Real-Time Optimization of a CO Preferential Oxidation Reactor Temperature with Extremum Seeking Control Techniques
Jea Pil Heo, Su Whan Sung, and Jietae Lee*

ABSTRACT: A hydrogen (H2)-rich gas mixture is used as the fuel of the proton exchange membrane fuel cell (PEMFC). A small amount of the carbon monoxide (CO) gas in the gas mixture can significantly deactivate the catalyst of the PEMFC, resulting in a reduction in the efficiency of power generation. Preferential oxidation is used to reduce the CO concentration less than 10 ppm in the gas mixture. It has an optimal reaction temperature at which the reaction shows the minimum exit CO concentration with minimum consumption of H2. This optimal temperature continuously changes under the varying conditions of operation and catalyst deactivation. In this study, two modified extremum seeking control (ESC) methods were proposed to continuously seek and maintain this optimal reaction temperature, guaranteeing CO concentration under 10 ppm even under time-varying conditions. The proposed methods have a smaller number of design parameters than the conventional extremum seeking approaches so that tuning the ESC method is much easier, more intuitive, and efficient. In addition, the proposed method can additionally use the secondary measurement to improve the performance of the ESC method by removing the possibility that the modeling error of the linear dynamic block can deteriorate the accuracy of calculating the gradient of the nonlinear static block. The experimental results confirmed that the proposed methods can track the optimal temperature within a short time compared to the conventional approach while successfully maintaining the CO concentration below 10 ppm.

INTRODUCTION

Hydrocarbon reforming is a method for H2 production to provide fuel for fuel cell power generation systems.1 Methanol (CH3OH) reforming in particular is a very promising process to provide hydrogen for proton exchange membrane fuel cells (PEMFCs) used in portable devices because methanol is liquid at room temperature, and its reforming temperature is much lower than that of natural gas reforming, which offers easy storage and manipulation.2 The hydrogen-rich gas mixture produced by methanol reforming typically contains a small amount of CO that can poison the anode catalyst of the PEMFC, resulting in significant degradation in the power efficiency. Thus, it is very important to maintain the CO concentration below 10 ppm.1–6

CO preferential oxidation (CO PROX) is used to maintain a low level of CO by oxidizing CO to CO2, which is inert to the anode catalyst. However, it suffers from the side reaction of H2 oxidation that consumes H2, which is the fuel of the fuel cell. The gas reactions are given as follows:

\[
\begin{align*}
\text{CO} + 0.5 \text{O}_2 & \rightarrow \text{CO}_2 \\
\text{H}_2 + 0.5 \text{O}_2 & \rightarrow \text{H}_2\text{O}
\end{align*}
\]

Hence, it is required to oxidize CO while suppressing the H2 oxidation as much as possible. One of the catalysts that have high selectivity for the CO oxidation over the H2 oxidation is a mixed oxide of copper and cerium (CuO-CeO2).4–6 Operation at a limited oxygen feed flow rate can also contribute to suppressing the H2 oxidation. However, using the high-selectivity catalyst under these preferable operating conditions is not enough to maintain the optimal reaction conditions in real application.

Note that the PROX reaction has an optimal reaction temperature at which the reaction shows the minimum exit CO concentration with minimum H2 consumption, and the optimal temperature is continuously changing as a result of the catalyst deactivation and changes in the operating conditions. Thus, it is very important to continuously track this optimal reaction temperature and maintain the PROX reaction within the optimal reaction temperature window,7 which ensure the CO concentration below 10 ppm even under time-varying conditions. To achieve these control objectives, Lee et al.7 have obtained a Wiener-type nonlinear dynamic model composed of a linear dynamic block and a nonlinear static block for a real
PROX reactor. They then designed a conventional extremum seeking control (ESC) method based on this Wiener-type model and applied it to the chosen real PROX reactor. The conventional ESC method was successful in finding and maintaining the optimal reaction temperature of the CO PROX reactor. Meanwhile, the conventional feedback control systems cannot achieve these results because they cannot manipulate the input multiplicity.9 Several theoretical works10,11 and applications12 of the ESC methods are available.

In this study, two modified ESC methods were proposed to improve the performance of the conventional ESC method. It was successfully demonstrated that the proposed methods are superior to the conventional one through their application in a real PROX reactor. The proposed methods have a smaller number of design parameters than the conventional extremum seeking approach, and hence, they offer a simpler, more intuitive, and more efficient design of the ESC. In addition, the proposed method utilizes a secondary measurement to improve the performance of the ESC by removing the possibility that the modeling error of the linear dynamic block can deteriorate the accuracy of calculating the gradient of the nonlinear static block. Experiments were conducted to confirm that the proposed methods can track the optimal temperature faster than the conventional approach and successfully maintain the CO concentration below 10 ppm.

THE CO PROX REACTOR SYSTEM AND A WIENER-TYPE MODEL

CO PROX Reactor System. Figure 1 shows the schematic diagram of the experimental CO PROX reactor used. Table 1 shows the specifications of the CO PROX reactor system. The system input of the CO PROX reactor system is the heating signal of $u$, and the system outputs are the reaction temperature of $T$ and the CO concentration of $c_{CO}$. The control objective is to seek and maintain the optimal reaction temperature corresponding to the minimum CO concentration by manipulating the input signal of $u$ in an automatic way. Refer to Lee et al.8 and Heo16 for details.

Wiener-Type Model. Lee et al.8 obtained the following simplified dynamic model in the form of Wiener-type nonlinear dynamics from the input signal of $u$ to the reactor temperature of $T$ and the exit CO concentration of $c_{CO}$.

$$\tau_T \frac{dT(t)}{dt} = -T(t) + T_{bias} + k_T u(t - \theta_M)$$

(3)

$$c_{CO}(t) = q(T(t))$$

(4)

where $\tau_T$, $k_T$, and $T_{bias}$ are the time constant, steady-state gain, time delay, and the bias of the linear dynamic block, respectively, and the function $q$ is the nonlinear static block that defines the relationship from the reactor temperature to the CO concentration. The model parameters $\tau_T = 600 \text{ s}$, $T_{bias} = 50.4 \ ^\circ\text{C}$, $k_T = 37.9 \ ^\circ\text{C/V}$, and $\theta_M = 23 \text{ s}$ were estimated empirically by fitting the model parameters to the process data graphically, and they were then used to design the control system, giving the transfer function from the input signal to the reactor temperature as

$$G_T(s) = \frac{37.9e^{-23s}}{600s + 1}$$

(5)

The nonlinear static function from the reactor temperature to the exit CO concentration was estimated by a polynomial fitting as follows:

$$c_{CO} = q(T) = 2.4892 + 0.13534(T - 160) + 0.15272(T - 160)^2$$

(6)

The Wiener-type model given in eqs 3–6 and Figure 2 accurately describes the dynamics of the CO PROX reaction as shown in the study by Lee et al.8

The good fitting of the CO PROX reactor dynamics to the Wiener-type model was justified as follows: in the CO PROX

![Figure 1. Schematic diagram of the experimental CO PROX reactor.](image)

| Table 1. Specifications of the CO PROX Reactor System |
|---------------------------------------------|
| Specification | Description |
| feed | CO (0.5 mL/min), O₂ (0.5 mL/min, 1 mL/min), H₂ (90 mL/min), CO₂ (10 mL/min), and N₂ (400 mL/min, 900 mL/min) |
| reactor size | 1/4 in. aluminum tube (6.35 mm OD, 4.8 mm ID) |
| catalyst | 1 g of CuO-CeO₂ catalyst (0.15–0.18 mm φ in size) |
| heating | heating band around the aluminum tube (input signal u: 0–5 V) |
| reactor temperature range | 25–250 °C |
| reactor temperature measurement | K-type thermocouple |
| CO concentration measurement | infrared (IR) gas analyzer (Teledyne model 7500) |
| data acquisition system | National Instruments data acquisition system (model NI cDAQ-9172) with several modules including a high-resolution thermocouple module (National Instruments model NI 9211) for detecting temperature and an analog output module (National Instruments model NI 9263) |
| control software | LabView (National Instruments) |
| reactor size | 1/4 in. aluminum tube (6.35 mm OD, 4.8 mm ID) |
reactor used, it can be assumed that the reactor tube and catalyst are at the same temperature (isothermal conditions) because the tubular reactor is made of a thin-walled highly thermally conductive aluminum tube. Under these assumed conditions, the energy balance equation of the reactor temperature becomes

\[ mC_p \frac{dT}{dt} = -Q_G - Q_E - Q_C + Q_R + P_i u(t) \]  

(7)

where \( mC_p \) is the heat capacity of the reactor. \( Q_G \), \( Q_E \), \( Q_C \), and \( Q_R \) are the heat flux due to the gas flow, the convective heat flux to the environment, the conductive heat flux through the reactor connections, and the heat generation by the reactions, respectively. \( P_i u(t) \) is the heat added to the reactor through the heater. \( Q_G \), \( Q_E \), and \( Q_C \) are linear functions of the reactor temperature \( T \). \( Q_R \) is the integration of the heat generated by the CO and \( H_2 \) oxidations along the reactor. It is a weak function of the reactor temperature and can be approximated to a constant. \(^8\) Thus, the model structure of eq 7 will be the same as those of eqs 3 and 4. The static nonlinear relationship in eqs 3 and 4 can be justified by the assumption that the concentrations are in a pseudo-steady state for the given reactor temperature because of the high activity of the catalyst.

### Extremum Seeking Control

ESC methods are used to track the optimal reactor temperature in real time.\(^{10-12}\) They first calculate the gradient between the input and the output with a small continuous perturbation signal, and then, they find the optimum point corresponding to the minimum or maximum output by reducing the gradient to zero using an integral controller.

#### Gradient Estimation

Consider a static nonlinear system

\[ y = f(x) \]  

(8)

ESC methods\(^{10-12}\) based on continuous perturbations and averaging are used to find the maximum point at which \( dy/dx = f'(x) = 0 \) at the steady state. To find the maximum point, the ESC methods use a continuous perturbation given as follows

\[ x(t) = \bar{x} + \delta \sin \left( \frac{2\pi t}{p} \right) \]  

(9)

where \( \delta \) and \( p \) are the perturbation size and period, respectively. For an analytic function \( f(x) \) and a constant \( \bar{x} \), its Taylor series is

\[ y(t) = f(x(t)) = f(\bar{x}) + f'(\bar{x}) \delta \sin \left( \frac{2\pi t}{p} \right) + \frac{1}{2} f''(\bar{x}) \delta^2 \sin \left( \frac{2\pi t}{p} \right) + \cdots \]  

(10)

Then, the average value of \( y(t) \sin(2\pi t/p) \) becomes

\[ \xi_1 = \frac{1}{p} \int_{-p}^{p} y(t) \sin \left( \frac{2\pi t}{p} \right) dt = \frac{\delta}{2} f''(\bar{x}) + O(\delta^3) \approx \frac{\delta}{2} \frac{dy}{dx} \]  

(11)

Thus, the gradient \( f'(x) \) can be directly calculated based on the average value of eq 11 estimated by numerical integration to be used in the ESC method. Alternatively, eq 12 can be used to calculate \( f'(x) \) as follows:

\[ \xi_2 = \frac{p}{2} \int_{-p}^{p} x(t) y(t) dt - \frac{1}{2} \int_{-p}^{p} x(t)^2 dt \left( \int_{-p}^{p} x(t) dt \right)^2 = \frac{1}{2} \delta^2 p^2 f'(\bar{x}) + O(\delta^4) \approx \frac{1}{2} \delta^2 p^2 f'(\bar{x}) + O(\delta^2) \approx \frac{dy}{dx} \]  

(12)

If \( f(x) \) is close to quadratic and \( \bar{x} \) is the optimum point corresponding to \( f'(\bar{x}) = 0 \) at which the frequency of the output response \( y(x(t)) \) will be doubled as shown in Figure 3.

![Figure 3. Input and output at \( \xi_1 = 0 \) or \( \xi_2 = 0 \).](image)

\[ \xi_1 = 0 \text{ or } \xi_2 = 0. \]  

Thus, the optimum point is equal to the point corresponding to \( \xi_1 = 0 \) or \( \xi_2 = 0 \). The ESC method tracks the optimum point by controlling the feedback signal of \( \xi_1 \) or \( \xi_2 \) to be zero using an integral controller.

**Conventional ESC.** Because the conventional ESC method finds the optimum point corresponding to the maximum output, the following process output is defined so that the ESC method would seek the optimum point corresponding to the minimum CO concentration.

\[ y(t) = 1 - c_{CO}(t)/1000 \]  

(13)

Although the ESC method is derived for the static process, it can be applied to nonlinear dynamic processes. Figure 4 shows the conventional ESC system applied to the CO PROX reactor system. Its main elements are given as follows:

\[ u(t) = \bar{u} + a \sin \left( \frac{2\pi t}{p} \right) \]  

(14)

\[ \bar{u} = k_i \int_{0}^{t} y(t) \sin \left( \frac{2\pi t}{p} \right) dt \]  

(15)

The integral controllers of eqs 14 and 15 are supposed to derive \( \bar{u} \) to the optimum point corresponding to \( dy/dx = 0 \). The parameter \( \phi \) is used to compensate for the phase lag of the linear dynamics of \( G(s) \).

To achieve closed-loop stability, the period \( p \) of the continuous perturbation signal should be large enough to ensure that the timescale of the process dynamics is negligible compared to that of the real-time optimization by the ESC method. To improve the transient responses, the high-pass filter (HPF(s) = \( \tau_{HP}/(\tau_{HP} + 1) \)) and low-pass filter (LPF(s) = \( 1/(\tau_{LP} + 1) \)) in Figure 4 are typically included. The high-pass
The approximate values of these parameters can be determined via simulations. A square wave instead of the sinusoidal signal is used instead of the high-pass and low-pass filters developed by Haring et al. Conventional ESC system for the CO PROX reactor. Figure 4. Conventional ESC system for the CO PROX reactor.

Figure 5. Proposed discrete-time ESC system.
al.\textsuperscript{15} By implementing these integrations at each sampling time of the perturbation period, the whole data of $y(t)$ between $t - p$ and $t$ are not required. The adverse effects of the previously mentioned high- and low-pass filters that deteriorate the transient responses while searching for the optimum point can be avoided in the proposed discrete-time ESC method.\textsuperscript{16}

In the conventional ESC, the phase lag $\phi$ is used to compute the feedback signal $\xi_2$ in the form of $y(t) \sin (2 \pi t / p - \phi)$. It means that the parameter $\phi$ directly affects the magnitude of the feedback signal, and the performances of the conventional ESC method can be very poor if the estimation of the parameter $\phi$ is not good.\textsuperscript{13} Meanwhile, the phase lag $\phi$ is used to determine only the sign of the feedback signal $\xi_2$ in the form of $\text{sgn} (b_i \cos \phi - b_k \sin \phi)$ so that it does not affect the magnitude of the feedback signal, resulting in the proposed ESC method being more robust to the modeling error of $\phi$ than the conventional ESC method.

**ESC with a Secondary Measurement.** Because the reactor temperature of the PROX reactor is measurable, it can be directly used to calculate the derivative of $dy/dT$ at the steady state. Because the process output $y$ is a static function of the reactor temperature $T$, eq 12 with $x = T$ can be used to estimate the derivative $dy/dT$ as follows:

$$\xi_2 = \frac{p \int_{-p}^{t} T(i)y(i)di - \int_{-p}^{t} T(i)di \int_{-p}^{t} y(i)di}{p \int_{-p}^{t} T(i)^2 di - (\int_{-p}^{t} T(i)di)^2} \approx \frac{dy}{dT}$$

(27)

It contains the derivative information of $dy/du$. Based on this, the proposed ESC method with a secondary measurement of $T$ can be obtained as follows:

$$u(t) = \bar{u} + a \sin \left(\frac{2 \pi s}{p} t\right)$$

(28)

$$\bar{u} = \frac{k_i}{p} \sum b_k$$

(29)

$$b_r = \int_{-p}^{t} T(i)di$$, $$b_y = \int_{-p}^{t} y(i)di$$

(30)

$$b_{ry} = \int_{-p}^{t} T(i)y(i)di$$, $$b_{T} = \int_{-p}^{t} T(i)^2 di$$

(31)

$$b_k = \frac{ph_{iy} - b_r b_y}{ph_{iT} - b_T^2}$$, $t = kp$

(32)

Figure 6 shows the block diagram of the proposed ESC method with a secondary measurement. It has only three parameters $p$, $a$, and $k_i$, and they can be tuned an intuitive way. Also, it is free from the adverse effects of the modeling error of the phase lag parameter. This method shares all the merits of the discrete-time ESC method.

### RESULTS AND DISCUSSION

The CO PROX reactor eliminates CO from the hydrogen-rich gas mixtures by oxidizing CO. This treated gas can be used as a fuel suitable for the PEM fuel cell because it does not deactivate the anode catalyst. The exit CO concentration should be less than 10 ppm. Thus, the ESC was applied to maintain the minimum CO concentration (preferably below 10 ppm). Since catalysts are gradually deactivated, ESC methods can extend the lifetime of the CO PROX reactor compared to the case in which the operation is performed at a fixed reactor temperature. To track and maintain a minimum CO concentration, the ESC\textsuperscript{10–12} can be used. The ESC adds a continuous excitation at the range of below 10 ppm is allowed in the CO concentration.

**Conventional ESC.** A CO PROX reactor system that has the following feed rates of CO (0.5 mL/min), O$_2$ (0.5 mL/min), H$_2$ (90 mL/min), CO$_2$ (10 mL/min), and N$_2$ (400 mL/min) was utilized first. The conventional ESC method with $\phi = 0$, $a = 1$, $k_i = 0.15$, $t_L = 5$ s, and $t_{RL} = 10$ s is applied to this reactor. The approximate values of these ESC parameters are determined from simulations using the ESC model shown in Figure 2.\textsuperscript{8} The process input $u$ is limited to the range of 1.5–4.9. For this CO PROX system, it is known that the optimal reaction temperature is around 160 °C. The ESC is enabled when the reaction temperature is within the range of 120–200 °C; otherwise, the ESC is disabled by setting $\bar{u}$ to 3.

Figure 7 shows the experimental results of the first three runs.\textsuperscript{16} For all the three initial reactor temperatures, this ESC method found and maintained the optimal reactor temperature. However, compared to a process time constant of 600 s, their convergence rates were rather slow. This can be attributed to the mismatch in the phase parameter $\phi$. 

Figure 6. Proposed ESC system with a secondary measurement.

Figure 7. Experimental results of the conventional ESC method (run 1: $p = 125.66$ s, run 2: $p = 125.66$ s, and run 3: $p = 62.83$ s).
Figure 8 shows the experimental results after changing the phase parameter $\phi$ from 0 to 1.57. Other ESC parameters were $a = 1$, $p = 62.83$ s, $\tau_L = 5$ s, and $\tau_H = 10$ s. The parameter $\phi$ was set to $\pi/2$ ($\phi = 1.57$) because the time constant of the CO PROX reactor was very large compared to the perturbation period $p$. Here, the N$_2$ flow rate was changed from 400 to 900 mL/min. As in the response of run 4, the convergence rate was increased. However, the exit CO concentration seems too high. Thus, in run 5, the feed concentration of O$_2$ was increased from 0.5 to 1 mol/L. With this increment of the O$_2$ flow rate, the optimal exit CO concentration close to zero is obtained. It was very tedious and difficult to tune the control parameters of the conventional ESC because the number of parameters is too many, and they show significantly coupled behaviors.

**Discrete-Time ESC.** For a CO PROX reactor system with the following feed flow rates was used: CO (0.5 mL/min), O$_2$ (1 mL/min), H$_2$ (90 mL/min), CO$_2$ (10 mL/min), and N$_2$ (900 mL/min). The proposed discrete-time ESC method with $\phi = 1.57$, $p = 100$ s, $a = 0.5$, and $k_i = 0.01$ was applied. Figure 9 shows the experimental results of the proposed discrete-time ESC method. These results indicate that the discrete-time ESC method controls and maintains the CO concentration at near zero, and the convergence rate to the minimum CO concentration is faster than that of the conventional ESC. Furthermore, the tuning was easier and more intuitive than that in the conventional ESC.

**ESC with a Secondary Measurement.** The proposed ESC method with a secondary measurement is applied to a CO PROX reactor system with the following feed flow rates: CO (0.5 mL/min), O$_2$ (1 mL/min), H$_2$ (90 mL/min), CO$_2$ (10 mL/min), and N$_2$ (900 mL/min). Here, the design parameters were $p = 100$ s, $a = 0.3$, and $k_i = 0.01$. Figure 10 shows the experimental results of the proposed ESC method with a secondary measurement for the CO PROX reactor.

![Figure 8](image1.png)  
![Figure 9](image2.png)  
![Figure 10](image3.png)

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**CONCLUSIONS**

A CO PROX reactor made of an aluminum tube with CuO-CeO$_2$ catalysts can be used to reduce the CO level in the hydrogen-rich gas produced by hydrocarbon steam reforming. The dynamics between the exit CO concentration and the heater input signal has been well described by the Wiener-type nonlinear process where a linear dynamic block is followed by a static nonlinear block. The exit CO concentration can be minimized by controlling the reactor temperature.

ESC methods can be used to track and maintain the optimal heater input or the optimal reactor temperature. The conventional ESC method has too many design parameters. It is not easy to understand the effects of the parameters on the closed-loop dynamics, making the tuning very confusing and difficult. Also, the modeling errors of the phase lag can degrade the accuracy of the gradient and significantly deteriorate the convergence rate to the minimum CO concentration in the conventional ESC.

The first proposed ESC method has a smaller number of design parameters and much more robust to the modeling error than the conventional ESC method. The second
proposed ESC method can be totally free from adverse effects of the modeling error of the phase lag parameter by utilizing a secondary measurement of the reactor temperature. Also, it has only three parameters $p$, $a$, and $k_p$, and they can be tuned in an intuitive way. The two proposed ESC methods were experimentally proven to be very promising in tracking and maintaining the optimal reactor temperature. The convergence rate to the minimum CO concentration was faster, and the tuning is easier and more efficient than that in the conventional ESC. The proposed ESC methods can be applied to many similar chemical processes, which reduce toxic components in gas mixtures via oxidation or hydration.

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Notes
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