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A model free adaptive control method based on self-adjusting PID algorithm in pH neutralization process

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ABSTRACT

In this paper, a new model free adaptive control method based on self-adjusting PID algorithm (MFAC-SA-PID) is proposed to solve the problem that the pH process with strong nonlinearity is difficult to control near the neutralization point. The MFAC-SA-PID method also solves the problem that the parameters of the model free adaptive control (MFAC) method are not easy to be adjusted and the effect is not obvious by introducing a fuzzy self-adjusting algorithm to adjust the controller parameters. Then the convergence and stability of the MFAC-SA-PID method are proved in this paper. In the simulation study, the control performance of the MFAC-SA-PID method proposed in this paper is compared with the traditional MFAC method and the improved model free adaptive control (IMFAC) method, respectively. The results show that the proposed MFAC-SA-PID method has better control effect on the pH neutralization process. The MFAC-SA-PID control performance also outperforms the traditional MFAC method and IMFAC method when step input disturbances are added, which indicates that the MFAC-SA-PID method has better robustness and stability.

1. Introduction

As an important indicator in industrial processes, pH is widely used in chemical production, wastewater treatment and biological processes. Since the quality of pH process control can directly affect the safe operation of the whole system, its precise control has important practical significance [1–5].

Since the beginning of the 20th century, scholars have been studying pH process control. With the rapid development of computer technology, more control methods have been proposed. But taking into account the economic cost, production environment and other conditions, PID control method are mostly used in industrial practice. Because the pH process has serious exponential nonlinearity, large hysteresis and high sensitivity near the neutralization point, and there are also uncertainties in the flow and concentration of the neutralized liquid in the actual process, the traditional PID control method is limited by the problem of parameter tuning, so it is sometimes difficult to achieve a satisfactory control effect. As a result, the control of pH process has been a difficult problem in industrial process control at present [6–13].

For many years, pH process as a typical representative of nonlinear system, the research on pH process control method has received extensive attention from scholars at home and abroad. A. Nejati *et al.* compared the performance of adaptive inverse controllers and global linearized controllers in pH process control [14]. Chen *et al.* [15] proposed a sliding mode control method based on the closed-loop transfer function recovery method for the equivalent first-order model of the pH neutralization process. Although the above control methods achieve stable control of the pH process to some extent, most of them adopt model linearization treatment and the design of the controller relies on an accurate mathematical model, which results in poor control performance when the system has strong nonlinearity and uncertainty.

Predictive control is a new class of computerized control algorithms developed in recent years, which is also widely used in pH process control. Wu *et al.* [16] designed a model predictive control (MPC) method based on decentralized fuzzy inference to solve the problems of strong nonlinearity, large hysteresis and uncertainty in pH process. Estofanero *et al.* [17] designed a predictive controller applied to pH neutralization process and determined the model predictive controller by trial. Zou *et al.* [18] extended the model algorithm control to nonlinear pH processes, and simulations showed that the proposed method has good control effect even with large modeling errors. However, the complex nonlinear programming techniques used in these nonlinear predictive control algorithms are time-consuming in the solution process, and it is

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difficult for them to meet the real-time requirements in industrial processes.

With the development of computer science and technology, data-driven modeling methods, represented by BP and RBF neural networks, are powerful tools for controlling nonlinear systems [19–21]. However, these data-driven control methods, especially when modeling based on neural networks, will lose some of the information of the original data and tend to overfit the model, which in turn makes the control performance poor.

Considering the strong nonlinearity and uncertainty of the pH neutralization process, it is difficult to establish an accurate mathematical model. Therefore, in this paper, the MFAC method was firstly adopted to establish the dynamic linearization model of the controlled system. However, the traditional MFAC method also has the problem of difficult parameter tuning and optimization, model-free adaptive control methods based with self-adjusting algorithms have also been studied by some scholars. The adopted self-tuning algorithms can be mainly classified into two categories: (1) intelligent optimization algorithm-based parameter adjustment strategies, such as particle swarm optimization algorithm [22]; (2) neural network-based parameter adjustment strategies, such as radial basis function neural network [23]. Although the use of these self-adjusting algorithms can achieve good control results to a certain extent, but when using intelligent optimization algorithms, the convergence of their algorithms will sometimes be too long; and the parameter adjusting algorithm based on neural networks is very much dependent on the network structure as well as the selection of the initial parameters, which also has a certain impact on the stability of the algorithm.

Therefore, then this paper proposes a MFAC-SA-PID method, which changes the system error of MFAC input to the proportion plus integral plus differential of system error. Meanwhile, MFAC-SA-PID method tunes the parameters of the controller by introducing a fuzzy self-adjusting algorithm. This fuzzy self-adjusting algorithm has less computation and complexity than the above two self-adjusting algorithms, and has better real-time performance and stability. Then, taking pH process control as an example, the control performance of proposed method, traditional MFAC method and IMFAC method [24] is compared respectively, and the anti-disturbance capability of the three methods under step disturbance is analyzed.

The paper is organized as follows, in Section 1, the background and current state of research on the control of pH neutralization process is presented. In Section 2, the mathematical model of the pH neutralization process is presented. In Section 3, the MFAC-SA-PID method proposed in this paper is introduced and its stability is analyzed. Then the simulation study is carried out in Section 4. Finally, the conclusion of this study is given in Section 5.

2. Description of the pH Process

The pH process for the acid-base neutralization reaction in the continuous stirred tank reactor (CSTR) is shown in Fig. 1.

In Fig. 1, the neutralization stream q_1 represents the nitric acid (HNO_3) solution, the buffer stream q_2 represents the sodium bicarbonate ($NaHCO_3$) solution, the process stream q_3 represents the mixed solution of sodium hydroxide (NaOH) and sodium bicarbonate ($NaHCO_3$), and the effluent solution q_4 represents the solution after its neutralization reaction.

Let W_{a1}, W_{a2}, W_{a3}, W_{a4} denote the ionization amount of the solution respectively, then at equilibrium equation is obtained as follows

$$W_{ai} = [H^+] - [NO_3^-] - [HCO_3^-] - 2\left[CO_3^{2-}\right] \tag{1}$$

Similarly, let W_{b1}, W_{b2}, W_{b3}, W_{b4} denote the amount of carbide ions in solution respectively, and at equilibrium we have

$$W_{bi} = [H_2CO_3] + [HCO_3^-] + \left[CO_3^{2-}\right] \tag{2}$$

where, $i = 1, 2, 3, 4$, H_2CO_3 is a weak acid, let its primary ionization constant K_{a1} and secondary ionization constant K_{a2} be

$$K_{a1} = \frac{[HCO_3^-][H^+]}{[H_2CO_3]} \tag{3}$$

$$K_{a2} = \frac{\left[CO_3^{2-}\right][H^+]}{HCO_3^-} \tag{4}$$

From Eqs. (1)–(4), the mathematical model of the pH process of the acid-base neutralization reaction can be established as follows.

$$\dot{W}_{a4} = \frac{1}{V}[(W_{a1} - W_{a4})q_1 + (W_{a2} - W_{a4})q_2 + (W_{a3} - W_{a4})q_3] \tag{5}$$

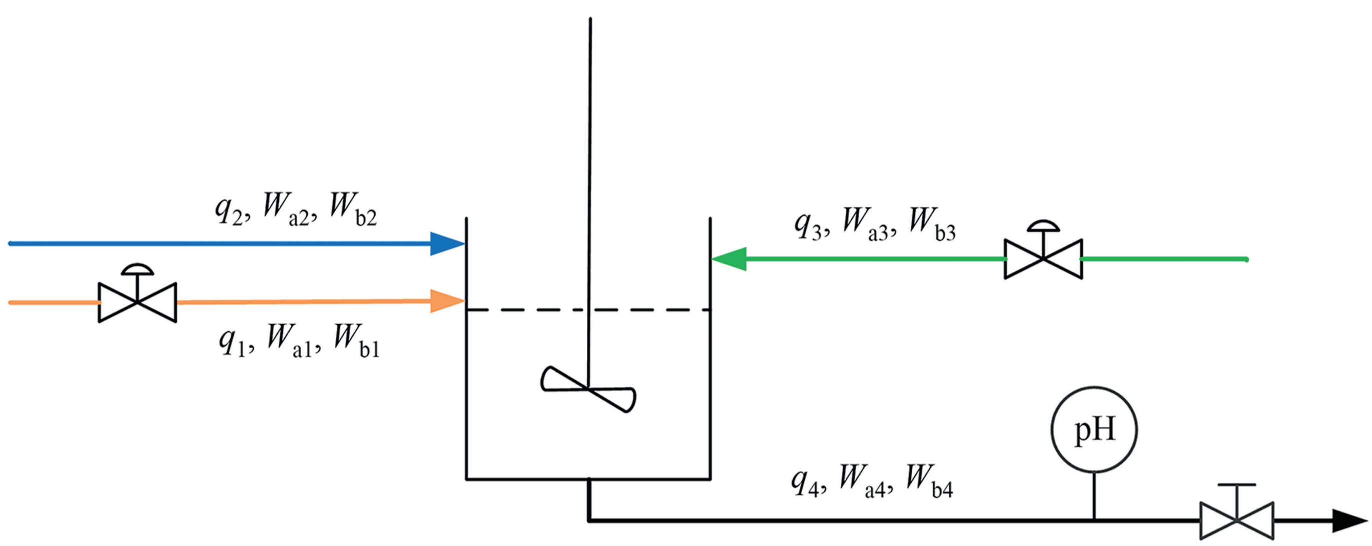

Fig. 1. pH process of acid-base neutralization reaction.

$$\dot{W}_{b4}=\frac{1}{V}[(W_{b1}-W_{b4})q_1+(W_{b2}-W_{b4})q_2+(W_{b3}-W_{b4})q_3] \quad (6)$$

$$W_{a4}=10^{pH-14}+W_{b4}\frac{1+2\times10^{pH-pK_2}}{1+10^{pK_1-pH}+10^{pH-pK_2}}-10^{-pH}=0 \quad (7)$$

where V is the volume of the continuous stirred tank reactor, $pK_1 = -\lg K_{a1}$, $pK_2 = -\lg K_{a2}$, considering q_1 as the control input and pH as the output, the single-input, single-output pH process model can be obtained. Table 1 gives the model parameters and initial operating conditions of the single-input, single-output pH control system.

3. Model Free Adaptive Control with Self-adjusting PID Method

3.1. Introduction to traditional model free adaptive control method

Consider a general single-input single-output nonlinear discrete-time system as follows:

$$y(k+1)=f\left(y(k),\cdots,y\left(k-n_y\right),u(k),\cdots,u(k-n_u)\right) \quad (8)$$

where $u(k)$, $y(k)$ are the input and output of the system at k, n_u, n_y are the input order and output order of the system, $k = 0, 1, 2, \cdots$, $f(\cdot)$ are unknown nonlinear functions.

The following assumptions are made about the system:

Assumption 1: There exists a continuous partial derivative of $f(\cdot)$ with respect to the (n_y+2) nd variable except for finite moment points.

Assumption 2: System (8) satisfies the generalized Lipschitz condition that for any $k_1\neq k_2$, $k_1, k_2 \geq 0$ and $u(k_1)\neq u(k_2)$ with

$$|y(k_1+1)-y(k_2+1)| \leq \beta|u(k_1)-u(k_2)| \quad (9)$$

where $y(k_i+1) = f(y(k_i),\cdots,y(k_i-n_y),u(k_i),\cdots,u(k_i-n_u))$, $\beta > 0$ is a constant.

Theorem 1: For the nonlinear system (8) satisfying Assumptions 1 and 2, there must exist a time-varying parameter $\phi_c(k)\in R$, called the pseudo partial derivative (PPD), when $\Delta u(k)\neq 0$, such that the system (8) can be transformed into the following tight-form dynamic linearized model:

$$\Delta y(k+1)=\phi_c(k)\Delta u(k) \quad (10)$$

where $\phi_c(k)$ is bounded for any moment k, $\Delta u(k) = u(k) - u(k-1)$ is the input increment at two adjacent sampling moments, and $\Delta y(k+1) = y(k+1) - y(k)$ is the output increment at two adjacent sampling moments.

Consider the following two control performance criterion functions:

Table 1
Model parameters and initial operating conditions of the pH process

Parameters	Value	Parameters	Value
pK_1	6.35	pK_2	10.25
W_{a1}	0.003 mol	W_{a2}	−0.03 mol
W_{a3}	−0.00305 mol	W_{b1}	0.0 mol
W_{b2}	0.03 mol	W_{b3}	0.00005 mol
q_1	16.6 ml·s^{-1}	q_2	0.55 ml·s^{-1}
q_3	15.6 ml·s^{-1}	V	2900 ml

$$J(\Delta u(k))=(y_d(k+1)-y(k+1))^2+\lambda(u(k)-u(k-1))^2 \quad (11)$$

$$\begin{aligned}J(\widehat{\phi}_c(k))=&(y(k)-y(k-1)-\widehat{\phi}_c(k)\Delta u(k-1))^2+\\&\mu(\widehat{\phi}_c(k)-\widehat{\phi}_c(k-1))^2\end{aligned} \quad (12)$$

The MFAC method based on tight-format dynamic linearization can be obtained as follows [25]

$$u(k)=u(k-1)+\frac{\rho\phi_c(k)}{\lambda+\phi_c(k)^2}(y_d(k+1)-y(k)) \quad (13)$$

$$\hat{\phi}_c(k)=\begin{cases}\hat{\phi}_c(1), \text{When } |\hat{\phi}_c(k)|\leq\varepsilon \text{ or} |\Delta u(k-1)|\leq\varepsilon \text{ or} \\ \qquad \operatorname{sgn}\left(\hat{\phi}_c(k)\right)\neq\operatorname{sgn}\left(\hat{\phi}_c(1)\right) \\ \hat{\phi}_c(k-1)+\dfrac{\eta\Delta u(k-1)}{\mu+\Delta u(k-1)^2}\Big(\Delta y(k)- \\ \qquad \hat{\phi}_c(k-1)\Delta u(k-1)\Big), \text{ Otherwise}\end{cases} \quad (14)$$

where $\mu>0$, $\lambda>0$, $\rho\in(0,1]$, $\eta\in(0,1]$ are the step factors, $\hat{\phi}_c(1)$ is the initial value of $\hat{\phi}_c(k)$, and $\varepsilon>0$ is a sufficiently small positive number. The step factor is introduced to increase the generality of the MFAC method, while the PPD reset algorithm is introduced to make $\hat{\phi}_c(k)$ have a stronger tracking capability for time-varying systems.

3.2. Introduction to model free adaptive control based on self-adjusting PID algorithm

The traditional MFAC method can be regarded as a time-varying integral controller, which is difficult to get the ideal control effect for some complex systems with large lags and nonlinearities due to the simplicity of the control law. Therefore, this paper improves the above traditional MFAC method, further extends the expression form of MFAC control law by combining incremental PID algorithm, and introduces fuzzy self-adjusting algorithm to adjust the controller parameters. Finally this paper gives the stability and convergence proof of the MAFC-SA-PID method.

Let y_d be a given constant expectation signal, and note the error $e(k) = y_d - y(k)$ at the moment k, then Eq. (13) can be rewritten as

$$\Delta u(k)=\frac{\rho\phi_c(k)}{\lambda+\phi_c(k)^2}e(k) \quad (15)$$

From Eq. (15), it can be found that the control law of the traditional MFAC method is essentially doing dynamic integration of the error $e(k)$ with an integration gain of $\rho\phi_c(k)/(\lambda+\phi_c(k)^2)$, which can be extended with reference to the incremental PID control algorithm for MAFC.

The control expression for the incremental PID control algorithm is written as

$$K_{Fp}(e(k)-e(k-1))+K_{Fi}e(k)+K_{Fd}(e(k)-2e(k-1)+e(k-2)) \quad (16)$$

Therefore the input error $e(k)$ of Eq. (15) can be changed to $e(k)$ or $\Delta e(k)$ as input and combined with the control mode of incremental PID, the control law of MFAC-SA-PID can be obtained as follows:

$$u(k) = u(k-1) + \frac{\rho\phi_c(k)}{\lambda + \phi_c(k)^2}\left[K_{Fp}(e(k) - e(k-1)) + K_{Fi}e(k) + K_{Fd}(e(k) - 2e(k-1) + e(k-2))\right] \tag{17}$$

The performance of the control system can be improved by adjusting the parameter K_{Fp}, K_{Fi}, K_{Fd} without adjusting the five parameters μ, λ, ρ, η, ε, which only need to be satisfied: $\mu > 0$, $\lambda > 0$, $\rho \in (0,1]$, $\eta \in (0,1]$, $\varepsilon > 0$. The online adjustment equation for K_{Fp}, K_{Fi}, K_{Fd} is as follows:

$$\begin{cases} K_{Fp} = K_{p0} + K_{\Delta p}\cdot\Delta K_p \\ K_{Fi} = K_{i0} + K_{\Delta i}\cdot\Delta K_i \\ K_{Fd} = K_{d0} + K_{\Delta d}\cdot\Delta K_d \end{cases} \tag{18}$$

In Eq. (18), K_{p0}, K_{i0}, K_{d0} is the initial value, $K_{\Delta p}$, $K_{\Delta i}$, $K_{\Delta d}$ is the scale factor of each component, $K_{\Delta p}$, $K_{\Delta i}$, $K_{\Delta d}$ are the adaptive control quantities. In this paper, the inputs of the fuzzy self-adjuster are the system error $e(k)$ and the error variation $\Delta e(k)$. Where the fuzzy self-adjuster affiliation function is in the form of a triangle with uniform symmetric distribution, and the fuzzy self-adjusting rules are shown in Table 2.

The SA-PID-MAFC method is composed of Eqs. (14), (17) and (18), and its system control structure is shown in Fig. 2. From Eq. (17), we can see when $K_{Fi} = 1$ and K_{Fp}, K_{Fd} are zero, the MFAC-SA-PID method is consistent with the expression of the traditional MFAC method, which also shows that the traditional MFAC method is a special case of the MFAC-SA-PID method. Therefore, the proposed MFAC-SA-PID method has wider applicability than the traditional MFAC method, and The fuzzy self-adjusting algorithm is introduced to make the MFAC-SA-PID method have better control accuracy and anti-disturbance performance.

3.3. Stability of the system

The following assumptions continue to be made on the basis of Assumptions 1 and 2:

Assumption 3: The pseudo partial derivatives $\phi_c(k)$ of the nonlinear system (8) are constant in sign and non-intrusive. In this paper, we assume $0 < \phi_c(k) \le \phi_0$, where ϕ_0 is the upper limit of the pseudo partial derivatives $\phi_c(k)$.

Assumption 4: K_{Fp}, K_{Fi}, K_{Fd} are all greater than zero and that the following conditions are satisfied: $K_{Fd} < \frac{1}{2}K_{Fi}$, $2K_{Fp} + 3K_{Fi} < \frac{4\sqrt{\lambda}}{\phi_0}$.

For the nonlinear system (8) satisfying Assumptions 1–4, there are two theorems when the MFAC-SA-PID method proposed in this paper is used as follows:

Table 2
Fuzzy self-adjusting rules base for $K_{\Delta p}$, $K_{\Delta i}$, $K_{\Delta d}$

e	Δe						
	NB	NM	NS	ZO	PS	PM	PB
NB	PB/NB/PS	PB/NB/NS	PM/NM/NB	PM/NM/NB	PS/NS/NB	ZO/ZO/NM	ZO/ZO/PS
NM	PB/NB/NS	PB/NB/NS	PM/NM/NB	PS/NS/NM	PS/NS/NM	ZO/ZO/NS	NS/ZO/ZO
NS	PM/NB/ZO	PM/NM/NS	PM/NS/NM	PS/NS/NM	ZO/ZO/NS	NS/PS/NS	NS/PS/ZO
ZO	PM/NM/ZO	PM/NM/NS	PS/NS/NS	ZO/ZO/NS	NS/PS/NS	NM/PM/NS	NM/PM/ZO
PS	PS/NM/ZO	PS/NS/ZO	ZO/ZO/ZO	NS/PS/ZO	NS/PS/ZO	NM/PM/ZO	NM/PB/ZO
PM	PS/ZO/PB	ZO/ZO/PS	NS/PS/PS	NM/PS/PS	NM/PM/PS	NM/PB/PS	NB/PB/PB
PB	ZO/ZO/PB	ZO/ZO/PM	NM/PS/PM	NM/PM/PM	NM/PM/PS	NB/PB/PS	NB/PB/PB

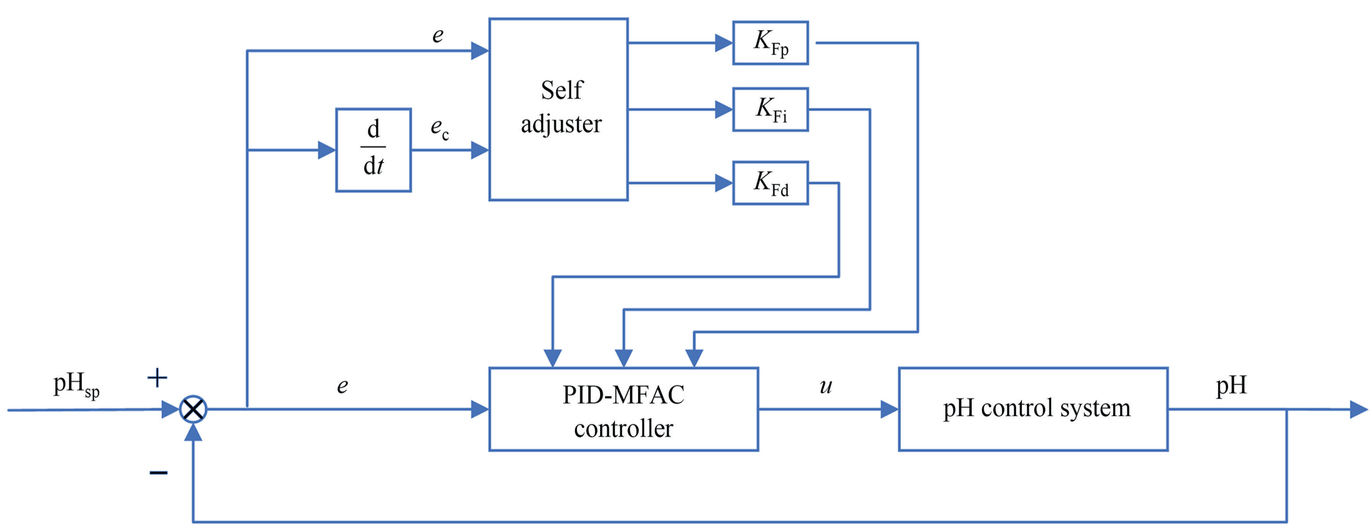

Fig. 2. System structure of MFAC-SA-PID method based on pH process.

Theorem 2: The output tracking error of the system is convergent, that is $\lim_{k\to\infty}|e(k)| = \lim_{k\to\infty}|y_d - y(k)| = 0$. Where y_d is the given desired value.

Theorem 3: The system control input sequence $\{u(k)\}$ and output sequence $\{y(k)\}$ are bounded and the closed-loop system is BIBO stable.

Proof of Theorem 2:

Noting $\phi(k) = \dfrac{\phi_c(k)^2}{\lambda+\phi_c(k)^2}$, we have

$$\begin{aligned} e(k+1) &= y_d - y(k+1) = y_d - (y(k) + \phi_c(k)\Delta u(k)) \\ &= (y_d - y(k)) - \phi_c(k)\Delta u(k) \\ &= e(k) - \phi(k)\left[K_{Fp}(e(k)-e(k-1)) + K_{Fi}e(k) + K_{Fd}(e(k)- \right. \\ &\quad \left. 2e(k-1)+e(k-2))\right] \\ &= \left[1-\phi(k)(K_{Fp}+K_{Fi}+K_{Fd})\right]e(k) + \phi(k)(K_{Fp}+ \\ &\quad 2K_{Fd})e(k-1) - \phi(k)K_{Fd}e(k-2) \end{aligned} \tag{19}$$

From $0<\phi_c(k)\le\phi_0$, we have $0<\phi(k)\le\dfrac{\phi_0{}^2}{\lambda+\phi_0{}^2}$. And further from $\lambda+\phi_0^2\ge 2\sqrt{\lambda}\phi_0$ we have $0<\phi(k)\le\dfrac{\phi_0}{2\sqrt{\lambda}}$ by deflating $\phi(k)$.

Let: $\alpha_1(k) = \phi(k)(K_{Fp}+K_{Fi}+K_{Fd})$, $\alpha_2 = \phi(k)(K_{Fp}+2K_{Fd})$, $\alpha_3 = \phi(k)K_{Fd}$. $\alpha_1(k)>\alpha_2(k)+\alpha_3(k)>\alpha_2(k)>\alpha_3(k)>0$ can be obtained from $K_{Fi}>2K_{Fd}>0$ in Assumption 4.

Taking absolute values for $e(k+1)$ and using the absolute value inequality as well as the maximum value inequality to deflate there is

$$\begin{aligned} |e(k+1)| &= |(1-\alpha_1(k))e(k) + \alpha_2(k)e(k-1) - \alpha_3(k)e(k-2)| \\ &\le |1-\alpha_1(k)||e(k)| + \alpha_2(k)|e(k-1)| + \alpha_3(k)|e(k-2)| \\ &\le (|1-\alpha_1(k)| + \alpha_2(k) + \alpha_3(k))\max(|e(k)|, |e(k-1)|, \\ &\quad |e(k-2)|) \end{aligned} \tag{20}$$

Let: $\beta(k) = |1-\alpha_1(k)| + \alpha_2(k) + \alpha_3(k)$, obviously $\beta(k) > 0$, then

$$|e(k+1)| \le \beta(k)\max(|e(k)|, |e(k-1)|, |e(k-2)|) \tag{21}$$

Two scenarios are discussed:

(1) When $1-\alpha_1(k)\ge 0$

Since the previous analysis yields $\alpha_1(k)>\alpha_2(k)+\alpha_3(k)$, then

$$\begin{aligned} \beta(k) &= |1-\alpha_1(k)| + \alpha_2(k) + \alpha_3(k) = 1-\alpha_1(k) + \alpha_2(k) + \alpha_3(k) \\ &< 1-(\alpha_2(k)+\alpha_3(k)) + \alpha_2(k) + \alpha_3(k) = 1 \end{aligned} \tag{22}$$

(2) When $1-\alpha_1(k)<0$

Construct the intermediate function $\gamma(k) = \alpha_1(k)+\alpha_2(k)+\alpha_3(k)$, and since $K_{Fd}<\dfrac{1}{2}K_{Fi}$, $2K_{Fp}+3K_{Fi}<\dfrac{4\sqrt{\lambda}}{\phi_0}$ and $0<\phi(k)\le\dfrac{\phi_0}{2\sqrt{\lambda}}$ in Assumption 4, we have

$$\begin{aligned} \gamma(k) &= \alpha_1(k)+\alpha_2(k)+\alpha_3(k) = \phi(k)(K_{Fp}+K_{Fi}+K_{Fd}) + \phi(k) \\ &\quad (K_{Fp}+2K_{Fd}) + \phi(k)K_{Fd} = \frac{\phi_c(k)^2}{\lambda+\phi_c(k)^2} \\ &\quad (2K_{Fp}+K_{Fi}+4K_{Fd}) \le \frac{\phi_0}{2\sqrt{\lambda}}(2K_{Fp}+K_{Fi}+4K_{Fd}) \\ &\quad < \frac{\phi_0}{2\sqrt{\lambda}}(2K_{Fp}+3K_{Fi}) < \frac{\phi_0}{2\sqrt{\lambda}}\times\frac{4\sqrt{\lambda}}{\phi_0} = 2 \end{aligned} \tag{23}$$

This leads to $\gamma(k) = \alpha_1(k)+\alpha_2(k)+\alpha_3(k)<2$, at which point we have

$$\begin{aligned} \beta(k) &= |1-\alpha_1(k)| + \alpha_2(k) + \alpha_3(k) = \alpha_1(k)+\alpha_2(k)+\alpha_3(k) - 1 \\ &= \gamma(k) - 1 < 1 \end{aligned} \tag{24}$$

In summary, from the Eq. (22), Eq. (24) can be obtained $\beta(k)$ constant less than 1, let $b_1 = \max(\beta(k))$.

Then we have $\beta(k)\le b_1<1$, at this time by the Eq. (20) have

$$|e(k+1)| \le b_1 \max(|e(k)|, |e(k-1)|, |e(k-2)|) \tag{25}$$

The recurrence of Eq. (25) for $|e(k)|$, $|e(k-1)|$, and $|e(k-2)|$ gives

$$|e(k)| \le b_1 \max(|e(k-1)|, |e(k-2)|, |e(k-3)|) \tag{26}$$

$$|e(k-1)| \le b_1 \max(|e(k-2)|, |e(k-3)|, |e(k-4)|) \tag{27}$$

$$|e(k-2)| \le b_1 \max(|e(k-3)|, |e(k-4)|, |e(k-5)|) \tag{28}$$

From Eqs. (26)–(28) and the maximum value inequality, we have

$$\begin{aligned} &\max(|e(k)|, |e(k-1)|, |e(k-2)|) \le \\ &b_1 \max(|e(k-1)|, |e(k-2)|, |e(k-3)|, |e(k-4)|, |e(k-5)|) \end{aligned} \tag{29}$$

And since $b_1<1$, obviously $|e(k-1)|>b_1|e(k-1)|$, $|e(k-2)|>b_1|e(k-2)|$, then Eq. (29) is further organized to obtain

$$\begin{aligned} &\max(|e(k)|, |e(k-1)|, |e(k-2)|) \\ &\quad \le b_1 \max(|e(k-3)|, |e(k-4)|, |e(k-5)|) \end{aligned} \tag{30}$$

Substituting Eq. (30) into Eq. (25) yields

$$\begin{aligned} |e(k)| &\le b_1 \max(|e(k-1)|, |e(k-2)|, |e(k-3)|) \\ &\le \begin{cases} b_1^{k/3} \max(|e(0)|, |e(1)|, |e(2)|), \\ \quad \text{When } k \bmod 3 = 0; \\ b_1^{(k-1)/3} \max(|e(0)|, |e(1)|, |e(2)|), \\ \quad \text{When } (k-1) \bmod 3 = 0; \\ b_1^{(k-2)/3} \max(|e(0)|, |e(1)|, |e(2)|), \\ \quad \text{When } (k-2) \bmod 3 = 0. \end{cases} \\ &= b_1^{[k/3]} \max(|e(0)|, |e(1)|, |e(2)|) \end{aligned} \tag{31}$$

where $[x]$ denotes the integer for $x\in R$ and k=3, 4, 5,$\cdots$, such that $c_1 = \max(|e(0)|, |e(1)|, |e(2)|)$, c_1 is a constant value greater than zero, then we get

$$\lim_{k\to\infty}|y_{\rm d}-y(k)|=\lim_{k\to\infty}|e(k)|\le\lim_{k\to\infty}b_1^{[k/3]}c_1=0 \tag{32}$$

In summary, the output tracking error of the system is convergent, and Theorem 2 is proved.

Proof of Theorem 3: From Eq. (17), we get

$$\begin{aligned}|\Delta u(k)|=&\frac{\phi_{\rm c}(k)}{\lambda+\phi_{\rm c}(k)^2}\left|\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)e(k)-\left(K_{\rm Fp}+2K_{\rm Fd}\right)e(k-1)\right)+\\ &K_{\rm Fd}e(k-2)|\le\frac{\phi_{\rm c}(k)}{2\sqrt{\lambda}\phi_{\rm c}(k)}|\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)e(k)\\ &-\left(K_{\rm Fp}+2K_{\rm Fd}\right)e(k-1)+K_{\rm Fd}e(k-2)|\\ &\le\frac{1}{2\sqrt{\lambda}}\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)|e(k)|+\left(K_{\rm Fp}+2K_{\rm Fd}\right)|e(k-1)|\\ &+K_{\rm Fd}|e(k-2)|\le\frac{1}{2\sqrt{\lambda}}\left(2K_{\rm Fp}+K_{\rm Fi}+4K_{\rm Fd}\right)\max(|e(k)|,\\ &|e(k-1)|,|e(k-2)|)<\frac{1}{2\sqrt{\lambda}}\left(2K_{\rm Fp}+3K_{\rm Fi}\right)\max(|e(k)|,\\ &|e(k-1)|,|e(k-2)|)\end{aligned} \tag{33}$$

Taking the absolute value of Eq. (34) and using the inequality to deflate according to the known conditions we have

$$\begin{aligned}|\Delta u(k)|=&\frac{\phi_{\rm c}(k)}{\lambda+\phi_{\rm c}(k)^2}\left|\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)e(k)-\right.\\ &\left(K_{\rm Fp}+2K_{\rm Fd}\right)e(k-1)\big)+K_{\rm Fd}e(k-2)|\\ &\le\frac{\phi_{\rm c}(k)}{2\sqrt{\lambda}\phi_{\rm c}(k)}|\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)e(k)-\\ &\left(K_{\rm Fp}+2K_{\rm Fd}\right)e(k-1)+K_{\rm Fd}e(k-2)|\\ &\le\frac{1}{2\sqrt{\lambda}}\left(K_{\rm Fp}+K_{\rm Fi}+K_{\rm Fd}\right)|e(k)|+\\ &\left(K_{\rm Fp}+2K_{\rm Fd}\right)|e(k-1)|+K_{\rm Fd}|e(k-2)|\\ &\le\frac{1}{2\sqrt{\lambda}}\left(2K_{\rm Fp}+K_{\rm Fi}+4K_{\rm Fd}\right)\max(|e(k)|,|e(k-1)|,\\ &|e(k-2)|)\\ &<\frac{1}{2\sqrt{\lambda}}\left(2K_{\rm Fp}+3K_{\rm Fi}\right)\max(|e(k)|,|e(k-1)|,|e(k-2)|)\end{aligned} \tag{34}$$

Let $b_2=\frac{1}{2\sqrt{\lambda}}(2K_{\rm Fp}+3K_{\rm Fi})$, then we have

$$|\Delta u(k)|<b_2\max(|e(k)|,|e(k-1)|,|e(k-2)|) \tag{35}$$

where k=2, 3, 4, $\cdots$, then $|u(k)|$ can be expressed as follows

$$\begin{aligned}|u(k)|&=|(u(k)-u(k-1))+(u(k-1)-u(k-2))+\cdots\\ &\quad+(u(2)-u(1))+u(1)|\\ &=|\Delta u(k)+\Delta u(k-1)+\cdots+\Delta u(2)+u(1)|\\ &\le|\Delta u(k)|+|\Delta u(k-1)|+\cdots+|\Delta u(2)|+|u(1)|\end{aligned} \tag{36}$$

Let $\varsigma(k)=|\Delta u(k)|+|\Delta u(k-1)|+|\Delta u(2)|$, then from Eqs. (31), (32) and (36), we have

$$\begin{aligned}\varsigma(k)&=|\Delta u(k)|+|\Delta u(k-1)|+\cdots+|\Delta u(2)|\\ &\le b_2\max(|e(k)|,|e(k-1)|,|e(k-2)|)+b_2\max(|e(k-1)|,\\ &|e(k-2)|,|e(k-3)|)+\cdots+b_2\max(|e(2)|,|e(1)|,|e(0)|)\\ &\le 3b_2\left(b_1^{[k/3]}+b_1^{[k/3]-1}+\cdots 1\right)\max(|e(2)|,|e(1)|,|e(0)|)\end{aligned} \tag{37}$$

From $b_1<1$ and the properties of the isoperimetric series we have

$$\begin{aligned}\varsigma(k)&\le 3b_2\left(b_1^{[k/3]}+b_1^{[k/3]-1}+\cdots 1\right)\max(|e(2)|,|e(1)|,|e(0)|)\\ &<\frac{3b_2}{1-b_1}c_1\end{aligned} \tag{38}$$

where $b_2=\dfrac{1}{2\sqrt{\lambda}}(2K_{\rm Fp}+3K_{\rm Fi})\le\dfrac{2}{\phi_0}$, $c_1=\max(|e(0)|,|e(1)|,|e(2)|)$ are constant values that do not vary with k, it is obtained that $\varsigma(k)$ is bounded and from Eq. (37) we have

$$|u(k)|\le\varsigma(k)+|u(1)|<\frac{b_2}{1-b_1}c_1+|u(1)| \tag{39}$$

Therefore, $|u(k)|$ is also bounded, so the control input sequence $\{u(k)\}$ and output sequence $\{y(k)\}$ of the system are both bounded, and Theorem 3 is proved.

4. Simulation Study

4.1. pH neutralization control

Considering the large gain of the pH process near the neutralization point, the set value of the pH process control is 7 in order to verify the effectiveness of the MFAC-SA-PID method.

Firstly, considering the case without adding disturbances, the MFAC-SA-PID control method proposed in this paper is compared with the traditional MFAC method and the IMFAC method in the Ref. [24], respectively.

The initial conditions for all three methods are set to $\phi_{\rm c}(1)=0.01$, $\varepsilon=0.00001$, and the controller parameters for all three methods are set to: $\eta=1$, $\lambda=1$, $\mu=1$, $\rho=0.4$.

The initial parameters of the fuzzy self-adjuster for the MFAC-SA-PID method are $K_{p0}=0.1$, $K_{i0}=1.2$, $K_{d0}=0.01$, and the scaling factors are $K_{\Delta p}=0.1$, $K_{\Delta i}=3$, and $K_{\Delta d}=0.01$, respectively.

The simulation results are shown in Fig. 3, Fig. 4, and Fig. 5. Fig. 3 shows the control effect of pH, Fig. 4 shows the variation of input acid flow rate q_1, and Fig. 5 shows the variation of MFAC-SA-PID fuzzy self-adjusting parameters.

From the simulation results, the overshoot of MFAC-SA-PID method is smaller than that of traditional MFAC method and IMFAC method, and MFAC-SA-PID method has faster regulation time, so MFAC-SA-PID method has better control performance for pH process with serious nonlinearity.

4.2. Adding 1% step input disturbance

Consider the step disturbance of adding 1% ($\Delta q_1=0.166\ {\rm m\cdot s^{-1}}$) to the input acid flow rate q_1 at 60 min, at which time the control effect of pH value under the three methods, the variation of acid flow rate, and the fuzzy self-adjusting parameters of MFAC-SA-PID method are shown in Fig. 6, Fig. 7, and Fig. 8.

From Fig. 6, the MFAC-SA-PID method can overcome the disturbance faster than the traditional MFAC method and IMFAC

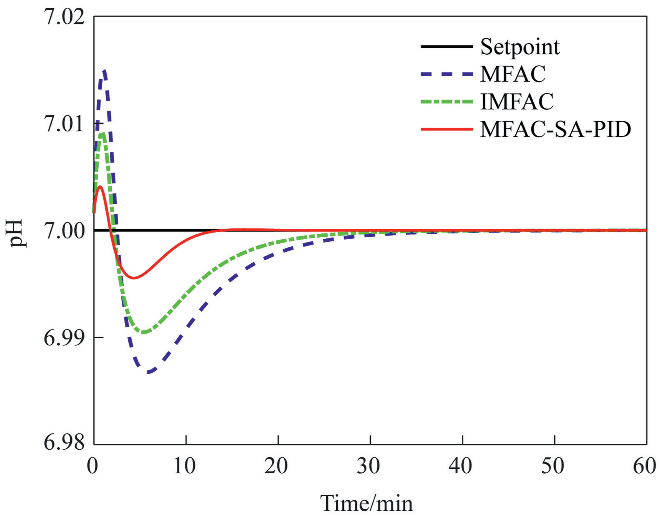

Fig. 3. Control effect of the three methods.

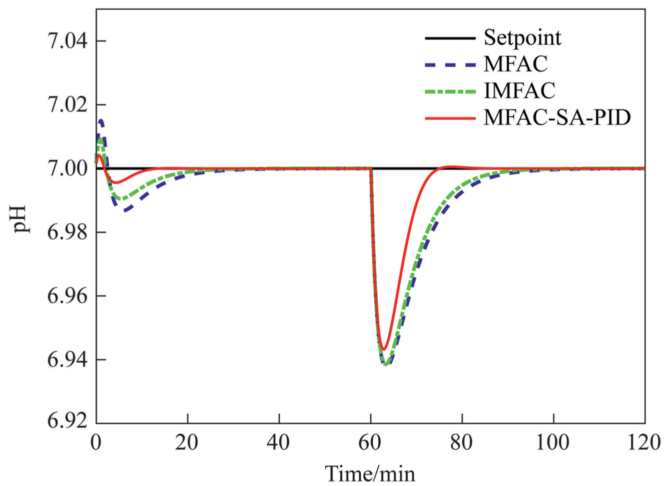

Fig. 6. The control effect under the addition of 1% step interference.

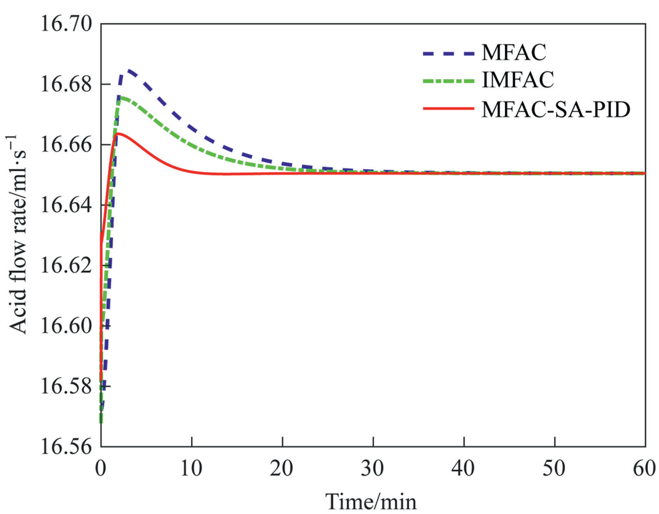

Fig. 4. Input acid flow q_1.

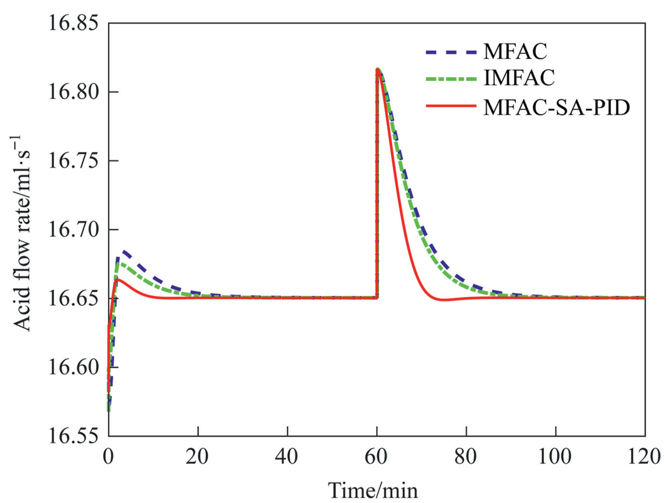

Fig. 7. Acid flow rate of the input with the addition of 1% step disturbance q_1.

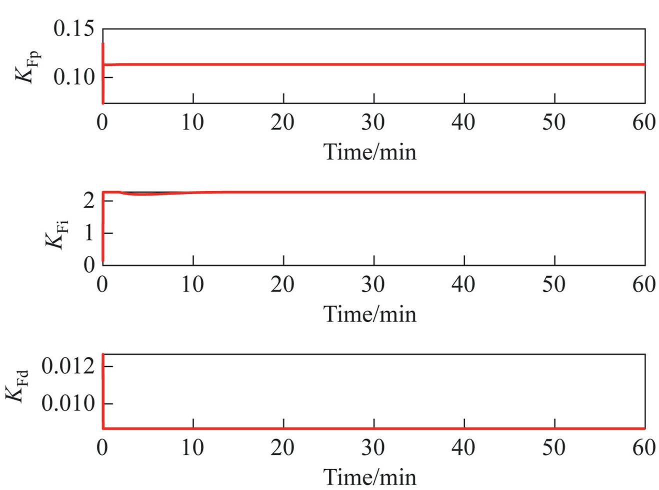

Fig. 5. Fuzzy self-adjusting parameters of MFAC-SA-PID.

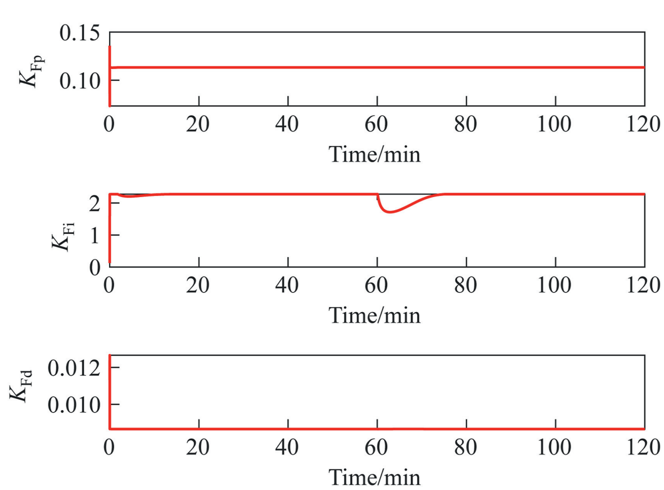

Fig. 8. Fuzzy self-adjusting parameters of SA-PID-MFAC.

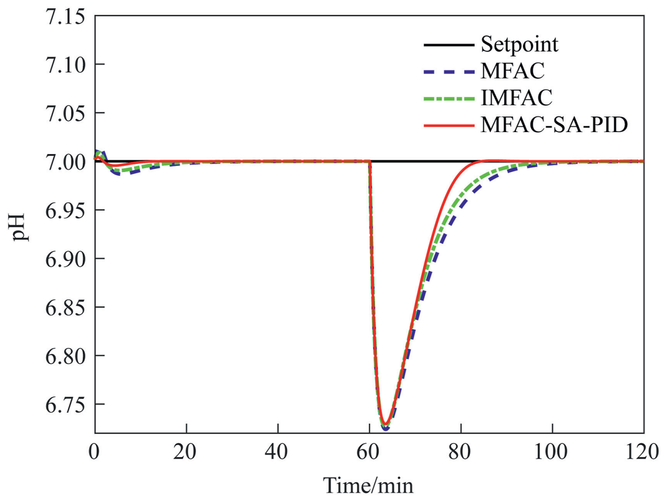

Fig. 9. Control effect with 5% step disturbance added.

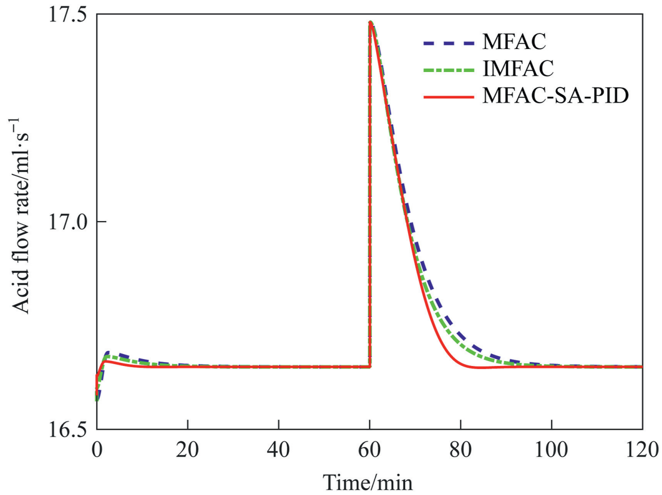

Fig. 10. Acid flow rate of input with 5% step disturbance added.

method when a 1% step input disturbance is added at 60 min. And its control performance is better than the traditional MFAC method and IMFAC method, which also shows that the MFAC-SA-PID method proposed in this paper has better anti-disturbance performance. From Fig. 7, it is easy to find that the acid flow input is regulated faster under MFAC-SA-PID method when there is a step input disturbance. Fig. 8 shows the adjustment process of the fuzzy self-adjusting parameters of the MFAC-SA-PID method under the step disturbance.

4.3. *Adding 5% step input disturbance*

To further compare the effectiveness of the three methods, consider adding a step input disturbance of 5% ($\Delta q_1 = 0.83\ \text{m}\cdot\text{s}^{-1}$) to the input acid flow rate q_1 at 60 min, at which time the control effect of pH value under the three methods, the variation of acid flow rate, and the fuzzy self-adjusting parameters of MFAC-SA-PID method are shown in Fig. 9, Fig. 10, and Fig. 11.

From the above simulation results, even under the case of large step input disturbance ($\Delta q_1 = 0.83\ \text{m}\cdot\text{s}^{-1}$), the MFAC-SA-PID method can overcome the disturbance faster and its control effect is better than the traditional MFAC method and IMFAC method,

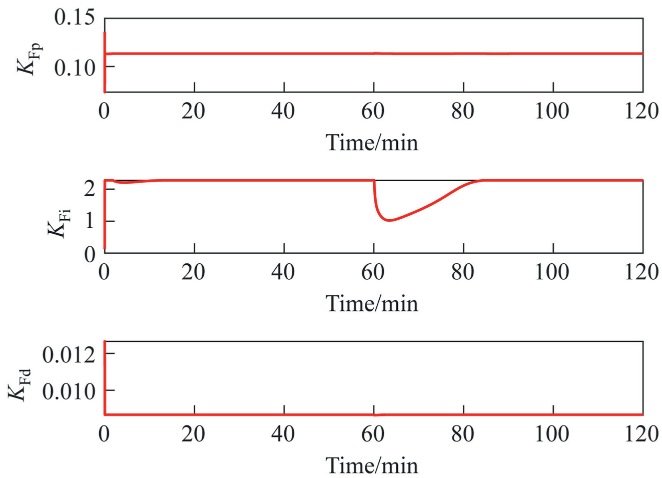

Fig. 11. Fuzzy self-adjusting parameters of MFAC-SA-PID.

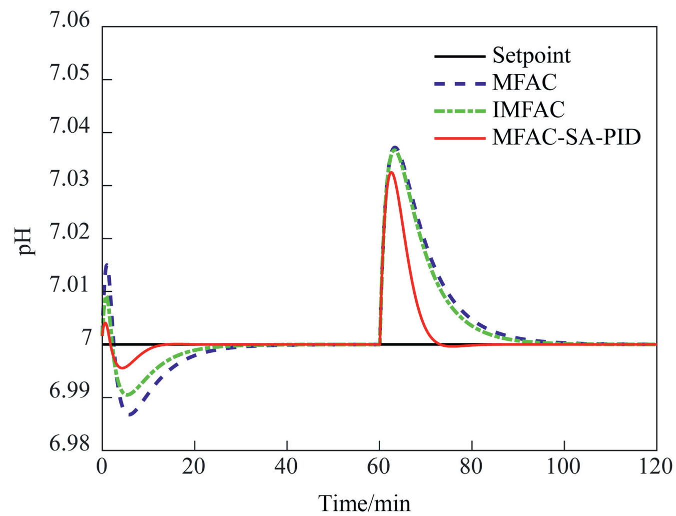

Fig. 12. Simulation results of the three methods with 10% increase in q_2.

which further shows that the MFAC-SA-PID method proposed in this paper also has better anti-disturbance performance and stability under different degrees of disturbance.

4.4. *Uncertainty of model parameters*

To verify the robustness of the proposed method, additive uncertainty is added to the system, *i.e.*, the parameter uncertainty of the controlled process objects. Consider the pH neutralization process when the model parameter q_2 is changed to further compare the effectiveness of the above three methods. Assuming that the flow rate of the buffer stream q_2 is increased by 10% ($\Delta q_2 = 0.055\ \text{m}\cdot\text{s}^{-1}$), 50% ($\Delta q_2 = 0.275\ \text{m}\cdot\text{s}^{-1}$), and 100% ($\Delta q_2 = 0.55\ \text{m}\cdot\text{s}^{-1}$) at 60 min respectively. The results of pH control under the three methods at this time are shown in Fig. 12, Fig. 13, and Fig. 14.

As shown in Figs. 12–14, the control effect of the MFAC-SA-PID method is still better than that of the conventional MFAC and IMFAC methods at 60 min when the model parameter q_2 of the pH neutralization process is changed. This further shows that the

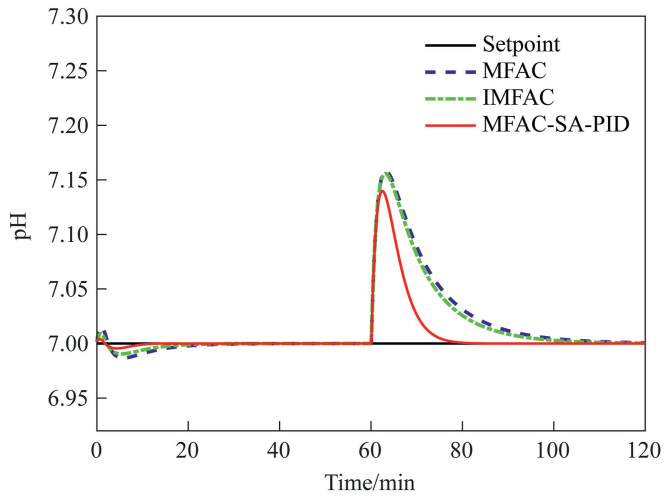

Fig. 13. Simulation results of the three methods with 50% increase in q_2.

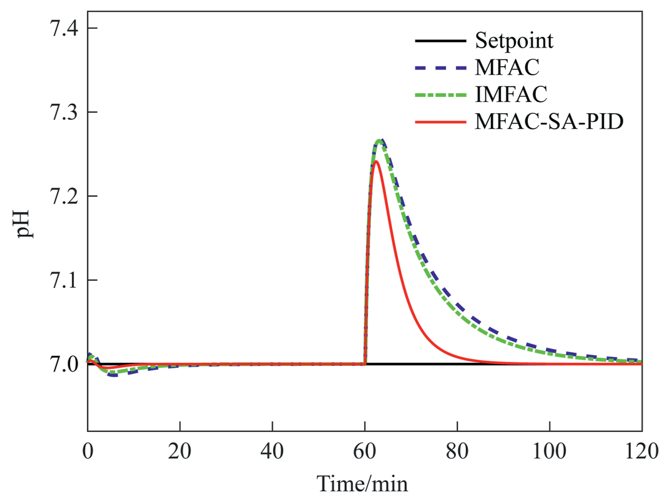

Fig. 14. Simulation results of the three methods with 100% increase in q_2.

MFAC-SA-PID method has better robustness when the model parameters are changed.

5. Conclusions

In this paper, a MFAC-SA-PID method is proposed for the problem that the pH process with strong nonlinearity and large time lag is difficult to control near the neutralization point. The MFAC-SA-PID method, which solves the drawbacks of difficult parameter adjustment and poor control performance of MFAC method by adding self-adjusting PID-parameters online. And the stability and convergence of this method are proved in this paper. Simulation studies show that the dynamic performance of the MFAC-SA-PID method is better than that of MFAC and IMFAC methods. It also shows that the MFAC-SA-PID method can better utilize the error information so that its fuzzy self-adjusting parameters can be adjusted online according to the fuzzy rules, which solves the problem that the MFAC method has a large impact on the control performance by adjusting the parameters. Therefore, the MFAC-SA-PID method has good control performance and robustness.

CRediT Authorship Contribution Statement

Kang Liu: Data curation, Formal analysis, Investigation, Methodology, Project administration, Validation, Visualization, Writing – original draft, Writing – review & editing. You Fan: Data curation. Juan Chen: Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Nomenclature

$e(k)$	the error at moment k
$\Delta e(k)$	the amount of error change
$f(\cdot)$	nonlinear systems
K_{a1}	primary ionization constant of carbonic acid
K_{a2}	secondary ionization constant of carbonic acid
K_{Fp}	fuzzy self-adjusting parameters
K_{Fi}	fuzzy self-adjusting parameters
K_{Fd}	fuzzy self-adjusting parameters
K_{d0}	initial values of fuzzy self-adjusting parameters
K_{i0}	initial values of fuzzy self-adjusting parameters
K_{p0}	initial values of fuzzy self-adjusting parameters
$K_{\Delta d}$	scale factor
$K_{\Delta i}$	scale factor
$K_{\Delta p}$	scale factor
ΔK_d	fuzzy self-adjusting volume
ΔK_i	fuzzy self-adjusting volume
ΔK_p	fuzzy self-adjusting volume
n_u	input order of the system
n_y	output order of the system
q_1	nitric acid solution flow rate, $ml\cdot s^{-1}$
q_2	sodium bicarbonate solution flow rate, $ml\cdot s^{-1}$
q_3	flow rate of mixed solution of sodium hydroxide and sodium bicarbonate, $ml\cdot s^{-1}$
q_4	solution flow rate after neutralization reaction, $ml\cdot s^{-1}$
Δq_1	the amount of change in the nitric acid solution, $ml\cdot s^{-1}$
$u(k)$	the input of the system at moment k
V	volume of continuous stirred tank reactor, $ml\cdot s^{-1}$
W_{ai}	the amount of ionization in solution, mol
W_{bi}	the amount of carbide ions in solution, mol
$y(k)$	the output of the system at moment k
y_d	desired signal
$\phi_c(k)$	pseudo partial derivative
$\hat{\phi}_c(1)$	initial value of the pseudo-partial derivative
ϕ_0	upper limit of the pseudo partial derivative
μ	step factor of MFAC
ε	a sufficiently small positive number
η	step factor of MFAC
λ	step factor of MFAC
ρ	step factor of MFAC

References

[1] H.J. Sena, F.V. da Silva, A.M.F. Fileti, ANN model adaptation algorithm based on extended Kalman filter applied to pH control using MPC, *J. Process. Contr.* 102 (2021) 15–23.

[2] C.X. Fu, X. Ma, L.X. Zhang, Fuzzy-PID strategy based on PSO optimization for pH control in water and fertilizer integration, *IEEE Access* 10 (2022) 4471–4482.
[3] D. Rathnayake, K.C. Bal Krishna, G. Kastl, A. Sathasivan, The role of pH on sewer corrosion processes and control methods: a review, *Sci. Total Environ.* 782 (2021) 146616.
[4] X. Wang, Y.B. Li, Y. Zhang, Y.R. Pan, L. Li, J.X. Liu, D. Butler, Stepwise pH control to promote synergy of chemical and biological processes for augmenting short-chain fatty acid production from anaerobic sludge fermentation, *Water Res.* 155 (2019) 193–203.
[5] İ.H. Vural, A. Altinten, H. Hapoğlu, S. Erdoğan, M. Alpbaz, Application of pH control to a tubular flow reactor, *Chin. J. Chem. Eng.* 23 (1) (2015) 154–161.
[6] A. Pawlowski, J.L. Guzmán, M. Berenguel, F.G. Acién, Control system for pH in raceway photobioreactors based on wiener models, *IFAC-PapersOnLine* 52 (1) (2019) 928–933.
[7] E.A. Herrera, T.A. Silva, Anticipative pH control of sugarcane juice, in: 2021 IEEE 5th Colombian Conference on Automatic Control (CCAC), IEEE, Ibague, Colombia, 2021.
[8] S.K. Sunori, G.S. Jethi, A.S. Bhakuni, P.K. Juneja, Neuro-fuzzy modeling of PH neutralization process in sugar mill, in: 2020 International Conference on Advances in Computing, Communication & Materials (ICACCM), IEEE, Dehradun, India, 2020.
[9] M.C. Heredia-Molinero, J. Sánchez-Prieto, J.V. Briongos, M.C. Palancar, Feedback PID-like fuzzy controller for pH regulatory control near the equivalence point, *J. Process. Contr.* 24 (7) (2014) 1023–1037.
[10] Y.J. Zhang, Y. Jia, T.Y. Chai, D.H. Wang, W. Dai, J. Fu, Data-driven PID controller and its application to pulp neutralization process, *IEEE Trans. Contr. Syst. Technol.* 26 (3) (2018) 828–841.
[11] Z.Y. Zou, D.D. Zhao, X.H. Liu, Y.Q. Guo, C. Guan, W.Q. Feng, N. Guo, Pole-placement self-tuning control of nonlinear Hammerstein system and its application to pH process control, *Chin. J. Chem. Eng.* 23 (8) (2015) 1364–1368.
[12] D. Alves Goulart, R. Dutra Pereira, Autonomous pH control by reinforcement learning for electroplating industry wastewater, *Comput. Chem. Eng.* 140 (2020) 106909.
[13] A.L. E, pH control using PI control algorithms with automatic tuning method, *Chem. Eng. Res. Des.* 79 (5) (2001) 611–620.
[14] A. Nejati, M. Shahrokhi, A. Mehrabani, Comparison between backstepping and input–output linearization techniques for pH process control, *J. Process. Contr.* 22 (1) (2012) 263–271.
[15] J. Chen, D.T. Chao, Q. Guo, Sliding mode control based on LTR observer for pH neutralization process, 2017 6th Data Driven Control and Learning Systems (DDCLS), IEEE, Chongqing, China, 2017.
[16] H. Wu, F. Yan, G.J. Wang, C. Lv, A predictive control based on decentralized fuzzy inference for a pH neutralization process, *J. Process. Contr.* 110 (2022) 76–83.
[17] L. Estofanero, R. Edwin, G. Claudio, Predictive controller applied to a pH neutralization process, *IFAC-PapersOnLine* 52 (1) (2019) 202–206.
[18] Z.Y. Zou, M. Yu, Z.Z. Wang, X.H. Liu, Y.Q. Guo, F.B. Zhang, N. Guo, Nonlinear model algorithmic control of a pH neutralization process, *Chin. J. Chem. Eng.* 21 (4) (2013) 395–400.
[19] E.H.K. Alkamil, S. Al-Dabooni, A.K. Abbas, R. Flori, D.C. Wunsch, Learning from experience: an automatic pH neutralization system using hybrid fuzzy system and neural network, *Procedia Comput. Sci.* 140 (2018) 206–215.
[20] P. Mehrpour, S. Ahmad Mirbagheri, M. Kavianimalayeri, A.H. Sayyahzadeh, M. Ehteshami, Experimental pH adjustment for different concentrations of industrial wastewater and modeling by artificial neural network, *Environ. Technol. Innov.* 31 (2023) 103212.
[21] Y. Yang, Q. Wu, A neural network PID control for PH neutralization process, in: 2016 35th Chinese Control Conference (CCC), IEEE, Chengdu, China, 2016.
[22] J. Ren, Y.S. Ding, K.R. Hao, Model-free adaptive control based on particle swarm optimization algorithm for water bath drawing process of carbon fiber, *Appl. Mech. Mater.* 303–306 (2013) 1180–1184.
[23] Y.M. Zhu, Z.S. Hou, F. Qian, W.L. Du, Dual RBFNNs-based model-free adaptive control with aspen HYSYS simulation, *IEEE Trans. Neural Netw. Learn. Syst.* 28 (3) (2017) 759–765.
[24] Z. Pang, B. Ma, W. Song, G. Liu, An improved tight-format model-free adaptive control method, *Control Decis.* 36 (2) (2021) 436–442. (in Chinese)
[25] Z. Hou, S. Jin, Model Free Adaptive Control: Theory and Applications, CRC Press, Boca Raton, FL,USA, 2013.