Korean J. Chem. Eng., 40(6), 1284-1296 (2023)
DOI: 10.1007/s11814-023-1378-2

pISSN: 0256-1115
eISSN: 1975-7220

Multiple linear regression and GRU model for the online prediction of catalyst activity and lifetime in counter-current continuous catalytic reforming

Yongbeom Shin*,‡, Myeongnam Park*,‡, Jaewook Lee*, Kibyung Kang, Sungki Wang**, Eunsung Lee**, Seongkeun Song**, and Dongil Shin*,†**

*Department of Chemical Engineering, Myongji University, 116 Myongjiro, Cheoingu, Yongin, Gyeonggido 17058, Korea
**Hanwha TotalEnergies Petrochemical, 103 Doggod2-ro, Daesan-eup, Seosan-si, Chungcheongnam-do 31900, Korea
(*Received 18 July 2022 • Revised 2 November 2022 • Accepted 16 December 2022*)

Abstract−In the catalytic reforming process, aromatic yield is a standard for evaluating the production quality of the process, and studies are underway to improve productivity by optimizing the cost and energy. In particular, the activity and lifetime prediction of reforming catalysts can contribute to overall process efficiency improvement, such as product quality, productivity, and predictive maintenance. However, it is difficult to predict real-time catalyst activity and lifetime according to changes in process operation with the existing method that requires experimental data. In this study, a multiple linear regression (MLR) model and GRU model with the real process operating data are proposed for long-term plant operation and optimization in the counter-current continuous catalytic reforming. The MLR-GRU model predicts catalyst performance degradation and lifetime according to operating conditions by defining a new variable, reforming catalyst activity. The proposed model can predict the future reformate yield with an error of less than 1%. As a result of predicting the catalyst lifetime according to various operating temperatures, feed flow patterns, and feed quality, the feed flow rate had the greatest influence on the catalyst lifetime profile. In terms of the amount of produced reformate oil, the case with maximum feed rate is the worst (−25.6%); on the other hand, the case with minimum feed rate is the best (+11.4%). Thus, it is important to establish an appropriate production plan of the produced reformate oil. The model proposed in this study can predict the reformate yield and lifetime, reflecting the degradation of catalyst performance according to the operating profile in real-time, which is expected to improve productivity by production scheduling, optimization, and predictive maintenance.

Keywords: Counter-current Continuous Catalytic Reforming, Reformate Yield, Catalyst Activity, Catalyst Lifetime Prediction, Scheduling and Optimization

INTRODUCTION

Catalytic reforming, which converts naphtha of low octane range into hydrocarbons of high octane range, is a vital process in refinery plants that produce high-value-added products such as high-octane gasoline and aromatics [1,2]. In particular, aromatic hydrocarbons such as benzene, toluene, and xylene are crucial substances used as raw materials for petrochemical products and polymers, and many studies on converting naphtha to aromatic hydrocarbons have been conducted for a long time. Currently, a large amount of naphtha is converted industrially into aromatic hydrocarbons through catalytic reforming [3-5]. In catalytic reforming, naphtha is converted to aromatics by a catalyst in a series-connected continuous reactor, and the catalyst is regenerated by a continuous catalyst regeneration (CCR) unit [6]. CCR is used in most catalytic reforming processes because of the high purity of aromatic and hydrogen produced, and high efficiency due to low operating cost [7]. The catalyst flowing through the CCR unit can be divided into co-current continuous reforming that flows in the same direction as the reactants and counter-current continuous catalytic reforming that flows in the opposite direction to the reactants; counter-current continuous catalytic reforming shows excellent efficiency and has been mainly used recently [8].

In catalytic reforming, aromatic yield is an important variable as a criterion for evaluating the production quality of the process; studies to improve productivity by optimizing the cost and energy consumption are in progress. In addition, research on optimization of the reforming process applied with the CCR unit, which is receiving attention recently, is required [9]. Many researchers have proposed a mathematical model of the CCR reforming process with complex reaction kinetics through modeling and simulation studies for optimizing the operation of the co-current continuous reforming process, which is a traditional technology, and they carried out studies to achieve improving energy efficiency and productivity [10-17]. However, counter-current continuous catalytic reforming, which has been recently applied to commercial processes due to its efficiency, has a unique structure in which the flow of reactants and catalyst flow are in opposite directions, and the difference in catalyst activity depending on the degree of reaction in each reactor; consequently, it is difficult to simulate with commercial simulation software such as the existing Aspen Plus, KBC, and Schneider [18]. A molecular-level kinetic study of the catalytic reforming reaction was conducted for modeling and simulation of the counter-current

†To whom correspondence should be addressed.
E-mail: dongil@mju.ac.kr
‡Both authors contributed equally to this work.

continuous catalytic reforming process, and a counter-current continuous catalytic reforming reactor model was developed based on the reaction kinetics [8,19]. A counter-current continuous catalytic reforming process model was proposed by the existing reactor model, and it was shown that it can be utilized for process real-time optimization (RTO) [18].

Because the flow of reactants and the flow of catalyst are opposite, which is the characteristic of counter-current continuous catalytic reforming, the activity of the catalyst is different for each reactor; therefore, it is necessary to reflect the activity of the catalyst, which greatly affects the reaction kinetics. In addition, it is important to monitor the catalyst activity and lifetime to improve the productivity of the process, and the catalyst activity and lifetime interact with many parts of the chemical plant, such as product quality, production volume, and maintenance period [20]. Hence, many researchers have investigated mathematical modeling for predicting the activity and lifetime of catalysts, and in the case of catalytic reactions, artificial neural network-based modeling studies using data with complex reaction kinetics are also being conducted [21,22]. Furthermore, catalyst activity and lifetime prediction model using a hybrid modeling method that compensates for the high computing cost, which is a disadvantage of mathematical modeling, and the artificial neural network model, which is a block box model, have been proposed [23-28].

Catalyst activity and lifetime are greatly affected by changes in process operating conditions and external factors [29,30]. Nevertheless, it is difficult to predict catalyst activity and lifetime in real-time according to process operation changes in the model proposed in previous studies, because experimental data obtained by collecting catalyst samples are absolutely necessary for predicting catalyst activity and lifetime. In this study, we propose a counter-current continuous catalytic reforming process model for long-term plant operation and optimization. The model includes a catalyst performance degradation and lifetime online prediction according to operating conditions that have not been reflected in the previous. To make online predictions according to the process operation pattern of the catalytic reforming, the proposed model handles the actual counter-current continuous catalytic reforming process data, not the experimental data by sample. The online data, reformate yield, is used to predict the degradation of the catalyst using process data, and the reformate yield is more affected by operating conditions than degradation of the catalyst; accordingly, reforming catalyst activity reflecting the used date and operating pattern of the catalyst is defined as a new variable. A multiple linear regression (MLR) model is proposed to define the reforming catalyst activity, and a gated recurrent unit (GRU) model is proposed to predict the future reforming catalyst activity. The reforming catalyst activity predicted using the GRU model is converted into a reformate yield through the MLR model, and finally the future reformate yield is predicted. A model that predicts the future reformate yield according to various operating patterns and predicts the catalyst replacement time when the reformate yield is less than 80% based on the reformate

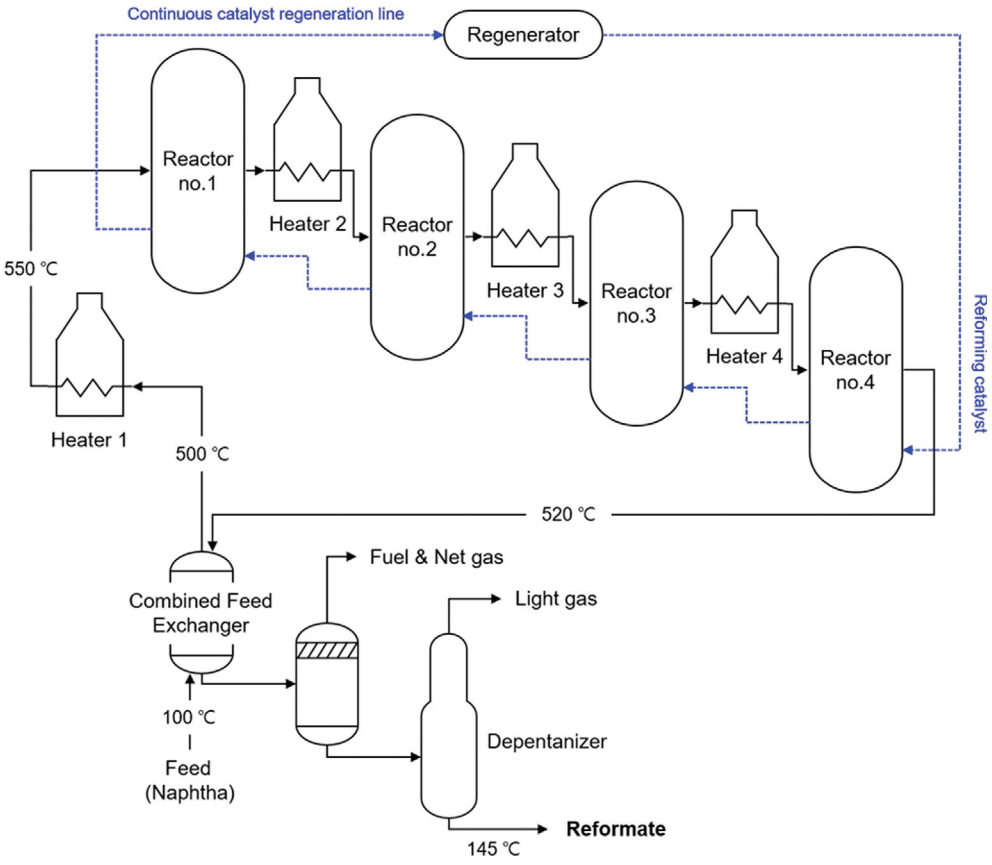

Fig. 1. Process flow diagram of catalytic reforming.

Table 1. Description of the dataset in catalytic reforming

Features	Value
Time span	August 1, 2014-December 31, 2020
Number of data point	2267
Number of variables	22
Variables	Use date, feed flow rate, cumulative feed flow rate, feed quality (concentration of naphthene, sulfur, nitrogen, water), boiling point (IBP, EP), temperature (average, reactor 1 to 4), pressure (average, reactor 1 to 4), reactor recycle gas flow rate, catalyst chloride, reformate yield

yield is proposed in real-time according to the operation pattern changes. In the next section, the target process and secured data of this study are explained, and then the MLR model and GRU modeling process are illustrated. Afterward, the reformate yield prediction results using the MLR-GRU model and the catalyst lifetime prediction results according to the operating profile is explained in Results and Discussion.

DESCRIPTION OF CATALYTIC REFORMING AND MODEL STRUCTURE

1. Catalytic Reforming

Catalytic reforming is a chemical process that converts naphtha with low octane range into reformate with high octane range. Fig. 1 shows the process flow diagram of catalytic reforming. Naphtha, the feed of this process, is introduced into the combined feed exchanger (CFE) at about 100 °C, and is supplied to a heating furnace (Heater 1) at 500 °C through heat exchange with the product of the fourth reactor (Reactor no. 4). Naphtha is heated to 550 °C in the heater 1 and then supplied to the first catalytic reactor (Reactor no. 1), and the reaction product is reheated through the heater and supplied to the next reactor. Naphtha goes through a total of four catalytic reactors, and the product produced through the fourth reactor is supplied to CFE and heat-exchanged with the new feed, and then passes through a separator and depentanizer to produce the final product, liquid reformate. The catalyst used for the reaction is UOP R-264, which is mainly used for naphtha reforming, and the catalyst has a continuous flow in the opposite direction to the feed flow, and the catalyst is regenerated by a counter-current continuous catalytic catalyst regeneration method.

Table 1 is a description of the data set of the collected catalytic reforming process. For the process, a dataset of 2267 data points and 22 operating variables of catalytic reforming was obtained for about 6 years and 5 months from August 1, 2014 to December 31, 2020. The data was measured in units of days, and the variables consisted of the number of catalyst using days, the flow rate and composition of the feed, the concentration of the catalyst poison, the reactor temperature and pressure, the recycle gas flow of the reactor, the catalyst chloride, and the reformate yield.

2. Model Structure

The reformate yield of catalytic reforming is process data that can be continuously obtained, and through the reformate yield the performance of the catalyst used in catalytic reforming can be continuously predicted without experimentation. However, there are two problems in predicting long-term performance degradation of catalysts by reformate yield. First, since the outlet condition of the previous reactor is connected to the inlet condition of the next reactor when using a continuous reactor like catalytic reforming, the outlet condition of each reactor must be accurately predicted for the yield prediction of the final product [31]. In addition, although the catalyst performance decreases as the process operation time increases, it is difficult to predict the reaction yield of a long-term continuous reactor using a mathematical model, because the nonlinearity is large through the regeneration process with continuous flow. Second, when the catalyst performance deteriorates due to the extended period of use of the catalyst, the yield is maintained within a certain operating range by adjusting operating variables such as operating temperature and pressure. There is a limit to accurately predicting the actual degradation of the catalyst performance using the reaction yield.

In order to solve the two problems, in this study, reforming catalyst activity indicating the yield of the current process compared to the yield using the initial fresh catalyst under specific operating conditions was defined as a new variable, and the GRU model was proposed to predict long-term catalyst performance degradation from actual process data. Fig. 2 is the model structure proposed in this study to predict the future reformate yield. We first proposed an MLR model that predicts the yield dependent only on feed and operating conditions when using a fresh catalyst without deterioration of catalyst performance by data at the beginning of the catalytic reforming process operation. Using the MLR model, the maximum yield from a specific feed and reactor operating condition was predicted. The reforming catalyst activity, which was obtained by dividing the yield of the actual data by the maximum yield, was proposed as a new learning variable. The reforming catalyst activity is based on the maximum yield that can come out of the condition, even if the process conditions are adjusted due to the degradation of the catalyst; as a result, the degradation of the catalyst can be reflected through the yield. Afterward, a GRU model for predicting reforming catalyst activity was proposed using the number of use days, feed flow rate and quality, and reactor operating conditions as inputs. Through the calculation of the output of the MLR model and the output of the GRU model under specific feed and reactor operating conditions in the future, the reformate yield that can be obtained by reflecting the degradation of catalyst performance and operating conditions of the process was predicted. The learning process of the MLR model that predicts the yield of fresh catalyst and the GRU model that predicts the future reforming catalyst activity is divided into 'Construction of the Multiple Linear Regression' and 'Construction of the GRU Model' in this paper.

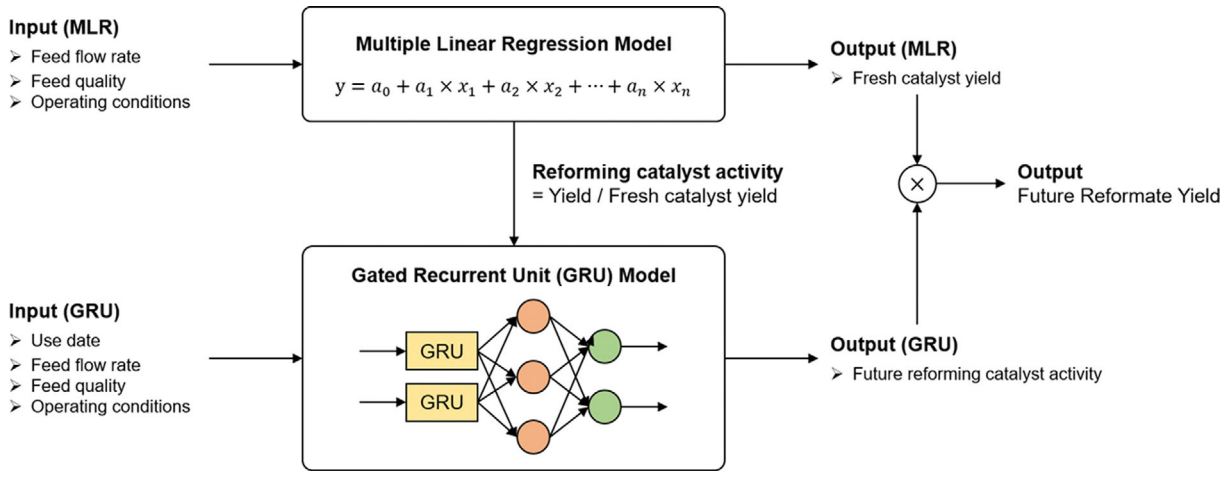

Fig. 2. Structure of the proposed model for future reformate yield prediction.

CONSTRUCTION OF THE MULTIPLE LINEAR REGRESSION MODEL

1. Data Preprocessing and Feature Selection

Since the catalytic reforming process dataset used in this study is the actual process operation data, there are many missing values and outliers; thus data preprocessing was performed at first. Since the dataset has units of days rather than continuous data in minutes or hours, missing values were deleted without filling in through interpolation, and outliers were corrected through interpolation using a standard score (Z-score) based on a Z value of 2.5. Pearson correlation coefficient (PCC) analysis was performed to select a feature of the MLR model predicting reformate yield when using the fresh catalyst through data processed with missing and outli-

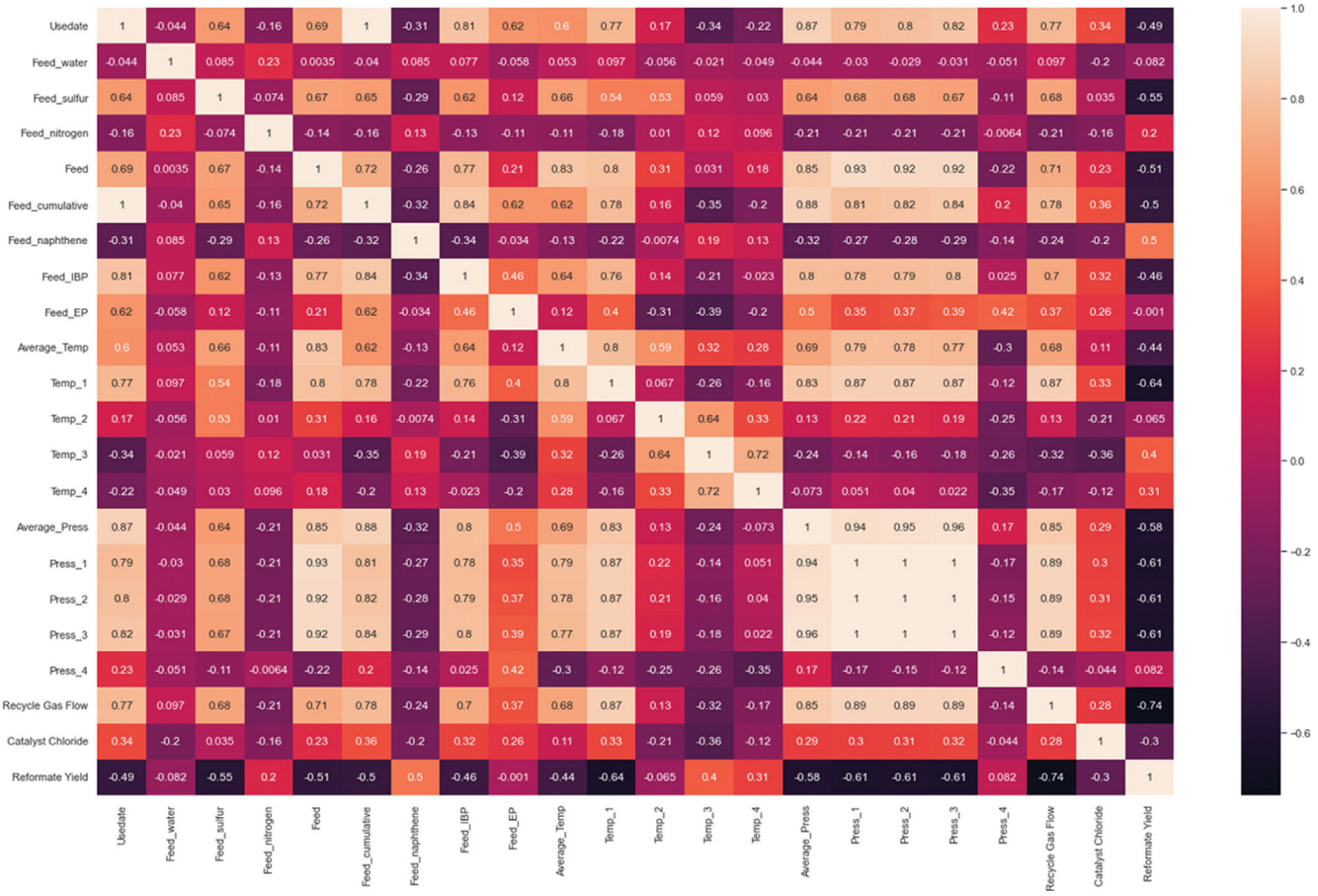

Fig. 3. Pearson correlation coefficient for determining the relationship.

ers. Fig. 3 is a heatmap showing the PCC analysis results for all features, and features were selected based on the PCC analysis result, which has a 'moderate correlation' over PCC of 0.4 [32]. As a result, variables with an absolute value of 0.4 or more of correlation coefficients such as 'Feed_sulfur', 'Feed', and 'Feed_IBP' were selected as variables to explain the flow rate and quality of the feed. As the operating parameters of the reactor, since the temperature is a variable directly related to the yield of a chemical reaction and the activity of a catalyst, 'Temp_1', 'Temp_2', 'Temp_3', and 'Temp_4' were all selected. For pressure, only 'Press_1' was used, which showed that the correlation coefficients of 'Press_1', 'Press_2', and 'Press_3' were 1, indicating that the dataset was completely consistent, and 'Press_4' was excluded because the pressure change in the reactor was insignificant. Additionally, reactor recycle gas flow, which has a high correlation with the yield of the reaction, was selected as an input variable. The selected variables were equally converted to a scale between 0-1 through the min-max normalization method.

2. Multiple Linear Regression Model

When using fresh catalyst, the MLR model for yield prediction according to the change of nine variables (Feed_sulfur, Feed, Feed_IBP, Temp_1, Temp_2, Temp_3, Temp_4, Press_1, Recycle Gas Flow) was defined as follows:

$$\text{yield}_{base}=a_0+a_1x_1+a_2x_2+a_3x_3+a_4x_4+a_5x_5+a_6x_6+a_7x_7+a_8x_8+a_9x_9$$

x_1: Feed_sulfur x_6: Temp_3
x_2: Feed x_7: Temp_4 (1)
x_3: Feed_IBP x_8: Press_1
x_4: Temp_1 x_9: Recycle Gas Flow
x_5: Temp_2

To estimate the parameters of the MLR model, the following reformate yield prediction model was obtained using data from the initial 90 days of operation using fresh catalyst:

$$\text{yield}_{base}=83.940+0.950x_1+1.533x_2+0.352x_3+0.358x_4 +0.728x_5+1.443x_6-1.816x_7-0.830x_8-0.056x_9 \quad (2)$$

The MLR model was validated using data from after 60 days (91 to 150 days). Fig. 4 is the reformate yield prediction result using the MLR model when using the fresh catalyst at the initial stage of operation, and the prediction error of the MLR model is 1.385 based on MAE and 1.413 based on RMSE, showing a 1% level of prediction error in the prediction of reformate yield.

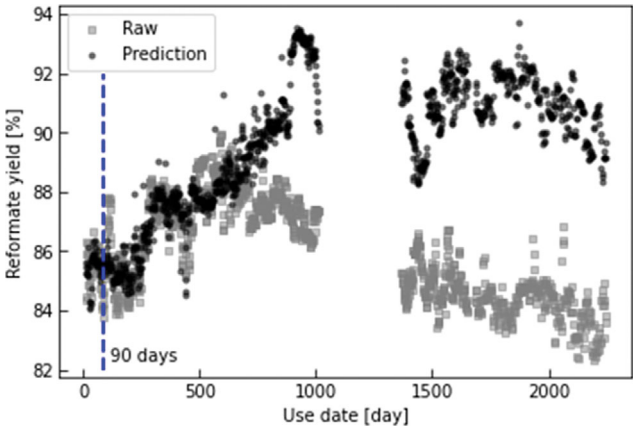

Fig. 4. Reformate yield prediction results when using fresh catalyst by MLR model.

3. Reforming Catalyst Activity

Using the output of the MLR model, which is a yield prediction model of the fresh catalyst under specific operating conditions, reforming catalyst activity, which is the ratio of reformate yield when using a fresh catalyst and catalyst with reduced performance, was defined as follows:

$$\text{Reforming catalyst activity}=\frac{\text{yield}}{\text{yield}_{base}} \quad (3)$$

where yield is the reformate yield at the time of prediction, and yield_{base} is the reformate yield when a fresh catalyst is used under the same operating conditions.

Fig. 5 shows the reformate yield of the existing data and the reforming catalyst activity obtained using the results of the MLR model. The reformate yield did not decrease until about four years even if the process operating time was increased, but rather increased, which is a result of the increase in the reactor operating temperature. On the other hand, reforming catalyst activity is the ratio of reformate yield when a fresh catalyst is used under the same

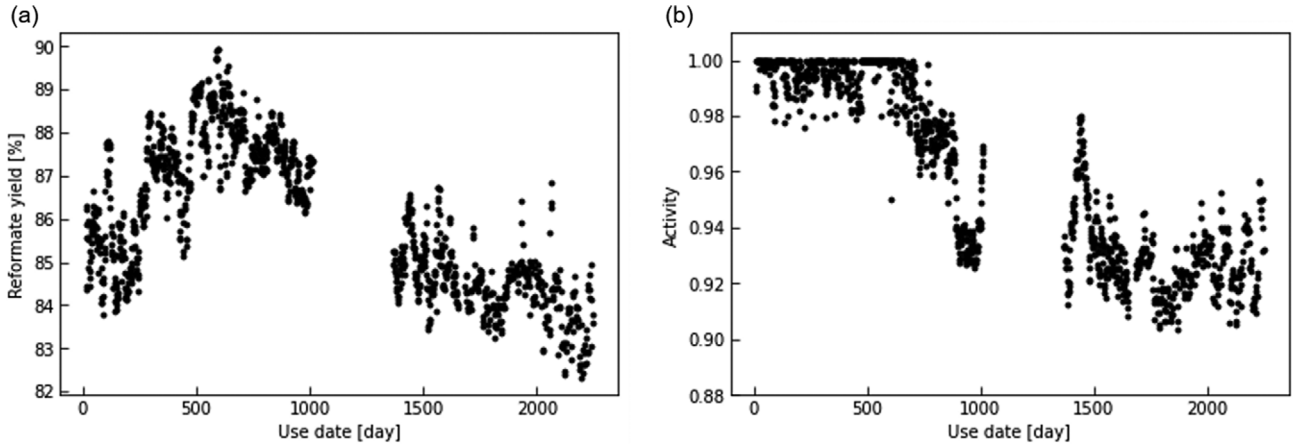

Fig. 5. Data comparison between reformate yield and activity. (a) Reformate yield, and (b) Reforming catalyst activity.

operating conditions, so it can indicate the state of the catalyst. As the process operation time increases, reforming catalyst activity tends to decrease. A new variable, reforming catalyst activity, was defined using the MLR model, which is a yield prediction model of the fresh catalyst, and a deep learning model for predicting catalyst performance degradation was proposed by learning the reforming catalyst activity defined in this study.

CONSTRUCTION OF THE GRU MODEL

1. Data preprocessing and Feature Selection

A deep learning model was designed for predicting reforming catalyst activity defined in the previous section; Fig. 6 is a flowchart showing the data preprocessing to train the model with the raw data obtained from the actual process. Missing values of the data (22 variables, 2267 data points) obtained from the catalytic reforming process were removed, and the outliers were judged based on Z-score 2.5 and corrected through linear interpolation. After missing values and outliers were processed, PCC analysis was used to select the model features. Since the reforming catalyst activity is a variable defined from the yield of the existing data, the PCC analysis result (Fig. 3) that was previously conducted was used to select the feature. To include reforming catalyst activity information for various process condition changes in the model, features with a correlation coefficient of |0.3| or higher were selected as input features. As a result, 12 variables (Usedate, Feed_sulfur, Feed, Feed_cumulative, Feed_naphthene, Feed_IBP, Temp_1, Temp_3, Temp_4, Press_1, Recycle Gas Flow, Catalyst Chloride) were selected as input for the prediction model. Additionally, since the catalyst usage profile affects the catalyst performance degradation, a total of 13 variables were determined as inputs by adding the past reforming catalyst activity as a feature of the model. After the features of the predictive model were selected, the min-max normalization method was used to match the data scale equally. The training was carried out by dividing the training dataset and the test dataset sequentially in a ratio of 8 to 2, which was set to the first 916 data points (to 1,714 days) for the training dataset and the remaining 229 data points (1,717 days to 2,246 days) for the test dataset. Because our dataset does not cover the whole period of the lifetime of catalyst, we set the last 20% data, which was not used for model training, for the test dataset to confirm the possibility of the performance degradation and lifetime prediction in the future.

Table 2. Results of Bayesian optimization for searching the optimal model

Model	Structure	Input sequence	Number of searches	RMSE
FNN	5 Dense	-	300	0.832
LSTM	2 LSTM - 1 Dense	20	40	1.847
GRU	3 GRU - 5 Dense	20	40	0.699

2. Hyperparameters and Model Structure Tuning

For deep learning modeling, the feed-forward neural network (FNN) model, which is the basic structure of artificial neural networks, and since the usage pattern of the existing catalyst affects the degradation of catalyst performance, we compared the performance of long short-term memory (LSTM) and gated recurrent unit (GRU) models, which are time series models based on recurrent neural network (RNN), to determine the reforming catalyst activity. Table 2 shows the results of the optimal model structure searching through Bayesian optimization to select the optimal model among FNN, LSTM, and GRU models, and 300 for FNN

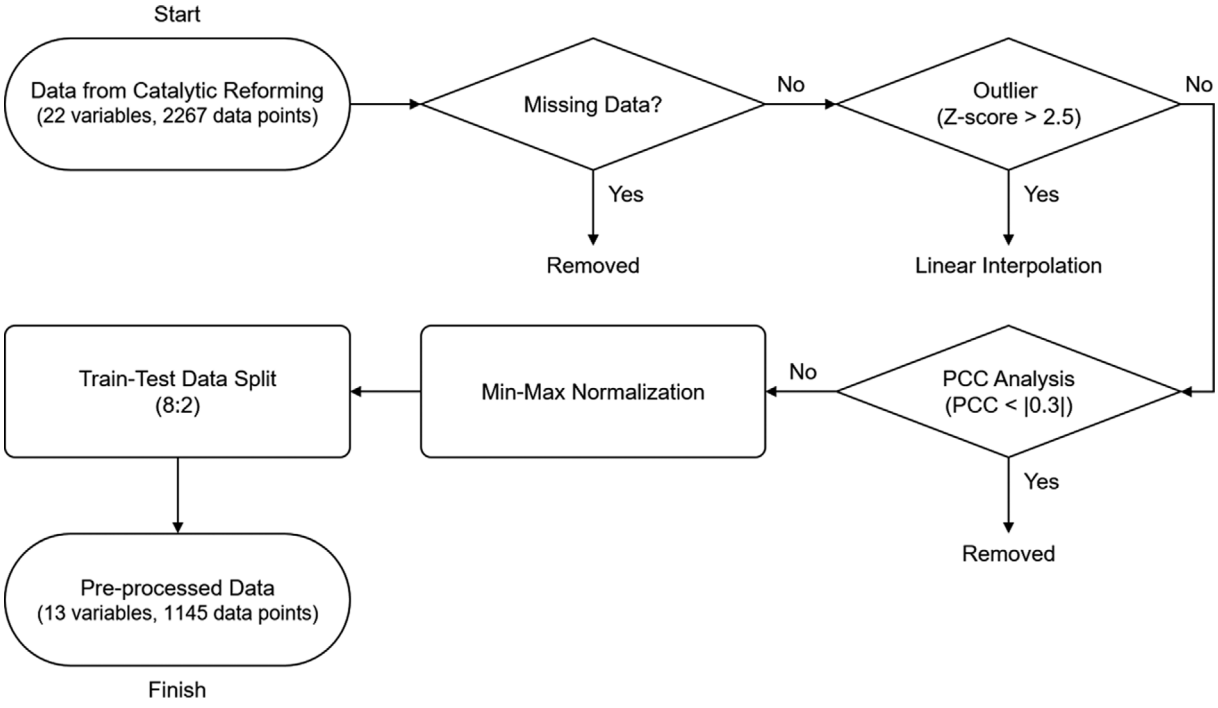

Fig. 6. Flow diagram of data preprocessing through dealing with missing and outlier, PCC analysis, min-max normalization, and data split.

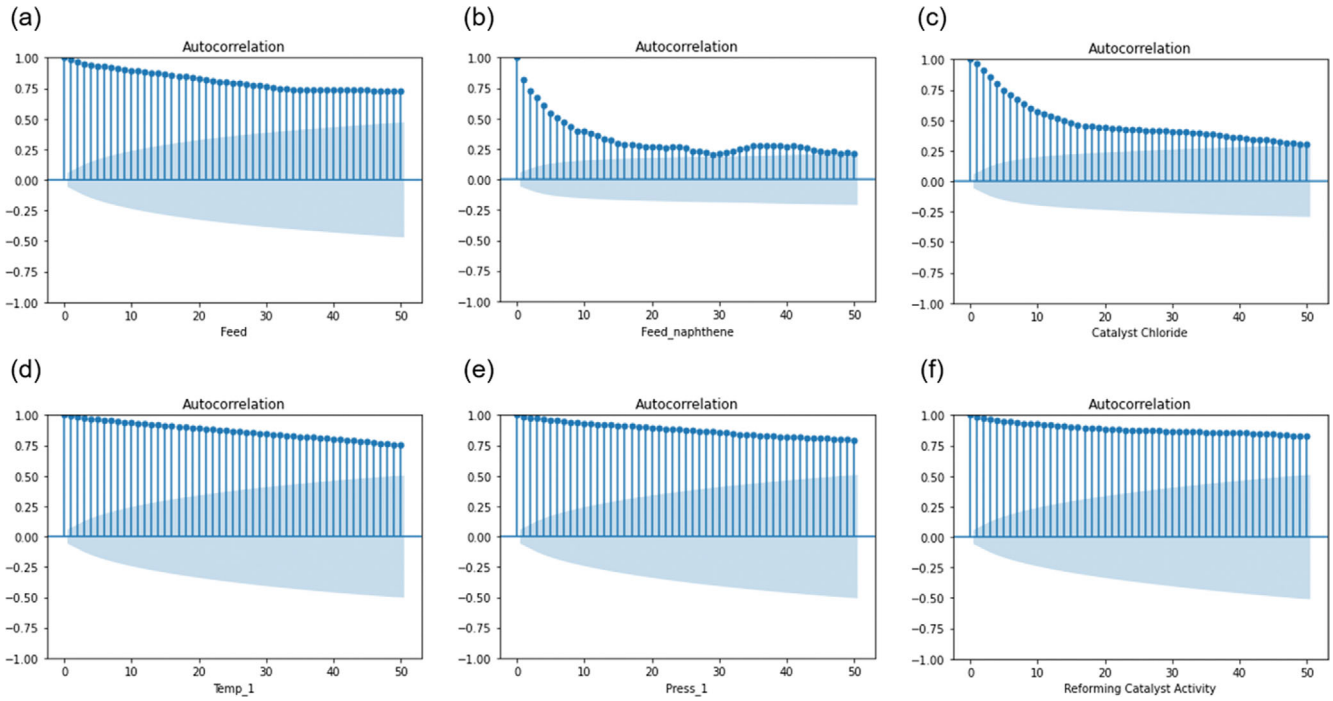

Fig. 7. Autocorrelation according to the variables. (a) Feed, (b) Feed_Naphthene, (c) Catalyst chloride, (d) Temp_1, (e) Press_1, and (f) Reforming catalyst activity.

Table 3. Model performance comparison according to the input sequence

	Input sequence	Number of searches	Optimal model structures (G: GRU layers)	RMSE
Case 1	3	1,000	70G-53G-118-418-304-1	0.056
Case 2	5	1,000	10G-63G-77G-498-386-258-1	0.007
Case 3	10	1,000	7G-55G-215-100-126-292-14-1	0.063
Case 4	15	1,000	18G-22G-499-223-423-1	0.064
Case 5	20	1,000	14G-26G-92-314-1	0.053

and 40 for LSTM and GRU respectively. As for the model performance, the GRU model showed the highest performance with an RMSE of 0.699, hence GRU was selected as the model for predicting the reforming catalyst activity.

Since the GRU model is a time series model of the RNN series, it is necessary to determine the sequence of the input data. To determine the input sequence, the autocorrelation function (ACF) was used to calculate the autocorrelation coefficient of each feature, and the results are shown in Fig. 7. Process control variables such as feed flow rate, reactor temperature, and pressure have high autocorrelation, and the current data showed a high correlation value up to a long time interval (more than 50 days). However, 'Feed_naphthene' and 'Catalyst Chloride' have a small lag of about 25 to 40 days compared to other variables. Since the input of the reforming catalyst activity prediction model has a sequence, in order to ensure that the sequences of all input variables have autocorrelation, the maximum input sequence is based on 'Feed_naphthene' with the smallest lag, up to 20 selected in this study. Afterward, the optimal input sequence from a time difference of 3 days to a maximum of 20 days was searched through Bayesian optimization to reduce the dimension of the model input, the complexity of the model, and the learning time. Table 3 shows the results of searching for 1000 models each through Bayesian optimization, selecting the case where the input sequence is 3, 5, 10, 15, and 20 days as each case. As a result of the optimization, the model consisting of 3 GRU layers, 3 dense layers, and output layers with a time sequence of 5 days showed the highest performance with an RMSE of 0.007, and was selected as the reforming catalyst activity prediction model.

RESULTS AND DISCUSSION

1. Prediction of Reforming Catalyst Yield

The future reformate yield can be predicted using the MLR model and GRU model proposed in this study. Fig. 8 shows the two proposed model structures and the flow diagram for predicting the future reformate yield. Through the MLR model, the reformate yield in the case of using a fresh catalyst according to specific operating conditions was predicted, and the yield of the actual data was converted into reforming catalyst activity, a newly defined variable in

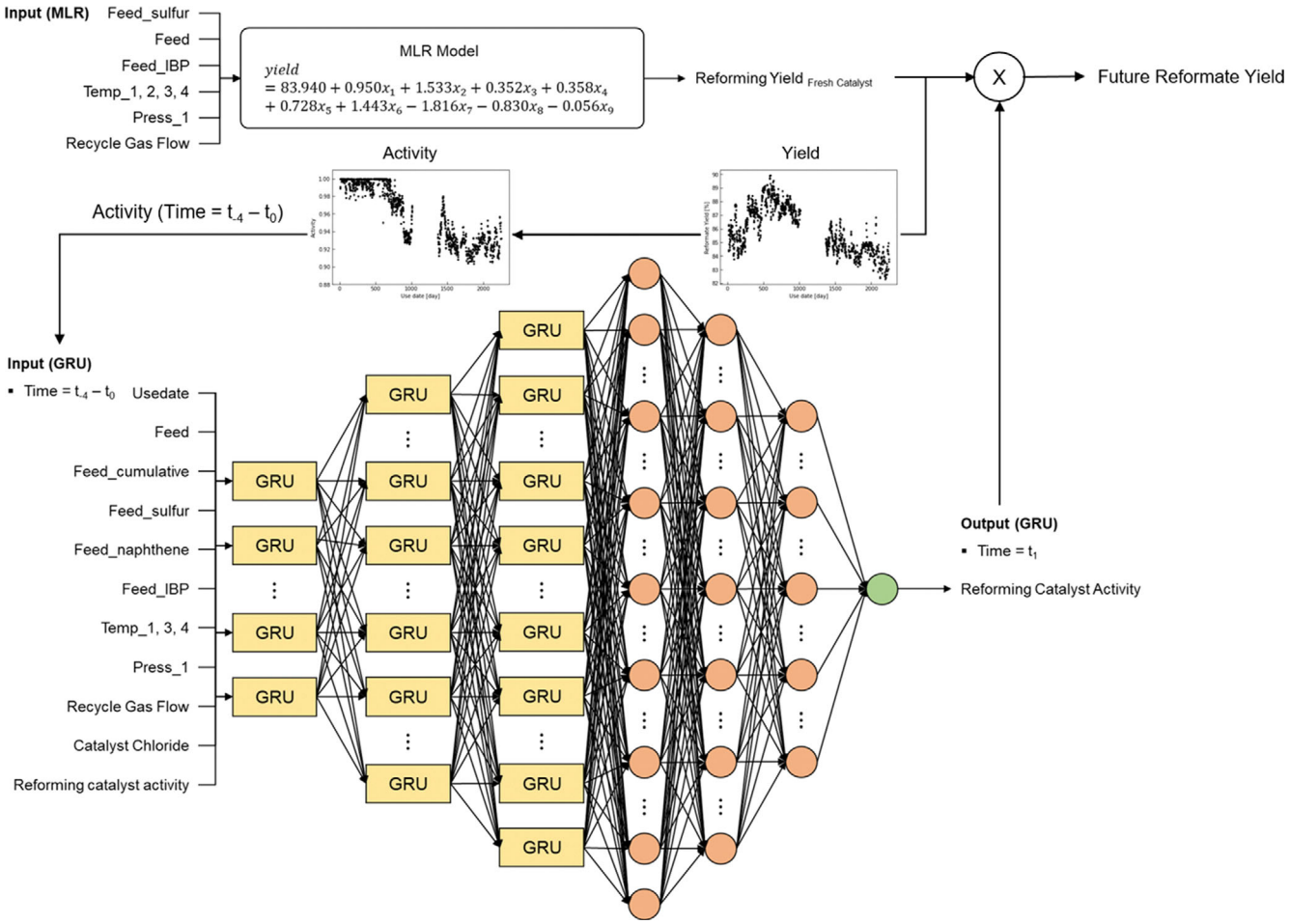

Fig. 8. Multiple linear regression and GRU modeling results for prediction of future reformate yield.

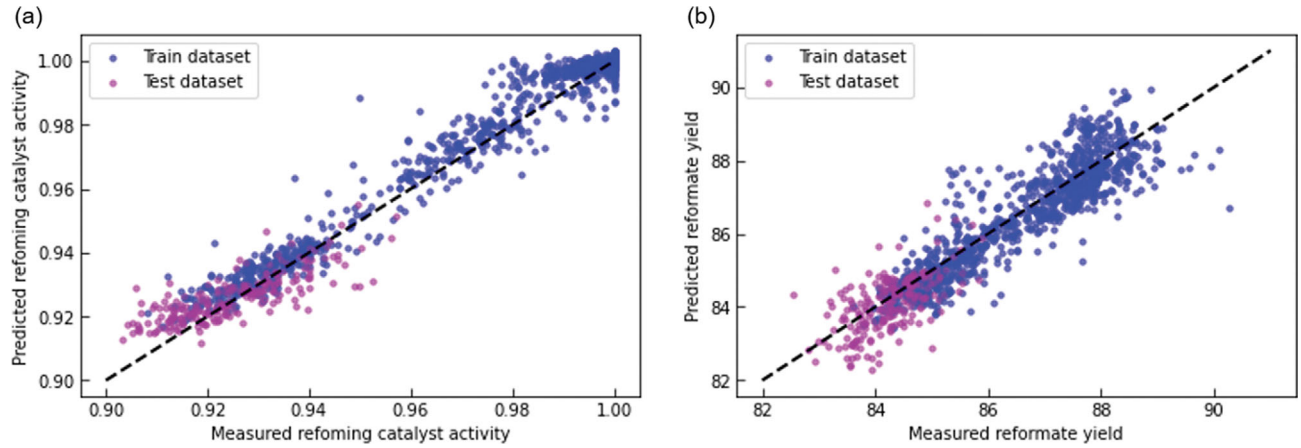

Fig. 9. Prediction results of reforming catalyst activity and reformate yield. (a) Reforming catalyst activity, and (b) Reformate yield.

this study. After that, the future reforming catalyst activity was predicted through the GRU model with the operational data profile for five days as input. Finally, the yield in the case of using the fresh catalyst under the operating conditions at the time of prediction was predicted through the MLR model, the future reformate yield was predicted by multiplying the predicted yield through the MLR model by the reforming catalyst activity obtained through the GRU model.

Fig. 9 is the result of predicting reforming catalyst activity and reformate yield using the model proposed in this study. Fig. 9(a) is the result of the predicting reforming catalyst activity through the GRU model. The prediction error was 0.0058 based on the RMSE

for the training dataset and 0.0069 for the test dataset, showing a low error, and high prediction accuracy was obtained for both the training and test dataset not used for training, which is shown that the model learned the data well without overfitting and underfitting. Fig. 9(b) is the result of predicting the reformate yield with the combination of the MLR and GRU model. The reformate yield prediction also showed high performance at 0.669 RMSE for the training dataset and 0.628 for the test dataset. It was shown that the reaction yield of the actual catalytic reforming process can be predicted with high performance through the combination of the MLR model and the GRU model proposed in this study, which can be usefully used to predict the lifetime of the reforming catalyst by predicting the future yield according to the process operation pattern.

2. Prediction of Reforming Catalyst Lifetime

Using the MLR-GRU reformate yield prediction model proposed in this study, the future reformate yield according to the process operation pattern was predicted, and the catalyst replacement time when the reformate yield decreased to less than 80% that does not satisfy economic feasibility was predicted. Fig. 10 is the reformate yield prediction result for case 1, assuming that the process operation conditions of the last day of the process data (Dec. 30, 2020) continue, and case 2, where the process operation conditions for the last 500 days are repeated. Fig. 10(a) and (b) are the reformate yield according to the number of catalyst using days and feed throughput for case 1, respectively. After 4,775 days of catalyst use and 933,000 tons of feed throughput, the reformate yield falls below 80% and the catalyst needs to be replaced.

Fig. 10(c) and (d) are the reformate yield according to the number of catalyst using days and feed throughput for case 2, and catalyst replacement should take place after 4,740 days (963,000 tons), which is similar to case 1. The reformate yield profile is greatly influenced by the process operating conditions and shows a similar shape to the profile of the change in operating conditions. In the case of an actual commercial process, since it is operated within a certain range of operation conditions, the catalyst performance degradation and replacement timing are similar.

Fig. 11 is the reformate yield prediction result for case 3, which is a case of continuously processing the maximum feed flow rate, and case 4, which continues to operate at the maximum reactor temperature among the past operation data. Fig. 11(a) and (b) are the reformate yield of case 3 according to the number of catalysts using days and the cumulative feed throughput. Case 3 has the same operating conditions as in case 1, except feed flow rate which is assumed to be 306 t/h, the maximum flow rate in the past dataset. When the feed throughput per hour is increased, the performance

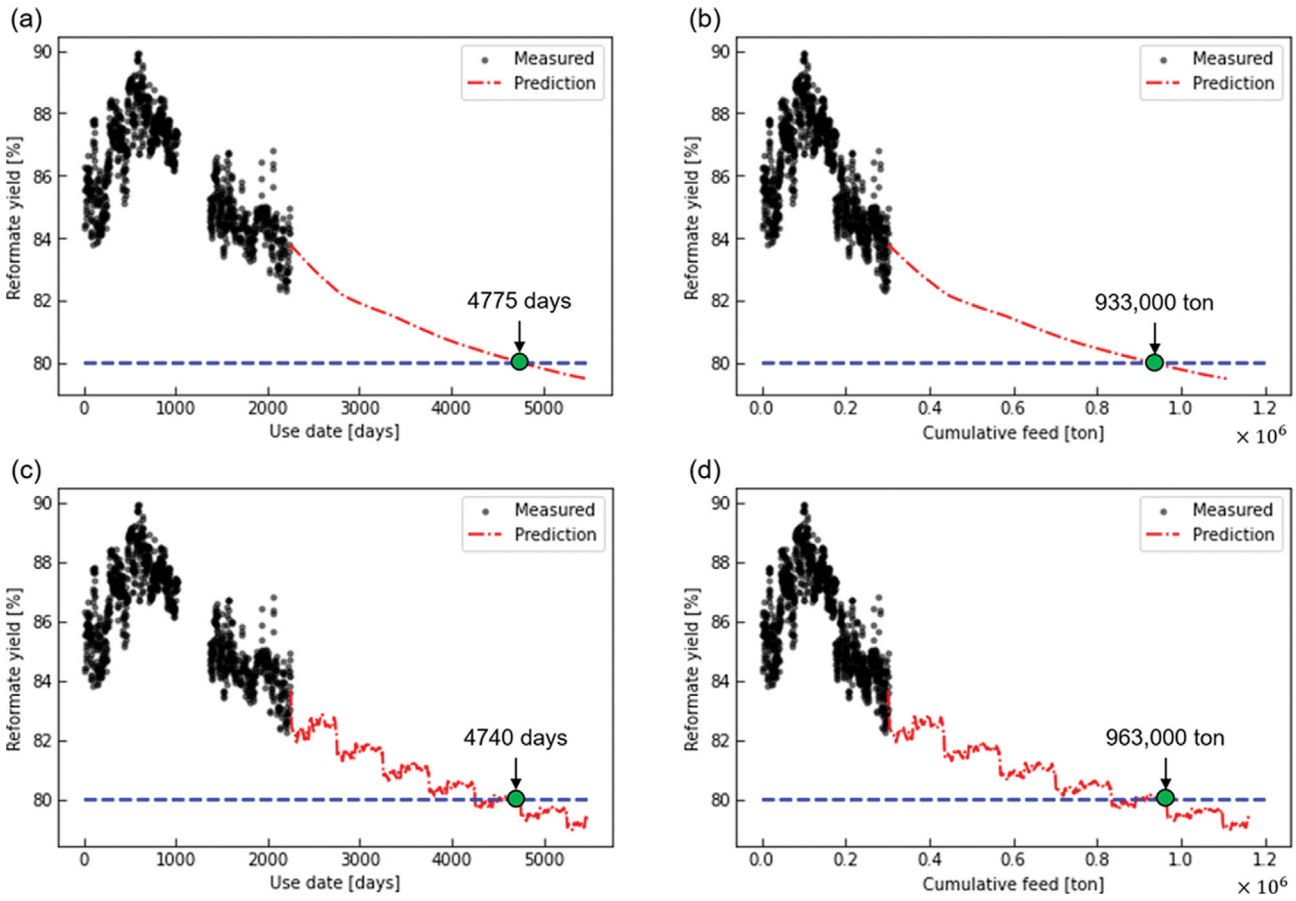

Fig. 10. Prediction results of reforming catalyst lifetime according to the use date and cumulative feed. (a) Case 1: Same as last day operating conditions - use date, (b) Case 1: Same as last day operating conditions - cumulative feed, (c) Case 2: Repeating last 500 days operating conditions - use date, and (d) Case 2: Repeating last 500 days operating conditions - cumulative feed.

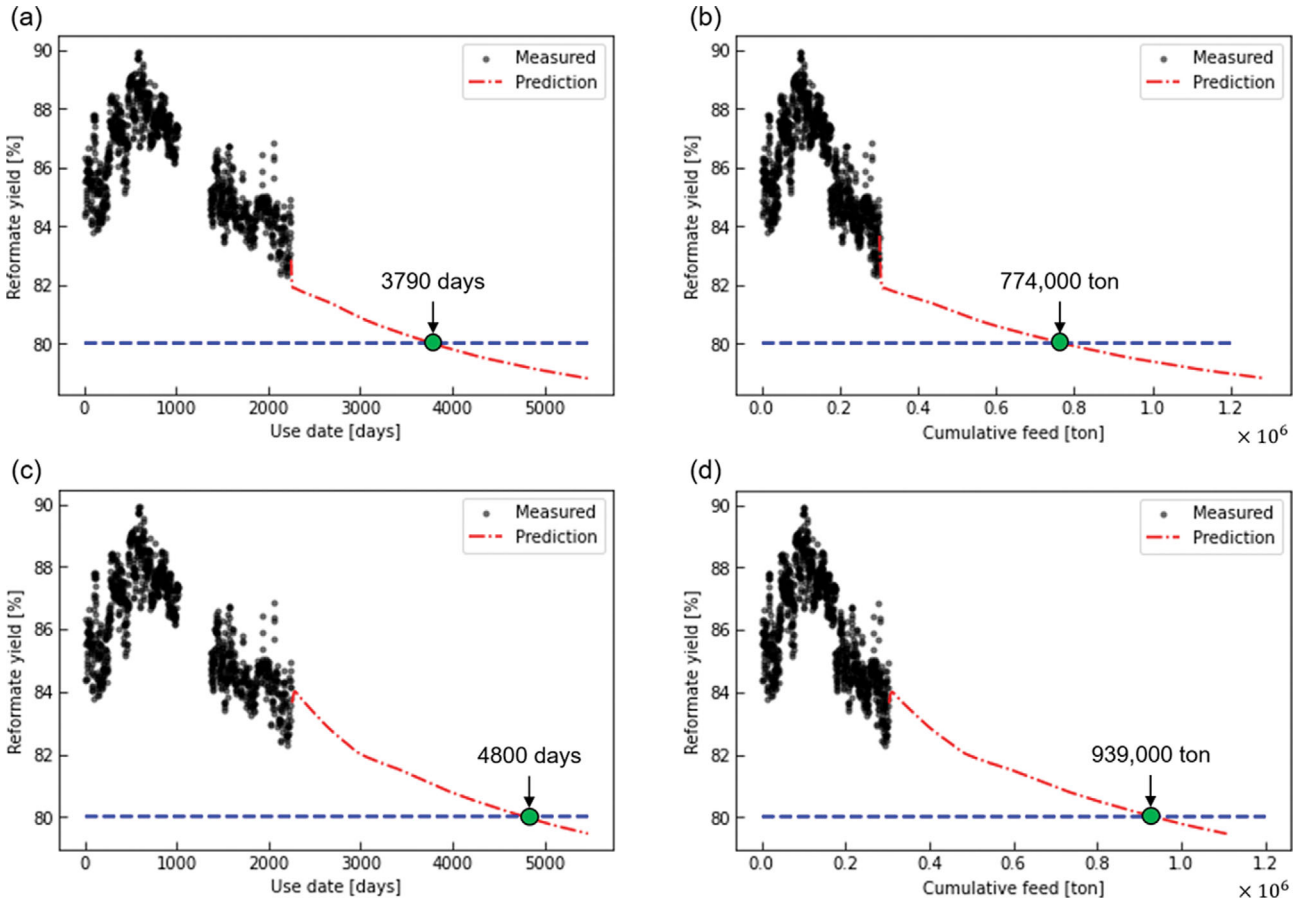

Fig. 11. Prediction results of reforming catalyst lifetime according to the use date and cumulative feed. (a) Case 3: Max feed rate operation - use date, (b) Case 3: Max feed rate operation - cumulative feed, (c) Case 4: Max temperature operation - use date, and (d) Case 4: Max temperature operation - cumulative feed.

Table 4. Prediction results of catalyst lifetime according to the operating conditions

No. Case	Description	Lifetime [day]	Lifetime [tons]
Case 1	Same as last day (2021-12-30) operating conditions	4,775	933,000
Case 2	Repeating last 500 days operating conditions	4,740	963,000
Case 3	Max feed rate (306 t/h)	3,790	774,000
Case 4	Max reactor temperature (545 °C)	4,800	939,000

degradation is faster than in other cases, and the cumulative throughput also decreases, which ultimately reduces the life of the catalyst.

Fig. 11(c) and (d) are the results of the reformate yield prediction according to the number of catalyst using days and cumulative feed throughput assuming the maximum reaction temperature among the past data, and 4,800 days (939,000 tons) were predicted as the catalyst replacement time. The results were similar to those of the existing cases 1 and 2, because there is no significant change in operating conditions in the case of actual commercial process operation. The average maximum temperature of the four reactors operated for about 6 years and 5 months was 542.6 °C, showing only about 2.7 °C (0.5%) difference from the reaction temperature of case 1, 539.9 °C. In addition, the lowest average temperature is 534.3 °C, showing a difference of about −1.04%, and when the process is operated within a certain allowable range, the change in reaction temperature is substantially insignificant; therefore, the reaction temperature does not significantly affect the catalyst performance degradation profile. On the other hand, the feed flow rate is a factor that greatly affects catalyst performance degradation, as it changes significantly to a maximum of 306 t/h (22%) and a minimum of 220 t/h (−12%) compared to the 250 t/h of case 1. The feed of the process is treated naphtha through the previous process, and the quality of the feed is also kept constant without any major change; thus, the catalyst lifetime is highly related to the flow rate of the feed. Table 4 shows the characteristics of each case, and the prediction results according to the catalyst lifetime and the cumulative feed.

As a result of predicting the catalyst lifetime according to the process operation pattern, the effect of the feed flow rate was found to be the greatest, hence the catalyst lifetime according to the feed

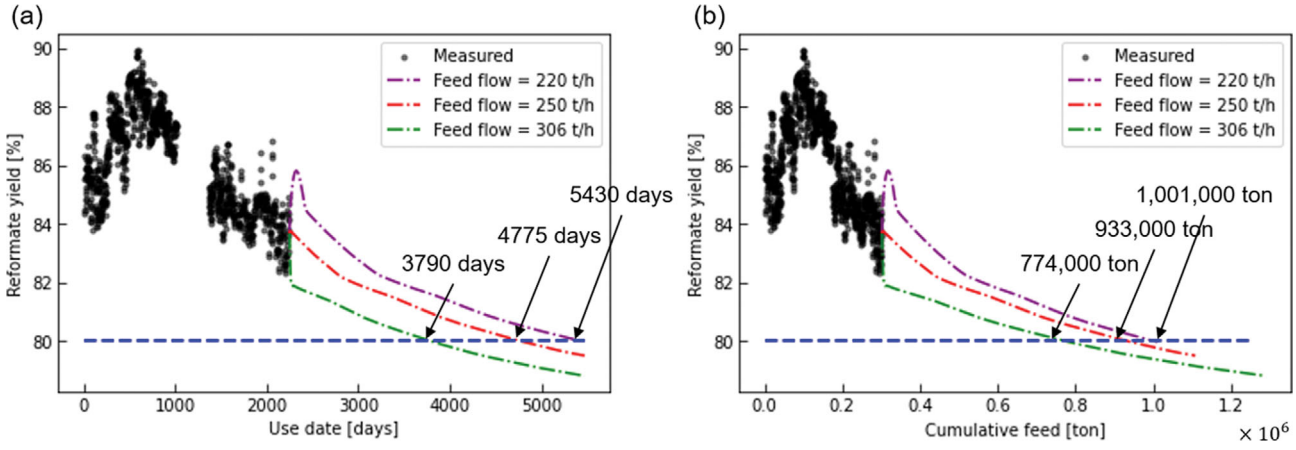

Fig. 12. Prediction results of catalyst lifetime change according to the feed flow rate. (a) Use date, and (b) Cumulative feed.

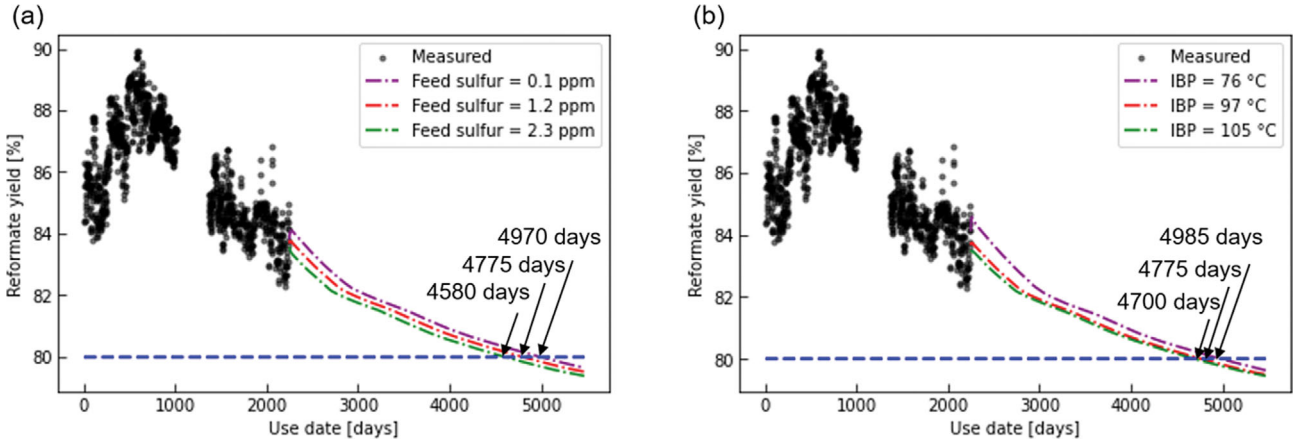

Fig. 13. Prediction results of catalyst lifetime change according to the feed flow quality. (a) Catalyst poison (Sulfur), and (b) Initial boiling point (IBP).

flow rate was analyzed. Fig. 12 is the result of predicting the lifetime according to the number of catalyst using day and the cumulative feed throughput for three cases of feed flow of 220, 250, and 360 t/h. When the hourly feed throughput of the catalyst increases, the cumulative feed throughput increases rapidly, and the catalyst lifetime is reduced from a maximum of 5,430 days to 3,790 days. The performance degradation rate increases, and the cumulative feed amount that can be processed is reduced from 1,001,000 tons to 774,000 tons. Therefore, although the low load of the catalyst can extend the catalyst lifetime, it is important to achieve the target production according to the demand.

Fig. 13 is the prediction result of catalyst lifetime according to the feed quality. Fig. 13(a) indicates the effects of concentration of sulfur, which behaves as a catalyst poison, on the catalyst lifetime. The concentration of sulfur in the feed was analyzed in the range from 0.1 to 2.3 ppm, the minimum and maximum concentration in the previous operation dataset. Absolutely, the increase of sulfur concentration induced the decrease of the catalyst lifetime from 4,970 to 4,580 days; however, the influence of the sulfur concentration was insignificant when compared to the case with various feed flow rate. Because the feed in our target process is the treated naphtha transferred from the previous process, the feed spec such as the concentration of sulfur and impurities is managed in a specific region. Therefore, the concentration of sulfur in the feed is small enough not to act as a catalyst poison, which leads to only a slight difference in catalyst lifetime. Fig. 13(b) illustrates the effects

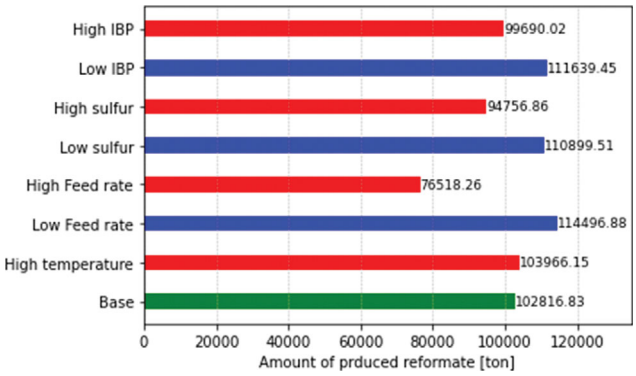

Fig. 14. Comparison of the amount of produced reformate after 2,246 days according to the various operating conditions.

Table 5. Operating conditions after end of dataset for each case

	Feed rate [t/h]	Temperature [°C]	Sulfur concentration [ppm]	IBP [°C]
Base	250	539.9	1.2	97
High temp.	250	545	1.2	97
High feed rate	306	539.9	1.2	97
Low feed rate	220	539.9	1.2	97
High sulfur	250	539.9	2.3	97
Low sulfur	250	539.9	0.1	97
High IBP	250	539.9	1.2	105
Low IBP	250	539.9	1.2	76

of initial boiling point (IBP), in the range from 76 to 105 °C, on the catalyst lifetime, which means the effect of the feed quality such as heavy or light feed. For the same reason as the concentration of sulfur, the catalyst lifetime shows an imperceptible difference in this study.

Fig. 14 is the amount of produced reformate oil from last day (2,246 days) in the dataset to catalyst lifetime of each case, according to the various operating conditions, such as distributions of temperature, feed rate, concentration of sulfur, and IBP, and the base is the profile of the last day operating conditions (case 1). In Table 5, detailed information for operating conditions in simulation cases is given. As a result, the worst is the case when the feed rate is maximum, which lowers the amount of product by 25.6%. On the other hand, the best is the case when the feed rate is minimum, which increases the amount of product, production.

CONCLUSIONS

We have proposed a model for predicting catalyst activity and reformate yield according to real-time process data and operating patterns to maximize productivity and reducing plant shutdown time by predicting the catalyst replacement in the counter-current continuous catalytic reforming. The reformate yield used to predict the degradation of the catalyst was affected more by the operating conditions than the degradation of the catalyst; the reforming catalyst activity was introduced as a new variable. A multiple linear regression model that predicts the reformate yield when using the initial fresh catalyst was proposed, and the reforming catalyst activity was calculated by the reformate yield of the fresh catalyst. Afterward, a GRU model for predicting the future reforming catalyst activity was proposed and, finally, the future reformate yield was predicted by the predicted reforming catalyst activity and the reformate yield of the fresh catalyst according to the process conditions.

The performance of the reformate yield prediction model showed high performance as RMSE of 0.628, showing that the reaction yield of the actual catalytic reforming process can be predicted with high accuracy. The catalyst replacement time that would be less than 80% of the reformate yield was analyzed by the model according to the various operating conditions such as temperature, feed rate, concentration of sulfur in the feed, and feed IBP. As a result, since the actual commercial process is operated within a certain operating range, the catalyst performance degradation and replacement timing were similar in most cases. If the hourly feed throughput of the catalyst is increased, the cumulative feed throughput increases rapidly, hence the number of days the catalyst can be used decreases. In addition, as a result of increasing the performance degradation rapidly, the cumulative feed amount that can be processed decreases. In terms of the amount of produced reformate oil, the case with maximum feed rate is similarly the worst (−25.6%), on the other hand, the case with minimum feed rate is the best (+11.4%). The results mean that the feed flow rate is the most influential variable in the commercial catalytic reforming, and it is important to establish a production plan of the produced reformate oil. Therefore, the model proposed in this study can be used for production scheduling and optimization to achieve the target production according to the demand in counter-current continuous catalytic reforming. In addition, instead of the method of determining the activity and remaining life of the catalyst through sampling, it is possible to predict the life of the catalyst in real-time according to the operation pattern through the continuous process data.

ACKNOWLEDGEMENTS

This work was partly supported by Korea Institute for Advancement of Technology (KIAT) grant funded by the Korea Government (MOTIE). (P0008475, Development Program for Smart Digital Engineering Specialist) and Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government (MOTIE) (20207200000070, Development of performance standardization and operation risk estimation for renewable energy-linked alkaline water electrolysis hydrogen production systems using digital twins). Authors are grateful for the sharing of data and ideas of Hanwha TotalEnergies Petrochemical.

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