Design of ethylene oxide production process based on adaptive design of experiments and Bayesian optimization

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Abstract

In process design, the values of design variables X for equipment and operating conditions should be optimized for entire processes, including all unit operations, such as reactors and distillation columns, to consider effects between unit operations. However, as the number of X increases, many more simulations are required to search for the optimal X values. Furthermore, multiple objective variables Y , such as yields, make the optimization problem difficult. We propose a process design method based on adaptive design of experiments and Bayesian optimization. Optimization of X values that satisfy target values of multiple Y variables are searched, and simulations for the optimized X values are then repeated. Therefore, X will be optimized by a small number of simulations. We verify the effectiveness of this method by simulating an ethylene oxide production plant.

Introduction

Process design involves the entire procedure from raw materials to products, combining multiple unit operations, such as reaction, heat exchange, and separation. In the development of new production processes, the design and performance of virtual plants are simulated. In this study, the objective is to design an efficient ethylene oxide (EO) production process.

EO is an important petrochemical product formed by the oxidation of ethylene. It is converted to various compounds such as ethylene glycol and ethanolamine, and is a starting material for fibers and detergents. EO production is about 9,000 t/y¹ in Japan. In the production process, the oxidation of ethylene is accompanied by side reactions:

Eq. (1) is the main reaction to produce EO and it requires silver catalysts (Ag_2O). Eqs. (2,3) are side reactions that produce carbon dioxide and water. The EO process has a risk of explosion because Eq. (2) has a heat reaction of +1323 kJ/mol at 523 K. Therefore, there is a restriction on reactor inlet compositions and conversions². As a result, unreacted gas is recycled, and the reactor exhibits nonlinear and complex behaviors. Here, we design the EO production process assuming that only these three reactions proceed.

Much research has been conducted to improve the performance of EO production. Zhou et al. used a reactor model to optimize design variables such as inlet compositions of ethylene, oxygen, and carbon dioxide, as well as reactor temperature and the amount of ethylene dichloride³. Lahiri et al. optimized design variables, such as inlet compositions and gas flow, using a support vector machine and a genetic algorithm for EO

recycling. They verified the results with an actual plant based on the optimized design variables, and reported improved EO production rate and catalyst selectivity⁴. Yang et al. performed modeling and validations for steady-state, dynamic, and start-up operation conditions with EO recycling. They speeded up the plant start-up with optimized start-up profiles for inlet compositions, temperature, and pressure⁵. Rahimpour et al. modeled catalyst deactivation using artificial neural networks to determine the optimal amount of ethylene dichloride⁶. Luo et al. integrated reaction kinetics, a reactor model, and a catalyst deactivation model of an industrial ethylene oxide reactor and compared the results with an actual EO plant⁷. Nawaz et al. used Aspen Custom Modeler to optimize several design variables, such as reactor temperature and inlet compositions⁸. Peschel et al. considered interactions between reaction concepts and the entire EO process, and modeled the physical and chemical principles. They reported reduced operating costs and reduced carbon dioxide emissions⁹. Distillation columns for the removal of carbon dioxide were optimized for several variables, such as temperature and pressure^{10, 11}.

Often, the values of design variables are optimized for each unit, such as a reactor and a distillation column traditionally. Hence, effects between units are not considered, and the optimized values of design variables from these methods are not always accurate when considering the entire process.

We propose a method for optimizing the entire process by considering effects between the units. Therefore, all units must be optimized simultaneously, with a very large number of design variables that need to be optimized. For example, the design variables X for the EO process, such as equipment and operating conditions, have 23 parameters. Assuming there are ten candidates for each X , the total number of combinations would be 10^{23} , which requires a huge number of simulations.

Hence, we propose a process design method based on Bayesian optimization¹² where X values that satisfy target values of multiple objective variables Y , such as yield, are searched. Then X will be optimized with a small number of simulations. We verify the effectiveness of the proposed method by simulating an EO plant.

Methods

First, the target EO process is described. Then, after the design of experiments (DoE) and the adaptive design of experiments are described, the procedure for combining these methods is discussed.

Ethylene Oxide (EO) process

The process flow diagram (PFD) of the designed EO process is shown in Figure 1. Here, the products are 99.5 wt-% EO and a 10.0 mol-% aqueous EO solution¹³. In the EO reactions, it is assumed that a silver catalyst is used. Ethylene and oxygen are supplied to a plug-flow reactor (R1) and catalyst at a high temperature and pressure, and are converted into EO. The produced EO is then absorbed by water in the absorption column (T1). As noted above, there is a risk of explosion in the reactor because of the large number of exothermic reactions; hence, there are restrictions on conversions and reactor inlet compositions. The unreacted gases are recycled and purged with raw materials, such as argon or ethane, and the carbon dioxide by-product is removed before they are supplied to the reactor again. Ethylene oxide absorbed in water is removed from the water (T2), and from light hydrocarbons (T3), and rectified in the distillation column (T4).

where $\mathbf{b}$ is a vector of regression coefficients. The prior distribution of $\mathbf{b}$ assumes a normal distribution with a zero mean and variance σ_b^2 . Then, the mean vector m_i of the $y^{(i)}$ and the covariance $\sigma_{y_i,j}^2$ of the $y^{(i)}$ and $y^{(j)}$ are calculated from:

$$m_i = E[y^{(i)}] = 0 \quad (4)$$

$$\sigma_{y_i,j}^2 = \text{cov}[y^{(i)}, y^{(j)}] = \mathbf{x}^{(i)} \mathbf{x}^{(j)T} \sigma_b^2 \quad (5)$$

The input $\mathbf{x}$ is transformed by the nonlinear function φ , and $\sigma_{y_i,j}^2$ is calculated from:

$$\sigma_{y_i,j}^2 = \varphi(\mathbf{x}^{(i)}) \varphi(\mathbf{x}^{(j)})^T \sigma_b^2 \quad (6)$$

Actual y have a measurement error. The i -th samples including the measurement error is $y_{\text{obs}}^{(i)}$ and its measurement error is $e^{(i)}$. Thus, $y_{\text{obs}}^{(i)}$ is given by:

$$y_{\text{obs}}^{(i)} = y^{(i)} + e^{(i)} \quad (7)$$

$e^{(i)}$ assumes a normal distribution with a zero mean and variance σ_e^2 , and $e^{(i)}$ is independent for each sample. Then, the covariance $\sigma_{y_{\text{obs},i,j}}^2$ between $y_{\text{obs}}^{(i)}$ and $y_{\text{obs}}^{(j)}$ is calculated as follows:

$$\sigma_{y_{\text{obs},i,j}}^2 = \varphi(\mathbf{x}^{(i)}) \varphi(\mathbf{x}^{(j)})^T \sigma_b^2 + \delta_{i,j} \sigma_e^2 = K(\mathbf{x}^{(i)}, \mathbf{x}^{(j)}) \quad (8)$$

where, K is a kernel function.

In the GPR method, if the output $\mathbf{y}_{\text{obs}} = (y_{\text{obs}}^{(1)} \dots y_{\text{obs}}^{(n)})^T$ corresponding to the past input vector $\mathbf{x}^{(1)} \dots \mathbf{x}^{(n)}$ is used as training data, the output for the new input vector $\mathbf{x}^{(n+1)}$ can be estimated as a normal distribution with mean $m(\mathbf{x}^{(n+1)})$ and variance $\sigma^2(\mathbf{x}^{(n+1)})$, and is estimated by:

$$m(\mathbf{x}^{(n+1)}) = \mathbf{k} \sum_n^{-1} \mathbf{y}_{\text{obs}} \quad (9)$$

$$\sigma^2(\mathbf{x}^{(n+1)}) = K(\mathbf{x}^{(n+1)}, \mathbf{x}^{(n+1)}) - \mathbf{k} \sum_n^{-1} \mathbf{k}^T \quad (10)$$

subject to:

$$\mathbf{k} = [K(\mathbf{x}^{(1)}, \mathbf{x}^{(n+1)}) \dots K(\mathbf{x}^{(i)}, \mathbf{x}^{(n+1)}) \dots K(\mathbf{x}^{(n)}, \mathbf{x}^{(n+1)})] \quad (11)$$

Probability in target range

For the evaluation of each candidate in Bayesian optimization, the probability P that each y is within the range ($y_{\min} \leq y \leq y_{\max}$) is calculated²⁵. It assumed that output $y^{(i)}$ is a normal distribution with mean $m(\mathbf{x}_{\text{new}}^{(i)})$ and variance $\sigma^2(\mathbf{x}_{\text{new}}^{(i)})$ when a new sample $\mathbf{x}_{\text{new}}^{(i)}$ is input into the GPR model. P is thus calculated by integrating the area of the normal distribution, as given by:

$$P(\mathbf{x}_{\text{new}}^{(i)}) = \int_{y_{\min}}^{y_{\max}} \frac{1}{\sqrt{2\pi\sigma^2(\mathbf{x}_{\text{new}}^{(i)})}} \exp\left\{-\frac{1}{2\sigma^2(\mathbf{x}_{\text{new}}^{(i)})} (y - m(\mathbf{x}_{\text{new}}^{(i)}))^2\right\} dy \quad (12)$$

Figure 2 shows the basic concept of P . When the predicted values are close to the target range of y , the probability of candidates like c_1 , which has $m(\mathbf{x})$ that are close to the target range of y , is large; and when the predicted values are far from the target range of y , the probability of candidates like c_2 with high $\sigma^2(\mathbf{x})$ is large.

Figure 2. The basic concept of probability P . Black dots is known data, red dots are unknown data, the black line is predicted values by the GPR method, and the blue lines are normal distributions for unknown data.

Proposed method

We propose a process design method based on Bayesian optimization, as shown in the outline in Figure 3. First, the EO process is designed, and one million candidates of X are generated by random numbers. Then, 50 candidates of X are selected by using D-optimal design, and the EO plant is simulated with those candidates, yielding Y values. The GPR model is constructed between X and Y with 50 samples, and P defined by Eq. (12) is calculated using output from the model. Because there are multiple Y variables, the acquisition function is the product of P for all Y variables. However, multiplying values less than one will result in smaller values; therefore, the acquisition function is the sum of $\log(P)$ values. In addition, because the target ranges of Y are different, the acquisition function is scaled by using the probabilities in the target range of the GPR model samples $[P(\mathbf{x}_{\text{train}})]$. This acquisition function P_{all} is calculated as follows:

$$P_{\text{all}}(\mathbf{x}_{\text{new}}(i)) = \log\left(\frac{P(\mathbf{x})_1 - \min(P(\mathbf{x}_{\text{train}})_1)}{\max(P(\mathbf{x}_{\text{train}})_1) - \min(P(\mathbf{x}_{\text{train}})_1)}\right) + \dots + \log\left(\frac{P(\mathbf{x})_k - \min(P(\mathbf{x}_{\text{train}})_k)}{\max(P(\mathbf{x}_{\text{train}})_k) - \min(P(\mathbf{x}_{\text{train}})_k)}\right) \quad (13)$$

where k is the number of Y . The EO plant is simulated with candidates of X having the highest values of P_{all} , and Y values are obtained. If the Y values do not achieve the targets, the GPR model is updated using simulation results, and this flow is repeated until all Y values are achieved within the targets.

Figure 3. Outline of the proposed method

Results and Discussion

Table 1 shows the design variables X and their ranges. The X were defined as 24 variables based on equipment and operating conditions, such as the flow rate, reactor specifications, and distillation column specifications. Table 2 show Y and their target ranges. The Y were defined as eight variables, such as reactor inlet composition, conversion ratio, and product limitations. The reactor inlet concentration and conversion ratio were set because of explosion limits.

Table 1. The design variables X and their ranges

X	Location	The design variables	Unit	Min	Max
X ₁	SRC1	The feed of ethylene	kmol/h	500	800
X ₂	SRC2	The feed of oxygen	kmol/h	550	850
X ₃	SRC3	The feed of methane	kmol/h	250	450
X ₄	R1	The number of tubes	-	500	900
X ₅	R1	Reactor length	m	20.0	60.0
X ₆	R1	Reactor temperature	K	498	523
X ₇	SRC4	The feed of water	kmol/h	1.00×10 ³	2.00×10 ³
X ₈	T1	The number of stages	-	13	15
X ₉	SP1	The rate of separation	-	0.00100	0.0500
X ₁₀	SP2	The rate of separation	-	0.00100	0.500
X ₁₁	T2	Pressure	bar	0.100	0.900
X ₁₂	T2	The number of stages	-	13	15
X ₁₃	T2	The number of feed stages	-	10	13
X ₁₄	T2	Bottom reflux ratio	-	0.0500	1.50
X ₁₅	T3	Pressure	bar	3.00	7.00
X ₁₆	T3	The number of stages	-	13	15
X ₁₇	T3	The number of feed stages	-	10	13
X ₁₈	T3	Bottom reflux ratio	-	0.500	1.50
X ₁₉	T3	Vapor fraction	-	0.100	0.700

X	Location	The design variables	Unit	Min	Max
X ₂₀	T4	Pressure	bar	0.100	0.900
X ₂₁	T4	The number of stages	-	13	15
X ₂₂	T4	The number of feed stages	-	10	13
X ₂₃	T4	Bottom reflux ratio	-	0.100	1.00
X ₂₄	T4	Top reflux ratio	-	0.500	2.00

Table 2. The objective variables **Y** and their target ranges

Y	Location	The design variables	Unit	Min	Max
Y ₁	S5	Reactor inlet concentration of ethylene	-	0.260	0.280
Y ₂	S5	Reactor inlet concentration of oxygen	-	0.0700	0.0800
Y ₃	S5	Reactor inlet concentration of CO ₂	-	0.0300	0.0600
Y ₄	R1	Conversion ratio of ethylene	-	0.0500	0.100
Y ₅	R1	Conversion ratio of oxygen	-	0.100	0.500
Y ₆	S36	The product 2 flow of EO	kmol/h	20.0	40.0
Y ₇	S43	The product 1 flow of EO	kg/h	4.38×10 ³	5.00×10 ³
Y ₈	S43	The product 1 concentration of EO	-	0.995	1.00

We used SimCentral²⁶ simulation software. Robustness is important when the results are obtained by inputting random data into the process simulator. With SimCentral, we receive an error flag and can discard the results when the solution is divergent and cannot be obtained. In an iterative process, a sequential simulator takes a long time to converge; but a convergent solution can be obtained in a short time with SimCentral. In addition, X can be set as needed in ADoE because the specifications can be freely changed. Furthermore, SimCentral can be performed using JavaScript and can be automated by association with Python programs, which means that the proposed method can be easily combined with SimCentral.

We succeeded in searching for X that achieved target ranges of Y using 139 simulations (D-optimal design required 50, and Bayesian optimization required 89). As a comparison method, X candidates were selected in a random search. In this case, the target ranges of the Y could not be achieved at all. Table 3 shows the result of 139 simulations, and Figure 4 shows the behavior of Y for 139 simulations. In Bayesian optimization, the number of samples within the target range of Y increased with the number of trials. The accuracy of the GP model improved as the number of samples increased with the number of trials. The Y₈ result, which has a narrow range, has a low probability and is not considered by P_{all} . Therefore, it was possible to treat each variable equally by performing range scaling, as shown in Eq (13).

Table 3. The results of Y that achieved targets

Y	Unit	Min	Max	Result 1
Y ₁	-	0.260	0.280	0.267
Y ₂	-	0.0700	0.0800	0.0710
Y ₃	-	0.0300	0.0600	0.0395
Y ₄	-	0.0500	0.100	0.0682
Y ₅	-	0.100	0.500	0.286
Y ₆	kmol/h	20.0	40.0	35.7
Y ₇	kg/h	4.38×10 ³	5.00×10 ³	4.87×10 ³
Y ₈	-	0.995	1.00	0.996

Y_1 (b) Y_2

(c) Y_3 (d) Y_4

(e) Y_5 (f) Y_6

(g) Y_7 (h) Y_8

Figure 4. Y values for 139 simulations. The blue circles indicate the Bayesian optimization results, the gray triangles indicate the random search results, the asterisks indicate the Bayesian optimization results that achieved the target ranges of Y, and the red lines indicate the target ranges.

Bayesian optimization depends on the initial samples because it starts with a small number. Therefore, we changed the random value of D-optimal design, and performed Bayesian optimization again. The result is shown in Figure 5. In 83 simulations (D-optimal design took 50, and Bayesian optimization took 33), we succeeded in searching for X values that achieved target ranges of Y. These X were the same as those before the random values were changed, indicating that the results of process design based on ADoE and Bayesian optimization were stable. A comparison of the results before and after changing the random values shows similar behavior, but the target was achieved faster after the change. The number of simulations decreased because there was a sample that could easily achieve the target in the initial samples. From the results shown in Figure 5, the reproducibility of the proposed method was confirmed.

We changed the candidates of X to one million samples and compared the results of the proposed method. Tables (4, 5) list the Y and X results, respectively. In the second set of results, 77 simulations (50 for D-optimal design, and 27 for Bayesian optimization) succeeded in searching for X that achieved the target ranges of Y. Similarly, we succeeded in the third set of results with 105 simulations (50 for D-optimal design, and 55 for Bayesian optimization). In Table 4, values of Y converged to relatively similar solutions. However,

in Table 5, the values of X were dissimilar. In actuality, the final X values will be selected on the basis of cost comparisons or chemical engineering knowledge.

Y_1 (b) Y_2

(c) Y_3 (d) Y_4

(e) Y_5 (f) Y_6

(g) Y_7 (h) Y_8

Figure 5. Y values for 83 simulations. The blue circles indicate the Bayesian optimization results before changing the random values of D-optimal design, the orange triangles indicate the Bayesian optimization results after changing the random values of D-optimal design, the asterisks indicate the Bayesian optimization results that achieved the target ranges of Y , and the red lines indicate the target ranges.

Table 4. Comparison of Y results

Y	Unit	Min	Max	Result_1	Result_2	Result_3
Y_1	-	0.260	0.280	0.267	0.276	0.277
Y_2	-	0.0700	0.0800	0.0710	0.0708	0.0732
Y_3	-	0.0300	0.0600	0.0395	0.0430	0.0438
Y_4	-	0.0500	0.100	0.0682	0.0589	0.0641
Y_5	-	0.100	0.500	0.286	0.260	0.274
Y_6	kmol/h	20.0	40.0	35.7	30.5	34.3
Y_7	kg/h	4.38×10^3	5.00×10^3	4.87×10^3	4.49×10^3	4.39×10^3
Y_8	-	0.995	1.00	0.996	0.995	0.995

Table 5. Comparison of X results

X	Unit	Min	Max	Result_1	Result_2	Result_3
X ₁	kmol/h	500	800	708	787	761
X ₂	kmol/h	550	850	727	736	808
X ₃	kmol/h	250	450	349	336	386
X ₄	-	500	900	829	581	778
X ₅	m	20.0	60.0	47.8	58.0	53.4
X ₆	K	498	523	517	519	518.3
X ₇	kmol/h	1.00×10 ³	2.00×10 ³	1.20×10 ³	1.01×10 ³	1.21×10 ³
X ₈	-	13	15	13	13	14
X ₉	-	0.00100	0.0500	0.0202	0.0423	0.0248
X ₁₀	-	0.00100	0.500	0.0662	0.0360	0.0512
X ₁₁	bar	0.100	0.900	0.400	0.270	0.241
X ₁₂	-	13	15	13	13	14
X ₁₃	-	10	13	10	12	11
X ₁₄	-	0.0500	1.50	0.159	0.359	0.133
X ₁₅	bar	3.00	7.00	5.42	3.97	4.306
X ₁₆	-	13	15	13	13	13
X ₁₇	-	10	13	11	12	11
X ₁₈	-	0.500	1.50	1.03	0.760	1.35
X ₁₉	-	0.100	0.700	0.131	0.108	0.132
X ₂₀	bar	0.100	0.900	0.301	0.394	0.336
X ₂₁	-	13	15	13	13	14
X ₂₂	-	10	13	11	12	10
X ₂₃	-	0.100	1.00	0.798	0.384	0.850
X ₂₄	-	0.500	2.00	0.911	0.937	1.28

Conclusion

We proposed a process design method that combines Bayesian optimization and a process simulator to search for design variables that satisfy the performances of an ethylene oxide plant with a small number of simulations. We verified the effectiveness of the method by comparing it with a random search. It was confirmed by the case study that the candidates for design variables that achieve the plant performances can be efficiently proposed. The reproducibility of the proposed method was also confirmed. Moreover, various candidates were obtained by increasing the number of Bayesian optimization trials. This method is expected to meet the needs of knowledgeable chemical engineers and to facilitate process designs of new plants.

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List of Figure Captions

Figure 1. PFD of the EO process.

Figure 2. The basic concept of P. Black dots is known data, red dots are unknown data, black line is predicted values by GPR method, blue lines are normal distribution for unknown data.

Figure 3. The outline of the proposed method

Figure 4. Y values for 139 times simulation. The blue points indicate the Bayesian optimization results, the gray points indicate the random search results, the asterisks indicate the Bayesian optimization result achieved the target ranges of Y, and the red lines indicate the target range.

Figure 5. Y values for 83 times simulation. The blue points indicate the Bayesian optimization results before changing the random value of D-optimal design, the orange points indicate the Bayesian optimization results after changing the random value of D-optimal design, the asterisks indicate the Bayesian optimization result achieved the target ranges of Y, and the red lines indicate the target range.