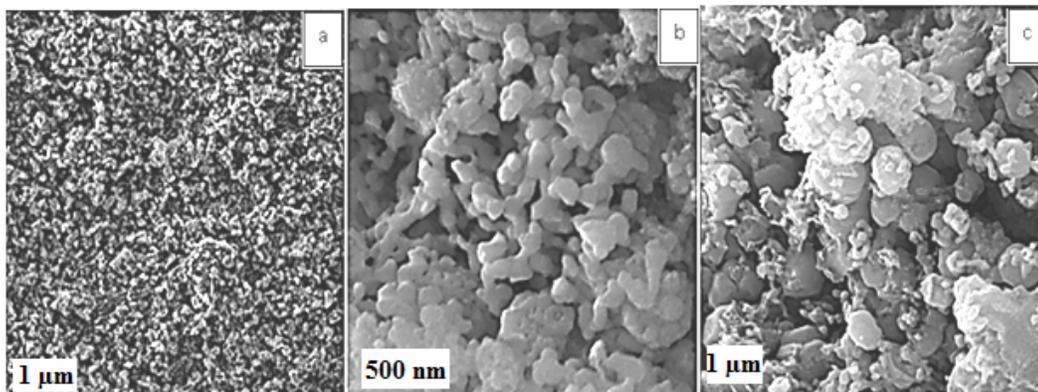
Fig. 4. The SEM observations of precursor and optimal calcined catalysts.

Table 1. BET Results of the Precursor and Calcined Catalysts (before and after the Test) Containing 80%Co/20%Ni/15wt%Al$_2$O$_3$ Prepared with Sol-Gel Procedure

Catalyst	Specific surface area (m^2 g^{-1})	Pore volume (cm^3 g^{-1})	Pore size (A°)
Precursor	57.13	3.91×10^{-2}	27.35
Fresh calcined	49.24	2.23×10^{-2}	28.28
Used calcined	19.82	1.46×10^{-2}	28.15

agglomeration in the tested catalyst (Fig. 4c), which may be due to the sintering after reactions.

The BET surface area, pore volume and average pore diameter of catalysts are tabulated in Table 1. As seen in this table, the catalyst precursor has a higher specific surface area (57.13 m^2 g^{-1}) than that in the calcined catalyst before (49.24 m^2 g^{-1}) and also after the test (19.82 m^2 g^{-1}). The obtained results in Table 1 show that there is no big variation among average pore size diameters of the catalyst in different stages of precursor and calcined samples (before and after the test). The high specific surface area of calcined catalyst before the test allows a high degree of metal dispersion [16].

The Effect of CO Conversion on the Products Selectivity

The effect of CO conversion on product distribution over a Co/Ni/Al$_2$O$_3$ catalyst was studied in a fixed-bed reactor at a wide range of operational condition (T = 200-
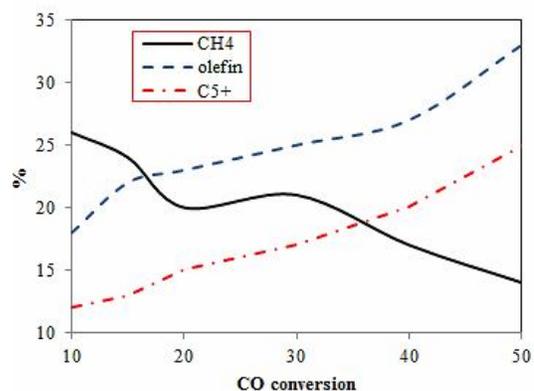
Fig. 5. Effect of CO conversion on the FT product selectivities.

250 ^{o}C, P = 1-10 bar, H_2/CO = 1-4 and $GHSV$ = 6000 h^{-1}. Figure 5 illustrates the effect of CO conversion on selectivities (CH$_4$, C$_{5+}$ and C$_2$-C$_4$ olefin) and shows that methane selectivity decreased (26-14%) as CO conversion

increased (10-50 %), whereas C_{5+} and C_2-C_4 olefin selectivities increased with increasing CO conversion.

LoLiMoT Development

LoLiMoT algorithm was established to predict CO conversion values as the target while appointing three variables namely partial pressure of CO (P_{CO}), partial pressure of H_2 (P_{H_2}) and H_2/CO feed ratio as the system input.

In the present study, by employing four-fold cross validation the data were partitioned into four equally sized segments (eight data in each segment). Four iterations of training and validation were performed during each of which; a different fold of the data was held-out for validation while the remaining three folds were used for training. To this end, 75% of the data was used to train the system and test its accuracy on the remaining 25% (Fig. 6). The MSE and the standard deviation values (7.4211e-004 and 5.5157e-004, respectively) proved that LoLiMoT shows a good ability for data prediction in this study. The curve plotted in Fig. 7 shows the error reduction process due to the rise in a number of neurons in the training process. The range and the type of the input and output variables are summarized in Table 2. It is necessary to be noted that the system is trained in the given range in Table 2. LoLiMoT has been used as a predictor in both the range of the experimental conditions (interpolation), and outside of training ranges (controlled extrapolation). The algorithm was also implemented by MATLAB.

As previously explained, the system was trained with 32 data in the given range in Table 2 to study the ability of LoLiMoT for data prediction within the trained range. The data was obtained in the following operational conditions: total pressure 1-10 bar; Temperature 200 ^{o}C; H_2/CO feed ratios 1-3 and $GHSV = 3000$ h^{-1}. The results are depicted in Fig. 8 (a). The obtained results indicate a good agreement between the experimental and predicted CO conversion ($R^2 > 0.99$).

The predictions were tested for the data within the trained range. In addition, the correlation between the derived data from output of the system and experimental data of CO conversion are illustrated in Fig. 8 (b). Regarding Fig. 8 (b), the LoLiMoT predicted data were in

the perfect agreement with experimental data ($R^2 > 0.93$). Therefore, this system could be employed for estimating the required data which were in the gained laboratory ranges but not experimentally available.

Data predicted beyond the training range (extrapolation) is a reliable and economical process of taking data. The data values at the given range and estimation value beyond the given range will always be necessary in real reaction for industry. However, just predicting based on the training data is not enough; we were to study the extrapolation ability of the model. The algorithm of controlled extrapolation has been illustrated in Fig. 9 and classified in the following steps:

a) Repeat the algorithm until the desirable number of data in a specific range is achieved.

b) Train LoLiMoT by all existing data samples as the input data.

c) Extrapolate the new data that has the closest range to the train data set (input data set).

d) Set the new input vector with the extrapolated data (step III) and existing data samples.

To achieve this purpose, two data sets for two steps (so-called extrapolation 1 and extrapolation 2) were selected to investigate the LoLiMoT ability for predicting data beyond the training range. Herein, as the same as interpolation step in predicting CO conversion values, obtained at following operational condition: $T = 200$ ^{o}C, $P = 1$-10 bars, H_2/CO $= 3/1$-3.5/1, $GHSV = 3000$ h^{-1}, the system was trained with 32 data in the specified range in Table 2 (extrapolation 1). This trend was continued until the desirable number of data in a specific range, adaptable to experimental data, was achieved. Consequently, in this section, 13 new estimated data have been achieved by controlled extrapolation. These results are shown in Fig. 10. Results indicate a good correlation between experimental and theoretical data ($R^2 > 0.86$).

Then we developed our study in the extrapolation (extrapolation 2) by LoLiMoT (*i.e.* H_2/CO $= 4$). The experimental data was obtained at following operational condition: $T = 200$ ^{o}C, $P = 1$-10 bars, H_2/CO $= 4$, $GHSV = 3000$ h^{-1}. Here, system was trained with 32 mentioned data as the same as the previous step and predicted data was extracted for H_2/CO $= 4$. The obtained results in the training step indicate a good agreement between the experimental
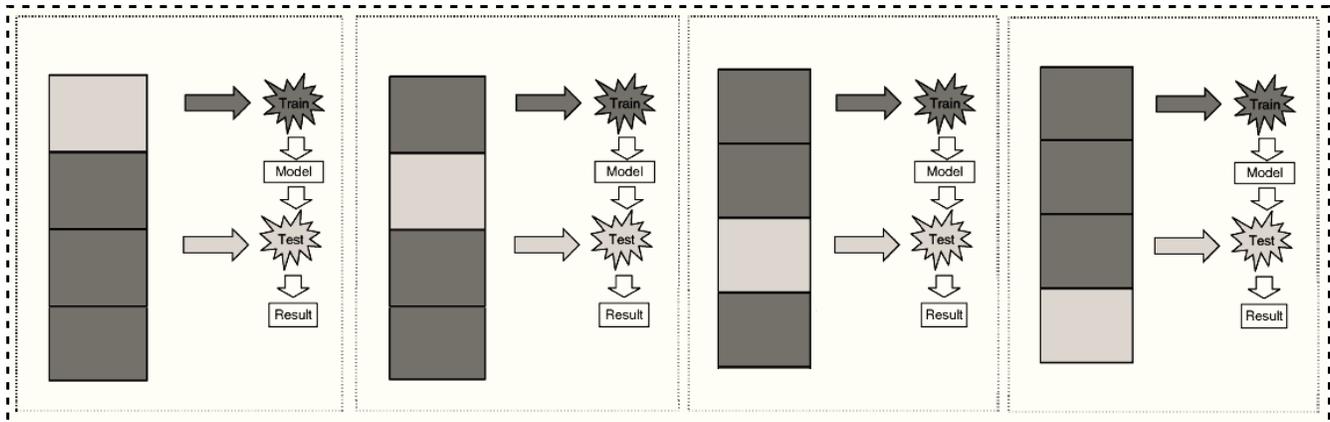
Fig. 6. Four-fold cross-validation.
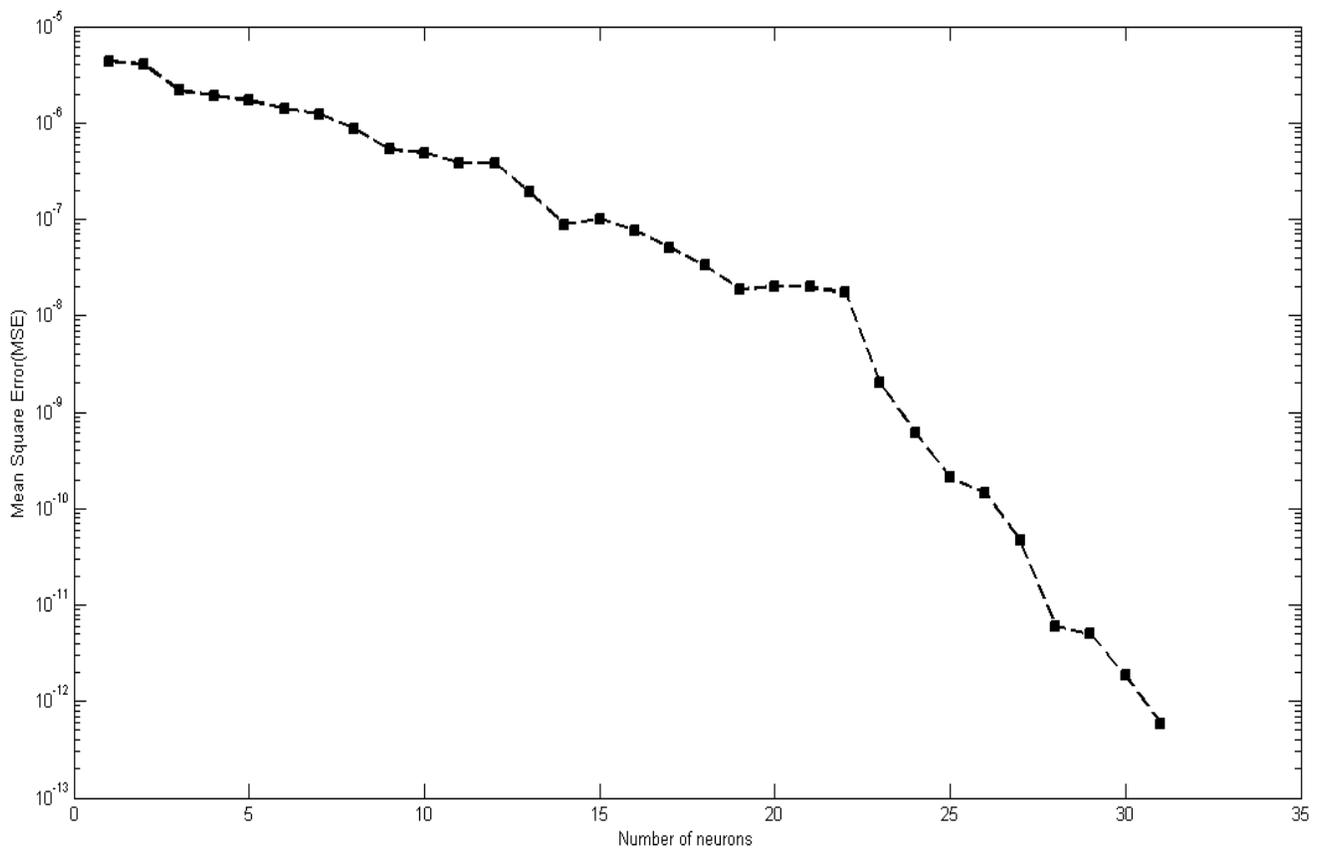
Fig. 7. Convergence curve in learning process by training data.

Table 2. Used Variables Range in Modeling Analysis

		Range
Input variables	P_{CO} (bar)	0.15-3.00
	P_{H_2} (bar)	0.30-4.50
	H_2/CO feed ratio	1/1-3/1
Output	CO conversion (%)	10-50
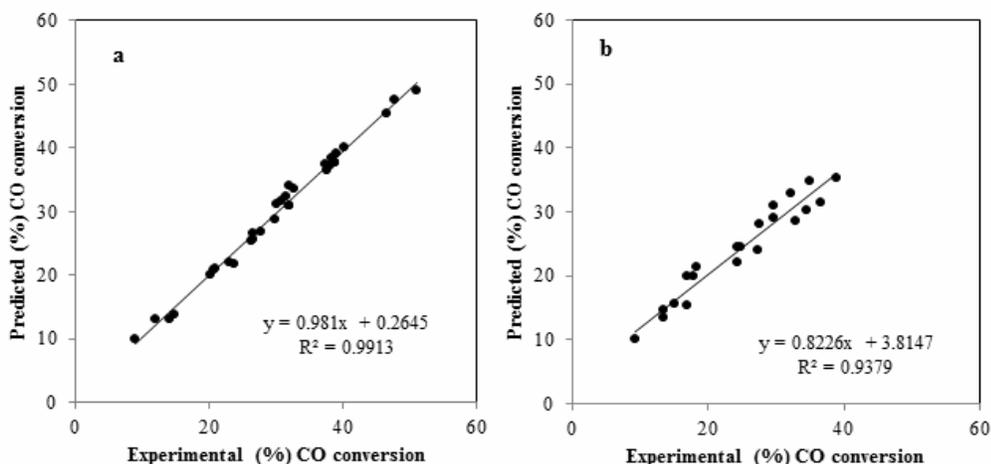
Fig. 8. Experimental *vs.* predicted CO conversion values for the training (a) and interpolation (b) data using LoLiMoT.
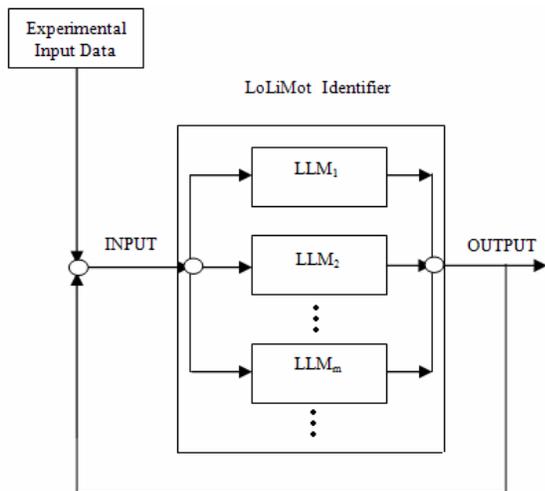
Fig. 9. Controlled extrapolation algorithm.
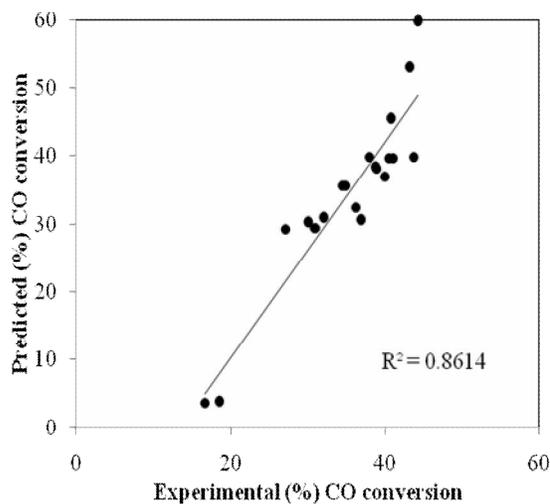
Fig. 10. Experimental *vs.* predicted CO conversion values for the extrapolation (1) data using LoLiMoT.

Table 3. Comparison of Correlated and Experimental C Values for FTS in Fixed Bed Reactor; Operational Condition: T = 200 ^{o}C, P = 1-10 bar, H_2/CO = 4, GHSV = 3000 h^{-1}

COconversion (*corr*) (%)	*COconversion* (exp) (%)	Δ *COconversion* (%)
44.1	41.0	-3.1
51.9	51.8	-0.1
52.0	49.0	-3.0
50.7	47.0	-3.7
48.6	45.5	-3.1
43.3	45.4	2.1

and predicted CO conversion. Among training step, LoLiMoT has the excellent accuracy with the correlation coefficient *(R^2 > 0.99)*. Therefore, for predicting the CO conversion values, LoLiMoT is a suitable system modeling. The Eq. (14) (based on Fig. 8a) is the corresponding correlation coefficient using LoLiMoT in the training step.

$$COconversion\ (corr) = 0.981\ COconversion\ (LoLiMoT) + 0.2645$$
$$(14)$$

where, the $COconversion\ (LoLiMoT)$ indicates the predicted CO conversion values for the training step by using the LoLiMoT.

Finally, the correlation Eq. (14) containing the empirical parameters, *i.e.*, slope and intercept in Eq. (14) was used to determine the CO conversion values of the investigated range. Table 3 shows a good agreement between the experimental and the predicted CO conversion values by LoLiMoT of investigated range in F-T process.

CONCLUSIONS

An alumina-supported cobalt nickel catalyst was prepared using sol-gel procedure, and it showed the high ability for light olefins synthesis in F-T process. The catalyst was characterized using XRD, SEM and BET surface area techniques, and it was concluded that the operating conditions had a marked effect on the morphology and texture of the catalysts. The obtained result from the investigation on the effect of CO conversion on product distribution showed that CH_4 selectivity monotonically

decreased, but C_{5+} and C_2-C_4 olefin selectivities increased with increasing in CO conversion. LoLiMoT algorithm was designed for predicting the CO conversion in the F-T synthesis. A Cross-validation technique was employed to assessment the performance of the system. The values of MSE (*i.e.* 7.4211e-004) and the standard deviation (5.5157e-004), indicated that LoLiMoT had an excellent accuracy for prediction of CO conversion over the mentioned catalyst in F-T process. It was also shown that LoLiMoT could predict data in the range of the experimental data (interpolation step) successfully, with a high accuracy from the trained model. The extrapolation ability of LoLiMoT was studied with a novel idea so-called controlled extrapolation. The results exhibited that predicted CO conversion values have a good agreement with the experimental data. In addition, this algorithm can be used in order to save time and to reduce the costs of experimental study of F-T reaction.

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