Relay Feedback Method for Tuning of Nonlinear pH Control Systems

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It is very difficult to control a pH process with conventional linear controller due to its severe nonlinearity. To reduce the nonlinearity of pH processes, many nonlinear methods which use empirical models or rigorous physico-chemical models of McAvoy et al. (1972) and Gustafsson and Waller (1983) have been proposed. However, due to the complexity of methods or the lack of theoretical background, just a few were used in industry. Recently Wright and Kravaris (1991) showed that the general reaction invariant model of pH process could be reduced to first-order and proposed a nonlinear pH control method called a strong acid-equivalent control. Their reduced-order model establishes the theoretical basis of many previous methods such as the piecewise linear PI controller of Shinskey (1974). The strong acid-equivalent control method is practical, because it can be implemented with only the nominal titration curve and it is robust for variations of the titration curve. For some processes, however, it is still time-consuming to obtain the nominal titration curve. Williams et al. (1990) devised a pH control system for both identification and control. It requires a special process with two strong base injections and three pH measurements.

Here, for easier implementation of nonlinear pH controllers, we propose an on-line tuning method using the relay feedback with hysteresis. Since just two or three parameters are sufficient to characterize pH processes for practical applications (Williams et al., 1990; Lee et al., 1992), we parameterize pH processes with three parameters and then identify them with the relay feedback testing. The relay feedback method is very popular in on-line tuning of PID controllers (Astrom and Hagglund, 1984; Seborg et al., 1989). This on-line tuning method would reduce the efforts to obtain titration curves with off-line experiments.

Reduced-Order Model

Consider a continuous stirred-tank mixer in Figure 1, in which the process stream is neutralized by a strong base such as sodium hydroxide (NaOH). Assuming that it has constant tank volume, perfect mixing, and fast acid/base reactions, following model equations can be obtained (McAvoy et al., 1972; Gustafsson and Waller, 1983; Wright and Kravaris, 1991):

$$V\frac{dx_i}{dt}=F(c_i-x_i)+(\alpha_i-x_i)u, \quad i=1,2,\ldots,n \tag{1}$$

$$\sum_{i=1}^{n} a_i([H^+])x_i+[H^+]-K_w/[H^+]=0 \tag{2}$$

$$pH=-\log_{10}[H^+]$$

where

c_i = total ion concentration of ith species in the process stream (mol/L)

α_i = total ion concentration of ith species in the titrating stream (mol/L)

$a_i([H^+])$ = coefficient of the ith state in Eq. 2 (Wright and Kravaris, 1991)

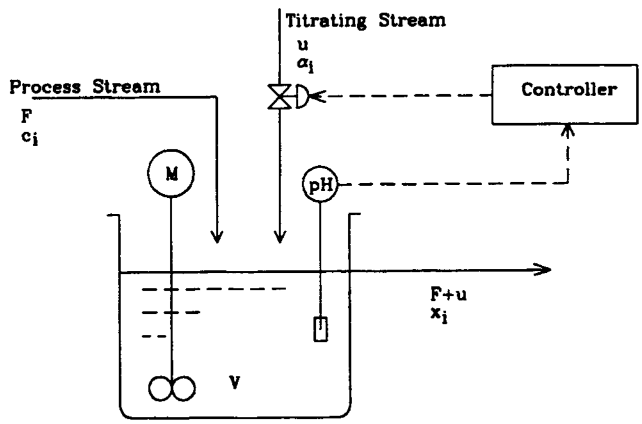

Figure 1. pH control system.

x_i = state variable, total ion concentration of ith species in the tank (mol/L)
V = volume of the tank (L)
F = flow rate of the process stream (L/s)
u = flow rate of the titrating stream (L/s)
K_w = ion product of water, 10^{-14}

Since Eq. 1 is distinct and structurally similar, it can be reduced to (Wright and Kravaris, 1991):

$$V\frac{dx}{dt} = -Fx + (1-x)u$$

$$x_i(t) = (\alpha_i - c_i)x(t) + c_i + e_i(t) \tag{3}$$

The error terms, $e_i(t)$'s, are due to mismatches in initial values of the states $x_i(t)$'s, whose magnitudes decay exponentially to zero. Equation 2 can be rewritten as:

$$x(t) = \frac{\sum_{i=1}^{n} a_i([\mathrm{H}^+])c_i + [\mathrm{H}^+] - K_w/[\mathrm{H}^+]}{\sum_{i=1}^{n} a_i([\mathrm{H}^+])(c_i - \alpha_i)} + (\text{error terms})$$

$$= \varphi(\mathrm{pH}) + (\text{error terms}) \tag{4}$$

With Eqs. 3 and 4, a pH control system can be constructed. Equation 4 contains many variables such as concentrations and dissociation constants of all species in the process stream which are very difficult to obtain. Fortunately, Wright and Kravaris (1991) showed that the righthand side of Eq. 4 requires just the information about the steady-state titration curve of process stream: that is, concentrations and dissociation constants of all detailed species in the process stream are not needed for calculating $x(t)$ from a pH measurement. For some processes, however, it may be still difficult or time-consuming to obtain the titration curves of process streams.

Parameterization of the Titration Curve

We consider a neutralization process of acid stream by a strong base. Recently, several authors proposed a model based on one fictitious weak acid and obtained very promising results for practical multicomponent processes (Parrish and Brosilow, 1988; Williams et al., 1990; Li et al., 1990). The model contains two parameters of concentration and dissociation constant of the fictitious weak acid whose combinations are linear. A more general model that assumes the process stream to consist of one strong acid and one weak acid with a dissociation constant K can be derived:

$$x(t) = \frac{-c_2 - c_3/(1+[\mathrm{H}^+]/K) + [\mathrm{H}^+] - K_w/[\mathrm{H}^+]}{-\alpha_1 - c_2 - c_3/(1+[\mathrm{H}^+]/K)} \tag{5}$$

where
α_1 = known concentration of the titrating strong base (mol/L)
c_2 = concentration of fictitious strong acid in the process stream (mol/L)
c_3 = total ion concentration of fictitious weak acid in the process stream (mol/L)
K = dissociation constant of the fictitious weak acid in the process stream

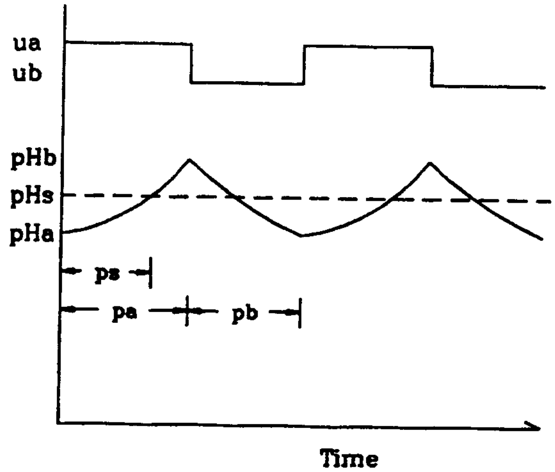

Figure 2. Typical response of a relay feedback system with hysteresis.

Three unknown parameters in Eq. 5 can be rearranged:

$$\{(x-1)[\mathrm{H}^+]\ \ [\mathrm{H}^+]^2 + \alpha_1 x[\mathrm{H}^+]\ \ -K_w\ \ (x-1)[\mathrm{H}^+]^2\}\begin{pmatrix} K(c_2+c_3) \\ K \\ c_3 \end{pmatrix} = \{-[\mathrm{H}^+]^3 + K_w[\mathrm{H}^+] - \alpha_1 x[\mathrm{H}^+]^2\} \tag{6}$$

Hence, because their combinations are linear, three unknown parameters can be estimated easily as in the two-parameter model; we propose an on-line method that estimates them conveniently.

Relay Feedback Identification

Relay with hysteresis produces a periodic response as shown in Figure 2. Since $u(t)$ of the relay output is piecewise constant, Eq. 3 becomes linear and can be solved analytically. That is, three states x_a, x_b and x_s corresponding to pH_a, pH_b and pH_s of Figure 2 are obtained as follows. x_a and x_b are the solution of:

$$x_a = x_b \exp\left(-\frac{F+u_b}{V}p_b\right) + \frac{u_b}{F+u_b}\left[1-\exp\left(-\frac{F+u_b}{V}p_b\right)\right]$$

$$x_b = x_a \exp\left(-\frac{F+u_a}{V}p_a\right) + \frac{u_a}{F+u_a}\left[1-\exp\left(-\frac{F+u_a}{V}p_a\right)\right]$$

and x_s is:

$$x_s = x_a \exp\left(-\frac{F+u_a}{V}p_s\right) + \frac{u_a}{F+u_a}\left[1-\exp\left(-\frac{F+u_a}{V}p_s\right)\right]$$

From these three pairs, (x_a,pH_a), (x_b,pH_b) and (x_s,pH_s), we estimate three unknown parameters, c_2, c_3 and K satisfying Eq. 6.

For some processes such as one weak acid solution, three parameters become overparameterized and meaningless parameters are obtained due to the numerical problem. So, we

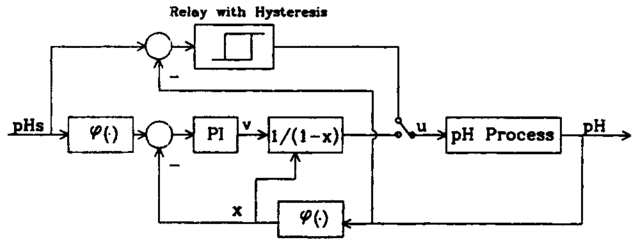

Figure 3. Nonlinear pH control system.

first find c_3 and K with $c_2=0$ from data pairs (x_a,pH_a) and (x_b,pH_b). If the righthand side is within 10% of the lefthand side of Eq. 6 for the pair (x_s,pH_s), we use them; otherwise, we find the three parameters satisfying Eq. 6 for all three pairs.

To evaluate the relay feedback identification method, we use a linearizing control system as shown in Figure 3, where the state $x(t)$ is used for feedback. The control scheme is different only in scaling from that of Wright and Kravaris (1991). Hence, both control performances for regulation of pH are the same. Gain and integral time of the inner PI controller are chosen as:

$$K_c = 1.414\omega V - F$$

$$T_I = K_c/[\omega^2 V]$$

Since the linearized system is:

$$V\frac{dx}{dt} = -Fx + v$$

above PI controller parameters make the two closed-loop poles at $\omega(-0.707 \pm 0.707j)$ recommended in the root-locus design. Faster responses can be obtained by increasing ω at the expense of higher sensitivity to the modeling error and measurement noises.

Simulation

Three processes are simulated. Two are such that our three parameter model can describe the nonlinearity of the titration curves well and one is a multicomponent process. In all simulations, we integrate Eq. 1 analytically as in Wright and Kravaris (1991) and solve the nonlinear equation (Eq. 2) by the bisection method. Table 1 shows the results obtained.

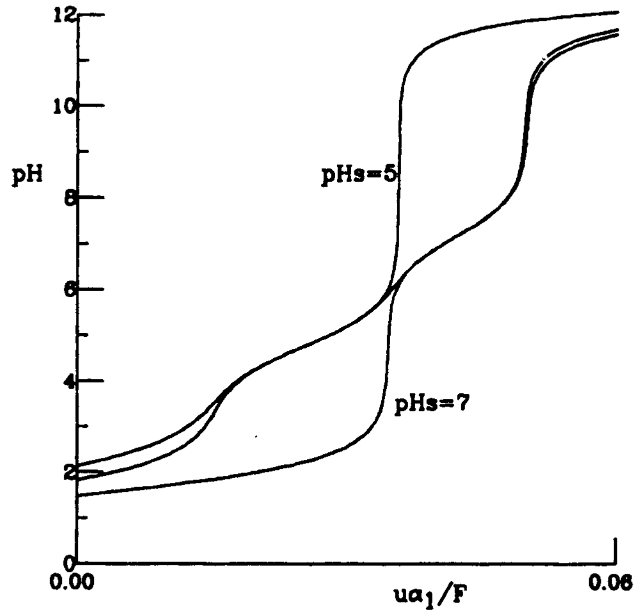

Figure 4. Titration curves for process iii and models of $\mathrm{pH}_s=5$ and $\mathrm{pH}_s=7$ without noise-free measurements.

For the first two processes, the identified models approximate the actual titration curves very well as expected, and good control responses are obtained. For the last process, it is difficult to approximate the entire titration curve very well by the three-parameter model. Figure 4 shows titration curves of the process and models. We can see that the models describe the process titration curves near the setpoints well and that they will be sufficient for regulation of the pH levels at the given setpoints. This is shown in Figure 5, in which $\omega=0.1$ is used. Reset windup of the PI controller is avoided simply by freezing the integration when the control input exceeds the upper or lower limit. As also shown in Figure 5, control performances are not so good for setpoints far from where models are evaluated. That is, in the case of frequent large setpoint changes, the entire titration curve by the off-line experiment or by the repeated applications of this method might be used.

The pH measurements are often corrupted by noises. Unbiased random noises decrease the periods p_a and p_b signifi-

Table 1. Results of the Relay Feedback Identification for Three pH Processes

Process*	pH_s	ΔpH	Noise	u_a	u_b	p_a	p_b	p_s	c_2	c_3	K
i) $CH_3COOH=0.025$ mol/L	7.0	0.5	0	0.02	0.01	28.5	4.54	21.0	0	0.025	1.79E-5
ii) $H_3PO_4=0.025$ mol/L	7.0	0.5	0	0.05	0.005	20.0	91.3	8.88	0.025	0.025	6.33E-8
iii) $CH_3COOH=0.02$ mol/L $H_3PO_4=0.015$ mol/L	5.0	0.5	0	0.02	0	67.8	65.1	36.3	0.0151	0.0204	1.69E-5
	5.0**	0.5	±0.1pH	0.02	0	69.8	66.3	38.0	0.0144	0.0213	1.44E-5
	5.0**	0.5	±0.5pH	0.02	0	70.2	67.5	36.6	0.0153	0.0208	1.53E-5
	7.0	0.5	0	0.02	0.01	97.2	81.3	37.4	0.0344	0.0154	7.01E-8
	7.0**	0.5	±0.1pH	0.02	0.01	98.0	81.8	38.7	0.0386	0.0164	6.85E-8
	7.0**	0.5	±0.5pH	0.02	0.01	91.3	76.0	35.8	0.0391	0.0153	6.94E-8

*$V=5$ L, $F=0.0188$ L/s, $\alpha_1=0.05$ mol/L and dissociation constants: $CH_3COOH=1.8E\text{-}5$, $H_3PO_4=(7.11E\text{-}3, 6.34E\text{-}8, 4.2E\text{-}13)$.
**First-order low-pass filter with the 1-second time-constant is used for smoothing the noise.

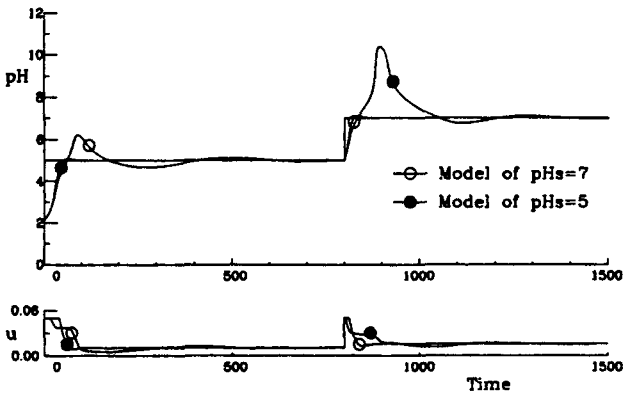

Figure 5. Control performance for process iii without measurement noise.

Models obtained with the relay feedback method for $pH_s = 5$ and $pH_s = 7$ are used.

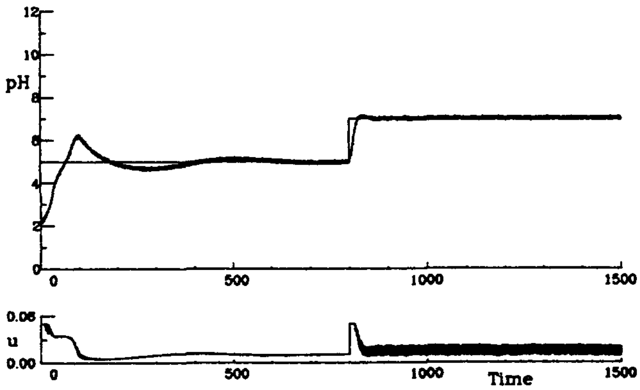

Figure 6. Control performance for process iii with measurement noise of ±0.1pH.

Model of $pH_s = 7$ is used.

cantly. A low pass filter can be used for reducing the effects of unbiased random noises. Table 1 shows the identification results that the pH measurements are corrupted with uniform random noises of ±0.1 pH and ±0.5 pH. Here, to smooth out the noise, the first-order exponential filter with 1-second time-constant is used. Although there are 15% deviations in the model parameters as compared with the noise-free ones, control performances are still good as shown in Figure 6. It is mainly due to the robustness of the pH control system of Figure 3.

Effects of incorrect information of α_1 and F are also simulated. The estimates are well within the accuracy of α_1 and F. Particularly, most important parameter K for characterizing the nonlinearity of the titration curve is well within ±4% for 10% variations of α_1 or F.

In summary, the relay feedback method can be used as an on-line tuner of nonlinear pH controller for regulating the pH level of processes. For the applications to be more reliable, further studies on robustness in various situations and process noises would be required.

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