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THESIS

CONTROLLER DESIGN OF CONTINUOUS pH PROCESSES USING MODEL-BASED CONTROL TECHNIQUE

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A Thesis Submitted in Partial Fulfillment of
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This work presents a control method designed for a continuous pH process with a fluctuation in influent pH. Two process configurations, a single mixing tank and two mixing tanks in series, are considered as case studies. The proposed method is capable of handling process uncertainties and coupling effects between the level and pH. In the control system, a state feedback controller is formulated by using an input-output linearization combined with an optimization to estimate the disturbances in the influent pH and unit interactions. A closed-loop compensator with predicted disturbance is applied to eliminate offset responses. A net proton–hydroxide ion estimated from measured pH is used in a developed model to improve the process prediction. The control objective is to handle the level and pH of a bench-scale, continuous pH process of HCl-NaOH system by adjusting their manipulated inputs. Performance of the proposed method is evaluated by setpoint tracking and disturbance rejection tests and is also compared with the proportional-integral controller. The results of both case studies showed that the developed controller can enforce the process with uncertainties to desired setpoints effectively while the outputs under the proportional-integral controller showed the oscillation and cannot achieve the desired target.

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LIST OF ABBREVIATIONS

F_W	=	Flow rate of influent stream
F_{A1}, F_{A2}	=	Flow rates of acid titrating stream
F_{B1}, F_{B2}	=	Flow rates of base titrating stream
F_{S1}, F_{S2}	=	Flow rates of effluent stream
$\tilde{F}_{S1}, \tilde{F}_{S2}$	=	Estimated flow rates of effluent stream
C_A, C_B	=	Concentration of acid stream and base stream
h_1, h_2	=	Levels of the mixing tank
$\tilde{h}_1, \tilde{h}_2$	=	Estimated levels of the mixing tank
$h_{1,sp}, h_{2,sp}$	=	Setpoints of the mixing tank level
ρ_W	=	Density of influent stream
ρ_{A1}, ρ_{A2}	=	Density of acid titrating stream
ρ_{B1}, ρ_{B2}	=	Density of base titrating stream
ρ_{S1}, ρ_{S2}	=	Density of effluent stream of tank 1 and tank 2
ρ_{m1}, ρ_{m2}	=	Density of titrating stream
A_{R1}, A_{R2}	=	Cross-sectional area of the tank 1 and tank 2
K_W	=	Dissociation constant of water
d_1, d_2	=	Process disturbances
$\bar{d}_1, \bar{d}_2$	=	Estimated process disturbances
u_{m1}, u_{m2}	=	Flow rates of titrating stream
$\tilde{u}_{m1}, \tilde{u}_{m2}$	=	Estimated flow rates of titrating stream
pH_{S1}, pH_{S2}	=	Outgoing pH of the mixing tank
y_1, y_2, y_3, y_4	=	Output
$\dot{x}, x$	=	Vector of states

LIST OF ABBREVIATIONS (Continued)

u	=	Vector of manipulated input
j	=	Objective function
U	=	Vector of manipulated input
$\tilde{U}$	=	Vector of estimated manipulated input
u_{lb}, u_{ub}	=	Lower bound and upper bound of manipulated input
$\dot{e}_1, \dot{e}_2, \dot{e}_3, \dot{e}_4$	=	Error dynamics
u_k, u_{k-1}	=	Manipulated input of the PI controller
K_c	=	Output gain of the PI controller
$y_{i,sp}$	=	Desired output setpoint
$y_{i,k} y_{i,k-1}$	=	Output of the PI controller
Δt	=	Sampling time
DGC	=	Differential geometric control
I/O	=	Input/output
MBC	=	Model-based control
MPC	=	Model predictive control
NI-cDAQ	=	National instruments compact data acquisition
NMBC	=	Nonlinear model-based control
PI	=	Proportional-integral
PID	=	Proportional-integral-derivative

LIST OF ABBREVIATIONS (Continued)

η_W	=	Net proton–hydroxide ion of influent stream
η_{S1}, η_{S2}	=	Net proton–hydroxide ion of effluent stream
$\tilde{\eta}_{S1}, \tilde{\eta}_{S2}$	=	Estimated of net proton–hydroxide ion of effluent stream
$\eta_{S1,sp}, \eta_{S2,sp}$	=	Net proton–hydroxide ion setpoint of effluent stream
η_{B1}, η_{B2}	=	Net proton–hydroxide ion of basic stream
η_{A1}, η_{A2}	=	Net proton–hydroxide ion of acid stream
$\beta_1, \beta_2, \beta_3, \beta_4$	=	Tuning parameters of the I/O controller
v_1, v_2, v_3, v_4	=	Reference setpoints
$\lambda_1, \lambda_2, \lambda_3, \lambda_4$	=	Tuning parameters of the first-order integrator
α_{m1}, α_{m2}	=	Number of dissociation titrating stream
α_{A1}, α_{A2}	=	Number of dissociation acid titrating stream
α_{B1}, α_{B2}	=	Number of dissociation base titrating stream
ψ	=	Compact form of the controller
$\tilde{\psi}$	=	Compact form of the closed-loop state estimator
τ_i	=	Tuning parameter of the PI controller

CONTROLLER DESIGN OF CONTINUOUS pH PROCESSES USING MODEL-BASED CONTROL TECHNIQUE

INTRODUCTION

Automatic pH control has been used extensively in various industries such as chemical, biochemical, pharmaceutical and water treatment (Shinskey, 1996; Guyot *et al.*, 2000; Li *et al.*, 1998; Williams *et al.*, 1990). A task of pH neutralization or pH adjustment particularly for a continuous process is quite difficult due to inherent nonlinearity and high sensitivity to input changes around a neutralization point. In past decades, there are various controller techniques designed for the continuous pH process. The controller syntheses are developed by using techniques such as a proportional-integral-derivative (PID)-based controller integrated with a sliding mode technique (Li *et al.*, 2001) and with an online model-based estimation of strong acid equivalent (Wright and Kravaris, 1991; Wright *et al.*, 1991), a controller with multi-model switching (Pishvaie and Shahrokhi, 2000), an adaptive input-output linearizing controller (Henson and Seborg, 1994) and internal model (Loh *et al.*, 2001), and a model predictive controller with Wiener model (Gomez *et al.*, 2004). These mentioned works have been addressed to a single stirred tank that pH of the influent stream is assumed to be a constant.

Furthermore, in real applications, there are many pH processes that have more than one mixing tanks connected in series and require to perform the pH control in several steps, for example, a reverse osmosis desalination process (Alatiqi *et al.*, 1999) and a continuous chemical/biological treatment of heavy metal in electroplating process (Van Hille *et al.*, 1999; Kurniawan *et al.*, 2006) and brine dechlorination of a chlor-alkali process (Melián-Martel *et al.*, 2010). The dynamics of the integrated processes are complicated because the uncertainty of effluent pH of a previous tank is consequently introduced and amplified in a following tank. The pH control strategies of the single mixing tank mentioned above may be not able to reject the uncertainties. The control of pH process in series has not been much reported in the literature compared with a pH control in the single tank. Perkins and Walsh (1996) applied the optimization technique with mixed integer nonlinear programming (MINLP) and nonlinear programming (NLP) to select proper control structure of pH process in series with the multiple PID controllers. Faanes and Skogestad (Faanes and Skogestad, 2004; Faanes and Skogestad, 2005) studied the effect of the number of stirred tanks in a multi-stage neutralization process. They proposed a scheme of a feedforward/feedback PI controller and a model predictive controller with an integral action that take into account for the pH uncertainty of influent fed to each tank of the system. Hurowitz *et al.* (2000) used a gain scheduling PI controller with a first-order plus dead-time (FOPDT) model to control a laboratory-scale pH process with an inline pH in series. Control performances of mentioned techniques are evaluated through a simulation under assumption of a constant influent flow rate except for Hurowitz's work. Futuremore, there are some configurations of an integrated pH process that both pH and level in each tank require to be handled simultaneously. The change in the tank level directly affects to amount of hydronium concentration in the current and consequent tanks.

OBJECTIVE

The objective of this research is to develop a model-based controller for uncertain continuous pH processes with a single mixing tank and two mixing tanks in series.

Scopes of thesis

The control system is developed for a pH process with inlet uncertainty under a consideration of two process schemes, a single mixing tank and two mixing tanks in series. The control system consists of a feedback controller, a closed-loop state estimator, and a first-order integrator. The feedback controller is formulated by using input-output linearization combined with optimization technique to calculate control actions and estimate process disturbances. The proposed control scheme is evaluated and compared with those of a PI controller by using a bench-scale pH process.

Impact of results

This research developed the control method for a continuous pH process with a single mixing tank and two mixing tanks in series. The presented method provides good tracking and disturbance rejection performance. An advantage of the proposed method is capable of handling pH and tank level of the continuous pH process simultaneously, which the coupled effect has not been mentioned yet in the literatures. The proposed method is practically to implement in a real-time control. It has a potential to extend a scope to a pH process with multiple tanks in series and the chemical/biological process with multiple mixing tanks in series as well.

LITERATURE REVIEW

1. Unmeasured disturbances

Unmeasured disturbances are uncertainty that cannot be measured directly during process operation. This uncertainty may occur from the flow rate and concentration deviations, unknown internal or external disturbances, and stream quality fluctuations obtained from online measurements (Pistikopoulos, 1995). If the controller cannot perform efficiently, it may cause severe deterioration in a closed-loop stability and control performance.

2. Nonlinear model-based control (NMBC)

During the past decades, the advanced control technique such as a nonlinear model-based control (NMBC) that a mathematical model is used into the controller formulation has been received more attentions from both industries and researchers due to high performance. NMBC for uncertain processes can be categorized into three main techniques: differential geometric control (DGC), Lyapunov-based control (LBC), and model predictive control (MPC).

2.1 Differential geometric control (DGC)

The DGC uses a method of a differential geometry to transform a nonlinear system into a linear one. The controller is obtained by analytical inversion of a mathematical model of which the input constraints are negligible (Kravaris and Kantor, 1990). Advantages of this technique are a direct stability analysis of a closed-loop system and few numbers of tuning parameters. However, the DGC cannot be applied for non-minimum phase processes because of unstable zero dynamics. The inversion of the model dynamics induces instability of the closed-loop system.

2.2 Lyapunov-based control (LBC)

LBC is a controller formulation based on a concept of Lyapunov's method. There are two dominant methods of Lyapunov stability are used to investigate the stability of nonlinear systems, called indirect and linearization methods. The linearization method or direct method is to determine the stability properties of a nonlinear process by constructing a Lyapunov function as real positive definite for a process and examining how this function develops in the time domain. The indirect method determines the stability of the nonlinear system though the eigenvalues of the Jacobian matrix of the linearized system around equilibrium. If the time derivative of Lyapunov function is a negative definite, then the system must eventually settle down to a normal equilibrium point. Although the Lyapunov-based control guarantees the closed-loop stability of nonlinear systems, it is difficult to find Lyapunov functions that guarantee stability. The LBC is not widely used due to a difficulty to find the Lyapunov function.

2.3 Model predictive control (MPC)

The MPC is an optimization-based strategy in which a process model is used to predict the effect of future control moves on future values of the outputs. A sequence of input moves is calculated by solving an open-loop optimal control problem at each time step. The constrained optimization problem can be continuous or discrete time problems (Mayne *et al.*, 2000). The control performance of MPC is depended on the number of time step of prediction and control horizons. The higher the number of time step, the better its control performance. However, if the large number of time step is applied, the computational time will increase. Although the MPC has ability to handle the multivariable control problem, it does not guarantee closed-loop stability.

3. Control of the continuous pH process

There are several controller strategies that have been proposed for the continuous pH process that can be categorized as follows:

3.1 Proportional-integral-derivative (PID)-based control

There are many works developing the controller under this direction. Li *et al.* (2001) presented controller system design by PID-based controller integrated with a sliding mode technique. The sliding mode controller is used to handle model uncertainties and process disturbances while the PID controller is used to solve the problem of discontinuous in switching signal. Wright and colleagues (Wright and Kravaris, 1991; Wright *et al.*, 1991) developed a mathematical model that information of inverse titration curve of the acid/ base pairs is interpreted as a strong acid equivalent. An online estimation of strong acid equivalent based on the proposed model is applied in a formulation of a PID controller. Pishvaie and Shahrokhi (2000) used the strong acid equivalent scheme with multi-model switching technique to capture the process nonlinearity. The concept of is then applied to the PID controller. However, the controller will provide a good robustness within a specific operating region.

3.2 Nonlinear model-based controller

The alternative direction for the controller synthesis is a use of a nonlinear model. Henson and Seborg (1994) developed an adaptive nonlinear controller for a neutralization process in the HNO_3-NaOH system by using an exact input-output linearization with an observer to estimate state variables and buffer concentration. Gomez *et al.* (2004) developed a nonlinear model predictive control (MPC) algorithm based on a neural wiener model. Barraud *et al.* (2009) applied a Lyapunov-based controller for the experiment of pH neutralization. However, the proposed method is applicable to a batch process only.

These mentioned research works have been design controller in the assumption of constant influent pH. There are only few methods that perform the actual test on control performance (Wright *et al.*, 1991; Pishvaie and Shahrokhi, 2000; Henson and Seborg, 1994; Loh *et al.*, 2001). In an industrial practice, the uncertainties in the influent pH and measurement sensors such pH and level sensors usually occur in the process. Many process schemes require handling both pH and level of the mixing tank simultaneously or have many mixing tanks connected in series to perform the pH control in several steps. The dynamics of the integrated pH processes are complicated because the uncertainty of effluent pH of the previous tank is introduced and amplified in a following tank. The examples of integrated pH processes are a reverse osmosis desalination process (Alatiqi *et al*., 1999), a continuous chemical/biological treatment of heavy metals in electroplating process (Van Hille *et al*., 1999; Low *et al*., 1995; Kurniawan *et al*., 2006) and a brine dechlorination of a chlor-alkali process (Melián-Martel *et al*., 2010). The method for these problems has not been mentioned in the literatures yet.

4. Input-output (I/O) linearization control

Input-output linearization is a controller synthesis technique that involves coordinate transformation to construct the relationship between the original system output (y) and the new input (v) in the linear form.

Consider the continuous-time multivariable nonlinear system in the compact form:

$$\begin{aligned} \dot{x} &= f(x,u) \\ y &= h(x) \end{aligned} \tag{1}$$

where $x(t) = [x_1, \ldots, x_n]^T \in \mathrm{X}$ is the vector of state variables, $u = [u_1, \ldots, u_m]^T \in \mathrm{U}$ is the vector of manipulated inputs, $f(x,u)$ is a nonlinear vector function, and $y = [y_1, \ldots, y_m]^T$ is the vector of controlled outputs.

The responses of the closed-loop process output are requested, having the linear form:

$$\begin{aligned} (\beta_1 D + 1)^{r_1} y_1 &= y_{sp_1} \\ &\vdots \\ (\beta_m D + 1)^{r_m} y_m &= y_{sp_m} \end{aligned} \qquad (2)$$

where D is the differential operator (i.e. $D \equiv d/dt$), $\beta_1, \ldots, \beta_m$ are positive, constant parameters that set the speed of the closed-loop response of outputs $y_1, \ldots, y_m$ respectively, and $y_{sp_1}, \ldots, y_{sp_m}$ are the output setpoints. The relative order of the controlled output y_i is denoted by r_i, where r_i is the smallest integer for which $(\partial / \partial u_i)(d^{r_i} y_i / dt^{r_i}) \neq 0$. The following notation is used:

$$\begin{aligned} y_i &= h_i^0(x) \\ \frac{dy_i}{dt} &= h_i^1(x) \\ \frac{d^2 y_i}{dt^2} &= h_i^2(x) \\ &\vdots \\ \frac{d^{r_i - 1} y_i}{dt^{r_i - 1}} &= h_i^{r_i - 1}(x) \\ \frac{d^{r_i} y_i}{dt^{r_i}} &= h_i^{r_i}(x, u_i) \end{aligned} \qquad (3)$$

This notation is made based on the following assumptions:

1) The relative orders, $r_1, \ldots, r_m$, are finite.

2) The characteristic matrix of the process, $\partial h_i^{r_i}(x, u_i) / \partial u_i \neq [0 \ldots 0]$, is non-singular on $\mathcal{X} \times \mathcal{U}$.

3) The process is controllable and observable locally around the nominal steady state.

4) The matrices $\partial f / \partial x$ and $\left(\partial h / \partial x\right)\left(\partial f / \partial x\right)^{-1}\left(\partial f / \partial u\right)$ evaluated at nominal steady-state pair, (x_{ss}, u_{ss}), are non-singular.

In case that the controlled output (y) does not have a finite relative order ($r = \infty$) the manipulated input (u) does not affects the controlled output. The feedback controller is obtained by solving equation (2). The compact form of the feedback controller denotes by:

$$u_i = \psi(x, y_{sp_i}) \tag{4}$$

MATERIALS AND METHODS

Materials

1. Personal computer with 2.13 GHz of Intel Core 2 Duo processor, 3.00 GB of RAM

2. National Instruments Compact Data Acquisition (National Instruments)

3. Bench scale pH process

4. Bench scale pH process in series

5. Software
   5.1 MATLAB Version R2010a (MathWorks, Inc.)
   5.2 LabVIEW version 8.2, LabVIEW Real-time module version 8.2, and LabVIEW FPGA module version 8.2 (National Instruments, Inc.)

Methods

This research proposed a control system for a pH process with a single mixing tank and two mixing tanks in series. The procedure of control system development is summarized in a flow diagram as shown in Figure 1. First, the feedback controllers for with a single mixing tank and two mixing tanks in series are designed by combining the I/O linearization and the optimization technique. Next, a closed-loop state estimator and a first-order integrator are designed. Finally, the control system is applied to real time control of the bench scale pH process of both configurations by using MATLAB and LabVIEW software. The performance of a control system is tested through the setpoint tracking and disturbance rejection. The details of each step are given in the following section.

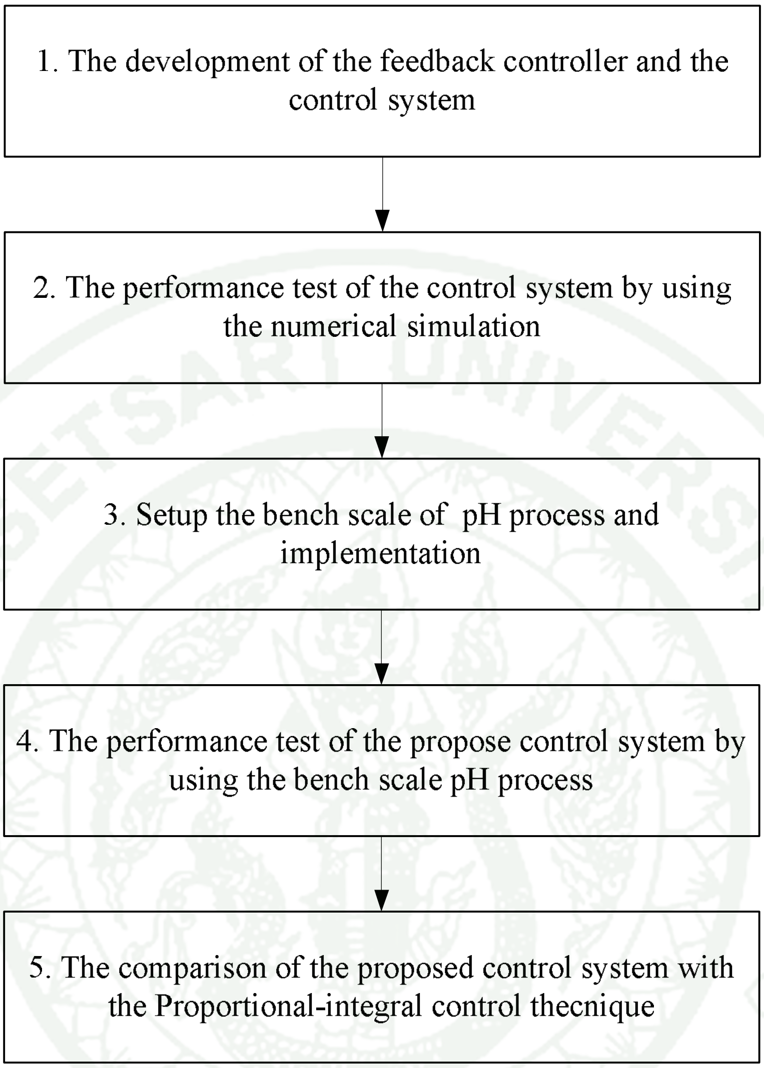


Figure 1 The development of the model-based controller for the continuous pH process

1. The development of the feedback controller and the control system

Consider a general class of multivariable processes with uncertainties described by a mathematical model of a form

$$\begin{aligned} \dot{x} &= f(x,u,d) \\ y &= h(x) \end{aligned} \tag{5}$$

where $x=[x_1,...,x_n]^T$ is the vector of state variables, $u=[u_1,...,u_m]^T$ is the vector of manipulated inputs, $y=[y_1,...,y_m]^T$ is the vector of controlled outputs, and $d=[d_1,...,d_n]^T$ is the vector of state unmeasured disturbances, respectively.

For the nonlinear system in Eq. (6), the relative order of the output, y_i is denoted by r_i where r_i is the smallest number for which $\frac{\partial}{\partial u}h_i^{r_i}(x,u)\neq 0$. It is assumed that the relative orders, $r_1,...,r_m$, are finite and that the process is controllable and observable locally (around the nominal steady state). The following notation is used:

$$
\begin{aligned}
y &= h\,(x)\\
\frac{dy}{dt} &\equiv \left[\frac{\partial h\,(x)}{\partial x}\right]\frac{\partial x}{\partial t} = h^1\left(x\right)\\
&\vdots\\
\frac{d^{\rho} y}{dt^2} &\equiv \left[\frac{\partial h^{\rho-1}(x)}{\partial x}\right]\frac{\partial x}{\partial t} = h^{\rho}\left(x,d\right)\\
\frac{d^{\rho+1} y}{dt^2} &\equiv \left[\frac{\partial h^{\rho}(x,d)}{\partial x}\right]\frac{\partial x}{\partial t} + \left[\frac{\partial h^{\rho}(x,d)}{\partial d}\right]\frac{\partial d}{\partial t} = h^{\rho+1}\left(x,d,d^{(1)}\right)\\
&\vdots\\
\frac{d^{r} y}{dt^r} &\equiv \left[\frac{\partial h^{r-1}(x)}{\partial x}\right]\frac{\partial x}{\partial t} + \left[\frac{\partial h^{r-1}(x,d)}{\partial d}\right]\frac{\partial d}{\partial t} = h^{r}\left(x,u,d^{(1)},...,d^{(r-\rho)}\right), \rho < r
\end{aligned}
\qquad (6)
$$

where ρ is the smallest number for which $\frac{\partial}{\partial d}h^{\rho}(x,d)\neq 0$.

1.1 Input-output (I/O) linearization

The setpoint tracking controller is formulated by using I/O linearization. The idea of I/O linearization is to find a direct relation between the output y and the input u. This is achieved by repeatedly differentiating the output y with respect to time, until it is explicitly related to the input u which is a called relative order.

Review of the input/output linearization for the process with uncertainties and definition of relative order can be found in Daoutidis and Kravaris (1989)

For implementing the input-output (I/O) linearization, the assumptions that the system in equation (5) is open-loop stable and internal dynamics (zero dynamics) is stable have been made. The close-loop output responses in equation (5) with the following form are requested:

$$\begin{aligned} (\beta_1 D+1)^{r_1}\, y_1 &= v_1 \\ &\vdots \\ (\beta_4 D+1)^{r_m}\, y_m &= v_m \end{aligned} \tag{7}$$

where D is the differential operator (i.e. $D \equiv d/dt$), $r_1,\ldots,r_m$ are the relative order of the controlled outputs, $y_1,\ldots,y_m$, with respect to the manipulated inputs, $v_1,\ldots,v_m$ are the reference output setpoints and $\beta_1,\ldots,\beta_m$ are the tuning parameters that adjust the speed of the responses of the outputs, $y_1,\ldots,y_m$, respectively.

1.2 Static state feedback design

We can induce to an unconstrained process of the form equation (7) by implementing the solution to the following constrained optimization problem at each time instant to estimate the control actions and unknown parameter, $\bar{d}$.

$$\begin{aligned} &j = \min_{u,\bar{d}} \sum_{i=1}^{m} \left(w_i (\beta_i D+1)^{r_i} y_i - v_i \right)^2 \\ &\text{subject to} \\ &u_{lb} < u < u_{ub} \\ &d_{lb} < \bar{d} < d_{ub} \end{aligned} \tag{8}$$

where $\bar{d}$ is the vector of estimated state disturbances, the subscript ub and lb denote upper and lower bounds respectively, and w is a weighting factor. By substituting the process dynamics in equation (5) into equation (8), using a definition in equation (6) and settling all time derivatives of $\bar{d}$ to be zero, a solution of the constrained optimization problem can be represented by:

$$\left[u,\bar{d}\right]=\psi\left(x,v\right) \tag{9}$$

1.3 Closed-loop state estimator

In this work, an integral action is added in the developed state feedback to ensure the offset-free closed-loop response. The method is similar to the concept of internal-model control. The estimated process outputs are calculated by using a closed-loop process model and the disturbances estimated from equation (9) that is assumed as the known disturbances in the closed-loop model. The state estimator is expressed by:

$$\begin{aligned}\dot{\tilde{x}}&=f\left(\tilde{x},\tilde{u},\bar{d}\right)\\ \tilde{y}&=h\left(\tilde{x}\right)\end{aligned} \tag{10}$$

where $\tilde{x}$ is the estimated states, $\tilde{y}$ is the estimated outputs, $\bar{d}$ is the disturbance obtained from equation (9) and $\tilde{u}$ is the vector of estimated inputs obtained by solving the following optimization:

$$\begin{aligned}&j=\min_{\tilde{u}}\sum_{i=1}^{m}\left((\beta_i\dot{D}+1)^{r_i}\,\tilde{y}_i-v_i\right)^2\\ &\text{subject to}\\ &u_{lb}\le\tilde{u}\le u_{ub}\end{aligned} \tag{11}$$

The control action obtained by solving the optimization problem in equation (11) can be represented by:

$$\tilde{u} = \tilde{\psi}\left(\tilde{y}, v, \bar{d}\right) \tag{12}$$

1.4 Integral action

To compensate for the offset due to the effects of model–process mismatch and the error in the estimate states in each loop-time instant, the following first-order error dynamics are introduced:

$$\begin{aligned} \dot{e}_i &= \lambda_i(\tilde{x}_i - x_i) \\ v_i &= x_{sp_i} + e_i \qquad , i = 1, \ldots, m \end{aligned} \tag{13}$$

where e, λ, and v are the error of the outputs, the positive constant of the first-order error dynamics, and the corrected setpoint of the outputs, respectively.

By combining the feedback I/O controller in equation (9), closed-loop compensators of equations (10), (12), and (13) the feedback control system can be described by:

$$\begin{aligned} \dot{x} &= f(x, u, d) \\ \left[u, \bar{d}\right] &= \psi\left(x, \upsilon\right) \\ \dot{\tilde{x}} &= f\left(\tilde{x}, \tilde{u}, \bar{d}\right) \\ \tilde{u} &= \tilde{\psi}\left(\tilde{y}, v, \bar{d}\right) \\ \dot{e} &= \lambda(\tilde{x} - x) \\ v &= y_{sp} + e \end{aligned} \tag{14}$$

A schematic diagram of the developed control system is shown in Figure 2.

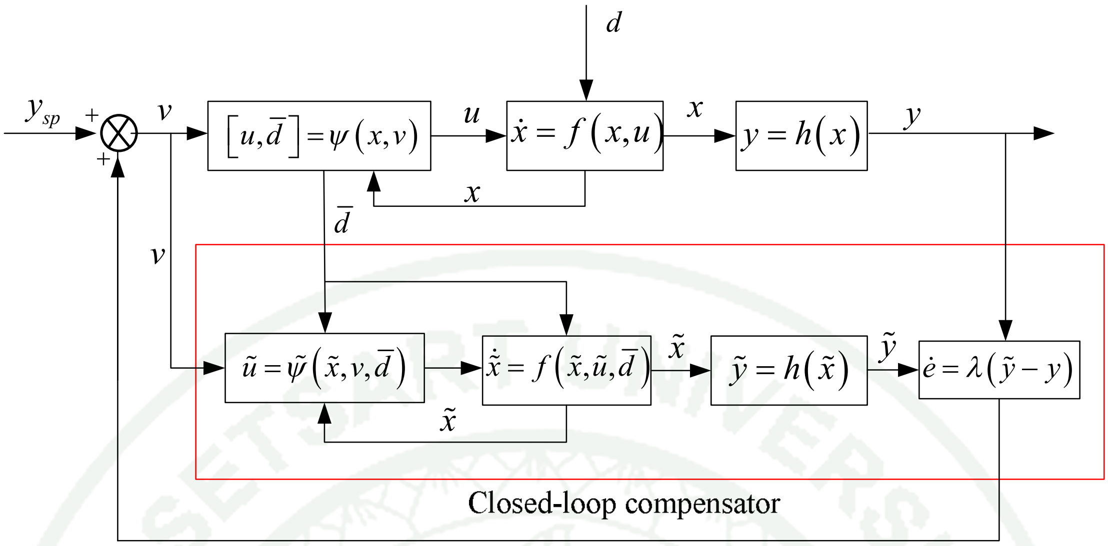


Figure 2 Schematic diagram of the proposed control system

2. Description of the bench-scale pH process

2.1 Setup for a continuous pH process with a single mixing tank

The figure of the bench-scale pH process with a single mixing tank is illustrated in Figure 3. The process comprises of the mixing tank, acid tank, base tank, ultrasonic level sensor, pH meter and peristaltic pumps. A simple diagram of the process is shown in Figure 4.

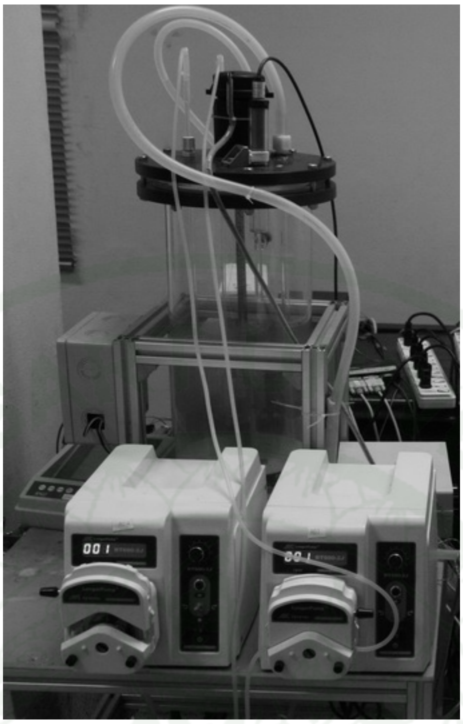


Figure 3 The bench-scale pH process with a single mixing tank

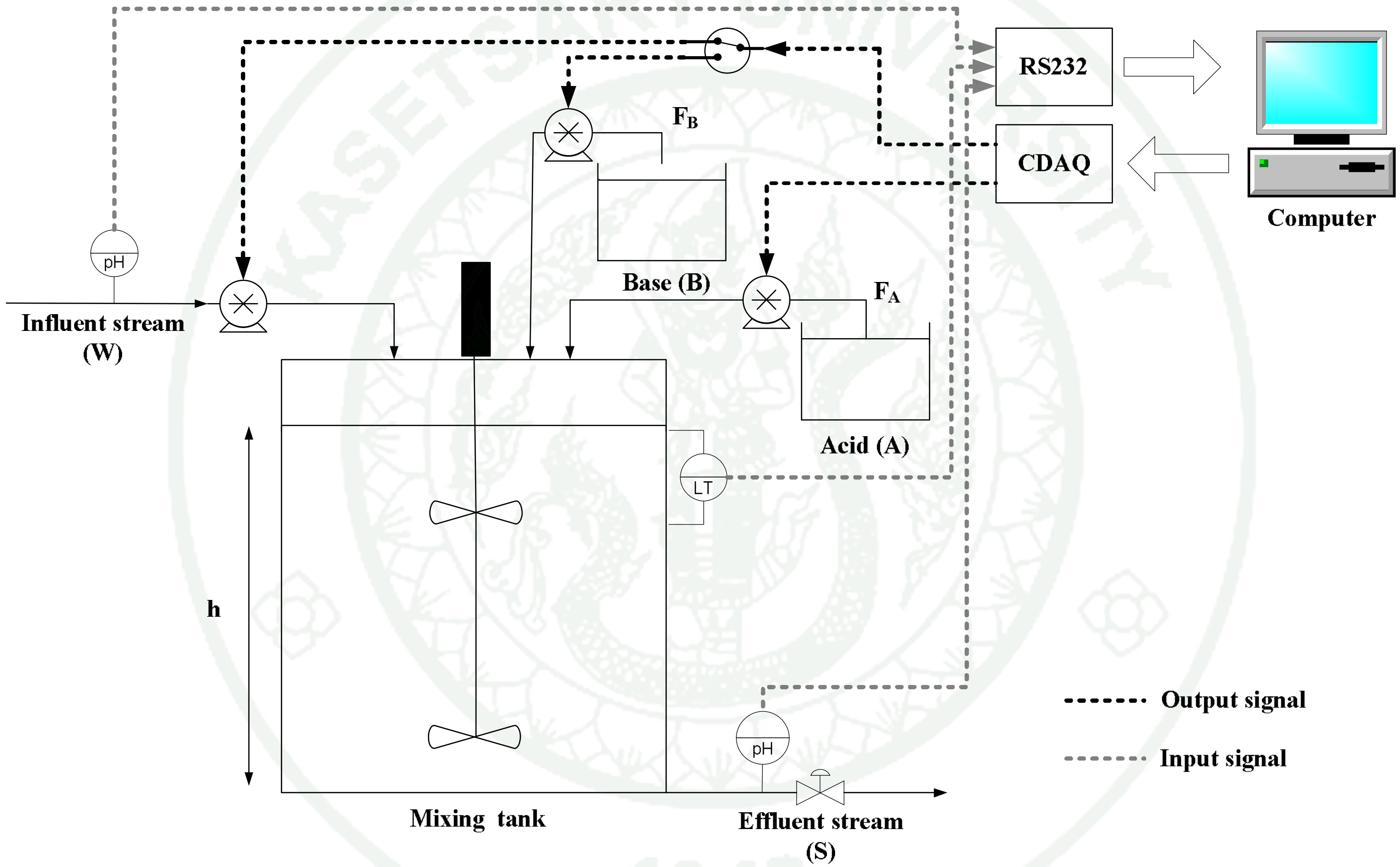
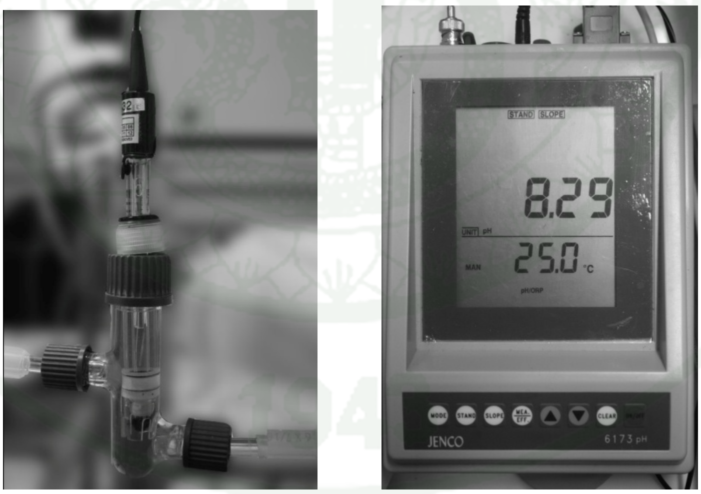


Figure 3 A simple diagram of the bench-scale pH process with a single mixing tank

The measured devices used in the experiment are comprised of an ultrasonic level sensor and a pH meter. The level sensor in Figure 5 is used to measure a liquid level in the mixing tank and to send the signal to the RS-232 hub.

Figure 5 The ultrasonic level sensor with RS-232 output

The pH meter consists of pH probes and electronic pH meter as shown in Figure 6. The pH probes are installed at two locations, which are at influent and effluent streams, for measuring pH values. The computer is received the value through the RS-232 hub.



(a) (b)

Figure 6 The picture of (a) pH probe, and (b) electronic pH meter

Figure 7 shows the peristaltic pump that is used for adjusting the feed corresponding to the analog signal received from NI-cDAQ. Three pumps are used to adjust the flow rate of the influent stream, acid stream, and the base stream fed to the mixing tanks.

Figure 7 The peristaltic pump used in the bench-scale pH process

2.2 Setup the bench-scale pH process with two mixing tanks in series

The bench-scale pH process with two mixing tanks in series is shown in Figure 8. The equipments mentioned in previous section which are the pH meter and peristaltic pump are also used in the process. Three pH meters are located at influent stream, the effluent stream of mixing tank1 and effluent stream of mixing tank2. Six peristaltic pumps are used to manipulate the feed flow rate, the outflow, the acid titrating stream, and the base titrating stream of the mixing tank 1 and mixing tank 2.

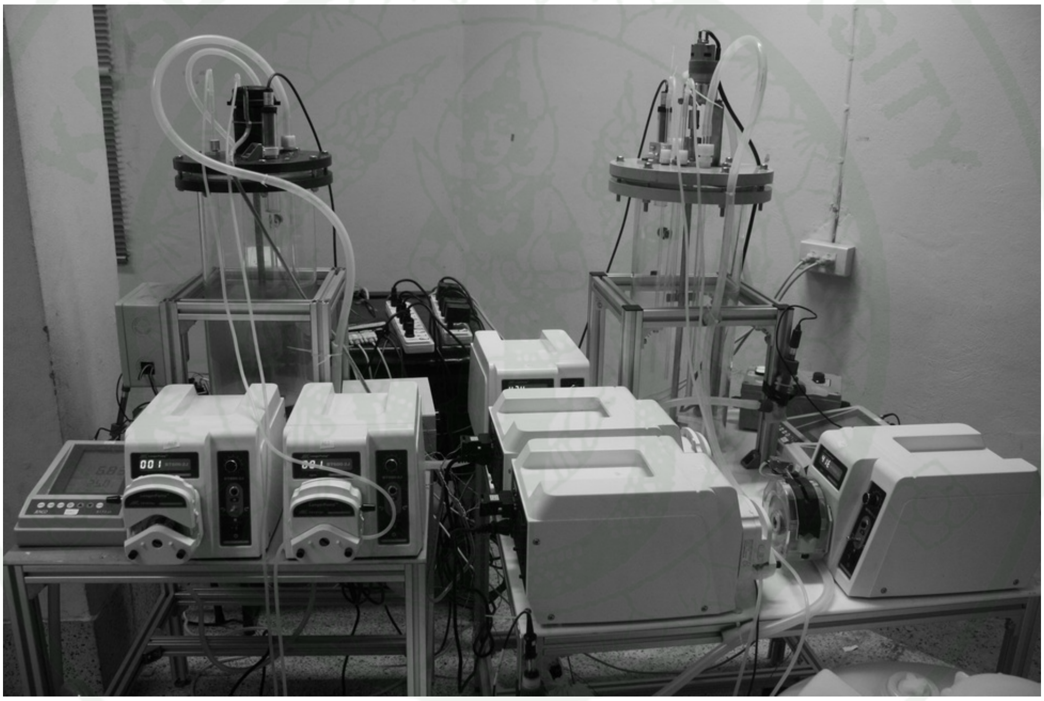


Figure 8 The bench-scale pH process with two mixing tanks in series

The liquid level of each tank was measured by ultrasonic level sensors that are located in tank 1 and tank 2. The ultrasonic level sensor with the analog signal is shown in Figure 9. A simple diagram of the bench scale pH process in series is shown in Figure 10.

Figure 9 Ultrasonic level sensor with the analog output

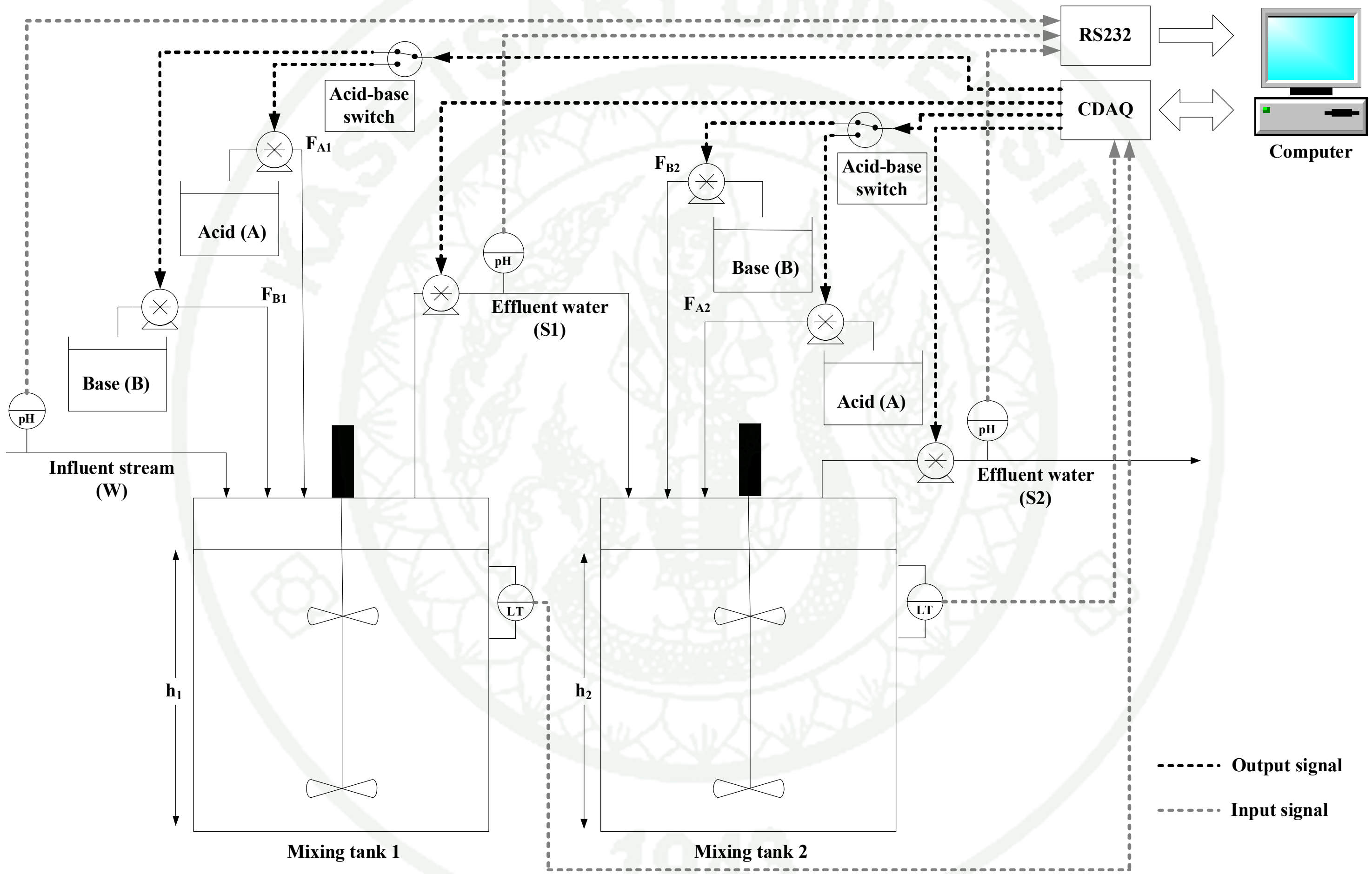


Figure 10 A simple diagram of the bench scale pH process with two mixing tanks in series

2.3 Description of National Instruments compact data acquisition (NI-cDAQ)

In this work, the NI-cDAQ is used as data acquisition hardware that consists of NI-c DAQ chassis and the I/O module. The NI-cDAQ chassis (NI-cDAQ 9174) is used for controlling the synchronization, timing and data transfer between a host computer and the I/O module. The I/O module is a device that interconnects the process signal, either the analog or digital signal, into the data streams. The modules of the analog voltage input (NI-9201), and output (NI-9263) are used in the experimental setup. Simplified diagram of the NI c-DAQ is shown in Figure 11.

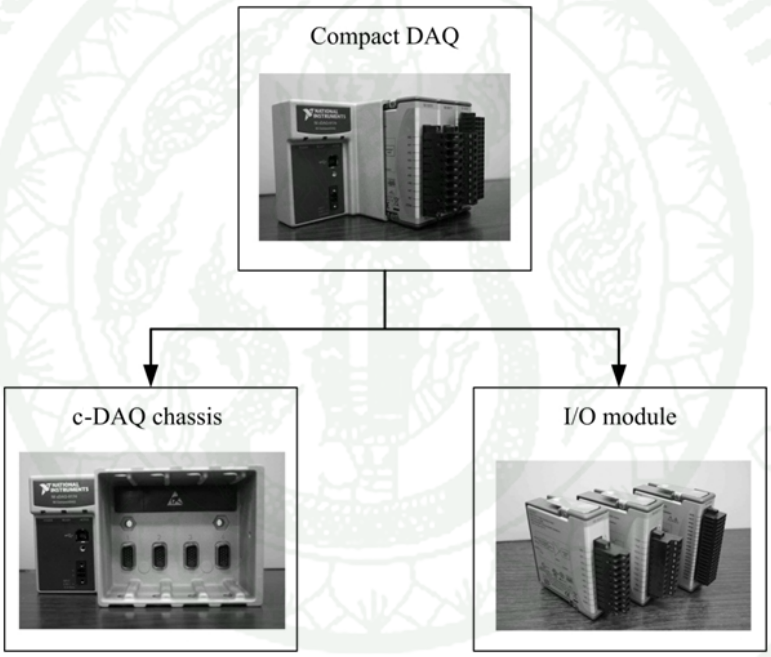


Figure 11 The data acquisition device NI-cDAQ consisting of NI-cDAQ chassis (NI-cDAQ 9174) and I/O modules from National Instruments

3. Test the of real-time control with bench-scale pH process and pH process in series

To setup a real-time control, the proposed control system is connected with bench-scale pH process. There are two configurations of the continuous pH process used in the experiment, a single mixing tank and two mixing tanks in series. The bench-scale pH process is configured corresponding to the desired experiment. Both setpoint tracking and disturbance rejection tests are applied to evaluate the performance of the control system. The recorded data of the input and output are used to plot and perform analysis. The performance of the proposed control system is compared with the digital PI controller with the same condition.

RESULTS AND DISCUSSION

Part I: Controller design and performance test of a bench-scale pH process with a single mixing tank

1 Process description

A pH process presented in Figure 12 consists of an influent stream, acid titrating stream and basic titrating stream that are fed into a continuous mixing tank. The flow of an effluent stream (F_S) purging out from the bottom exit is varied with a liquid level in the vessel (h). A height of liquid is measured by an ultrasonic level sensor. The pH of influent and effluent are measured by pH probes that are installed at the inlet and outlet of the tank. The influent feed (F_W) has a fluctuation in pH. Only acid or basic titrating stream is used to adjust pH of liquid in the mixing tank to a desired pH setpoint, of which the selection of titrating chemicals is depended on the influent pH and desired setpoint.

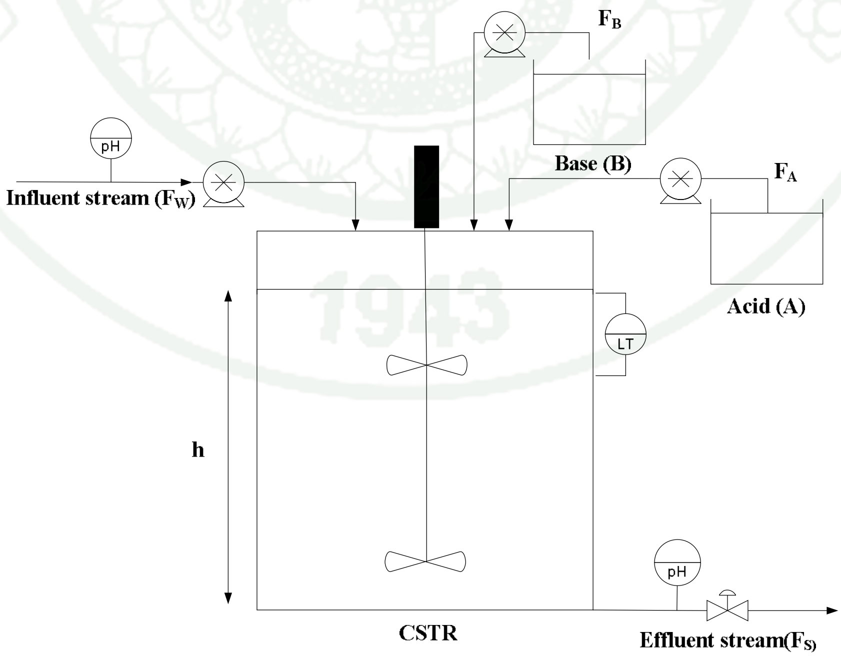


Figure 12 Schematic of a bench-scale pH process

A mathematical model of the continuous pH process is developed with the following assumptions:

1) Cross-sectional area of the mixing tank is constant.
2) Densities of the influent and outflow of both mixing tanks are approximately equal ($\rho_W = \rho_S = \rho$).
3) Densities of acid and base streams are constant.
4) The tank is well-mixed conditions.

From the process shown in Figure 11, the material balance can be obtained as follows:

$$\frac{dh}{dt} = \frac{\rho(F_W - f\sqrt{h}A_t) + \rho_A F_A + \rho_B F_B}{\rho A_R} \tag{15}$$

where h is the tank level, F_W is the influent stream, F_A and F_B are the titrating streams of acid and base, f is the friction coefficient of effluent stream, ρ_A is the density of acid stream, ρ_B is the density of basic stream A_t is the cross-sectional area of the exit pipe, and A_R is the cross-sectional area of the mixing tank.

From a definition of a potential of hydrogen ion ($pH = -\log H^+$), hydroxide ion ($pOH = -\log OH^-$), and correlation of $pH + pOH = 14$, we can define the net proton–hydroxide ions in the term of η as a following equation

$$\eta = \left(10^{-pH} - \frac{K_w}{10^{-pH}}\right) \tag{16}$$

where K_w is the equilibrium constant for the ionization of water ($K_w = 10^{-14}$ at STP and 25^oC). The component balance of the process in the term of η can be described as follows:

$$\frac{d\eta_S}{dt} = \frac{(\eta_W + d)F_W + \eta_A F_A + \eta_B F_B - \eta_S f\sqrt{h}A_t}{A_R h} - \eta_S \frac{\rho(F_W - f\sqrt{h}A_t) + \rho_A F_A + \rho_B F_B}{\rho A_R h} \quad (17)$$

where η_S, η_A, η_B are the net proton–hydroxide ions of the mixing tank, the acid stream and the base stream, and d is a disturbance of the dynamics of η_S. In the case of a titrating stream with a high concentration, the net proton–hydroxide ions of the acid and base stream defined in equation (16) can be approximated by following equations

For the high concentration acid: $\eta_A = \alpha_A C_A$ (18a)

For the high concentration base: $\eta_B = -\alpha_B C_B$ (18b)

where α_A and α_B are the coefficients of total ion concentration of acid and base streams, C_A and C_B are the concentration of acid and base streams.

The control objective is to control the pH and level in the mixing tank (pH_S and h) by manipulating the influent flow rate and acid/base titrating stream (F_W, F_A and F_B). In this work, we practically use either acid or base stream to adjust pH of a mixture in the mixing tank corresponding to the desired setpoint ($pH_{S,sp}$). Therefore, the manipulated inputs of the process can be reduced to two variables, which are F_W and u_m. The parameter u_m is the flow rates of a proper titrating stream fed in the mixing tank. The algorithm of a selection of the proper titrating stream is shown in Figure 13.

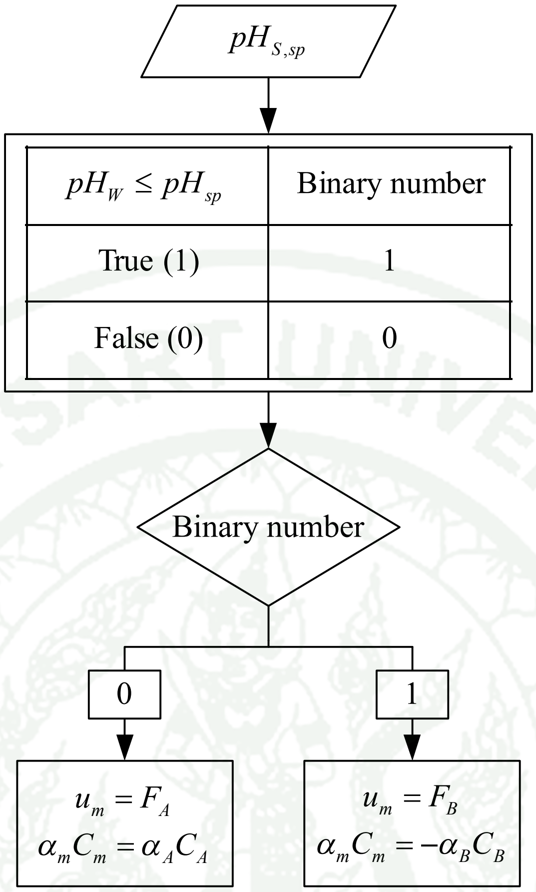


Figure 13 Flow diagram of the selection of the titrating stream for the continuous pH process in series

Then, the process model of the continuous pH process used in this work can be summarized as follows:

$$\frac{d\eta_S}{dt} = \frac{(\eta_W + d)F_W + \alpha_m C_m u_m - \eta_S f\sqrt{h}A_t}{A_R h} - \eta_S \frac{\rho(F_W - f\sqrt{h}A_t) + \rho_m u_m}{\rho A_R h}$$
$$\frac{dh}{dt} = \frac{\rho(F_W - f\sqrt{h}A_t) + \rho_m u_m}{\rho A_R} \tag{19}$$
$$y = [pH_S\ h]^T, \quad u = [u_m\ F_W]^T$$

Note that the equation (16) can be used to convert the relation between pH and η in the calculation.

2. Applying the proposed controller

2.1 Feedback controller

For implementing the input-output (I/O) linearization, the assumptions that the system in equation (6) is open-loop stable and internal dynamics (zero dynamics) is stable have been made. The close-loop output responses in equation (6) with the following form are requested:

$$\begin{aligned}(\beta_1 D+1)^{r_1} y_1 &= v_1 \\ (\beta_2 D+1)^{r_2} y_2 &= v_2\end{aligned} \tag{20}$$

where D is the differential operator (i.e. $D \equiv d/dt$), r_1, r_2 are the relative orders of the controlled outputs, y_1, y_2 with respect to the manipulated inputs, v_1, v_2 are the reference output setpoints and β_1, β_2 are the tuning parameters that adjust the speed of the responses of the outputs, y_1, y_2, respectively.

In this work, a minimized sum of squared errors between the requesting close-loop output responses and the reference output setpoints is applied to calculate the control actions (u) and to estimate the unmeasured disturbances ($\bar{d}$). With the relative order of the outputs, $r_1 = 1$ and $r_2 = 1$ an objective function of the optimization problem can be formulated in a following form:

$$j_1 = \min_{u,\bar{d}} \left(w_1 \left(\beta_1 \dot{\eta}_S + \eta_S - v_1 \right)^2 + w_2 \left(\beta_2 \dot{h} + h - v_2 \right)^2 \right)$$

subject to

$$\begin{aligned} u_{lb} &\le u \le u_{ub} \\ \bar{d}_{lb} &\le \bar{d} \le \bar{d}_{ub} \end{aligned} \tag{21}$$

where w is the weighting factor, $\bar{d}$ is the estimated disturbance, ub and lb are the upper bound and lower bound respectively. From a solution of the optimization problem in equation (21), the control action and estimate disturbance can be represented by:

$$[u,\bar{d}] = \{u_m, F_W, \bar{d}\} = \psi(\eta_S, h_1, v_1, v_2) \tag{22}$$

2.2 Closed-loop state estimator

The state estimator is used to ensure the offset response combine with using an integral action. The estimated process outputs are calculated by using a closed-loop process model and the estimated disturbances obtained from equation (10) that is assumed as the known disturbances in the closed-loop model. The closed-loop state estimator with the disturbance prediction is developed as shown in equation (23).

$$\begin{aligned} \frac{d\tilde{\eta}_S}{dt} &= \frac{\left(\eta_W + \bar{d}\right)\tilde{F}_W + \alpha_m C_m \tilde{u}_m - \tilde{\eta}_S f\sqrt{\tilde{h}}A_t}{A_R \tilde{h}} - \tilde{\eta}_{S1}\frac{\rho(\tilde{F}_W - f\sqrt{\tilde{h}}A) + \rho_m \tilde{u}_m}{\rho A_R \tilde{h}} \\ \frac{d\tilde{h}}{dt} &= \frac{\rho(\tilde{F}_W - f\sqrt{\tilde{h}}A_t) + \rho_m \tilde{u}_m}{\rho_S A_R} \end{aligned} \tag{23}$$

where $\tilde{\eta}_S$ is the estimated net proton–hydroxide ions in the mixing tank, $\tilde{h}$ is the estimated mixing tank level, and $\tilde{u}_m$, $\tilde{F}_w$ are the estimated inputs obtained by solving the following optimization:

$$\begin{aligned} & j_1 = \min_{\tilde{u}} \left(w_1 \left(\beta_1 \dot{\tilde{\eta}}_S + \tilde{\eta}_S - v_1 \right)^2 + w_2 \left(\beta_2 \dot{\tilde{h}} + \tilde{h} - v_2 \right)^2 \right) \\ & \text{subject to} \\ & u_{lb} \le \tilde{u} \le u_{ub} \end{aligned} \tag{24}$$

where $\tilde{u}$ is the set of the estimated inputs ($\tilde{u}_m$, $\tilde{F}_W$). The control action obtained by solving the optimization problem in equation (24) can be represented by:

$$\tilde{u} = \left\{ \tilde{u}_m, \tilde{F}_W \right\} = \tilde{\psi}(\tilde{\eta}_S, \tilde{h}, v_1, v_2, \bar{d}) \tag{25}$$

2.3 Integrator

To compensate for the offset due to the effects of the model–process mismatch and the error in the estimated states, the following error dynamics are introduced:

$$\begin{aligned} \dot{e}_1 &= \lambda_1(\tilde{\eta}_S - \eta_S) \\ \dot{e}_2 &= \lambda_2(\tilde{h} - h) \\ v_1 &= \eta_{S1,sp} + e_1 \\ v_2 &= h_{1,sp} + e_2 \end{aligned} \tag{26}$$

where e, λ, and v are the error of the output, the positive constant of the first-order error dynamics, and the corrected setpoint of the output, respectively. The full equations of control system are shown in Appendix A.

3 Real-time implementation

To evaluate control performance of the proposed method, the bench-scale pH process with single mixing tank shown in Figure 3 is used in the test. The liquid levels (h), the influent flow (F_W) and the titrating streams (u_m) of the mixing tank are operated within the range 10-45 cm, 0.8-4.5 L/min and 0-100 mL/min, respectively. All process outputs are measurable. The process is monitored and controlled by a desktop computer through the National Instruments compact data acquisition (NI-cDAQ) and a RS232 hub. The LabVIEW software is used to create the proposed control system and perform data acquisition. An interface diagram of the control system is shown in Appendix Figure C1. The parameters of the bench-scale pH process are given in Table 1.

Table 1 Parameter values of the bench-scale pH process with the single mixing tank

Variable	Description	Value	Unit
ρ_A	Density of hydrochloric acid	1087.8	kg/m^3
ρ_B	Density of sodium hydroxide	1055.4	kg/m^3
ρ	Density of the influent/effluent	1000	kg/m^3
A_R	Cross-sectional area of the mixing tank	0.0254	m^2
A_1	Cross-sectional area of the effluent pipe	2.83×10^{-5}	m^2
K_W	Dissociation constant of water	10^{-14}	$(mol/L)^2$
α_A	Coefficients of total ion concentration of hydrochloric acid	1	-
α_B	Coefficients of total ion concentration of sodium hydroxide	1	-
η_A	Net proton hydroxide ion of acid stream	1.164	mol/L
η_B	Net proton hydroxide ion of base stream	1.164	mol/L
f	Friction coefficient of effluent pipe	1.05	$m^{0.5}/s$

3.1 Control performance

The proposed controller is applied to the bench-scale pH process for evaluating the setpoint tracking performance. Three case studies are considered, which are pH setpoints in the range of acid ($pH_{S,sp}$ = 5), in the neutralization point ($pH_{S,sp}$ = 7) and in the range of base ($pH_{S,sp}$ = 8). The level setpoints are set to be $h_{sp} = 30$ cm., and the following tuning parameters, $\beta_1 = 40$, $\beta_2 = 60$, $\lambda_1 = 5$, $\lambda_2 = 5$, $w_1 = 1000$ and $w_2 = 1$, are used in the tests. The initial condition of the pH and the tank level are within the range of 11 < pH < 11.2 and 18 < h < 22.2 cm. Note that there is no disturbance applied to the influent pH.

Figure 14-16 show the experimental results of the setpoint tracking for three case studies. It is clear that the proposed method successfully brings the outputs to the desired setpoints. In all cases, the responses of the outgoing pH have less oscillation at desired setpoints compared to those of the tank level because of 1 inch/ resolution limitation of the level sensor. The controller has been tuned to have the response time of the level faster than those of the pH. There is much easier for the process to adjust the pH when the level is close to the desired setpoint. The control of the outgoing pH at pH7 takes a longest time to reach the desired setpoint compared to other cases due to a high sensitivity of the pH characteristics around the neutralization point. The flow rate of the titrating stream slowly changes when the pH of the process is closed to the setpoint as shown in Figure 15.

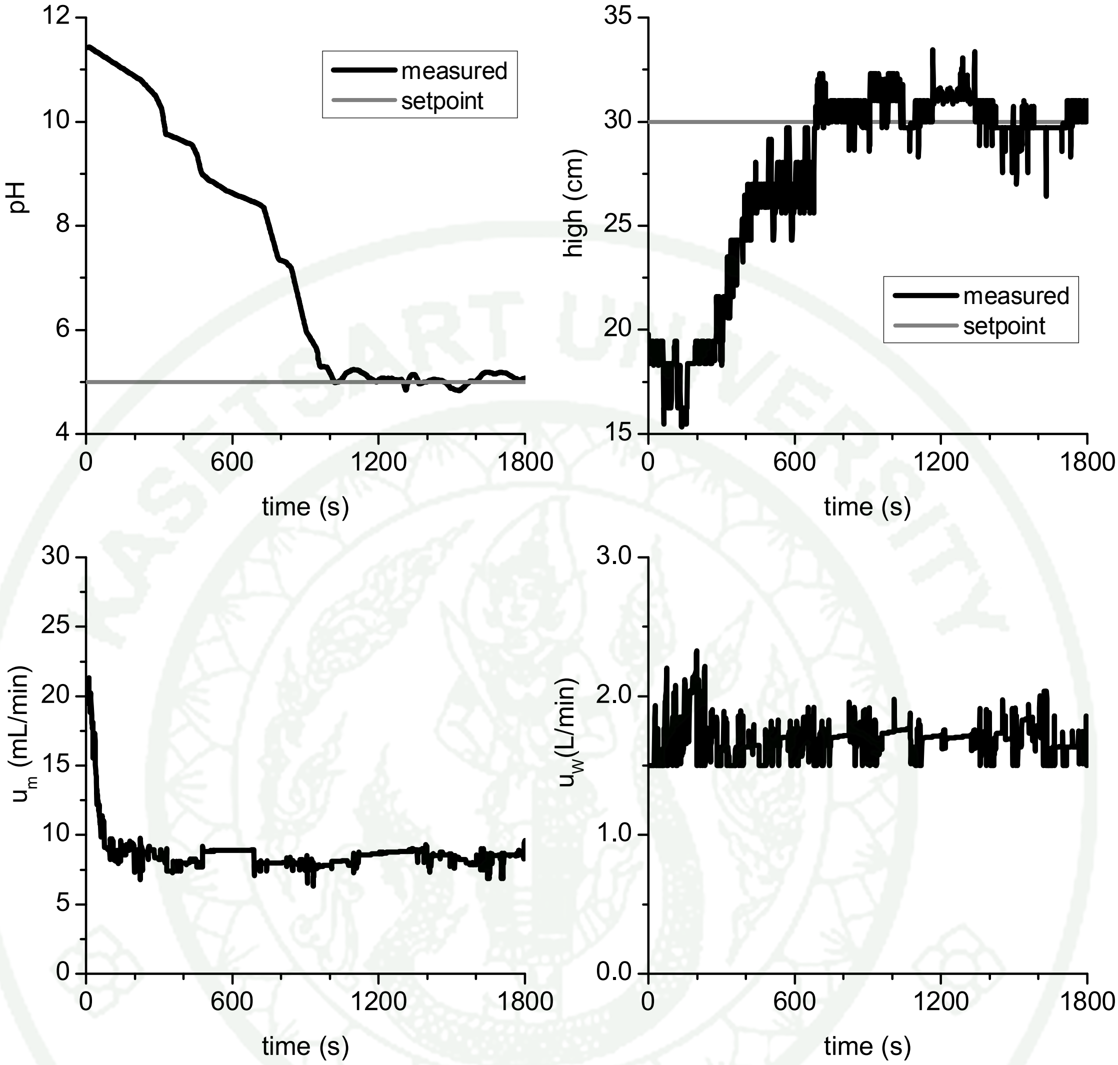


Figure 14 The process responses of the case of $pH_{S,sp} = 5$ under the proposed method

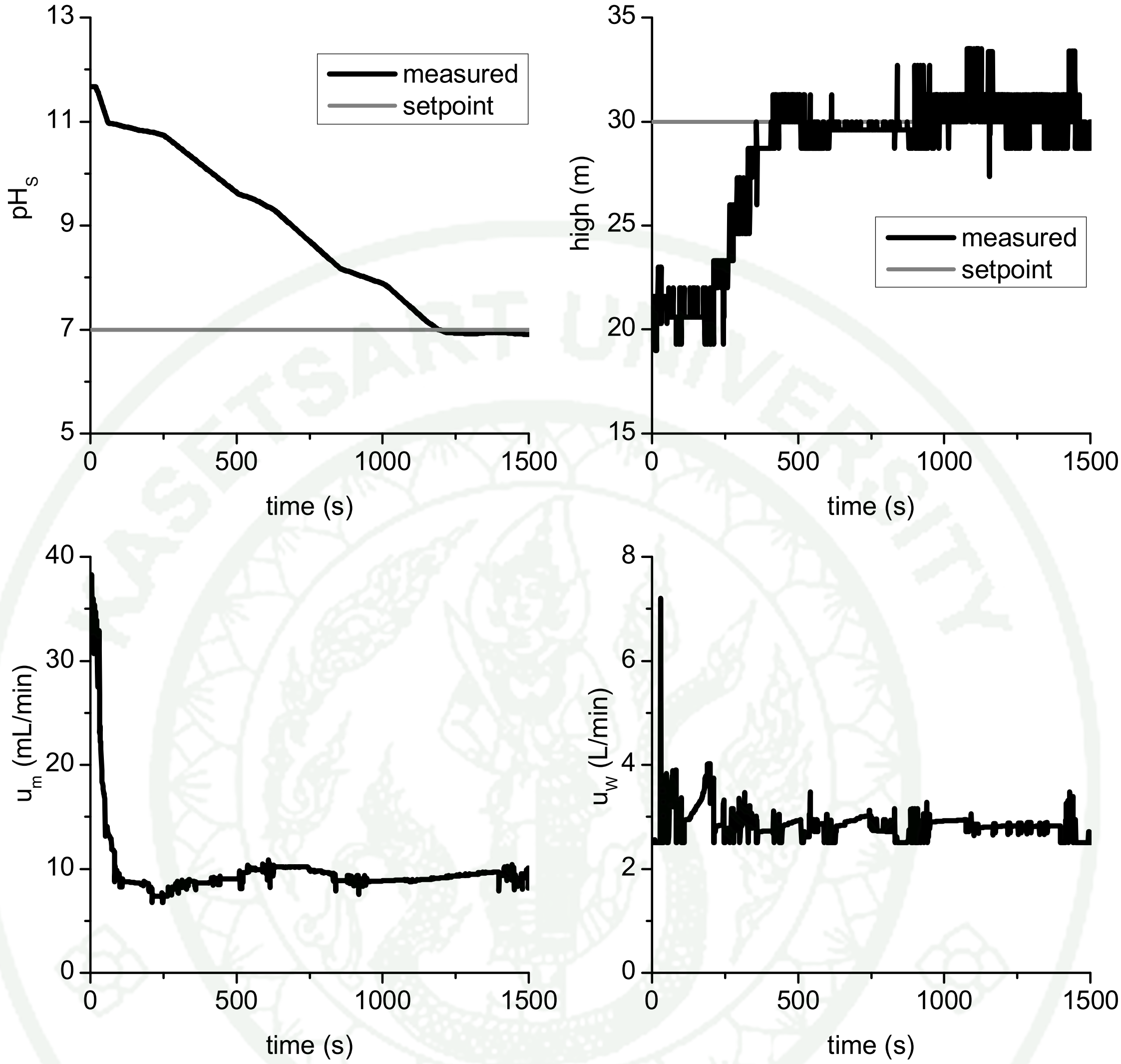


Figure 15 The process responses of the case of $pH_{S,sp} = 7$ under the proposed method

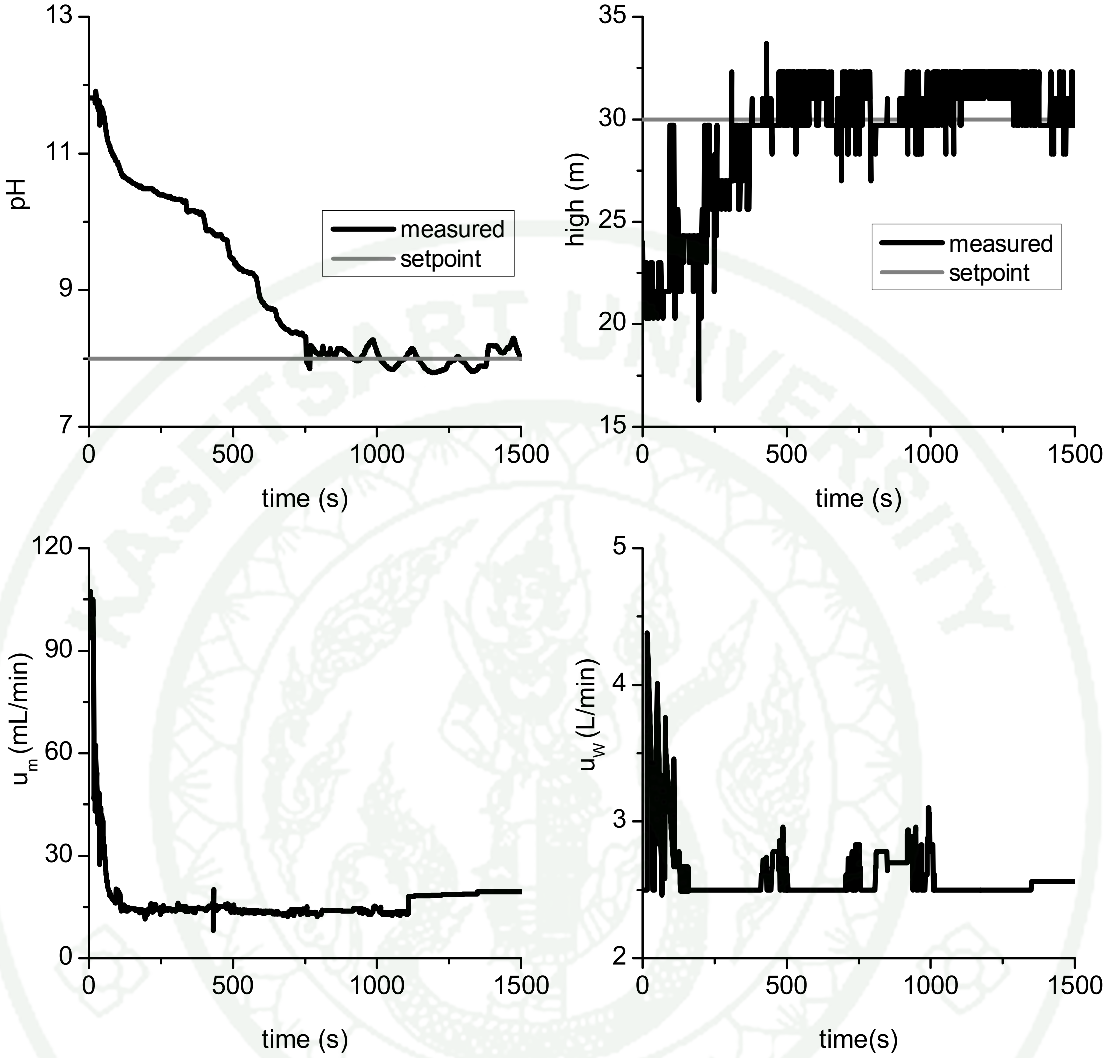


Figure 16 The process responses of the case of $pH_{S,sp}$ = 8 under the proposed method

3.2 Comparison the control performance with PI controller

To evaluate the performance and robustness of the proposed controller, a digital PI controller described in equation (27) is used for comparison purpose.

$$u_k = u_{k-1} + K_c\left[\left(y_{i,sp} - y_{i,k}\right) - \left(y_{i,sp} - y_{i,k-1}\right) + \frac{\Delta t\left(y_{i,sp} - y_{i,k}\right)}{\tau_i}\right] \tag{27}$$

where K_c denotes the proportional gain, τ_i denotes the integral gain, $u_{k\text{-}1}$ and $y_{k\text{-}1}$ are the input and the output at the previous step, u_k and y_k are the input and the output at the current step, y_{sp} is the output setpoint, and Δt is the time interval of the controller. The sets of parameter values, $\{K_c = 2.41, \tau_i = 306$ s$\}$, are applied for both setpoint tracking and disturbance rejection tests. The parameters of PI controller are calculated by using Internal Model Control tuning rule (Seborg *et al*., 2010). The tuning parameters of the proposed method using in the section 6.1 is also used in the test.

For the setpoint tracking, the control system is tested by setpoint tracking of two given sets of setpoints with no disturbance applied to the influent pH. The first set of desired setpoints is [$pH_{S,sp}$ = 7, h_{sp} = 30 cm.]. Then, the setpoint is changed to [$pH_{S,sp}$ = 7, h_{sp} = 31 cm.] at t = 1250 s. The results in Figure 17 show that the proposed controller successfully forces the outputs to the desired setpoints effectively while the outputs under the PI controller shows a high oscillation around the setpoints.

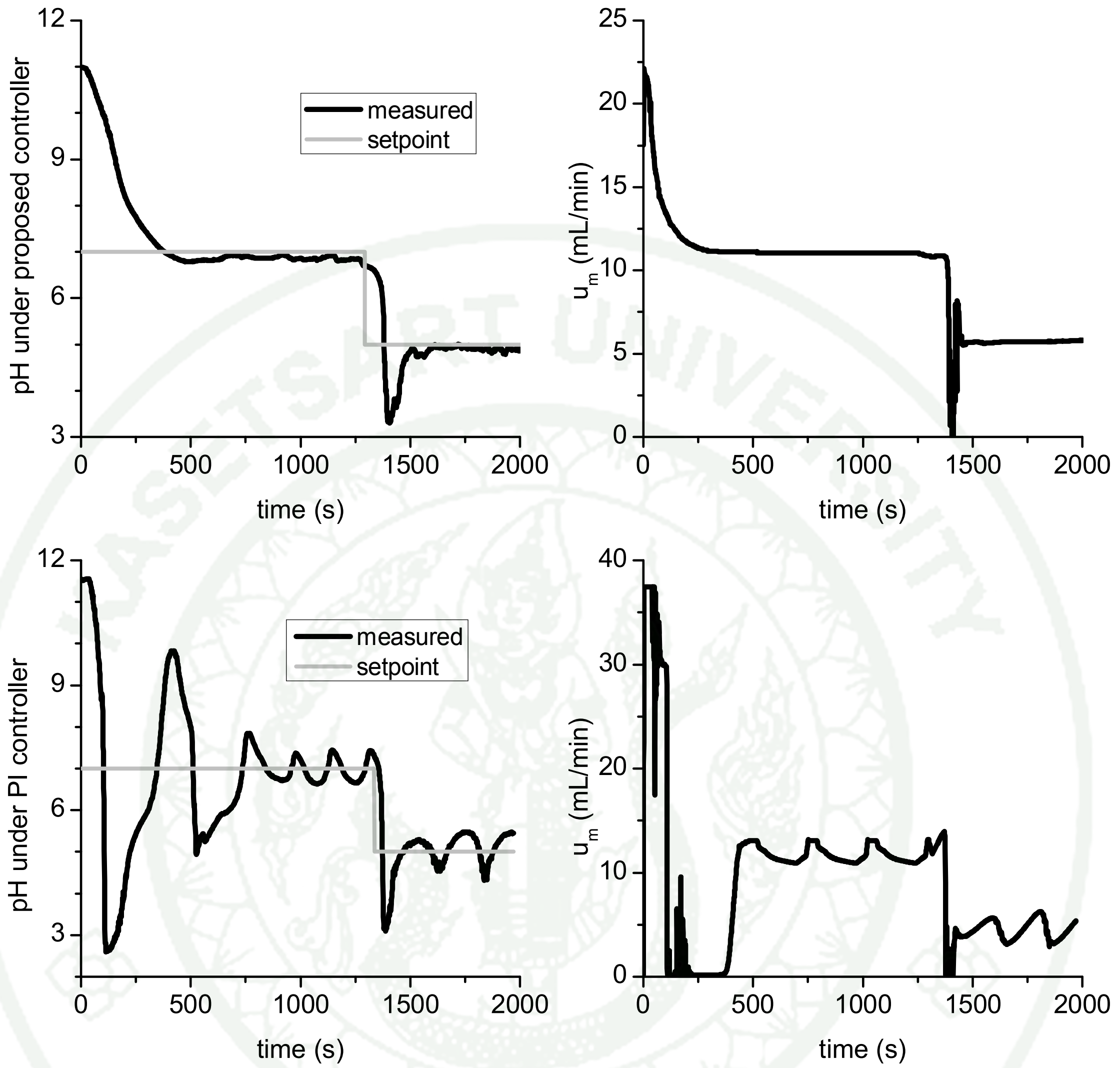


Figure 17 Process responses of the setpoint tracking test under the proposed method and the PI controller

The disturbance rejection performance is tested by introducing a pulse disturbance to the influent pH that creates the fluctuation pH (pH_w = 11-11.7). The process outputs are initially at [$pH(0)$, $h(0)$] = [11, 30 cm.] and controlled at [pH_{sp}, h_{sp}] = [5, 30 cm.]. The process response under the proposed controller and PI controller are illustrated in Figure 18 and 19, respectively. The results show that the proposed controller provides good results for stabilizing the pH in the mixing tank at pH = 5 in spite of a fluctuation in pH inlet. In contrast, the PI controller cannot force and stabilize the pH in the mixing tank to the desired setpoint.

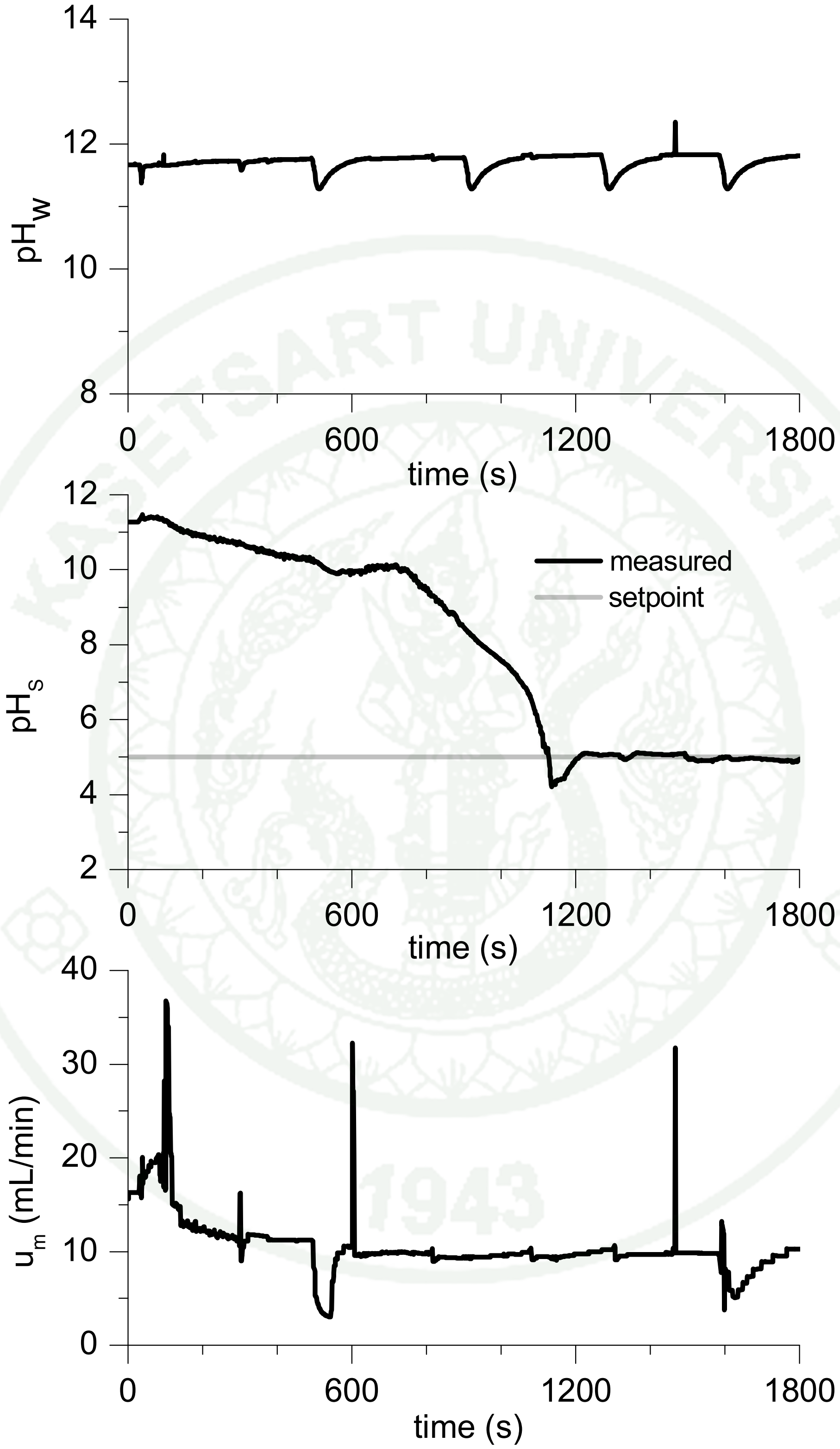


Figure 18 Process responses of the disturbance rejection test under the proposed method

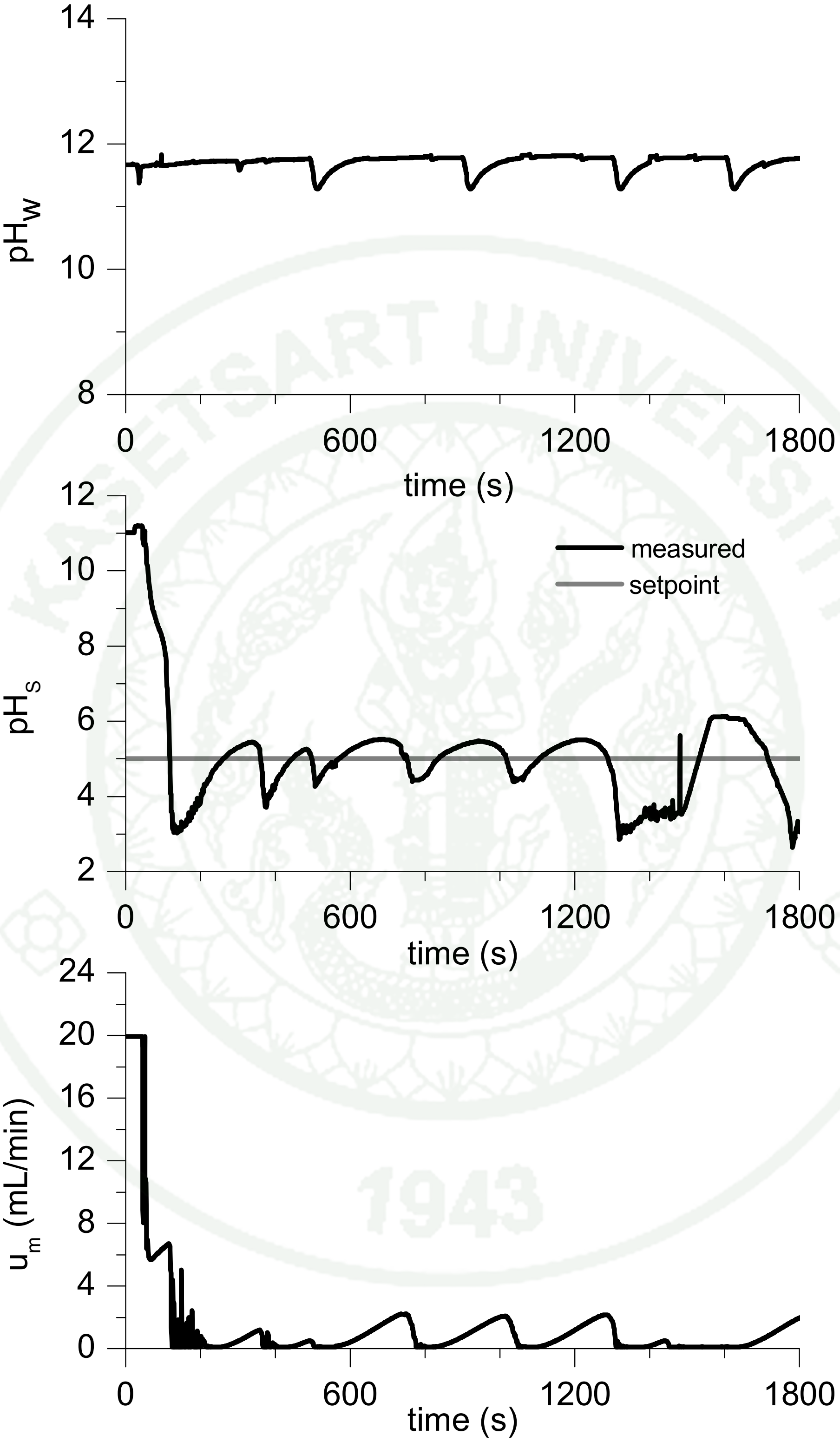


Figure 19 Process responses of the disturbance rejection test under the PI controller

Part II: Controller design and performance test applied to a bench-scale pH process with two mixing tanks in series

1. Process Description

Consider a pH process carried out using a series of continuous mixing tanks. Schematic diagram of the pH process is shown in Figure 20. An influent stream (F_W) has fed to a first mixing tank. Peristaltic pumps are used to feed acid and base streams (F_A and F_B) to control the outgoing pH of each tank around desired setpoints. The tank levels are controlled by adjusting their outgoing flows (F_{S1} and F_{S2}).

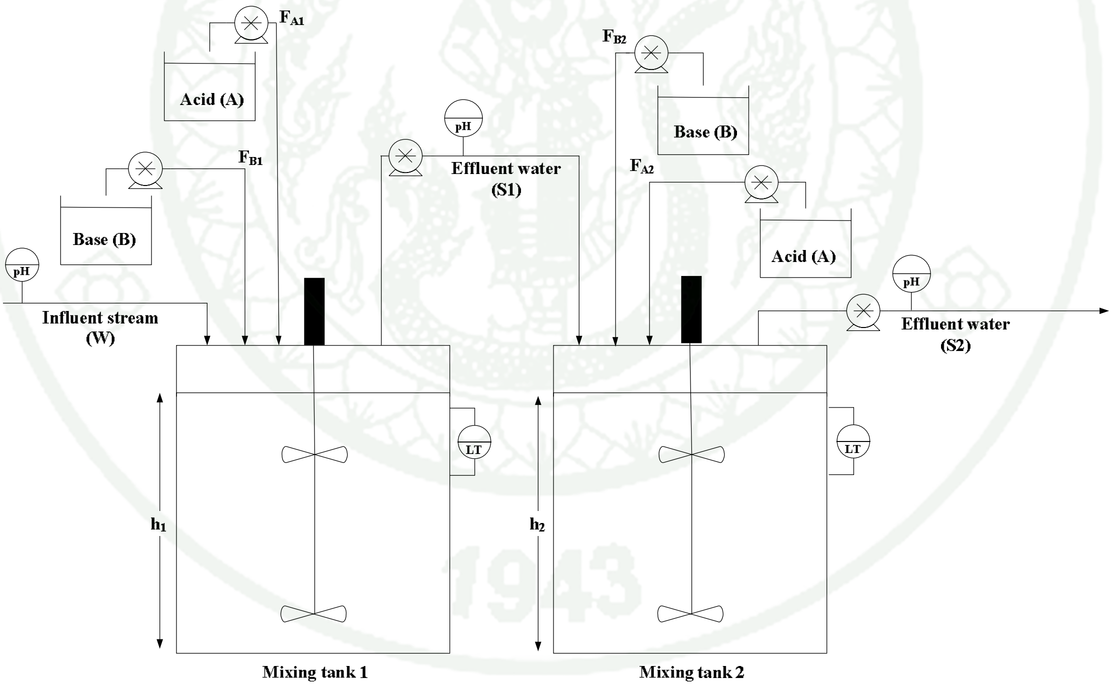


Figure 20 Schematic of a continuous pH process in series

Mathematical model of continuous pH process in series is developed with the following assumptions:

1) Cross sectional area of the mixing tanks are constant.

2) Densities of the influent and outflow of both mixing tanks are approximately equal ($\rho_W = \rho_{S1} = \rho_{S2} = \rho$).

3) Densities of acid and base streams are constant.

4) The tanks are well-mixed condition.

From the process shown in Figure 20, the material balance can be obtained as follows:

$$\begin{aligned} \frac{dh_1}{dt} &= \frac{\rho(F_W - F_{S1}) + \rho_{A1}F_{A1} + \rho_{B1}F_{B1}}{\rho A_{R1}} \\ \frac{dh_2}{dt} &= \frac{\rho(F_{S1} - F_{S2}) + \rho_{A2}F_{A2} + \rho_{B2}F_{B2}}{\rho A_{R2}} \end{aligned} \tag{28}$$

where h is the tank level, F_W is the influent stream, F_A and F_B are titrating streams of acid and base, and A_R is the cross-sectional area of the mixing tank.

The component balance of the process in the term of η defined in equation (16) can be described as follows:

$$\begin{aligned} \frac{d\eta_{S1}}{dt} &= \frac{(\eta_W + d_1)F_W + \eta_{A1}F_{A1} + \eta_{B1}F_{B1} - \eta_{S1}F_{S1}}{A_{R1}h_1} - \eta_{S1}\frac{\rho(F_W - F_{S1}) + \rho_{A1}F_{A1} + \rho_{B1}F_{B1}}{\rho A_{R1}h_1} \\ \frac{d\eta_{S2}}{dt} &= \frac{(\eta_{S1} + d_2)F_{S1} + \eta_{A2}F_{A2} + \eta_{B2}F_{B2} - \eta_{S2}F_{S2}}{A_{R2}h_2} - \eta_{S2}\frac{\rho(F_{S1} - F_{S2}) + \rho_{A2}F_{A2} + \rho_{B2}F_{B2}}{\rho A_{R2}h_2} \end{aligned} \tag{29}$$

where η_S, η_A, η_B are the net proton–hydroxide ions of the mixing tank, the acid stream and the base stream, and d is a disturbance of the dynamics of η_S. If a titrating stream has a high concentration, the net proton–hydroxide ions of acid and base stream in equation (16) can be approximated by following equation

For the high concentration acid:

$$\begin{aligned} \eta_{A1} &= \alpha_{A1} C_{A1} \\ \eta_{A2} &= \alpha_{A2} C_{A2} \end{aligned} \tag{30a}$$

For the high concentration base:

$$\begin{aligned} \eta_{B1} &= -\alpha_{B1} C_{B1} \\ \eta_{B2} &= -\alpha_{B2} C_{B2} \end{aligned} \tag{30b}$$

where α_A and α_B are the coefficients of total ion concentration of acid and base streams, C_A and C_B are the concentration of acid and base streams.

The control objective is to handle the pH and level in both mixing tank (pH_{S1}, pH_{S2}, h_1 and h_2) by manipulating the outgoing flows and acid/base stream of each tank (F_{S1}, F_{S2}, F_{A1}, F_{B1}, F_{A2}, and F_{B2}). In the implementation, we practically use either acid or base stream to adjust pH of a mixture in the mixing tanks corresponding to their desired setpoint ($pH_{S1,sp}, pH_{S2,sp}$). Therefore, the number of manipulated inputs of the process can be reduced to four variables, which are F_{S1}, F_{S2}, u_{m1}, and u_{m2}. The parameters, u_{m1} and u_{m2}, are the flow rates of a proper titrating stream fed in the mixing tank 1 and mixing tank 2. The selection of the titrating stream is based on the algorithm shown in Figure 21.

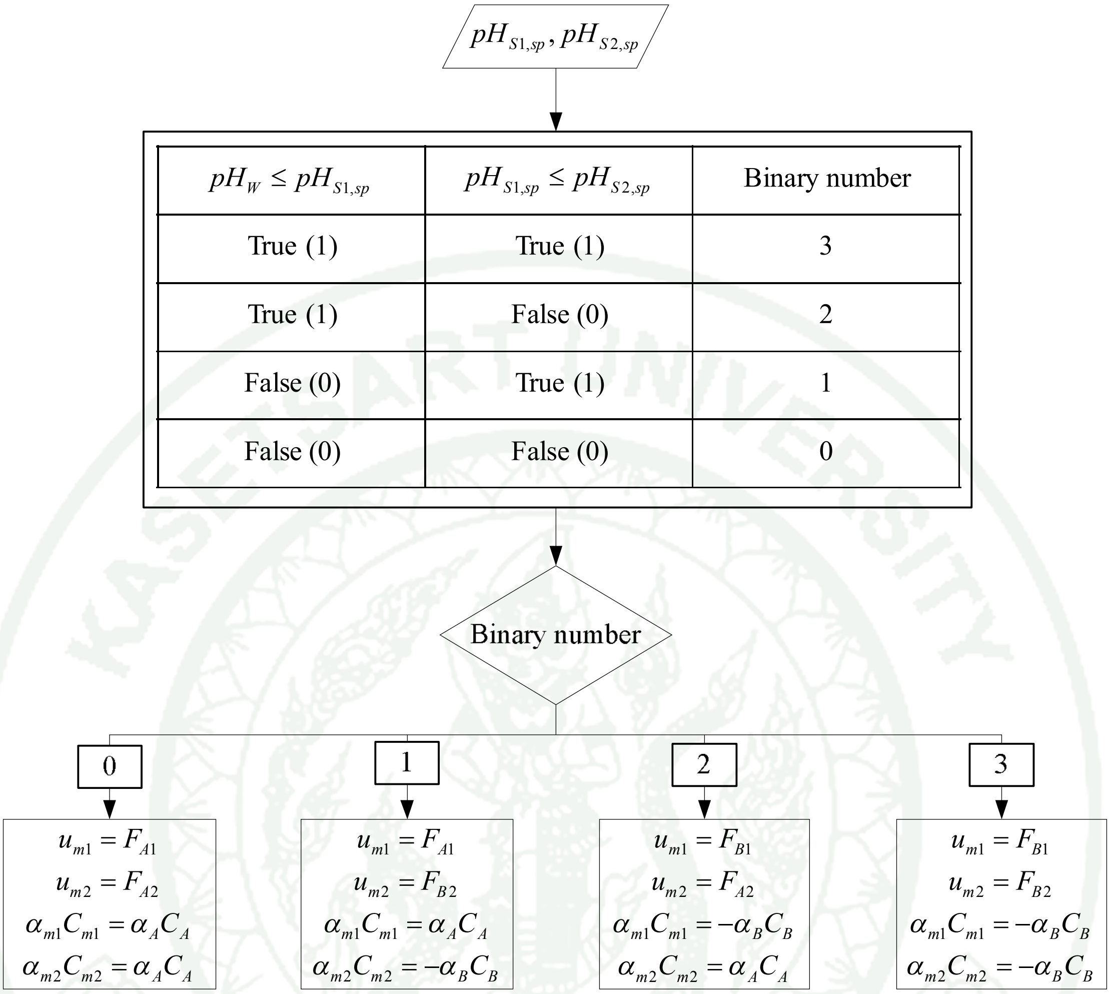


Figure 21 Flow diagram of the selection of the titrating streams for the continuous pH process in series.

From equation (28) and equation (29), the process model of the continuous pH process in series can be summarized as follows:

$$\frac{d\eta_{S1}}{dt} = \frac{(\eta_W + d_1)F_W + \eta_{m1}u_{m1} - \eta_{S1}F_{S1}}{A_{R1}h_1} - \eta_{S1}\frac{\rho(F_W - F_{S1}) + \rho_{m1}u_{m1}}{\rho A_{R1}h_1}$$
$$\frac{dh_1}{dt} = \frac{\rho(F_W - F_{S1}) + \rho_{m1}u_{m1}}{\rho A_{R1}}$$
$$\frac{d\eta_{S2}}{dt} = \frac{(\eta_{S1} + d_2)F_{S1} + \eta_{m2}u_{m2} - \eta_{S2}F_{S2}}{A_{R2}h_2} - \eta_{S2}\frac{\rho(F_{S1} - F_{S2}) + \rho_{m2}u_{m2}}{\rho A_{R2}h_2} \qquad (31)$$
$$\frac{dh_2}{dt} = \frac{\rho(F_{S1} - F_{S2}) + \rho_{m2}u_{m2}}{\rho A_{R2}}$$
$$y = [pH_{S1}\ h_1\ pH_{S2}\ h_2]^T, \quad u = [u_{m1}\ F_{S1}\ u_{m2}\ F_{S2}]^T, \quad d = [d_1, d_2]^T$$

The equation (14) can be used to convert the relation between pH and η.

2. Applying the proposed controller

2.1 Feedback controller

For implementing the input-output (I/O) linearization, the assumptions that the system in equation (6) is open-loop stable and internal dynamics (zero dynamics) is stable have been made. The close-loop output responses with the following form are requested:

$$\begin{gathered}(\beta_1 D + 1)^{r_1}\ y_1 = v_1 \\ \vdots \\ (\beta_4 D + 1)^{r_4}\ y_4 = v_4\end{gathered} \qquad (32)$$

where D is the differential operator (i.e. $D \equiv d/dt$), $r_1,...,r_4$ are the relative order of the controlled outputs, $y_1,...,y_4$, with respect to the manipulated inputs, $v_1,\ldots,v_4$ are the reference output setpoints and $\beta_1,\ldots,\beta_4$ are the tuning parameters that adjust the speed of the responses of the outputs, $y_1,\ldots,y_4$, respectively.

In this work, a minimized sum of squared errors between the requesting closed-loop output responses and the reference output setpoints is applied to calculate control actions (u) and to estimate unmeasured disturbances ($\bar{d}$). With the relative orders of the outputs, r_1 = 1, r_2 = 1, r_3 = 1 and r_4 = 1 an objective function of optimization problem can be formulated in a following form:

$$\begin{aligned} j_1 &= \min_{u,\bar{d}} \sum_{i=1}^{4} \left[w_i \left(\beta_i \dot{\eta}_{Si} + \eta_{Si} - v_i \right)^2 \right] \\ &\text{subject to} \\ &u_{lb} \le u \le u_{ub} \\ &\bar{d}_{lb} \le \bar{d} \le \bar{d}_{ub} \end{aligned} \tag{33}$$

where $\bar{d}$ is the vector of estimated state disturbances, the subscript ub and lb denote the upper and lower bounds respectively, and w is the vector of weighting factors. By substituting the process dynamics in equation (31) into equation (33), using a definition in equation (32) and settling all time derivatives of $\bar{d}$ to be zero, a solution of the constrained optimization problem can be represented by:

$$[u,\bar{d}] = \{u_{m1}, F_{S1}, u_{m2}, F_{S2}, \bar{d}_1, \bar{d}_2\} = \psi(\eta_{S1}, \eta_{S2}, h_1, h_2, v_1, v_2, v_3, v_4) \tag{34}$$

2.2 Closed-loop state estimator

In this work, an integral action is added in the developed state feedback to ensure the offset-free closed-loop response. The method is similar to the concept of an internal-model control. The estimated process outputs are calculated by using a closed-loop process model and the estimated disturbances obtained from equation (34) that is assumed as the known disturbances in the closed-loop model.

The closed-loop state estimator of the process is developed as shown in equation (35).

$$
\begin{aligned}
\frac{d\tilde{\eta}_{S1}}{dt} &= \frac{\left(\eta_W + \bar{d}_1\right)F_W + \alpha_{m1}C_{m1}\tilde{u}_{m1} - \tilde{\eta}_{S1}\tilde{F}_{S1}}{A_{R1}h_1} - \tilde{\eta}_{S1}\frac{\rho(F_W - \tilde{F}_{S1}) + \rho_{m1}\tilde{u}_{m1}}{\rho A_{R1}h_1} \\
\frac{d\tilde{h}_1}{dt} &= \frac{\rho(F_W - \tilde{F}_{S1}) + \rho_{m1}\tilde{u}_{m1}}{\rho_{S1}A_{R1}} \\
\frac{d\tilde{\eta}_{S2}}{dt} &= \frac{\left(\tilde{\eta}_{S1} + \bar{d}_2\right)\tilde{F}_{S1} + \alpha_{m2}C_{m2}\tilde{u}_{m2} - \tilde{\eta}_{S2}\tilde{F}_{S2}}{A_{R2}h_2} - \tilde{\eta}_{S2}\frac{\rho(\tilde{F}_{S1} - \tilde{F}_{S2}) + \rho_{m2}\tilde{u}_{m2}}{\rho A_{R2}h_2} \\
\frac{d\tilde{h}_2}{dt} &= \frac{\rho(\tilde{F}_{S1} - \tilde{F}_{S2}) + \rho_{m2}\tilde{u}_{m2}}{\rho A_{R2}}
\end{aligned}
\tag{35}
$$

where $\tilde{\eta}_S$ is the estimated net proton–hydroxide ions in the mixing tank, $\tilde{h}$ is the estimated mixing tank level, and $\tilde{u}_{m1}$, $\tilde{F}_{S1}$, $\tilde{u}_{m2}$, $\tilde{F}_{S2}$ are the estimated inputs obtained by solving the following optimization:

$$
\begin{aligned}
j_2 &= \min_{\tilde{u}} \sum_{i=1}^{4}\left[w_i\left(\beta_i\dot{\tilde{\eta}}_{Si} + \tilde{\eta}_{Si} - v_i\right)^2 \right] \\
&\text{subject to} \\
u_{lb} &\le \tilde{u} \le u_{ub}
\end{aligned}
\tag{36}
$$

where $\tilde{u}$ is the set of the estimated inputs $(\tilde{u}_{m1}, \tilde{u}_{m2}, \tilde{F}_{S1}, \tilde{F}_{S2})$. The control action obtained by solving the optimization problem in equation (36) can be represented by:

$$
\tilde{u} = \left\{\tilde{u}_{m1}, \tilde{F}_{S1}, \tilde{u}_{m1}, \tilde{F}_{S2}\right\} = \tilde{\psi}(\tilde{\eta}_{S1}, \tilde{\eta}_{S2}, \tilde{h}_1, \tilde{h}_2, v_1, v_2, v_3, v_4, \bar{d}_1, \bar{d}_2) \tag{37}
$$

2.3 Integrator

To compensate for the offset due to the effects of model–process mismatch and the error in the estimate states in each loop-time instant, the following first-order error dynamics are introduced:

$$\begin{aligned}
\dot{e}_1 &= \lambda_1(\tilde{\eta}_{S1} - \eta_{S1}) \\
\dot{e}_2 &= \lambda_2(\tilde{h}_1 - h_1) \\
\dot{e}_3 &= \lambda_3(\tilde{\eta}_{S2} - \eta_{S2}) \\
\dot{e}_4 &= \lambda_4(\tilde{h}_2 - h_2) \\
v_1 &= \eta_{S1,sp} + e_1 \\
v_2 &= h_{1,sp} + e_2 \\
v_3 &= \eta_{S2,sp} + e_3 \\
v_4 &= h_{2,sp} + e_4
\end{aligned} \tag{38}$$

where e, λ, and v are the error of the output, the positive constant of the first-order error dynamics, and the corrected setpoint of the output, respectively. The full description of control system are shown in Appendix B.

3. Experimental result

A real-time implementation of the proposed control system is carried out with the bench-scale pH process to evaluate setpoint tracking and disturbance rejection performances. The setpoint tracking is tested by applying a step change of the pH setpoint in the mixing tank 1 and mixing tank 2. The disturbance rejection is evaluated by introducing a sinusoidal disturbance to the measured influent pH of the mixing tank 1. A comparison of the proposed method with the PI controller under IMC tuning rules with a case of the step change in the pH setpoint is also discussed. The weighting parameters (w) for the optimization problem in equation (33) and equation (36) are chosen based on the ratio of the scale of the level to the scale of the net proton–hydroxide ions, which are w_1 = 1000, w_2 = 1, w_3 = 1000 and w_4 = 1, respectively. The experimental parameters of the bench-scale pH processes are given in Table 2.

Table 2 Parameter values of the bench-scale pH process with the two mixing tanks in series

Variable	Description	Value	Unit
ρ_A	Density of hydrochloric acid	1087.8	kg/m^3
ρ_B	Density of sodium hydroxide	1055.4	kg/m^3
ρ	Density of the influent/effluent	1000	kg/m^3
A_{R1}	Cross-sectional area of the first mixing tank	0.0254	m^2
A_{R2}	Cross-sectional area of the second mixing tank	0.0283	m^2
K_W	Dissociation constant of water	10^{-14}	$(mol/L)^2$
α_A	Coefficients of total ion concentration of hydrochloric acid	1	-
α_B	Coefficients of total ion concentration of sodium hydroxide	1	-
u_W	Influent flow rate	2.323	L/min

3.1 Setpoint tracking performance

The control system is tested for the tracking of two given sets of the setpoints. The first set of desired setpoints is [$pH_{S1,sp}$, $h_{1,sp}$, $pH_{S2,sp}$, $h_{2,sp}$ = 8, 31cm, 10, 31cm]. Then, the setpoint is changed to [$pH_{S1,sp}$, $h_{1,sp}$, $pH_{S2,sp}$, $h_{2,sp}$ = 8, 31cm, 4, 31cm] at t = 1000 s. In practice, the process is much easier to adjust the pH when the level is close to the desired setpoint. Therefore, the controller needs to be tuned to have the response time of the level faster than those of the pH. The following tuning parameter values, { β_1 = 0.3s, β_2 = 1s, β_3 = 0.3s, β_4 = 1s}, are applied in the test and the process conditions are summarized in Table 3. The experimental results of the process responses are illustrated in Figure 22 and Figure 23.

Table 3 Parameter values used in the setpoint tracking test.

Tank	1st Setpoint	2nd Setpoint	Tuning parameters	Initial condition	Acid/ base concentration (mol/L)
1	$pH_{S1,sp}=8$ $h_{1,sp}=31\text{cm}$	$pH_{S1,sp}=8$ $h_{1,sp}=31\text{cm}$	$\beta_1=0.3\text{s}$ $\beta_2=1\text{s}$	$pH_{S1}(0)=4$ $h_1(0)=19\text{cm}$ $\text{u}_{m1}(0)=0\frac{\text{mL}}{\text{min}}$ $\text{u}_{S1}(0)=0.8\frac{\text{L}}{\text{min}}$	$C_A=0.00758$ $C_B=0.00741$
2	$pH_{S2,sp}=10$ $h_{2,sp}=31\text{cm}$	$pH_{S2,sp}=4$ $h_{2,sp}=31\text{cm}$	$\beta_3=0.3\text{s}$ $\beta_4=1\text{s}$	$pH_2(0)=4$ $h_2(0)=19\text{cm}$ $\text{u}_{m2}(0)=0\frac{\text{mL}}{\text{min}}$ $\text{u}_{S2}(0)=0.8\frac{\text{L}}{\text{min}}$	$C_A=0.00758$ $C_B=0.00741$

The proposed controller successfully forces the pH of the mixing tanks to track the given setpoints. It is capable of selecting the proper titrating stream during the real-time operation. The base stream is initially chosen to be the titrating stream for the mixing tank 2 before switching to the acid stream due to the change of the pH setpoint as illustrated in Figure 23. Figure 22 shows that the proposed controller compensates the uncertainties in measured pH and level signals despite unchanged setpoints of mixing tank 1. The control action of the tank 2 has a bit aggressive compared with those in the tank 1 due to the consequent effect of disturbance rejection from the mixing tank 1.

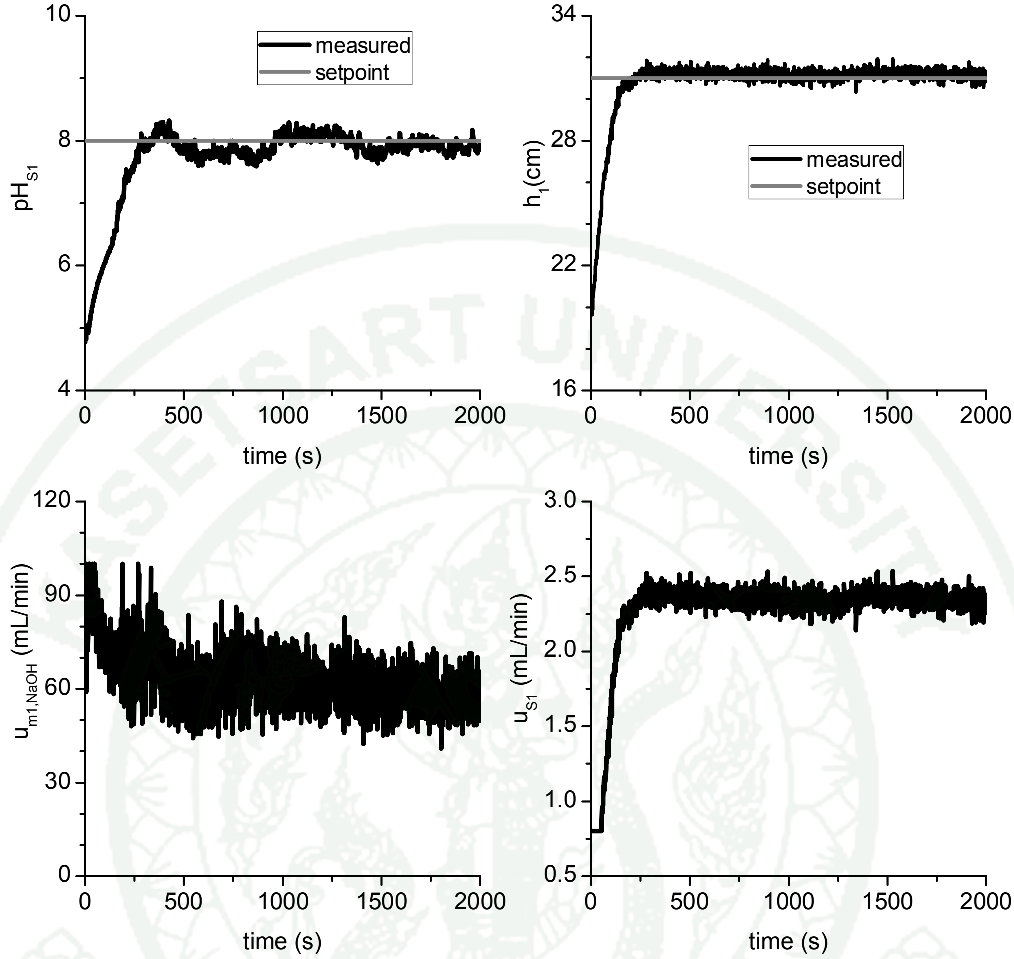


Figure 22 Process responses of the mixing tank 1 with a step change of the pH setpoint under the proposed method

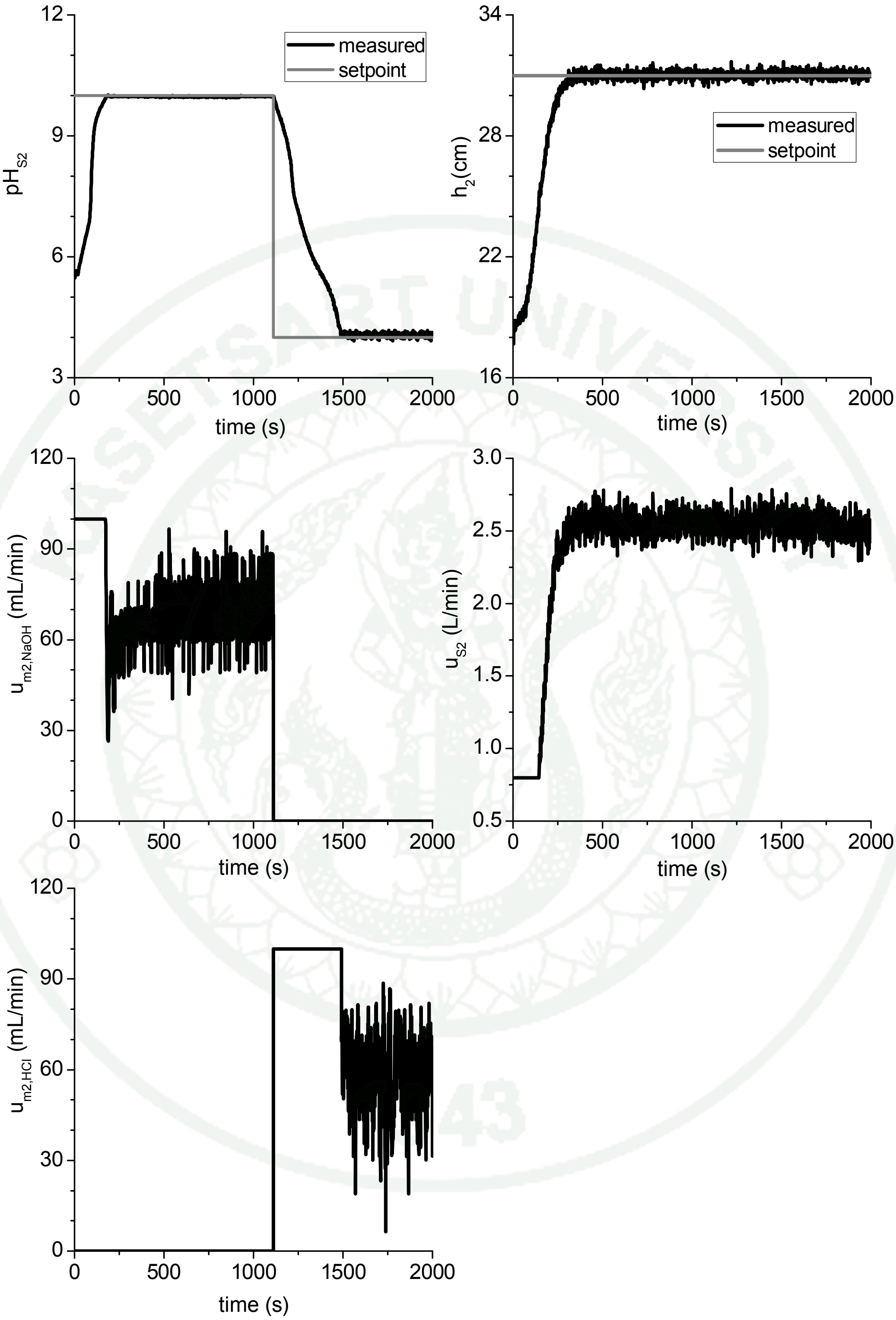


Figure 23 Process responses of the mixing tank 2 with a step change of the pH setpoint under the proposed method

3.2 Disturbance rejection performances

The pH fluctuation of the influent stream (F_W) due to the turbulent mixing effect of the feed flow and the sensitivity of pH probe is introduced in this example. The pH disturbance of a sine function, $d = \sin(10t)$, is added to the measured pH value of the influent at $t = 900\,s$. The set of tuning parameters in section 3.1 is also applied to this test and the initial conditions are summarized in Table 4.

Table 4 Parameter values used in the disturbance rejection test

Tank	Setpoint	Tuning parameters	Initial condition	Acid/ base concentration (mol/L)
1	$pH_{S1,sp} = 6$ $h_{1,sp} = 31$ cm	$\beta_1 = 0.3$ s $\beta_2 = 1$ s	$pH_{S1}(0) = 8$ $h_1(0) = 19$ cm $u_{m1}(0) = 0$ mL/min $u_{S1}(0) = 0.8$ L/min	$C_A = 0.00758$ $C_B = 0.00741$
2	$pH_{S2,sp} = 9$ $h_{2,sp} = 31$ cm	$\beta_3 = 0.3$ s $\beta_4 = 1$ s	$pH_2(0) = 8$ $h_2(0) = 19$ cm $u_{m2}(0) = 0$ mL/min $u_{S2}(0) = 0.8$ L/min	$C_A = 0.00758$ $C_B = 0.00741$

The experimental results are showed in Figure 24 and Figure 25. As can be seen, the proposed method successfully rejects the uncertainty in the influent pH. The acid stream for the mixing tank 1 has an aggressive action to compensate the sinusoidal disturbance of the influent pH as shown in Figure 24. The change of the titrating flow of the mixing tank 1 is relatively small compared to the outgoing flow. Therefore, there was no significant change in the mixing tank 2 as shown in Figure 25.

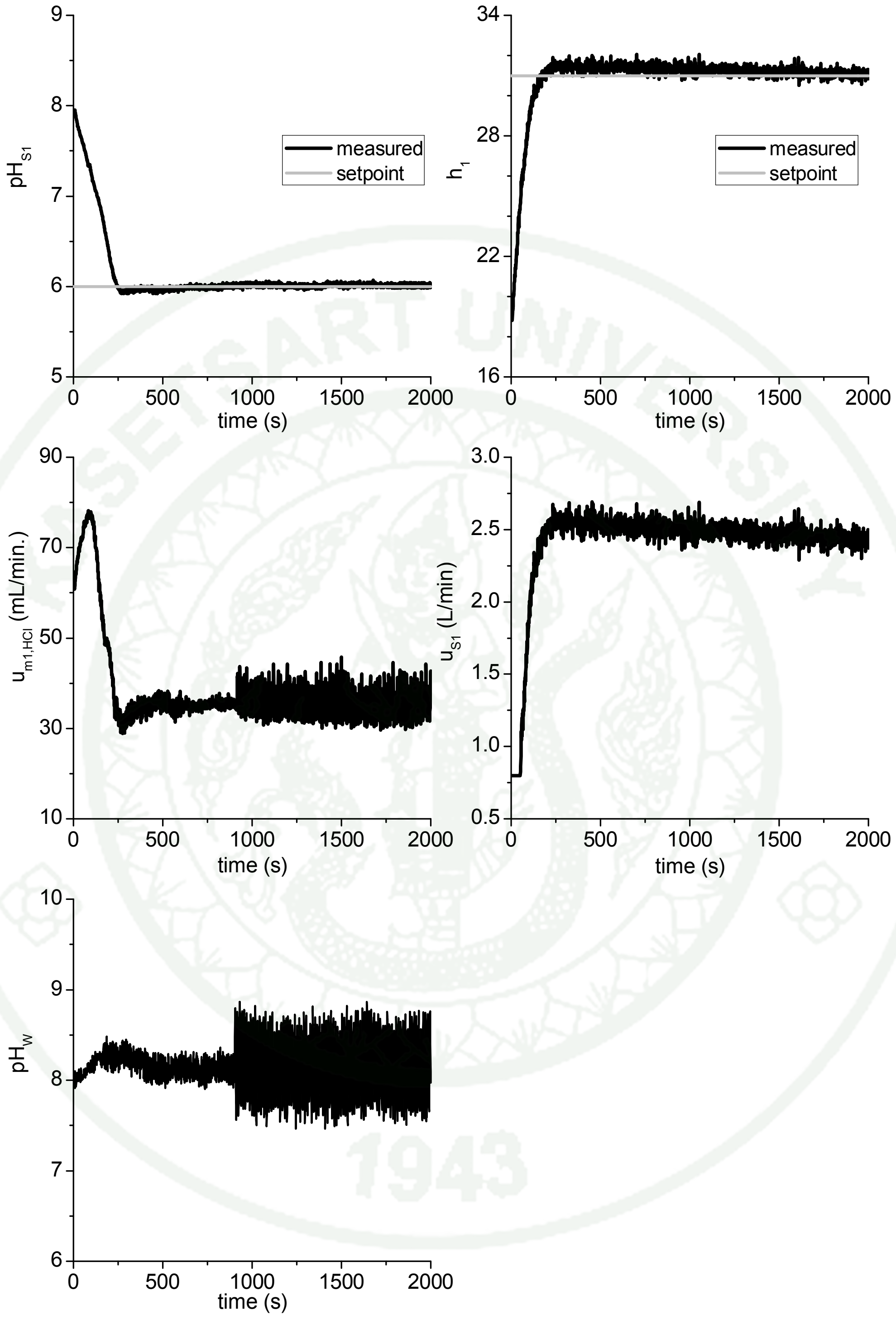


Figure 24 Process responses of the mixing tank 1 with a sinusoidal disturbance in the influent pH under the proposed method

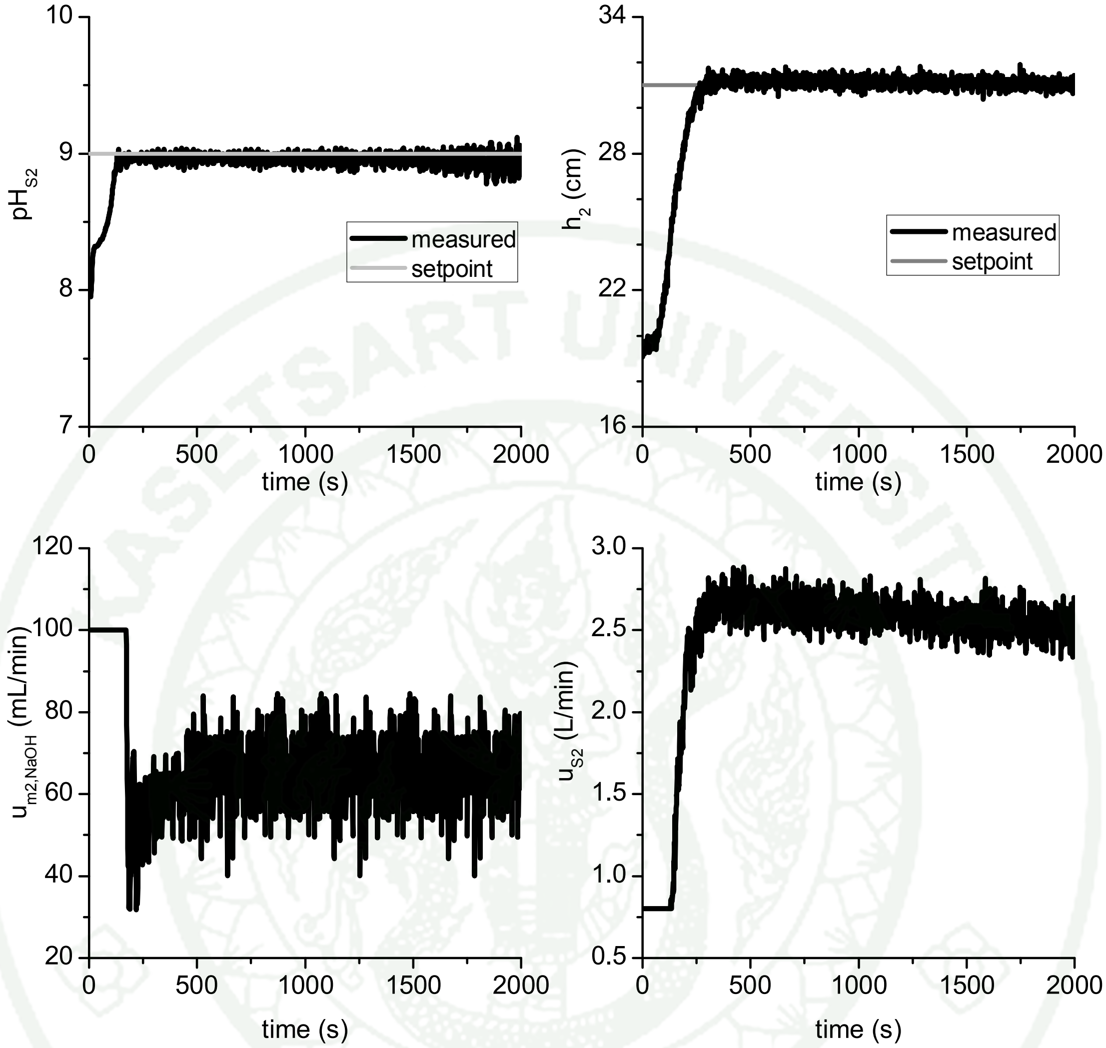


Figure 25 Process responses of the mixing tank 2 with a sinusoidal in the influent pH under the proposed method

3.3 Comparison of the control performance with the PI controller

In order to evaluate the control performance, a digital PI controller described in equation (39) was used for comparison purpose.

$$u_k = u_{k-1} + K_c\left[\left(y_{i,sp} - y_{i,k}\right) - \left(y_{i,sp} - y_{i,k-1}\right) + \frac{\Delta t\left(y_{i,sp} - y_{i,k}\right)}{\tau_i}\right] \quad (39)$$

where K_c denotes the proportional gain, τ_i denotes the integral gain, u_{k-1} and y_{k-1} are the input and the output at the previous step, u_k and y_k are the input and the output at the current step, y_{sp} is the output setpoint, and Δt is the time interval of the controller. The PI controller in equation (39) is applied formulating controllers of both tanks, of which the manipulated inputs and controlled outputs are $u_1 = u_{m1}$, $u_2 = u_{S1}$, $u_3 = u_{m2}$, $u_4 = u_{S2}$, $y_1 = pH_{S1}$, $y_2 = h_1$, $y_3 = pH_{S2}$, and $y_4 = h_2$. The PI controllers are tuned based on the IMC method (Seborg *et al.*,2010; Marlin,2000). The PI control actions are updated in every second (Δt = 1 s). The parameters and initial condition using in the test are given in Table 5. In the comparison, the objective is to force the pH of the mixing tank 1 to track the desired setpoints at t = 1000 s. The proposed controller also needs to stabilize the mixing tank 2 at the current setpoints while the step change of the first tank is occurred.

Table 5 Parameter values used in the comparison test

Parameter	Tank 1	Tank 2
1st Setpoint	$pH_{S1,sp} = 9$ $h_{1,sp} = 31\text{cm}$	$pH_{S2,sp} = 7$ $h_{2,sp} = 31\text{cm}$
2nd Setpoint	$pH_{S1,sp} = 10$ $h_{1,sp} = 31\text{cm}$	$pH_{S2,sp} = 7$ $h_{2,sp} = 31\text{cm}$
Tuning parameters of proposed method	$\beta_1 = 0.3\text{s}$ $\beta_2 = 1\text{s}$	$K_{c1} = 1\times10^{-2}\,\text{mL/s}$ $\tau_1 = 8.6\text{s}$ $K_{c2} = -6.17\text{mL/s}\cdot\text{cm}$ $\tau_2 = 18.346\text{s}$
Tuning parameters of PI controller	$\beta_3 = 0.3\text{s}$ $\beta_4 = 1\text{s}$	$K_{c3} = 1\times10^{-2}\,\text{mL/s}$ $\tau_3 = 8.6\text{s}$ $K_{c4} = -6.17\text{mL/s}\cdot\text{cm}$ $\tau_4 = 18.346\text{s}$
Initial condition	$pH_{S1}(0) = 7$ $h_1(0) = 19$ cm $\text{u}_{m1}(0) = 0$ mL/min $\text{u}_{S1}(0) = 0.8$ L/min	$pH_2(0) = 7$ $h_2(0) = 19$ cm $\text{u}_{m2}(0) = 0$ mL/min $\text{u}_{S2}(0) = 0.8$ L/min
Acid/ base concentration (mol/L)	$C_A = 0.00758$ $C_B = 0.00741$	$C_A = 0.00758$ $C_B = 0.00741$

The experimental results of the process responses under the proposed controller and under the PI controller are shown in Figures 26-27 and Figures 28-29, respectively. From Figures 26-27, it is clear that the proposed controller successfully to force the outputs of the mixing tank 1 to desired setpoints and to stabilize the process outputs of the mixing tank 2 simultaneously. The proposed controller provides very impressive results for stabilizing the pH of the second mixing tank at pH = 7 in spite of a large disturbance induced by the first tank. The process settled to the desired setpoints after 150 seconds. In contrast, the PI controller take approximately 1000 seconds to settle the pH in the mixing tank 1 and it cannot stabilize the pH of the mixing tank 2. The PI controller will provide good control performance within a small region. It is easy to drop in controllability when a large disturbance is present.

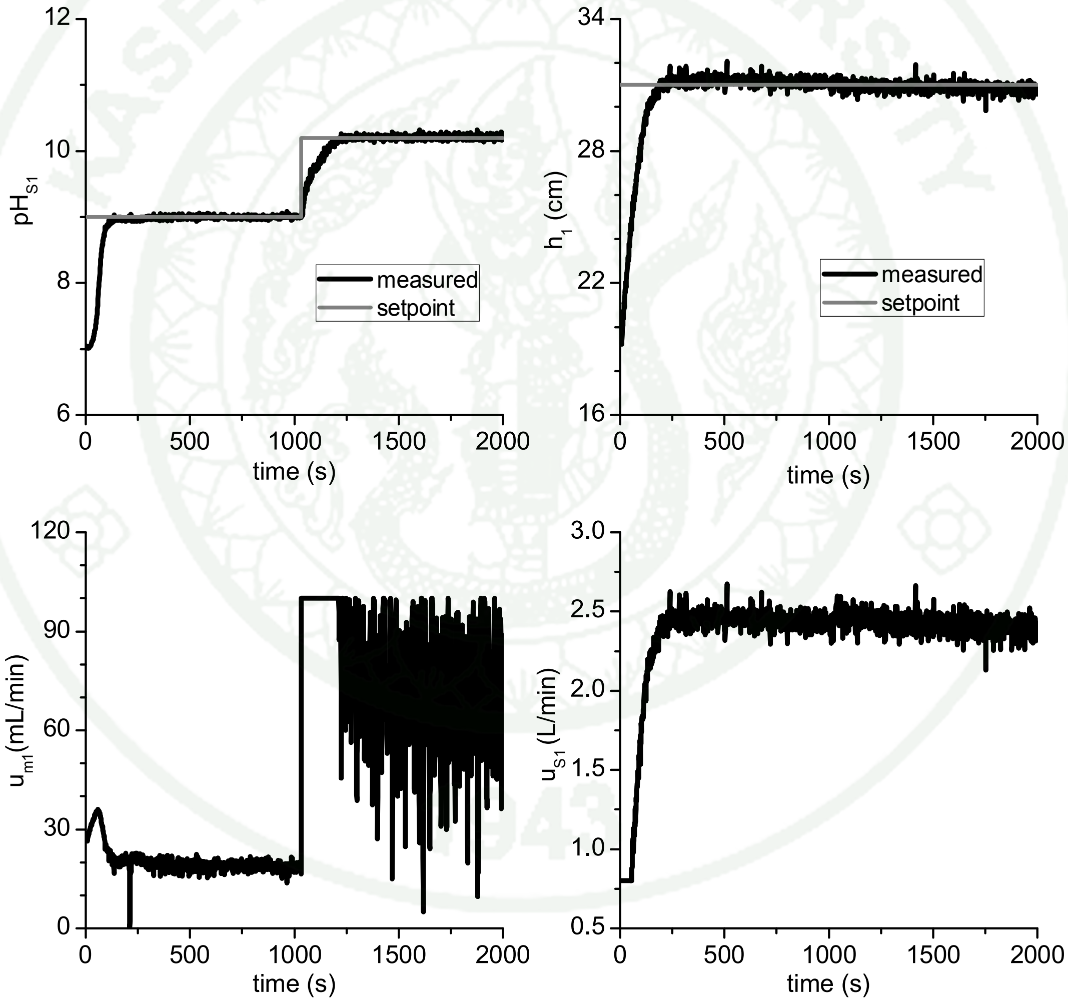


Figure 26 Process responses of the mixing tank 1 under the proposed method for a comparison case

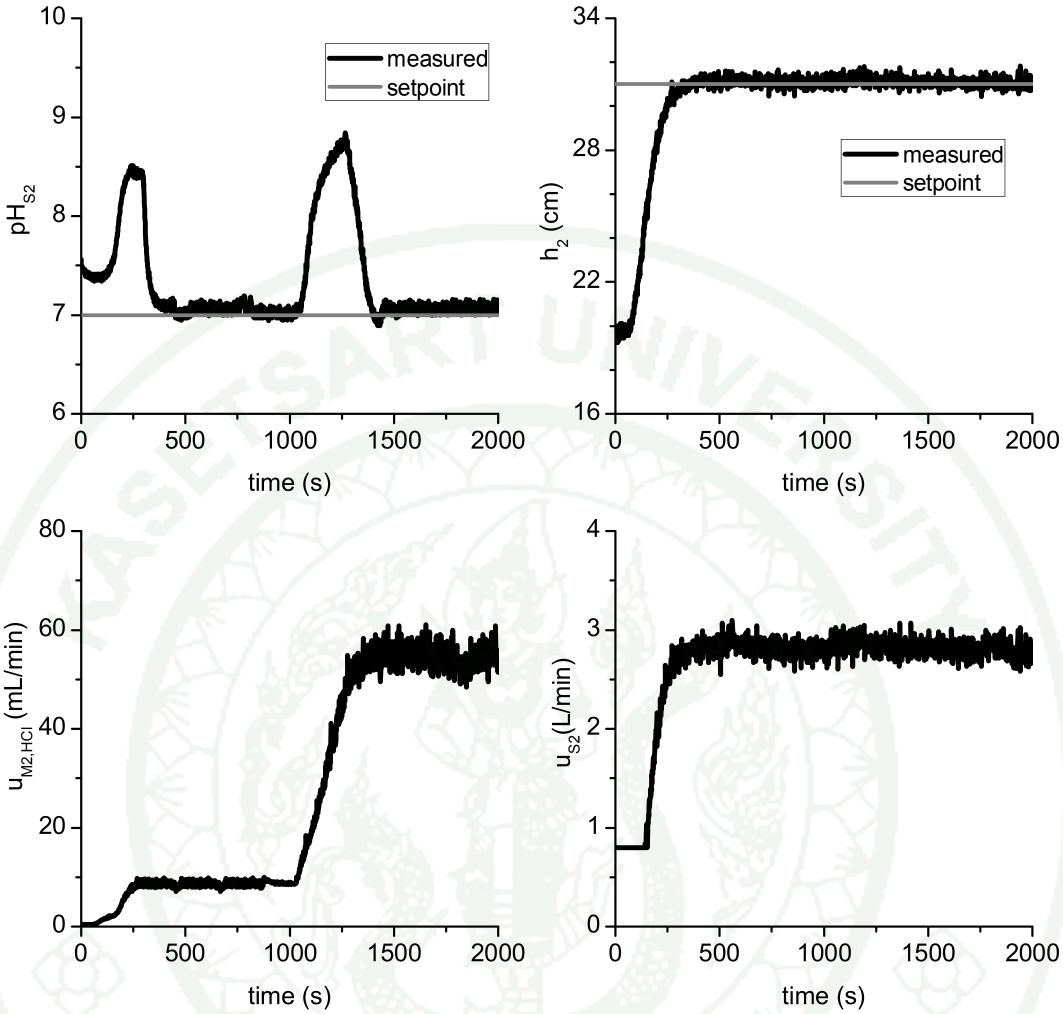


Figure 27 Process responses of the mixing tank 2 under the proposed method for comparison case

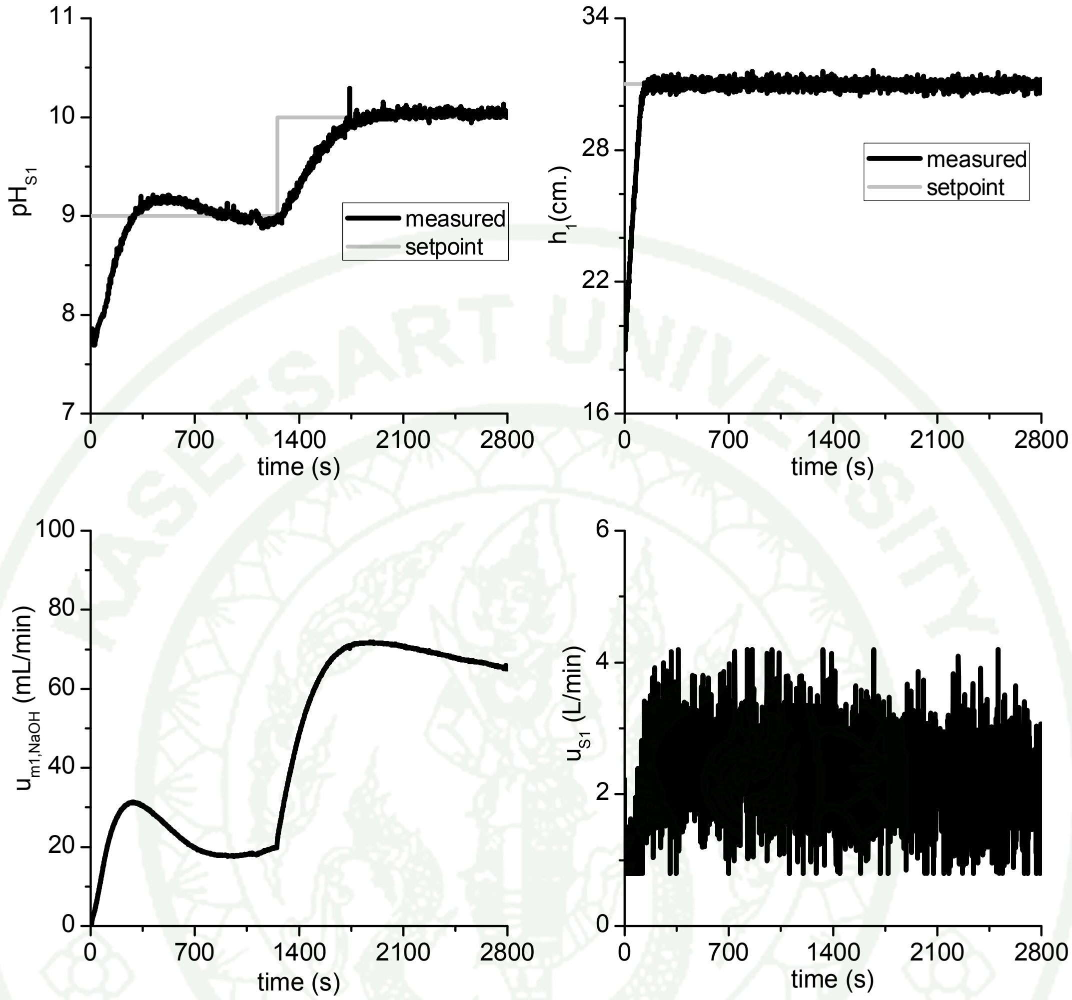


Figure 28 Process responses of the mixing tank 1 under the PI controller for comparison case

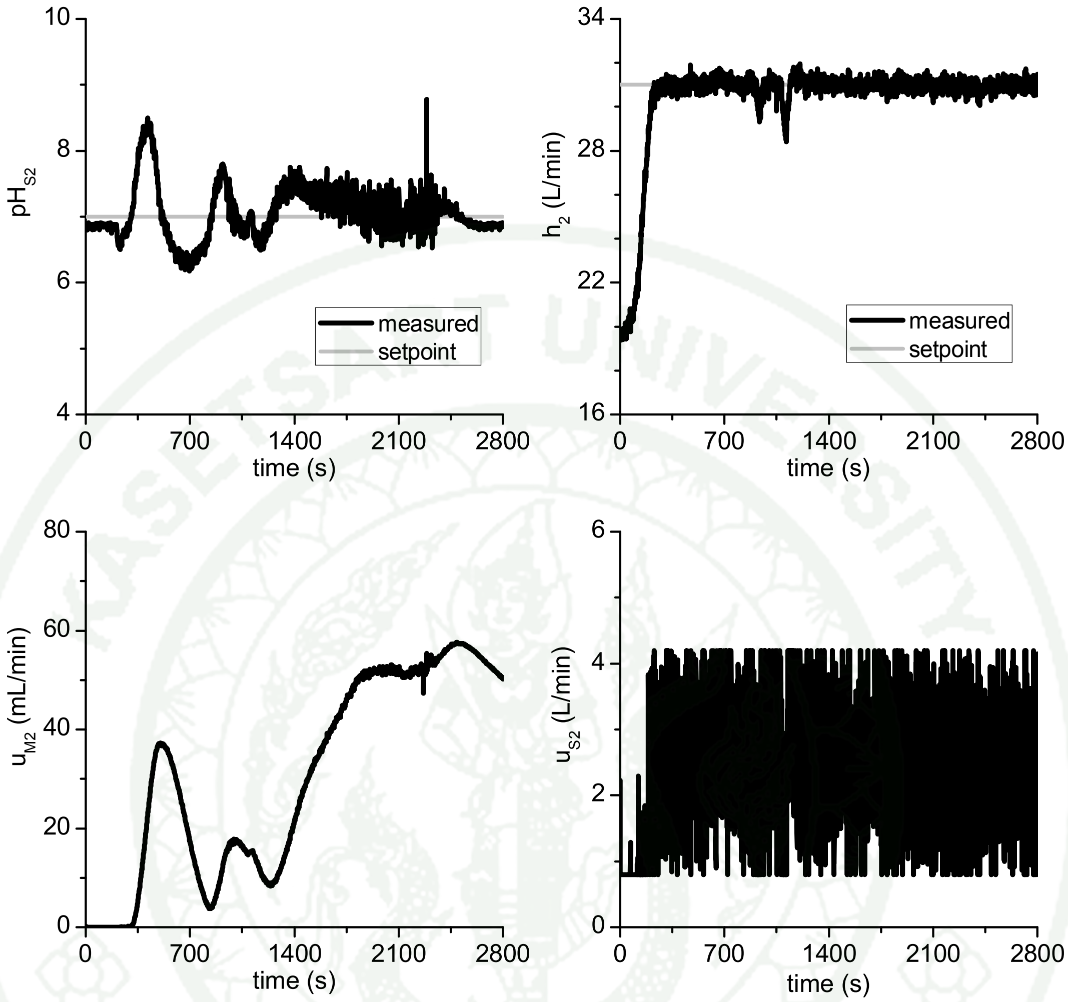


Figure 29 Process responses of the mixing tank 2 under the PI controller for comparison case

CONCLUSION AND RECOMMENDATION

Conclusion

A development of a control method for a pH process with a single mixing tank and two mixing tanks in series was presented in this work. The method consists of a feedback controller, a closed-loop state estimator with disturbance prediction, and a first-order integrator. An input-output linearization controller in the form of the optimization combining with the closed-loop state estimator and the first-order integrator is proposed to handle the process with the disturbances.

The proposed control system was investigated in the bench-scale pH process with two configurations, a single mixing tank scheme and two mixing tanks in series, to test the control performance. The experimental results of the pH process of both configurations show that the proposed control scheme successfully forces the outgoing pH and level of the mixing tank to the given setpoints asymptotically, whether changes in the operating range of pH and the level setpoints or the existence of the disturbances in influent pH. The proposed control system is effective for the pH process in series and provides good performance compared to the basic PI controller. It provides a strong capability to reject the effects of internal interaction and external uncertainties.

The output responses of the pH process with the configuration of two mixing tanks in series can reach the setpoints faster than those of a single mixing tank because of less coupled effect between pH and level. The process that manipulated input is the inlet flow to the tank has highly nonlinear dynamics than those of the outlet flow.

Recommendation

In this work, the control method for pH process with two configurations, a single mixing tank and two mixing tanks in series, has been developed. Although the proposed control method is successfully handle the bench-scale pH process, some points of improvement need to be considered in future studies. There are only two configurations of the bench-scale process that has been focused in this study. However, other configurations such as system with a pH-inline before a mixing tank, a system with multiple mixing tanks and a system with time delays in pH/level sensors are also needed to have further studies. In addition, the real-time signal filtering for the developed control system is also an interested topic to improve the aggressive of the control action. We have study a continuous pH process in series of the acetic acid/sodium hydroxide, the weak acid-strong base system. Preliminary results have been reported in Appendix D. The further study of the system and the study of other buffered pH processes, i.e., strong acid-weak base, weak acid-strong base and polyprotic/polyhydroxyl pH system are also interesting researches.

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APPENDICES

Appendix A

Control system for pH process with single mixing tank

The equations of the control system for the bench-scale pH process are described as follows:

- The objective function of feedback controller

$$j = \min_{F_W, u_m, \bar{d}} \left[\left(\beta_1 \left(\begin{array}{l} \dfrac{(\eta_W + d)F_W + \alpha_m C_m u_m - \eta_S g\sqrt{h}A_t}{A_R h} \\ -\eta_S \dfrac{\rho F_W + \rho_m u_m - \rho g\sqrt{h}A_t}{\rho A_R h} \end{array} \right) + \eta_S - v_1 \right)^2 + \left(\beta_2 \frac{\rho F_W + \rho_m u_m - \rho\beta\sqrt{h}A_t}{\rho A_R} + h - v_2 \right)^2 \right] \quad (A.1)$$

The closed loop state estimator

- The objective function of closed loop state estimator

$$j = \min_{\tilde{F}_W, \tilde{u}_m} \left[\left(\beta_1 \left(\begin{array}{l} \dfrac{(\eta_W + \bar{d})\tilde{F}_W + \alpha_m C_m \tilde{u}_m - \tilde{\eta}_S g\sqrt{\tilde{h}}A_t}{A_R \tilde{h}} \\ -\tilde{\eta}_S \dfrac{\rho \tilde{F}_W + \rho_m \tilde{u}_m - \rho g\sqrt{\tilde{h}}A_t}{\rho A_R h} \end{array} \right) + \tilde{\eta}_S - \upsilon_1 \right)^2 + \left(\beta_2 \frac{\rho \tilde{F}_W + \rho_m \tilde{u}_m - \rho\beta\sqrt{\tilde{h}}A_t}{\rho A_R} + \tilde{h} - \upsilon_2 \right)^2 \right] \quad (A.2)$$

- The first order integrator

$$\begin{aligned} \dot{e}_1 &= \lambda_1(\eta_S - \tilde{\eta}_S) \\ \dot{e}_2 &= \lambda_2(\tilde{h} - h) \\ v_1 &= \eta_{sp} + e_1 \\ v_2 &= h_{sp} + e_2 \end{aligned} \quad (A.3)$$

Appendix B

Control system for pH process with two mixing tanks in series

The equations of the control system for the bench-scale pH process with two mixing tanks in series are described as follows:

- The objective function of feedback controller

$$j_1 = \min_{u_{m1},u_{m2},F_{S1},F_{S2},\bar{d}_1,\bar{d}_1} \left[\begin{array}{l} \left(\beta_1 \left(\begin{array}{c} \dfrac{\left(\eta_W + \bar{d}_1\right) F_W + \alpha_{m1} C_{m1} u_{m1} - \eta_{S1} F_{S1}}{A_{R1} h_1} \\ -\eta_{S1} \dfrac{\rho F_W + \rho_{m1} u_{m1} - \rho F_{S1}}{\rho A_{R1} h_1} \end{array} \right) + \eta_{S1} - v_1 \right)^2 \\ + \left(\beta_2 \dfrac{\rho F_W + \rho_{m1} u_{m1} - \rho F_{S1}}{\rho A_{R1}} + h_1 - v_2 \right)^2 \\ + \left(\beta_3 \left(\begin{array}{c} \dfrac{\left(\eta_{S1} + \bar{d}_2\right) F_{S1} + \alpha_{m2} C_{m2} u_{m2} - \eta_{S2} F_{S2}}{A_{R2} h_2} \\ -\eta_{S2} \dfrac{\rho F_{S1} + \rho_{m2} u_{m2} - \rho F_{S2}}{\rho A_{R2} h_2} \end{array} \right) + \eta_{S2} - v_3 \right)^2 \\ + \left(\beta_4 \dfrac{\rho F_{S1} + \rho_{m2} u_{m2} - \rho F_{S2}}{\rho A_{R2}} + h_2 - v_4 \right)^2 \end{array} \right] \quad \text{(B.1)}$$

The closed loop state estimator

- The objective function of closed loop state estimator

$$j_2 = \min_{\tilde{u}_{m1},\tilde{u}_{m2},\tilde{F}_{S1},\tilde{F}_{S2}} \left[\begin{array}{l} \left(\beta_1 \left(\begin{array}{c} \dfrac{\left(\eta_W + \bar{d}_1\right) F_W + \alpha_{m1} C_{m1} \tilde{u}_{m1} - \tilde{\eta}_{S1} \tilde{F}_{S1}}{A_{R1} \tilde{h}_1} \\ -\tilde{\eta}_{S1} \dfrac{\rho F_W + \rho_{m1} \tilde{u}_{m1} - \rho \tilde{F}_{S1}}{\rho A_{R1} \tilde{h}_1} \end{array} \right) + \tilde{\eta}_{S1} - v_1 \right)^2 \\ + \left(\beta_2 \dfrac{\rho F_W + \rho_{m1} \tilde{u}_{m1} - \rho \tilde{F}_{S1}}{\rho A_{R1}} + \tilde{h}_1 - v_2 \right)^2 \\ + \left(\beta_3 \left(\begin{array}{c} \dfrac{\left(\eta_{S1} + \bar{d}_2\right) F_{S1} + \alpha_{m2} C_{m2} \tilde{u}_{m2} - \tilde{\eta}_{S2} \tilde{F}_{S2}}{A_{R2} \tilde{h}_2} \\ -\tilde{\eta}_{S2} \dfrac{\rho F_{S1} + \rho_{m2} \tilde{u}_{m2} - \rho \tilde{F}_{S2}}{\rho A_{R2} \tilde{h}_2} \end{array} \right) + \tilde{\eta}_{S2} - v_3 \right)^2 \\ + \left(\beta_4 \dfrac{\rho F_{S1} + \rho_{m2} \tilde{u}_{m2} - \rho \tilde{F}_{S2}}{\rho A_{R2}} + \tilde{h}_2 - v_4 \right)^2 \end{array} \right] \quad \text{(B.2)}$$

- The first order integrator

$$\begin{aligned} \dot{e}_1 &= \lambda_1(\eta_{S1} - \tilde{\eta}_{S1}) \\ \dot{e}_2 &= \lambda_2(\tilde{h}_1 - h_1) \\ \dot{e}_3 &= \lambda_3(\eta_{S2} - \tilde{\eta}_{S2}) \\ \dot{e}_4 &= \lambda_4(\tilde{h}_2 - h_2) \\ v_1 &= \eta_{S1,sp} + e_1 \\ v_2 &= h_{1,sp} + e_2 \\ v_3 &= \eta_{S2,sp} + e_3 \\ v_4 &= h_{2,sp} + e_4 \end{aligned} \quad \text{(B.3)}$$

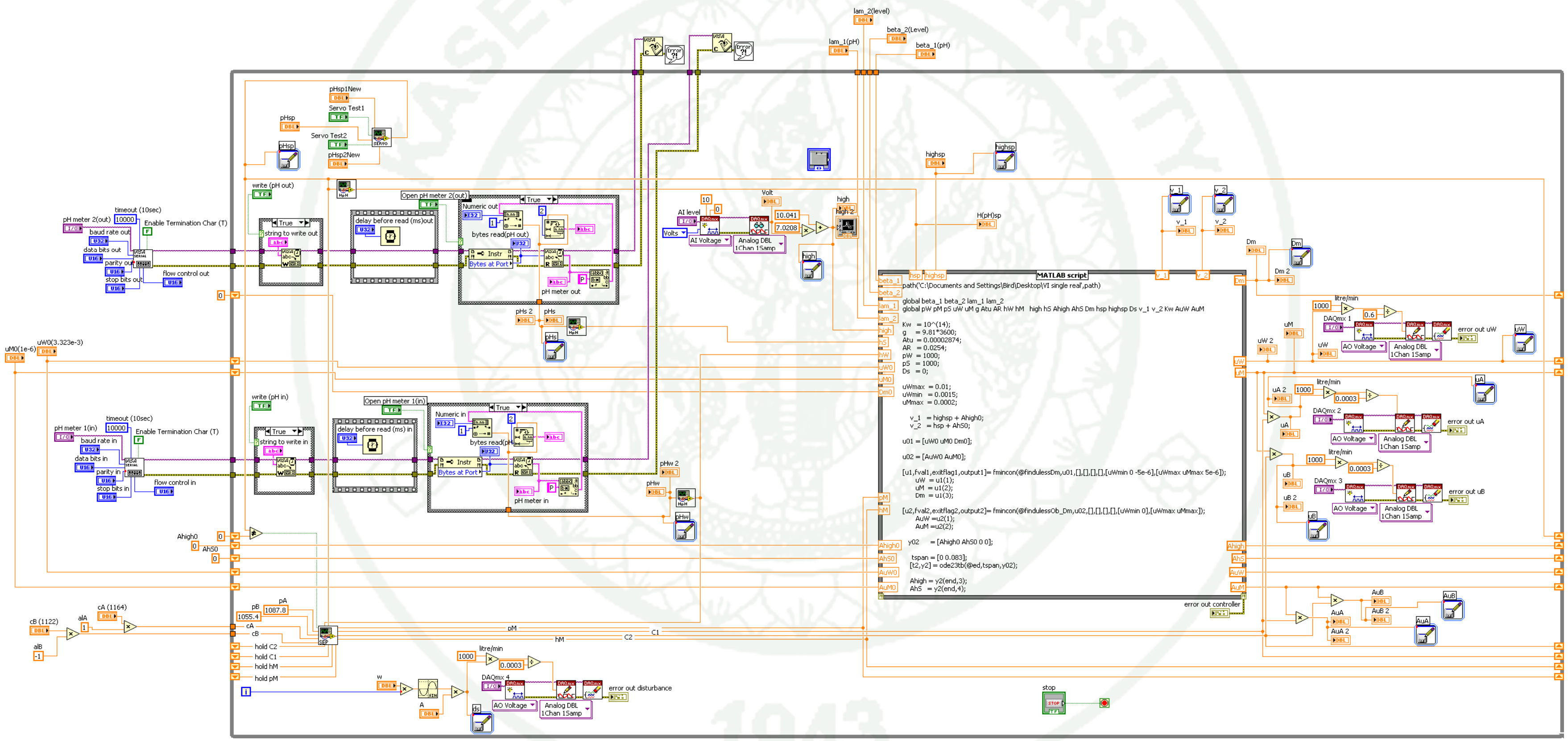
Appendix C

LabVIEW block diagram of the bench-scale pH process

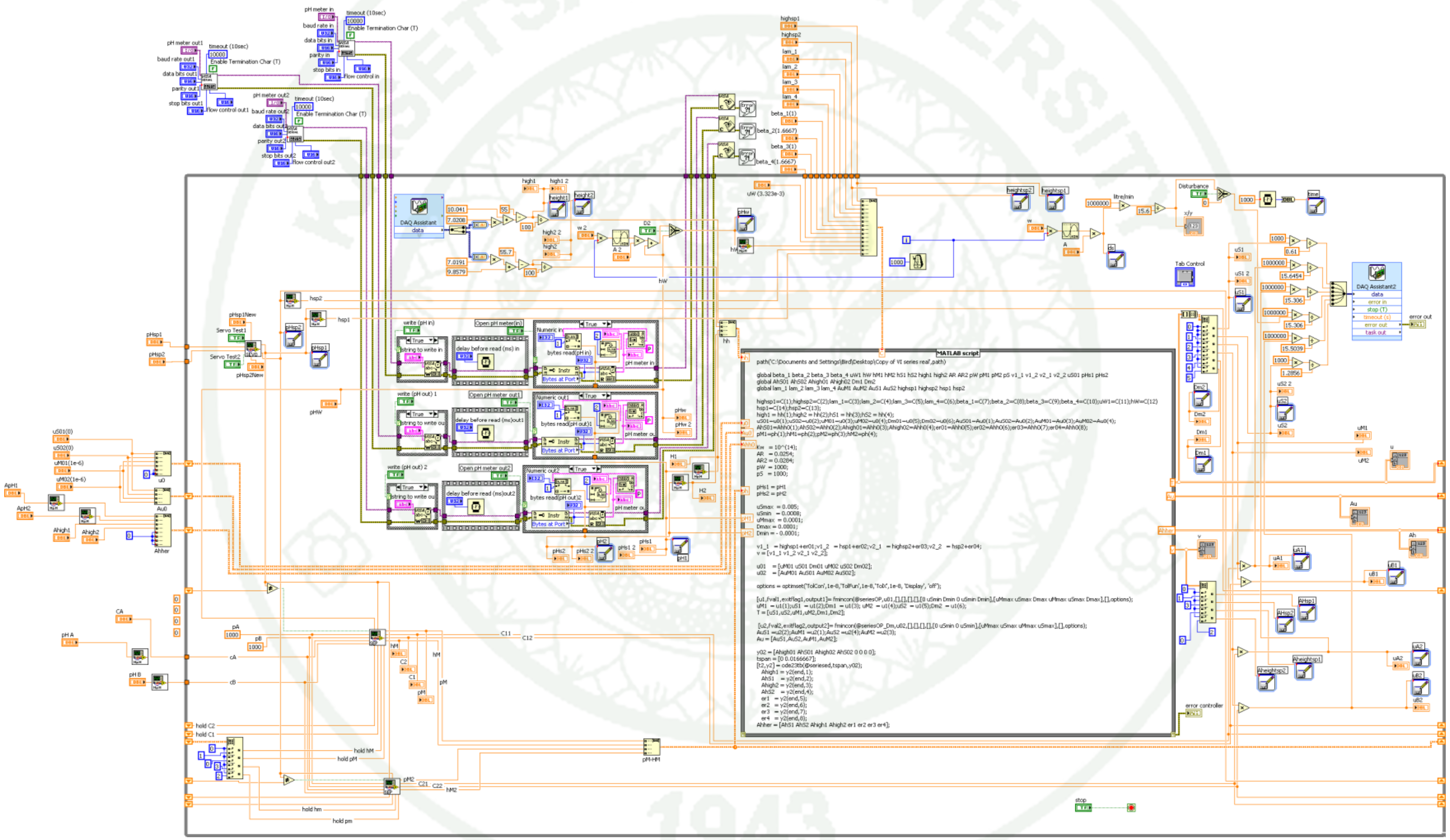
Appendix Figure C1 shows the LabVIEW block diagram of the developed control system for bench-scale pH process

Appendix Figure C2 shows the LabVIEW block diagram of the developed control system for bench-scale pH process in series

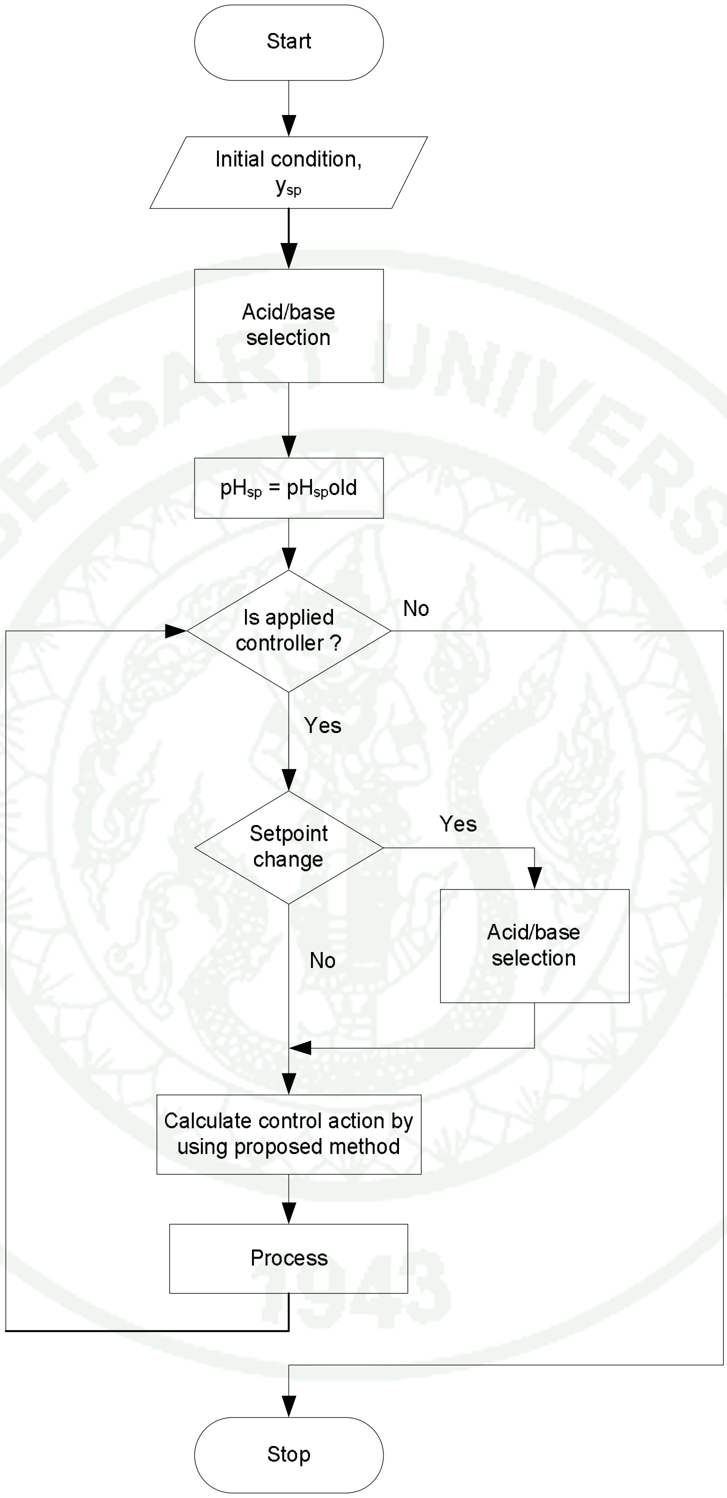
Appendix Figure C3 shows the flow diagram of the proposed method.



Appendix Figure C1 LabVIEW block diagram of the developed control system applied to the bench-scale pH process with the single mixing tank



Appendix Figure C2 LabVIEW block diagram of the developed control system applied with the bench-scale pH process with two mixing tanks in series



Appendix Figure C3 Flow diagram for the proposed method

Appendix D

Experimental result of the control performance of the bench-scale pH process with acetic acid/sodium hydroxide system

The bench-scale process with the configuration of two mixing tanks in series under the acetic acid/sodium hydroxide system is tested for the control performance. The control objective is to handle the pH and level of each mixing tank with the given setpoints. The set of tuning parameter and the initial conditions are summarized in Table 6. The coefficients of total ion concentration α of weak acid are summarized in Wright *et al.*, (1991) as shown in Table 7.

Appendix Table D1 Parameter values used in the setpoint tracking test

Mixing tank	1st Setpoint	Tuning parameters	Initial condition	Acetic acid concentration (mol/L)
1	$pH_{S1,sp} = 6$ $h_{1,sp} = 31$ cm	$\beta_1 = 0.3$ s $\beta_2 = 1$ s	$pH_{S1}(0) = 8$ $h_1(0) = 19$ cm $u_{m1}(0) = 0$ mL/min $u_{S1}(0) = 0.8$ L/min	$C_A = 0.174$
2	$pH_{S2,sp} = 4.5$ $h_{2,sp} = 31$ cm	$\beta_3 = 0.3$ s $\beta_4 = 1$ s	$pH_2(0) = 8$ $h_2(0) = 19$ cm $u_{m2}(0) = 0$ mL/min $u_{S2}(0) = 0.8$ L/min	$C_A = 0.174$

Appendix Table D2 Coefficient of total ion of species *i*

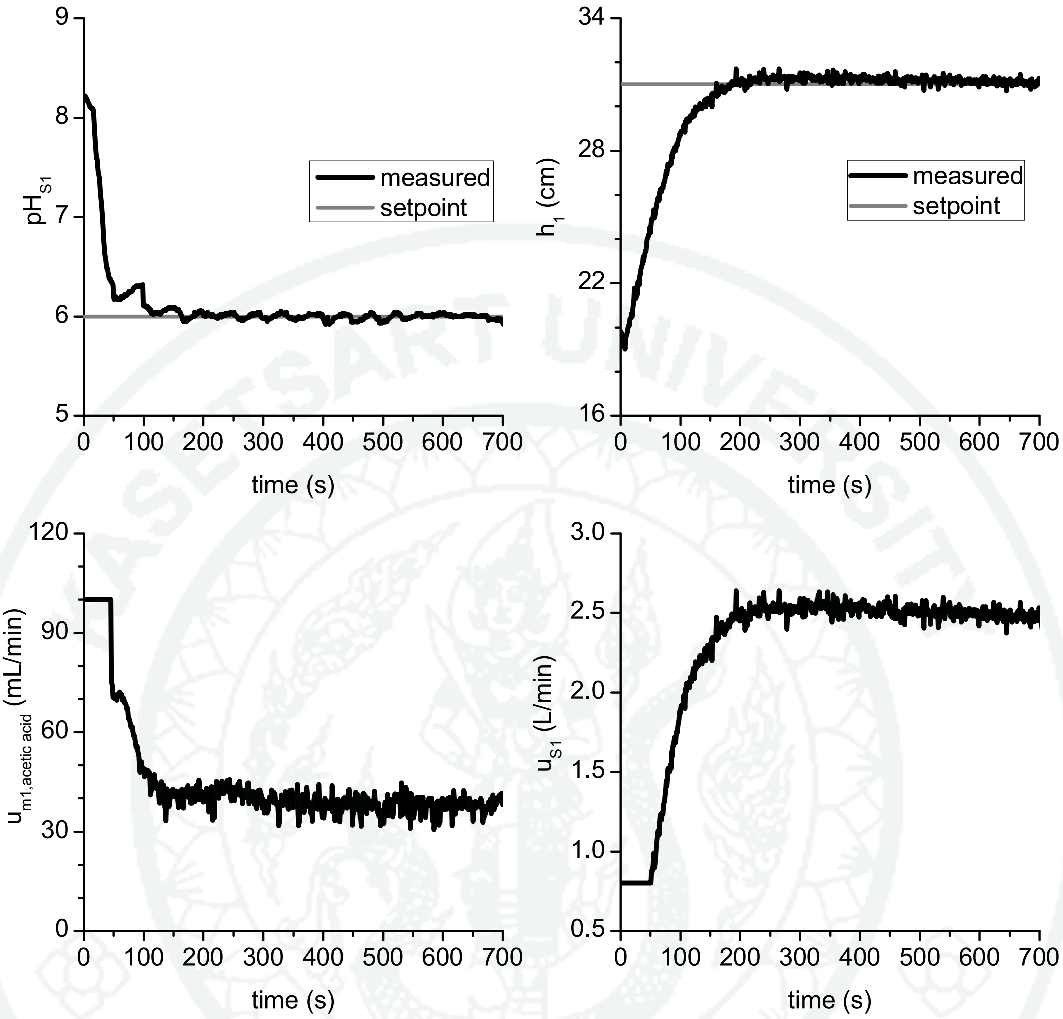
i ionic species	α_i
Anion of monoprotic acid	$\frac{1}{1+10^{pK_a-pH}}$
Anion of diprotic acid	$\frac{2+10^{pK_{a2}-pH}}{1+10^{pK_{a2}-pH}+10^{(pK_{a1}+pK_{a2}-2pH)}}$
Anion of triprotic acid	$\frac{3+2\times10^{pK_{a3}-pH}+10^{pK_{a2}+pK_{a3}-2pH}}{1+10^{pK_{a3}-pH}+10^{(pK_{a2}+pK_{a3}-2pH)}+10^{(pK_{a1}+pK_{a2}+pK_{a3}-3pH)}}$
Cation of monohydroxyl base	$-\frac{10^{(-pH)}}{10^{(-pH)}+10^{(pK_b-pK_w)}}$
Cation of dihydroxyl base	$-\frac{2\times10^{(-2pH)}+10^{(pK_{b2}-pK_w-pH)}}{10^{(-2pH)}+10^{(pK_{b2}-pK_w-pH)}+10^{(pK_{b1}+pK_{b2}-2pK_w)}}$
Cation of trihydroxyl base	$-\frac{3\times10^{(-3pH)}+2\times10^{(pK_{b3}-pK_w-2pH)}+10^{(pK_{b2}+pK_{b3}-2pK_w-pH)}}{10^{(-3pH)}+10^{(pK_{b3}-pK_w-2pH)}+10^{(pK_{b2}+pK_{b3}-2pK_w-pH)}+10^{(pK_{b1}+pK_{b2}+pK_{b3}-3pK_w)}}$

Source : Wright *et al.*, (1991)

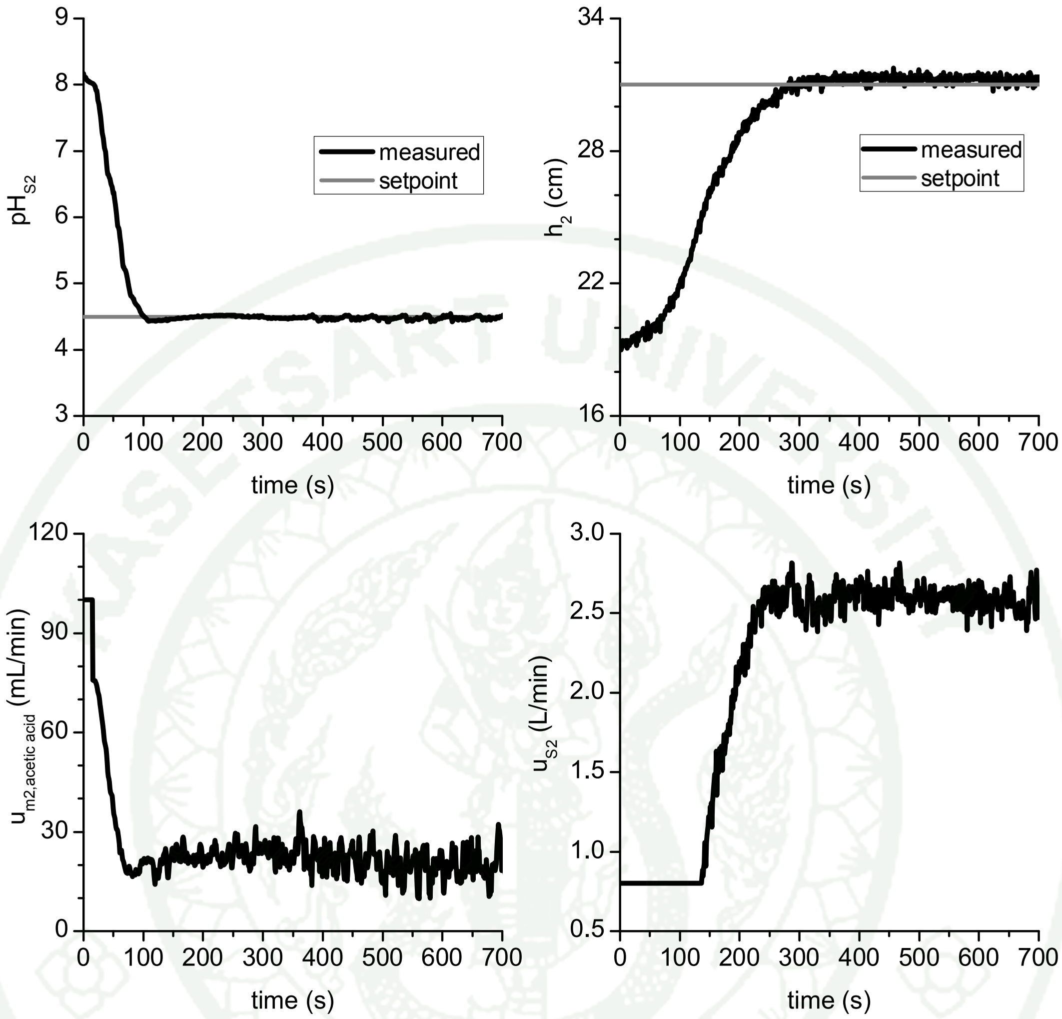
Acetic acid is a weak monoprotic acid with $pK_a = 4.792$. The α_m that is

$$\alpha_m = \frac{1}{1+10^{4.792-pH}} \tag{D.1}$$

is applied in the controller formulation. The experimental results are shown in Appendix Figure D1 and Appendix Figure D2. As can be seen, the proposed control method successfully forces the outputs to their desired setpoints. The responses of the outgoing pH have less oscillation at desired setpoints due to the dissociation effect of acetic acid that vary with pH of the solution.



Appendix Figure D1. The process responses of the mixing tank 1 under proposed controller in the system of acetic acid titrating stream.



Appendix Figure D2. The process responses of the mixing tank 2 under proposed controller in the system of acetic acid titrating stream.

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