Research Article

Input-output Linearizing Control of Strong Acid-base Neutralization Process with Fluctuation in Feed pH

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Abstract
This work presents a control system design for coupled control of pH and level with fluctuation in influent pH by manipulating influent and acid flow rates. A mathematical model based on the difference between proton and hydroxide ions estimated by the measured pH is introduced and applied in a formulation of the model-based control system. A feedback controller and estimated state disturbance are obtained by solving a minimization problem of squared errors between requesting input-output linearizing output responses and the reference setpoints. To eliminate the offset response, the estimated disturbance is applied in the calculation of the closed-loop compensator. The performance of the developed control system is evaluated in a bench-scale pH neutralization process of HCl-NaOH system, and it is also compared to that of a proportional-integral controller. The results show that the developed controllers could enforce a system with fluctuation in influent pH to the desired setpoints effectively, while the PI controller gave oscillation in outputs around the setpoints and cannot achieve the desired targets.

Keywords: pH Neutralization, Model-based control, I/O linearization, pH Feed Fluctuation

1 Introduction

Automatic pH control has been used extensively in chemical, biochemical, and pharmaceutical industries and water treatment [1]–[4]. However, pH neutralization or pH adjustment for a continuous process is quite difficult due to inherent nonlinearity, high sensitivity to input changes around a neutralization point and fluctuations in the feed pH and feed flow. There are several controller strategies that have been proposed for a continuous pH neutralization process.

Controller syntheses have been developed by using techniques such as a Proportional-Integral-Derivative (PID)-based controller integrated with a sliding mode technique [5], an online model-based estimation of strong acid equivalent [6], [7], and a gain scheduling technique based on multiple linear models [8], [9] or a neural network model [10]. However, the mentioned PID-based controllers have good robustness only within a specific operating region. Furthermore, many tuning parameters are required due to the implementation of multiple linear models.

Alternative methods include use of a nonlinear model-based controller to handle the neutralization process, for example, a controller with multi-model switching [11], an adaptive controller based on input-output linearization [12] and internal model [13], and a model predictive controller with Wiener model [14]. These mentioned methods are typically studied with constant influent pH. However in practice, uncertainty

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in the influent pH occurs in process operation. The real-time performances of the mentioned methods remain questionable for a neutralization process with fluctuation in the influent pH. To achieve the desired pH setpoint, the controller should have excellent robustness to reject such a disturbance.

Thus, this work presents a model-based controller design for a continuous neutralization process with fluctuation in a feed pH by using input-output linearization to provide trackability of the process outputs and to estimate the disturbance in the influent pH. A mathematical model representing the difference between proton and hydroxide ions–called the proton-hydroxide ions–estimated from the measured pH is introduced and applied in the formulation of the model-based controller. A feedback controller and estimated disturbance are obtained by solving a problem of minimization of squared errors between requesting input-output linearizing output responses and the reference setpoints. A closed-loop compensator with disturbance prediction is applied to eliminate the output offset. Performances of the developed control system are evaluated using the bench-scale pH process of hydrochloric acid-sodium hydroxide (HCl-NaOH) system under servo and regulatory tests. A Proportional-Integral (PI) controller is considered for performance comparison. The objective is to control the level and the pH in the reactor by manipulating the influent and acid feed rates. One advantage of the proposed method is a capability to handle the uncertainties of the influent pH and also the coupling effects between the level and pH, the first of which has not received attention in the literature.

2 Modeling of pH Neutralization Process

2.1 *Process description*

A schematic diagram of the bench-scale pH neutralization process of HCl-NaOH system is shown in Figure 1. A continuously stirred-tank reactor made of glass and 19 cm in diameter and 50 cm in height is used. Inside the reactor, a motor turbine is used for mixing the liquids. The liquid level in the reactor is measured by an ultrasonic sensor, and pH probes are installed at the inlet and outlet of the reactor to measure the pH values of influent and effluent streams. The reaction is carried out in a continuous mode of a base influent

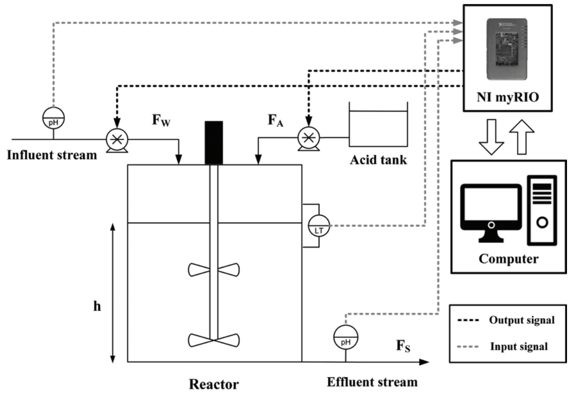

Figure 1: Schematic of a bench-scale pH process.

flow (NaOH). Both liquid level and pH are controlled. The flow rate of an influent stream (F_W) is used to adjust with the liquid level in the reactor (h), while any pH fluctuation at the influent feed can be handled by manipulating an acid stream (HCl) to the reactor.

2.2 *Mathematical model*

The dynamic model for the continuous pH process is derived from first principles and is developed using a lumped approach for the real-time control application. A mathematical model of continuous pH process is developed with the following assumptions:

1. The cross-sectional area of the reactor is constant.
2. Densities of the influent and effluent are approximately equal.
3. Concentration of the acid stream is constant.
4. The system is well mixed, and there are no delays in measured pH signals.

Dynamics of the reactor level can be described as follows:

$$\frac{dh}{dt} = \frac{\rho(F_W - k\sqrt{h}) + \rho_A F_A}{\rho A} \tag{1}$$

where h is the reactor level, F_W is the influent flow rate, F_A is an acid titrating flow rate and, k is flow constant of the effluent steam, ρ and ρ_A are densities of the effluent and acid streams, respectively, and A is the cross-sectional area of the reactor.

In this work, the pH is considered in the form of net proton–hydroxide ions (η), which can be defined by the following equation:

$$\eta = \left(10^{-pH} - \frac{K_w}{10^{-pH}}\right) \tag{2}$$

where K_w is the equilibrium constant for the ionization of water. The component balance take into account of the relationship for net proton-hydroxide ions in the reactor can be described as follows:

$$\frac{d\eta_S}{dt} = \frac{F_W}{Ah}\left(\eta_W + d - \eta_S\right) + \frac{F_A}{Ah}\left(\eta_A - \frac{\rho_A}{\rho}\eta_S\right) \tag{3}$$

where η_S and η_A are the net proton-hydroxide ion concentrations of the reactor liquid and the acid 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 acid stream defined in the Equation (3) can be approximated by the following equation:

$$\eta_A = \alpha C_A \tag{4}$$

where α is the coefficient of the total ion concentration of acid stream and C_A is the concentration of acid stream.

The objective of this work is to control the pH and level in the reactor by manipulating the flow rate of influent feed and acid stream.

Combing the Equations (1)–(4), the process model of the continuous pH process used in this work can be summarized as follows [Equation (5)]:

$$\begin{aligned} \frac{dh}{dt} &= \frac{\rho(F_W - k\sqrt{h}) + \rho_A F_A}{\rho A} \\ \frac{d\eta_S}{dt} &= \frac{F_W}{Ah}\left(\eta_W + d - \eta_S\right) + \frac{F_A}{Ah}\left(\alpha C_A - \frac{\rho_A}{\rho}\eta_S\right) \\ y_1 &= h,\ y_2 = \eta_S \\ u_1 &= F_W,\ u_2 = F_A \end{aligned} \tag{5}$$

Note that Equation (2) can be used to relate pH and η in the calculation.

3 Control System Design

3.1 *Feedback controller*

In this study, a feedback controller is formulated by I/O linearization technique. 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, where the number of differention is called the relative order. A review of I/O linearization and definition of relative order can be found in [15], [16].

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

$$\begin{aligned} \dot{x} &= f(x,u,d) \\ y &= h(x) \end{aligned} \tag{6}$$

where x is the vector of state variables, u is the vector of manipulated inputs, y is the vector of controlled outputs, and d is the vector of unmeasured state disturbances, respectively.

For implementing I/O linearization, the assumptions that the system in Equation (6) is open-loop stable and that the internal dynamics (zero dynamics) are stable have been made. Closed-loop output responses are prescribed in the following forms [Equation (7)]

$$\begin{aligned} (\beta_1 D + 1)^{r_1} y_1 &= v_1 \\ (\beta_2 D + 1)^{r_2} y_2 &= v_2 \end{aligned} \tag{7}$$

where D is the differential operator (i.e. $D = d/dt$), r_1 and r_2 are the relative orders of the controlled outputs, y_1 and y_2 with respect to the manipulated inputs, v_1 and v_2 are the reference output setpoints and β_1 and β_2 are the tuning parameters for adjusting the speed of the responses of y_1 and y_2, respectively. In this study, the level and net proton dynamics of the reactor are directly affected by the influent and acid titrating flows which implies that the relative orders of both outputs are equal to one (r_1 = 1 and r_2 = 1). Therefore, the following optimization problem that minimizes the sum of squared errors between the requesting close-loop output responses and the reference output setpoints is solved at each time instant to calculate the control actions (u) and estimated disturbance ($\bar{d}$):

$$\begin{aligned} & j_1 = \min_{u,\bar{d}} \left(w_1(\beta_1\dot{y}_1 + y_1 - v_1)^2 + w_2(\beta_2\dot{y}_2 + y_2 - v_2)^2 \right) \\ & \text{subject to} \\ & u_{lb} \le u \le u_{ub} \\ & \bar{d}_{lb} \le \bar{d} \le \bar{d}_{ub} \end{aligned} \tag{8}$$

where w is the weighting factor, $\bar{d}$ is the estimated disturbance, and ub and lb are the upper bound and lower bound, respectively. Consequently, the control action and estimate disturbance a solution of the optimization problem in Equation (8) can be represented by:

$$\left[u,\bar{d}\right]=\left\{u_1,u_2,\bar{d}\right\}=\psi(y_1,y_2,v_1,v_2) \tag{9}$$

3.2 *Closed-loop state estimator*

A closed-loop state estimator with an integral action is used to ensure the offset response. Using the process outputs estimated by a closed-loop process model and the estimated disturbances obtained from Equation (9), the closed-loop state estimator with disturbance prediction is developed as shown in Equation (10):

$$\begin{aligned}\frac{d\tilde{y}_1}{dt}&=\frac{\rho(\tilde{u}_1-k\sqrt{\tilde{y}_1})+\rho_A\tilde{u}_2}{\rho A}\\ \frac{d\tilde{y}_2}{dt}&=\frac{\tilde{u}_1}{Ah}(\eta_W+d-\eta_S)+\frac{\tilde{u}_2}{Ah}\left(\alpha C_A-\frac{\rho_A}{\rho}\tilde{y}_2\right)\end{aligned} \tag{10}$$

where $\tilde{y}_1$ is the estimated reactor level, $\tilde{y}_2$ is the estimated net proton–hydroxide ions concentration in the reactor, and $\tilde{u}_1$ and $\tilde{u}_2$ are the estimated inputs obtained by solving the following optimization:

$$j_2=\min_{\tilde{u}}\left(w_1(\beta_1\dot{\tilde{y}}_1+\tilde{y}_1-v_1)^2+w_2(\beta_2\dot{\tilde{y}}_2+\tilde{y}_2-v_2)^2\right)$$

subject to (11)

$$u_{lb}\le\tilde{u}\le u_{ub}$$

where $\tilde{u}$ is the set of the estimated inputs ($\tilde{u}_1$, $\tilde{u}_2$). The control action obtained by solving the optimization problem in Equation (11) can be represented by [Equation (12)]:

$$\tilde{u}=\left\{\tilde{u}_1,\tilde{u}_2\right\}=\tilde{\psi}(\tilde{y}_1,\tilde{y}_2,v_1,v_2,\bar{d}) \tag{12}$$

3.3 *Integrator*

To compensate for offset due to the effects of model-process mismatch and error in the estimated states, the following error dynamics are introduced [Equation (13)]:

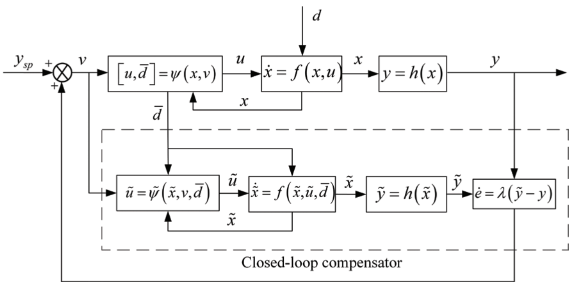

Figure 2: Schematic diagram of the developed control system.

$$\begin{aligned}\dot{e}_1&=\lambda_1(\tilde{y}_1-y_1)\\ \dot{e}_2&=\lambda_2(\tilde{y}_2-y_2)\\ v_1&=y_{1,sp}+e_1\\ v_2&=y_{2,sp}+e_2\end{aligned} \tag{13}$$

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

4 Experimental Setup

The laboratory bench-scale pH process in Figure 3 is at the Model-based Control Laboratory, Department of Chemical Engineering, Kasetsart University. The pH and level in the reactor are measured by a pH probe (model: ECFG7350401B; Eutech Instruments, 0–13 pH), a pH meter (model 6173; JENCO; 0.01pH resolution) and an ultrasonic sensor (model: P43-F4Y-2D-1C0-300E; PiL Sensoren GmbH; distance 100–800 mm, 0.25 mm resolution) for signal feedback. A titrant stream-hydrochloric acid (1.16 M of HCl) is used to adjust the pH in the reactor. The level, the influent flow and the titrating stream are operated within the ranges of 10–50 cm, 0–4.5 L/min and 0–100 mL/min, respectively. The process is monitored and controlled by a desktop computer through the embedded controller device (model: NI myRIO-1900, the National Instruments). LabVIEW and MATLAB software are used to implement the proposed control system and perform data acquisition. In MATLAB, the *fmincon* function is used for solving the control action and estimated disturbance in the proposed optimization problem. The parameters of the bench-scale pH process are given in Table 1.

Figure 3: The bench-scale pH process.

Table 1: Parameter values of the bench-scale pH process

Variable	Value	Units
ρ_A	1087.8	kg/m^3
ρ	1000	kg/m^3
A	0.0254	m^2
K_W	10^{-14}	(mol/L)^2
α	1	
C_A	1.164	mol/L
k	2.97×10^{-5}	m$^{2.5}$/s

5 Results and Discussion

5.1 *Closed-loop performance*

The proposed controller is applied to the bench-scale pH process for evaluating the closed-loop performance. As mentioned previously, pH and level in the reactor are controlled by manipulating the flow rate of the titrating stream and influent stream. The controller is designed to track the pH set point at the neutralization point ($pH_{S,sp} = 7$), while the level setpoint is set to be h_{sp} = 30 cm, and the following tuning parameters are used in the tests: $\beta_1 = 40$, $\beta_2 = 60$, $\lambda_1 = 5$, $\lambda_2 = 5$, $w_1 = 1$, and $w_1 = 1{,}000$. These parameters are the optimal values obtained by trial and error, and the initial guess values of tuning parameters were selected by the Internal Model Control (IMC) method. The initial condition of the pH and the level are $pH = 11.6$ and $h = 20$ cm without disturbance in the influent pH.

The experimental results of the closed-loop test for this case study are shown in Figures 4 and 5. The results indicate that the proposed method successfully enforces the outputs to the desired setpoints. Note that the proposed controller is tuned to have the response time of the level faster than those of the pH because

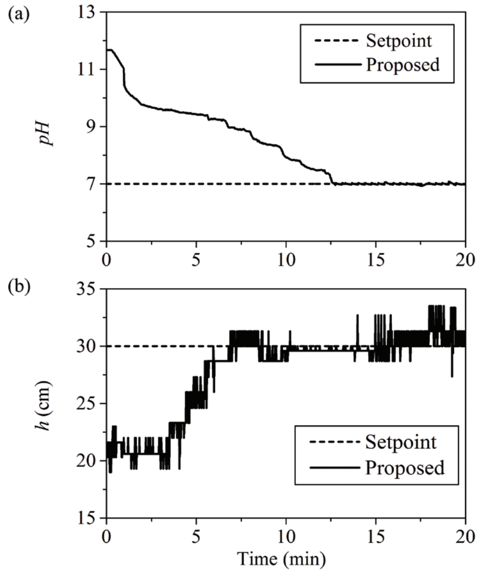

Figure 4: Closed-loop responses of (a) pH and (b) level in the reactor.

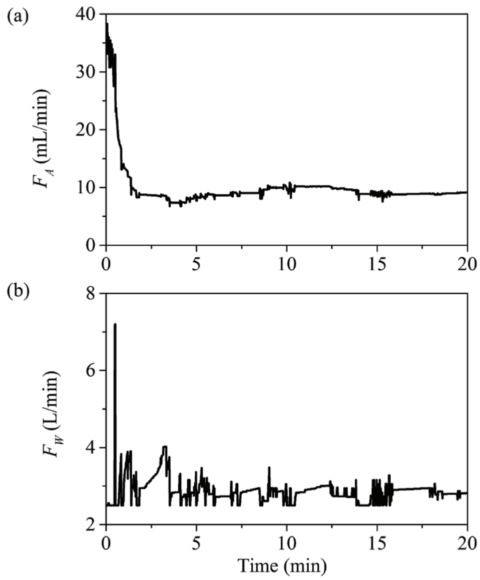

Figure 5: Profiles of (a) acid feed rate and (b) influent feed rate corresponding to the closed-loop responses in Figure 4.

the pH control of the reactor takes time to reach the desired setpoint due to a high sensitivity of the pH characteristics around the neutralization point. With the reactor level stabilized, adjusting the reactor pH becomes more effective. The pH control of the reactor takes time to reach the desired setpoint due to a high sensitivity of the pH characteristics around the neutralization point.

5.2 *Control performance with PI controller*

To evaluate the performance and robustness of the proposed controller, a digital PI controller, described in Equation (14), is used for comparison purposes:

$$u_k = u_{k-1} + K_c\left[(y_{i,sp} - y_{i,k}) - (y_{i,sp} - y_{i,k-1}) \dots \right. \\ \left. \dots + \frac{\Delta t(y_{i,sp} - y_{i,k})}{\tau_i}\right] \quad (14)$$

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 sets of parameter values, $\{K_c = 2.41, \tau_i = 306 \text{ s}, \Delta t = 1 \text{ s}\}$, are applied for both setpoint tracking and disturbance rejection tests; These parameters were calculated by using the IMC tuning rule [17].

5.2.1 Setpoint tracking performance

The control system is tested by setpoint tracking of two given sets of setpoints with no disturbance applied to the influent pH. Initially, the set of desired setpoints is [$pH_{S,sp}$ = 7, h_{sp} = 30 cm]. Then, the setpoint is changed to [$pH_{S,sp}$ = 5, h_{sp} = 30 cm] at t = 15 min. The results in Figure 6 show that the proposed controller successfully forces the outputs to the desired setpoints effectively, while the outputs under the PI controller oscillate greatly around the pH setpoints. The proposed controller provides smooth responses in the outputs and control actions with a shorter settling times to achieve the desired setpoint than those of the PI controller. The control action of the PI controller is proportional to the output error size. Given the large initial output error, the PI controller computes an aggressive action that causes oscillation in the reactor pH.

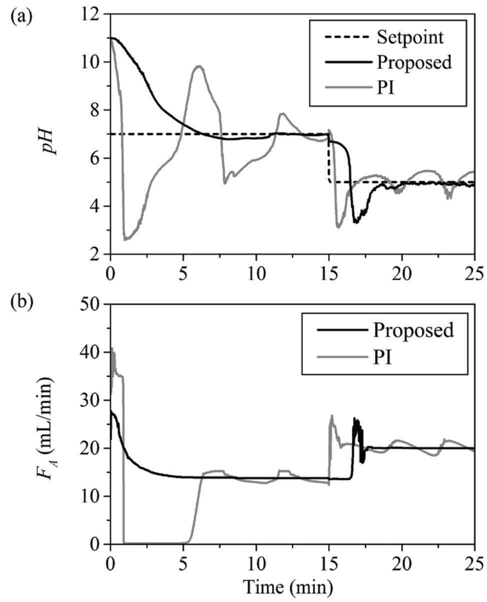

Figure 6: Profiles of (a) reactor pH and (b) acid feed rate under setpoint tracking.

5.2.2 Disturbance rejection performance

The disturbance rejection performance is tested by introducing a pulse disturbance to the influent pH that creates fluctuation in pH (pH_w = 11–11.7). The process outputs are initially at [pH (0), h(0)] = [11, 30 cm] and setpoints are given as [pH_{sp}, h_{sp}] = [5, 30 cm]. The process response under the proposed controller and PI controller are illustrated in Figures 7 and 8, respectively. The results show that the proposed controller provides noticeable improvement for stabilizing the pH in the reactor at pH = 5 despite a fluctuation in the influent pH. In contrast, the PI controller cannot force and stabilize the reactor pH to the desired setpoint. The proposed controller successfully handles uncertainty in the influent pH because the unmeasured disturbance in η_S has been predicted and applied to the state estimator.

6 Conclusions

This work has proposed a model-based control system design of continuous neutralization process for a strong acid-base system with fluctuation in the feed

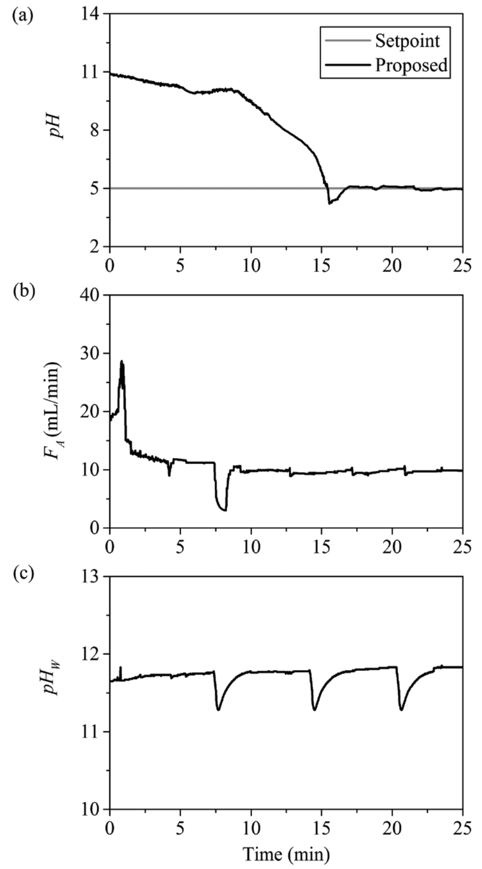

Figure 7: Profiles of (a) reactor pH, (b) acid feed rate and (c) influent pH under the proposed method with disturbances in the influent pH.

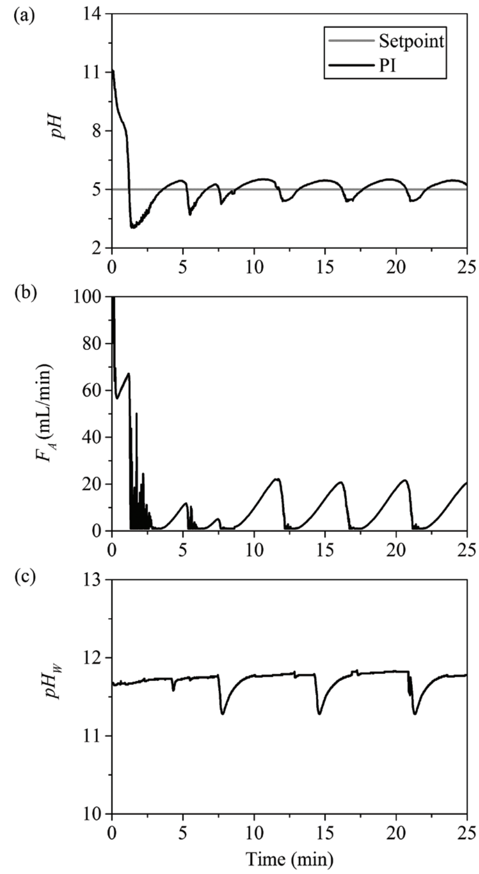

Figure 8: Profiles of (a) the reactor pH, (b) titrant feed rate and (c) influent pH under PI controller with disturbances in influent pH.

pH. An I/O linearization controller was formulated as the optimization problem to handle a coupling effect of pH and level by manipulating the influent flow rate and titrating stream. The proposed controller, which was integrated with a closed-loop state estimator for handling process disturbances and compensating for the output offset, was investigated in the bench-scale pH. Experimental results showed that the proposed control system is more efficient for both setpoint tracking and disturbance rejection when than PI controller. It also provides noticeable improvement for tracking the desired setpoints and stabilizing the reactor pH when disturbance in the influent occur. The proposed controller provides the optimal solution and avoids aggressive control action under the constraint as well.

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Nomenclature

A = cross-sectional area of tank
F_w = influent flow rate
F_A = acid flow rate
F_S = effluent steam flow rate

C_A = acid concentration
h = reactor level
h_{sp} = desired setpoint of reactor level
K_C = controller gain
K_W = dissociation constant of water
d = unmeasured process disturbance
$\overline{d}$ = estimated process disturbance
k = flow constant
pH = pH of effluent stream
pH_{sp} = desired setpoint pH
pH_W = pH of influent stream
t = time
u = manipulated input
w = weighting factor
y = process output
$\tilde{y}$ = estimated output
y_{sp} = output setpoint
α = coefficient of the total ion concentration
β = tuning parameter
η = net proton–hydroxide ions
η_W = net concentration of proton-hydroxide ions in influent steam
η_{sp} = setpoint of net concentration of proton–hydroxide ions
η_A = net concentration of proton-hydroxide ions in acid steam
v = reference setpoint
λ = tuning parameter
ρ = density of effluent steam
ρ_W = density of influent steam
ρ_A = density of acid steam
τ_i = integral gain

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