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Adaptive Neural Network Control of Hydrogen Production via Autothermal Reforming of Methanol

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Abstract: This study focuses on the control problem of hydrogen production via autothermal reforming of methanol. To deal with uncertain system dynamics and external disturbance, an improved adaptive neural network controller is designed to regulate hydrogen flow rate by manipulating methanol flow rate. Theoretical derivation and analysis demonstrate its adaptability to model mismatch and external disturbance. Furthermore, a variable ratio controller law is employed as the reforming temperature controller to achieve steady reforming temperature by adjusting the reforming air flow rate. Finally, the effectiveness of the entire system is testified by experimental means.

Key words: Hydrogen production, adaptive neural network control, autothermal reforming

INTRODUCTION

Driven by the increasing concern for the depletion of the traditional energy sources and also the environment pollution caused by them, hydrogen has become a hot research topic as a clean and efficient replacement. However, one of the major difficulties preventing the wide utilization of this clean energy is the lack of high-density onboard hydrogen storage solutions and the absence of a dense hydrogen distribution infrastructure (Williams, 1993). To overcome these problems, it is proposed that hydrogen can be produced continuously onboard from liquid hydrocarbons instead. Among these hydrocarbons, methanol is considered to be a better candidate compared with other counterparts, thanks to its advantages such as easy storage, high H:C ratio and no C:C ratio (Lindstrom and Pettersson, 2003). Among methods of methanol reforming, Autothermal Reforming (ATR) is an effective one to produce hydrogen from methanol as it does not require external heating (Mu *et al.*, 2007). Nevertheless, owing to a series of complicated reactions involved, ATR of methanol can be seen as a nonlinear multi-input multi-output dynamic process. Once integrated into a fuel cell vehicle, fast start-up and frequent hydrogen flow rate load changes will be required, which posts challenging control problem. Clearly, the inlet flow rates of methanol, water and air should be carefully selected and controlled in order to obtain desired hydrogen flow rate and appropriate reforming temperature. Moreover, uncertainty of model parameters and time delay issues further increase the difficulty in controlling the process (Hu *et al.*, 2008; Zheng *et al.*, 2012). Therefore, how to design an effective and robust control scheme has become important in practical applications.

In the recent decade, some efforts have been devoted to this end. Hu *et al.* (2008) proposed a feed-forward plus feedback control structure to regulate the process temperature. It was observed that the feedback control failed to achieve acceptable performance when the load changed. Although, the feed-forward controller can improve its performance, it was very sensitive to model mismatch. Hence, the authors concluded that advanced control should be applied to solve this problem. In this effort, Hu and Chmielewski (2009) introduced a nonlinear multivariable predictive controller to regulate the temperature and hydrogen flow rate. Only simulation results were obtained to demonstrate the feasibility of the controller. In addition, the reference trajectories were determined off-line. Alternatively, Zheng *et al.* (2012) designed a simple adaptive controller for ATR of methanol with acceptable experimental results but it did not take the reforming temperature into account. Kolavennu *et al.* (2008) proposed a model reference adaptive controller using the Lyapunov method, for tracking a time varying hydrogen load in automobile powered fuel cell but it just remained in simulation stage. Results of system identification indicate significant uncertainty of the model parameters, thus an advanced controller adaptive to the uncertainty is necessary. The

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conventional nonlinear adaptive control typically handles parametric uncertain systems whose parameters appear linearly in the equations and it is unsuitable to handle the levels of uncertainty normally associated with complex nonlinear systems (unknown functions in the systems or parameteric uncertain systems whose parameters appear nonlinearly). This leads to consider the field of adaptive neural network control (Chen and Liu, 1994; Huang *et al*., 2004), which has emerged as a powerful method for handling the higher levels of uncertainty typified by unknown functions instead of parameters.

Therefore, in this study, the control problem of hydrogen production via ATR of methanol is considered. By adjusting the methanol flow rate, an improved adaptive neural network controller is proposed for regulating the hydrogen flow rate, even with the presence of system uncertainty and external disturbance. In addition, a variable ratio controller is proposed to maintain the steady reforming temperature by manipulating the reforming air flow rate. To verify the performance of the proposed scheme, numbers of experimental studies have been conducted. The results substantiate that the control scheme can achieve satisfactory performance without the requirement of explicit modeling step.

This study is organized as follows. Section 2 introduces the process of methanol reforming and an experimental platform built for the research. In Section 3, the control scheme is designed for ATR of methanol. Section 4 focuses on performance of the proposed control scheme. Finally, the conclusion is given in Section 5.

PROBLEM FORMULATION

Currently, three major methods are available for hydrogen production from methanol: partial oxidation, steam reforming and autothermal reforming (Mu *et al*., 2007). Among these, Autothermal Reforming (ATR) of methanol:

$$CH_3OH + \beta O_2 + (1-2\beta)H_2O \rightarrow (3-2\beta)H_2 + CO_2 \ (0 \le \beta \le 0.5)$$
$$\Delta_r H^0_{298} = (50.19 - 483.64\beta) \ \ \text{kJmol}^{-1}$$

can reach an approximate thermal equilibrium when methanol, oxygen and water are fed in with an appropriate proportion. β is a coefficient that represents the ratio between oxygen and methanol involved in the reaction. Thus, ATR of methanol is an optimal choice in terms of generating hydrogen with an acceptable concentration without requiring additional heating or cooling devices.
In order to get a more applicable and validated control strategy, an experimental platform for hydrogen production via ATR of methanol is built in our lab as shown in Fig. 1.

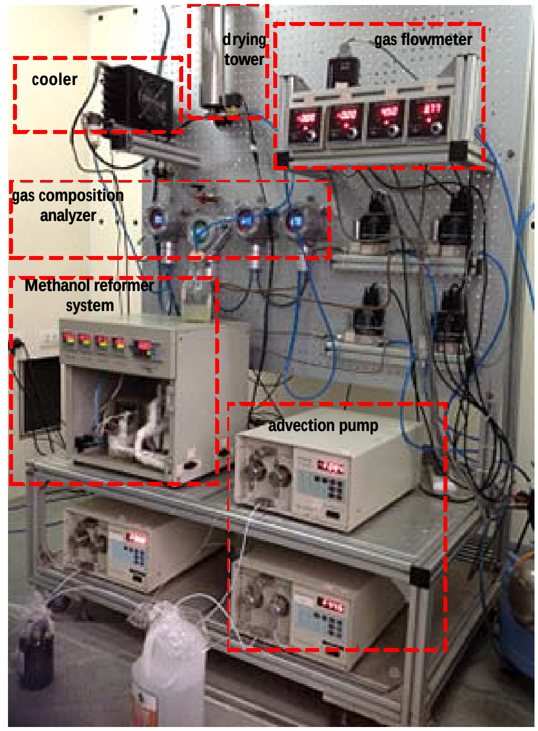

Fig. 1: Experimental platform for hydrogen production

In order to design an appropriate controller, extensive experiments have been conducted for identification of the system. With the system identification software, numerous models were obtained. For instance, two models between the hydrogen flow rate and the methanol flow rate were obtained as:

$$G_1(s) = \frac{56.925(368.48s+1)}{10654.6s^2 + 721.29s + 1} \tag{1}$$

$$G_2(s) = \frac{25.45(45.36s+1)}{5342.6s^2 + 356.78s + 1} \tag{2}$$

The identification results confirm that the system dynamics contain a lot of uncertainties and a simple constant control law may not fulfill the objective.

CONTROLLER DESIGN

Essentially, the objectives of the reformer controller are two-fold: (1) to generate the hydrogen in response to the demand from the consumer, such as fuel cell and (2) to maintain the reforming temperature to guarantee the safety of the reactor. To fulfill the two requirements, one should simultaneously manipulate all feed flows: methanol, water and air appropriately. Numerous experiments have demonstrated that the hydrogen flow rate depends heavily on the methanol flow rate and the water flow rate, while the reforming temperature is extremely sensitive to the reforming air flow rate in contrast to the other inputs. Therefore, the entire control scheme can be conveniently divided into hydrogen flow rate control loop and reforming temperature control loop.

Adaptive neural network controller: Firstly, recalling the dynamics between the hydrogen flow rate and the methanol flow rate, which can be seen as an uncertain parameter-varying dynamic system, adaptive NN control strategy is considered to be a proper but robust solution due to its order reduction and disturbance rejection.

Huang *et al*. (2004) designed a stable adaptive neural network controller for a class of unknown nonlinear systems, which introduced a modified Lyapunov function to eliminate the singularity issue completely and guaranteed uniform ultimate boundedness of the tracking error. In this part, we focus on the improvement and application of the work of Huang *et al*. (2004) for the autothermal methanol reforming process.

The adaptive neural network controller employs the methanol flow rate as the manipulated variable and the hydrogen flow rate as the control variable. The model between the hydrogen flow rate and the methanol flow rate has the following form:

$$\frac{Y(s)}{U(s)} = G(s) = \frac{K(Bs+1)}{A_2 s^2 + A_1 s + 1} \tag{3}$$

Considering the disturbance, the equation of state can be expressed as follows:

$$\begin{cases} \dot{x}_1 = x_2 \\ \dot{x}_2 = -a_1 x_1 - a_2 x_2 + b_1 u + b_2 \dot{u} + d \\ y = x_1 \end{cases} \tag{4}$$

where, $x = [x_1, x_2]^T$, u, y, d are the state variables, system input (the methanol flow rate), system output (the hydrogen flow rate) and disturbance, respectively and:

$$a_1 = \frac{1}{A_2}, a_2 = \frac{A_1}{A_2}, b_1 = \frac{K}{A_2}, b_2 = \frac{KB}{A_2}$$

Regard u as a state variable, define $\varphi = [-a_1, -a_2, b_1]^T$, $X_1 = [x_1, x_2, u]^T$, then the equation of state can be expressed as follows:

$$\begin{cases} \dot{x}_1 = x_2 \\ \dot{x}_2 = \varphi^T X_1 + b_2 \dot{u} + d \\ y = x_1 \end{cases} \tag{5}$$

Assumption 1: Disturbance d is unknown but bounded and $d_M>0$ exists such that $|d|\le d_M$.

Assumption 2: A known continuous function $h(X_1)>0$ and a unknown constant $b_{2m}>0$ exist such that $|\varphi^T X_1|\le h(X_1)$ and $0<b_{2m}\le b_2$.

Suppose that y_d is the desired output of the system, then $[x_1, x_2]^T$ should converge to $[y_d, \dot{y}_d]^T$ in the effect of the control strategy. Define the errors as follows:

$$\begin{cases} e_1 = x_1 - y_d \\ e_2 = x_2 - \dot{y}_d \end{cases} \tag{6}$$

and a filtered tracking error r as:

$$r = \lambda e_1 + e_2, \text{ with } \lambda>0 \tag{7}$$

We first present a desired control, u_d, such that under this control, the control performance is satisfied.

Lemma: For Eq. 5 satisfying Assumptions 1and 2, if the time derivative of a desired Lyapunov-based controller is designed as:

$$\dot{u}_d = -k(t)r - \left[k(t)\delta + k_v v^2\right]\text{sgn}(r) - \frac{1}{b_2}\varphi^T X_1 \tag{8}$$

where, $k(t)\ge k_0>0$ and $k_v>0$, $\delta>0$, sgn (r) is sign function, then the filtered tracking error r is bounded and the system is asymptotically stable when:

$$\delta \ge \frac{1}{k_0 b_{2m}}\left(\frac{1}{4k_v b_{2m}} + d_M\right)$$

Proof: The conclusion can be proved by taking a similar proof procedure as the Lemma 3.2 by Huang *et al*. (2004). Recalling Eq. 8, suppose that:

$$\dot{u}_{1d} = -\frac{1}{b_2}\varphi^T X_1$$

since φ and b_2 are unknown, $\dot{u}_{1d}$ is not available. In what follows, neural networks shall be applied for approximating the unknown nonlinearities in Eq. 8.

As the function $\dot{u}_{1d}$ is smooth, there exists a NN approximation such that:

$$\dot{u}_{1d} = W^T\Phi(V^T X)+\varepsilon \tag{9}$$

where, $X = [X_1^T, 1]^T = [x_1, x_2, u, 1]Y^T$ is the input of NN, V is the weight matrix from X to neural nodes, W is the ideal weight between the output of nodes and the output of NN $\dot{u}_{1d}$, $\Phi(V^TX)$ is NN basis function and has several choice.

Assumption 3: Bounded function approximation error ε satisfies $|\varepsilon|\le\varepsilon_M$ with positive constant ε_M.

Owing to the unknown ideal NN weight W, let W be an estimate of W. V| can be random generated and invariable. Therefore, we have:

$$\dot{u}_1 = \hat{W}^T\Phi(V^TX) \tag{10}$$

where, $\hat{W}$ is N×1 dimension vector, V is 4×N dimension matrix, $X = [x_1, x_2, u, 1]^T$, N is the number of neural nodes. The basis function adopted is:

$$\Phi(Z) = \begin{bmatrix}\phi(z_1) & \phi(z_2) & \cdots & \phi(z_N)\end{bmatrix}^T$$

$$\phi(z_i) = \frac{e^{z_i} - e^{-z_i}}{e^{z_i} + e^{-z_i}}, \quad i = 1, 2, \cdots, N$$

Theorem: For Eq. 5 satisfying Assumptions 1, 2 and 3, the time derivative of input is considered to be designed as:

$$\dot{u} = -k(t)r - \left[k(t)\delta + k_v v^2\right]\mathrm{sgn}(r) + \hat{W}^T\Phi(V^TX) \tag{11}$$

where, $k_v>0$, $\delta>0$, sgn(r) is sign function, the gain k(t):

$$k(t) = \sigma\left[1 + \frac{1}{\omega_1}\left|\hat{W}^T\Phi(V^TX)\right|^2 + \frac{1}{\omega_2}\left|h(X_1)\right|^2\right] \tag{12}$$

where $\sigma>0$, $\omega_1>0$, $\omega_2>0$, the weight update law:

$$\dot{\hat{W}} = -\eta\Phi(V^TX)r - \gamma\eta|r|\hat{W} \tag{13}$$

where $\eta>0$, $\gamma>0$.

If the initial weight:

$$\hat{W}(0)\in\Theta_w,\ \Theta_w = \left\{\hat{W}\,\middle|\,\|\hat{W}\|\le\frac{\phi_M}{\gamma}\right\},\ \left\|\Phi(V^TX)\right\|\le\phi_M$$

with ϕ_M constant, then the weight $\hat{W}(t)$ is bounded. Meanwhile, the filtered tracking error r is bounded and the system is asymptotically stable when:

$$\delta \ge \frac{1}{\sigma}\left(1 + \rho + \frac{\gamma}{4b_{2m}}\|W\|^2\right)$$

Where:

$$\rho = \left(\frac{\omega_1}{4} + \frac{\omega_2}{4b_{2m}^2}\right)\left(1 + \frac{1}{b_{2m}}\right)^2 + \frac{1}{4k_v b_{2m}^2} + \frac{d_M + \varepsilon_M}{b_{2m}} \tag{14}$$

Proof: Define a Lyapunov function:

$$V = \frac{1}{2}r^2 + \frac{1}{2\eta}\tilde{W}^T\tilde{W}$$

and the conclusion can be proved by taking a similar proof procedure as the Theorem 4.1 by Huang *et al.* (2004).

Remark: There are four major highlights in the proposed control scheme compared with the work of Huang *et al.* (2004). First, it can not only make the tracking error Uniformly Ultimately Bounded (UUB) but also achieve asymptotic stability under certain conditions. Second, the controller parameter k(t) is extended to more general form so that the choice of parameters is more flexible. Third, the effects of disturbance are considered to prove that the proposed controller is still valid when disturbance exists. And last but not least, the time derivative of input is chosen to design which can avoid high frequency oscillation of the control action and protect the actual actuator.

Since the gradient of hydrogen flow rate which is denoted by state variable x_2 can not be measured, a high-gain observer (Tornambe, 1992) is used to track it. The observer can be expressed as the following equations:

$$\begin{cases}\dot{\hat{x}}_1 = \hat{x}_2 + p_1k(y - \hat{y})\\ \dot{\hat{x}}_2 = p_2k^2(y - \hat{y})\\ \hat{y} = \hat{x}_1\end{cases} \tag{15}$$

where, $\hat{x} = [\hat{x}_1\ \hat{x}_2]$ and y are estimated values, k is the gain of the observer, p_1 and p_2 are coefficients that make the equation $s^2+p_1s+p_2 = 0$ have different roots.

In addition, the water flow rate also plays a crucial role on the hydrogen flow rate. In this study, according to the results of many experiments and the characteristics of the reformer, we conclude that water/methanol molar ratio of 1.2:1 is optimal for the hydrogen production. Therefore, the water flow rate is set as the methanol flow rate multiplied by the fixed ratio.

Variable ratio controller: Secondly, as the reactor will extinguish itself if the reforming temperature is too low, while the catalyst may be deactivated at high temperature, the reforming temperature of this miniature reactor should be strictly constrained in the range of 450-560°C. Furthermore, numerous experiments indicate that the optimum reforming temperature is within the range of 490 - 540°C, within which the catalyst for ATR of methanol performs excellent activity, long-term stability and no thermal degradation, while the optimum ratio of

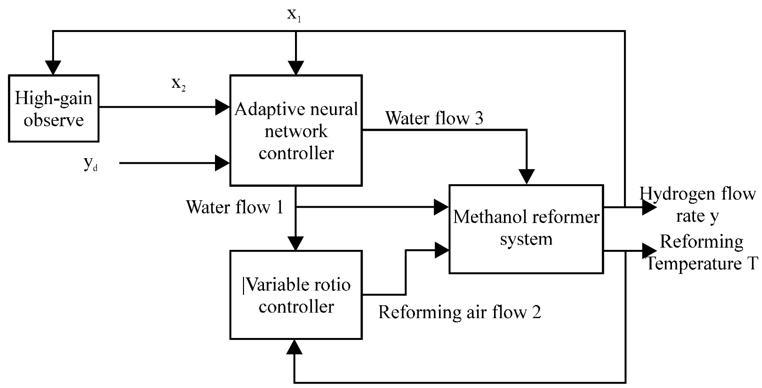

Fig. 2: The schematic diagram of the overall control scheme

reforming air flow rate (L/min) to methanol flow rate (mL/min) is roughly within the range of (0.4 - 0.6):1. Thus, it is necessary to adjust the flow ratio (air: methanol) due to the high sensitivity of the reforming temperature to the reforming air flow rate. In this study, a variable ratio controller is proposed to maintain an appropriate reforming temperature by manipulating reforming air flow rate.

The control law governing the air flow rate u_2 is given as:

$$u_2 = K(n)u_1 \tag{16}$$

where, u_1 is the methanol flow rate in discrete-time and K(n) is the varying ratio at n^{th} sampling time. The ratio is updated at each sampling time according to the real-time feedback of reforming temperature. The update law can be written as:

$$K(n) = K(n\text{-}1)+\Delta K \tag{17}$$

Where:

$$\Delta K = \begin{cases} -0.01, T(n) > 540 \\ 0.01, T(n) < 490 \\ -0.01\text{sgn}(\Delta T), |\Delta T| > 2, 490 \le T(n) \le 540 \\ 0, \text{otherwise} \end{cases} \tag{18}$$

and:

$$\Delta T = T(n)\text{-}T(n\text{-}1) \tag{19}$$

where T(n) represents the reforming temperature at time n.

Although, the update law proposed above is an empirical design based on operator experience, it has been proven to be a simple yet effective method to maintain a suitable and steady temperature.

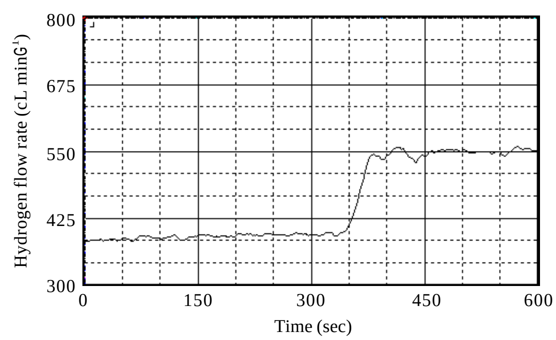

Fig. 3: The response curve of hydrogen flow rate

Combining the adaptive NN controller and the variable ratio controller, we get a control scheme which can be used for the actual methanol reformer system. The schematic diagram of the overall control scheme can be depicted in Fig. 2.

EXPERIMENTAL RESULTS

In order to evaluate performance of the proposed control scheme, a lot of experiments have been conducted on our testbed.

Parameters of the adaptive NN controller have been modulated through experiments. Initially, the hydrogen flow rate was kept at 385 cL min^{-1}. To imitate the varying load in actual practice, at time of 350 sec, the desired hydrogen flow rate was set to be 425 cL min^{-1}. A typical system response using the proposed hydrogen flow rate controller was shown in Fig. 3. With the presence of external disturbance and uncertain system dynamics, the hydrogen flow rate converged to the desired value with very small tracking error. Besides, even with the presence of rapid load changes, the reforming temperature was still maintained within the range of 515-535°C shown in Fig. 4.

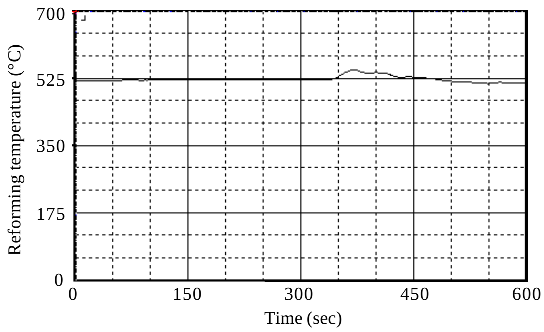

Fig. 4: The response curve of reforming temperature

In summary, experimental results substantiate that the methanol reformer system along with the proposed control scheme can respond to swift load change effectively, while the system remain stable. It can be further used for other applications, such as electric vehicles, consumer goods, etc.

CONCLUSION

In this study, an experimental platform is built for hydrogen production based on ATR of methanol. In order to overcome the difficulty of unknown system dynamics and external disturbance, an improved adaptive neural network controller is designed to regulate the hydrogen flow rate by manipulating methanol flow rate. Meanwhile, a variable ratio controller is proposed to achieve a steady reforming temperature by adjusting the reforming air flow rate. To verify the effectiveness the entire system, experiments have been conducted and the results demonstrate that the reactor can generate the desired hydrogen flow rate while maintaining the reforming temperature within the acceptable range.

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