

# Application of self-tuning PID control to a reactor of limestone slurry titrated with sulfuric acid

M. Alpbaz, H. Hapoğlu, G. Özkan\*, S. Altuntaş

*Department of Chemical Engineering, Ankara University, Tandogan 06100, Ankara, Türkiye*

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## Abstract

One of foremost air-polluting which can be determined as being deterioration in the quality of the air is the emission of sulfur-dioxide gas. In this study,  $\text{CaCO}_3$  solution which is one of the method used for the removal of the waste gas  $\text{SO}_2$  from air was used. The neutralization of  $\text{CaCO}_3$  with  $\text{H}_2\text{SO}_4$  was realized experimentally at the pH value that dissolution rate is maximum.

The neutralization of limestone with  $\text{H}_2\text{SO}_4$  was realized in a stirred continuous reactor. pH value of the medium was controlled by utilizing self-tuning PID (STPID) algorithm and the on-line computer control system. ARMAX was used as the system model. A pseudo-random binary sequence (PRBS) was utilized as a forcing function in order to identify the dynamics of the process to be controlled and the system output was measured. The model parameters were evaluated by using Bierman algorithm. The tuning parameters of the STPID controller were determined.

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## 1. Introduction

Pollution can be caused by pre-eminently pollutants such as particle,  $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{H}_2\text{S}$  in the earth's atmosphere. They play a major role in environmental pollution by both natural and artificial means. This situation is known as air pollution and it is defined as being deterioration in the quality of the air as a result of such phenomena. One of the foremost air-polluting emission is sulfur-dioxide gas ( $\text{SO}_2$ ).  $\text{SO}_2$  combines with water vapor in the air and then this gas forms the droplets of sulfuric acid, which fall to the ground as acid rain, causing harm to everything living and non-living.

Over 200 processes have been given in literature on the removal of  $\text{SO}_2$  from flue gases and among these processes about twenty of them have been used in power plants and in other industries. These processes can in general be classified as wet and dry processes [1,2]. In the wet limestone flue gas desulfurization process, powdered limestone dissolves and neutralizes acidity produced by  $\text{SO}_2$  absorption in the liquid phase [3]. A lot of studies have been carried out on the reactions of calcium carbonate with acidic solutions.

In the removal of  $\text{SO}_2$  from flue gases and among these processes, pH control is a very important phenomena [4–7]. The control of pH is recognized as a difficult problem in the literature due to its highly non-linear nature. On the other hand, the control of pH is industrially important for several reasons. The most common pH process is the neutralization of an acidic or basic waste stream, which may be necessary for any of the following reasons: prevent corrosion and or damage to construction materials, protect aquatic life and human welfare, as an initial treatment, allowing effective operation of biological treatment processes, provide neutral pH water for recycle, either as process water or as reboiler feed.

In the present study, the pH control of the neutralization process of limestone with  $\text{H}_2\text{SO}_4$  was realized in a stirred continuous reactor by utilizing self-tuning PID (STPID) algorithm. ARMAX was used as the system model. A pseudo-random binary sequence (PRBS) was utilized as a forcing function in order to identify the dynamics of the process to be controlled and the system output was measured. The model parameters were evaluated by using Bierman algorithm. The tuning parameters (e.g.  $t_1$ ) of the STPID controller were determined by ISE and IAE criteria. In the removal of  $\text{SO}_2$  from flue gases and among these processes, pH control is a very important phenomena. The control of pH is recognized as a difficult problem in the literature due to its highly non-linear nature. One of the purposes

\* Corresponding author. Tel.: +90 312 2126720 1302.  
E-mail address: gozkan@eng.ankara.edu.tr (G. Özkan).

### Nomenclature

$A(z^{-1})$	monic polynomial in the $z$ -domain representing the poles of the discrete-time
$a_i$	parameters of $A$ polynomial
$B(z^{-1})$	polynomial in the $z$ -domain representing the zeros of the discrete-time system
$b_i$	parameters of $B$ polynomial
$C(z^{-1})$	monic polynomial in the $z$ -domain representing the zeros of the process noise
$e(t)$	white noise
IAE	error absolute value integral
ISE	error square integral
$K_c$	steady-state gain for three term controller
$r(t)$	set point
$U(t)$	manipulated variable
$y(t)$	output variable

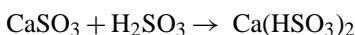
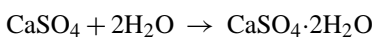
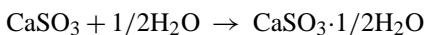
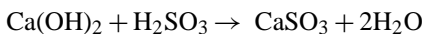
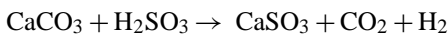
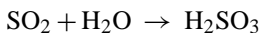
### Greek letters

$\varepsilon(t)$	difference between the measured variable and set point at time $t$
$\tau_D$	derivative constant coefficient
$\tau_I$	integral constant coefficient

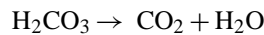
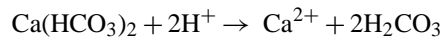
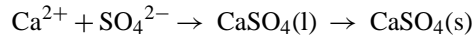
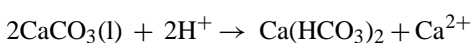
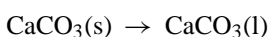
of this paper is to show that the assumption of a linear second order ARMAX model, together with self-tuning PID algorithm provides satisfactory pH control. The second purpose is to control process at the set point in which  $\text{CaCO}_3$  dissolution is the highest level.

## 2. Reaction of disulfurization

A short description of the chemical reactions are given below:  
Reaction for  $\text{SO}_2$ :



Dissolution of limestone into  $\text{H}_2\text{SO}_4$  for this study:



## 3. Self-tuning control

Process model used is generally a controlled auto regressive moving average model (CARMA) or auto regressive moving average exogenous (ARMAX) [8–10]. For a single-input single output system to be controlled, the equation:

$$A(z^{-1})y(t) = z^{-k}B(z^{-1})u(t) + C(z^{-1})e(t) \quad (1)$$

where  $A$ ,  $B$  and  $C$  are polynomials in the backward shift operator ( $z^{-1}$ ) and  $k$  is the system time delay associated with the control input.  $A$  and  $B$  represents the poles and zeros of the discrete time system, respectively.  $C$  contains the zeros of process noise and  $e(t)$  is an uncorrected random sequence.  $y(t)$  is system output at time  $t$  and  $u(t)$  is system input.

In self-tuning control, the model parameters are estimated on-line and the controller settings based on current parameter estimator are adjusted. The self-tuning approach has received more attention than any other adaptive control strategy. Process model used is generally a CARMA with a form of least square parameter estimation. CARMA model can be given as

$$y(t) = x^T(t)\theta^T + e(t) \quad (2)$$

Where  $x$  is the data vector,  $\theta$  the parameter vector defined as the collection of coefficients in the  $A$ ,  $B$ , and  $C$  polynomials, and  $e$  is random noise.  $\theta$  and  $x$  are given by:

$$\theta^T = [a_1, a_2, \dots, a_{na}, b_0, b_1, \dots, b_{nb}, d_0, c_1, c_2, \dots, c_{nc}] \quad (3)$$

$$x^T = [y(t-1), y(t-2), \dots, y(t-na), u(t-1), u(t-2), \dots, u(t-nb-1), 1, e(t-1), \dots, e(t-nc)] \quad (4)$$

The discrete form of the PID control algorithm can be converted into a self-tuning equivalent. The control equation is given as follows:

$$U(t) = \frac{S}{R} [r(t) - y(t)] \quad (5)$$

Here  $r(t)$  represents the set point, and:

$$S = s_0 + s_1z^{-1} + s_2z^{-2} \quad (6)$$

$$s_0 = K_c \left( 1 + \frac{\Delta t}{2\tau_I} + \frac{\tau_D}{\Delta t} \right) \quad (7)$$

$$s_1 = K_c \left( -1 + \frac{\Delta t}{2\tau_I} - \frac{2\tau_D}{\Delta t} \right) \quad (8)$$

$$s_2 = K_c \left( \frac{\tau_D}{\Delta t} \right) \quad \text{and} \quad R = (1 - z^{-1}) \quad (9)$$

Here  $\Delta t$  is the sampling interval. The PID constants can be found from the values of  $s_0$ ,  $s_1$  and  $s_2$ . Substituting the control equation

into CARMA, process model yields the following closed-loop response equation:

$$y(t) = \frac{z^{-1}BS}{AR + z^{-1}BS}r(t) + \frac{RC}{AR + z^{-1}BS}e(t) \quad (10)$$

The characteristic equation is called as Tailoring polynomial  $T$  and it is given by:

$$T(z^{-1}) = A(z^{-1})R + z^{-k}B(z^{-1})S(z^{-1}) \quad (11)$$

The properties of this closed-loop can be varied by placing the poles of the characteristic equation within the unit-circle in the  $z$  plane. The coefficients of the  $A$  and  $B$  polynomials are estimated from The Bierman UDU<sup>T</sup> algorithm [11] and the coefficients of the  $T$ -polynomial are defined by user.  $s_0$ ,  $s_1$  and  $s_2$  can be found from the characteristic Eq. (11).

The degrees of the polynomials in the characteristic equation are:

$$n_a + n_r = n_b + n_s + 1 = n_t \quad (12)$$

where  $n_s$  is the degrees of the  $s$  and it is taken as 2, and  $n_r$  is the degree of  $r$  polynomials its value must be 1, because of the polynomial representation of velocity form of the PID algorithm. This means that  $n_a = n_b + 2$  and  $n_t = n_b + 3 = n_a + 1$ . If a second order  $A$  polynomial ( $n = 2$ ,  $n_b = 0$  and  $n_t = 3$ ) is selected a unique set of PID controller coefficients can be obtained from the design. If the order of the  $A$  polynomial is three, i.e.  $n = 3$ ,  $n_b = 1$  and  $n_t = 4$ , all the coefficients of  $T$  polynomial should be user defined to placed the poles of the characteristic equation easily. In this case, the system transfer function chosen, is a third order  $T$  polynomial ( $n = 2$ ,  $n_b = 1$ ) and has the form:

$$y(t) = \frac{b_0z^{-1}}{1 + a_1z^{-1} + a_2z^{-2}}u(t) \quad (13)$$

The closed loop relationship is obtained by combining the system model equation (Eq. (13)) and the controller equation (Eq. (5)) as

$$y(t) = \frac{b_0z^{-1}S}{R(1 + a_1z^{-1} + a_2z^{-2}) + b_0z^{-1}S}r(t) \quad (14)$$

The equivalent chosen closed loop  $T$  polynomial is of the form:

$$T = 1 + t_1z^{-1} + t_2z^{-2} + t_3z^{-3} \quad (15)$$

By equating the denominator of Eq. (14) with Eq. (15), the following relationships are obtained:

$$s_0 = \frac{(t_1 - a_1 + 1)}{b_0} \quad (16)$$

$$s_1 = \frac{(t_2 - a_2 + a_1)}{b_0} \quad (17)$$

and:

$$s_2 = \frac{(t_3 - a_2)}{b_0} \quad (18)$$

The discrete form of the necessary incremental PID control law may be written in terms of the change in the control signal as

$$\Delta U = s_0\varepsilon(t) + s_1\varepsilon(t-1) + s_2\varepsilon(t-2) \quad (19)$$

The steps in the operation of the self-tuning used in this study can be given as

- (a) Apply a perturbation to the system as a forcing function and attain the plant output.
- (b) Estimate  $A$  and  $B$  from the CARMA model using Bierman U–D update algorithm.
- (c) Calculate  $s_0$ ,  $s_1$ , and  $s_2$  from equations.
- (d) Find  $K_c$ ,  $\tau_I$  and  $\tau_D$  from equations.
- (e) Obtain the incremental control signal from equations.
- (f) Output the updated control signal to the process.
- (g) Return to (c).

In this work, the form of the model of the system to be controlled is preserved to ensure that only one set of PID controller coefficients is produced from the design, and the integral action in the PID controller provides steady-state following even if the parameter values of the system or controller change.

#### 4. Experimental system

The dissolution rate of the limestone and dynamic properties of this system were observed in a jacketed batch reactor. The experiment was initiated by the addition of limestone to the pure water. A sulfuric acid solution ( $H_2SO_4$ ) was titrated into a limestone slurry of 2 L in the reactor to maintain the pH. The reactor temperature was kept constant with hot water passing through the reactor jacket. Feed flow rate is controlled by using a pump adjusted by on-line computer control system. All the dynamic properties such as pH, temperature of the reactor can be observed with this on-line computer control system (Fig. 1).

0.04 M  $H_2SO_4$  is used as acid source. During the titration, the rate of addition of acid is continuously adjusted so as to bring the pH of the slurry to the desired value just in time. Limestone used in the present work is high quality limestone. The total time of titration for one run is about 35 min which is believed to provide the product accumulation sufficient enough to see its effects. At each time step, the CPU time required by the STPID methods is enough. The sampling time is chosen as about system dead time. The dead time of the process is 1.0 s. The constraints of manipulated variable is added to on-line computer program.

#### 5. Results and discussion

In the first part of the work, process model parameters were observed. In this study, second order ARMAX model (Eq. (20)) was used:

$$y(t) = \frac{b_0}{1 + a_1z^{-1} + a_2z^{-2}}u(t-1) + e(t) \quad (20)$$

PRBS signal was given to the process in the open-loop. The changes in PRBS effects and the changes of pH with time were

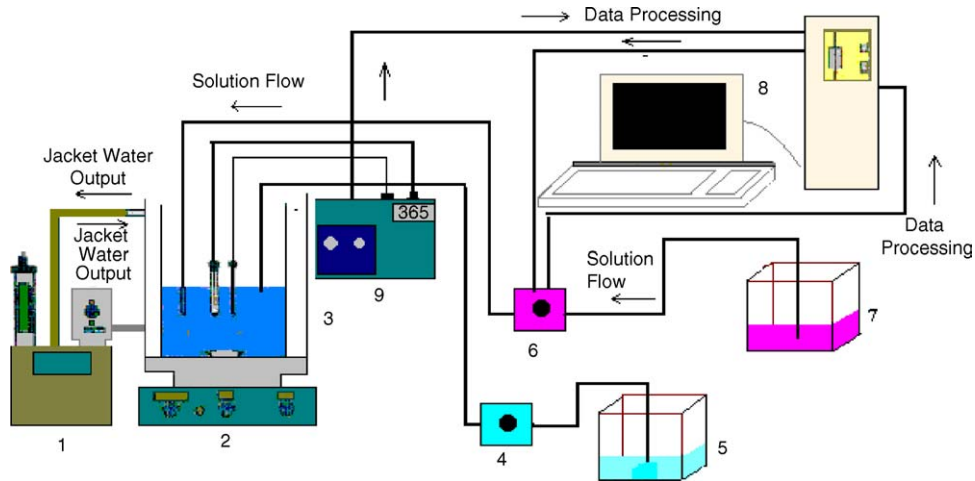


Fig. 1. Experimental system. (1) Thermo bath, (2) stirrer, (3) reactor, (4) base pump, (5) base tank, (6) acid pump, (7) acid tank, (8) computer, (9) pH meter.

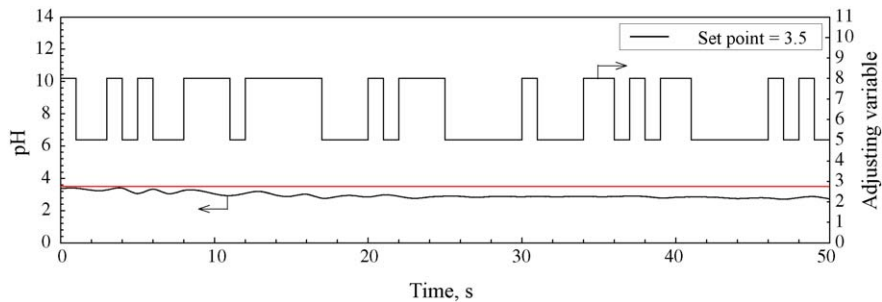


Fig. 2. PRBS signal and system result.

observed by on-line computer. These PRBS signal and the process pH values are given in Fig. 2. Model parameters were calculated from these data given in Fig. 2 by using Bierman algorithm. While base flow rate is constant, acid flow rate is used as manipulated variable and STPID control is realized at the set point of pH 3.5 without adding  $\text{CaCO}_3$ . The best control performance was obtained by using  $t_1 = 0.00005$  as a tuning

parameter. The oscillatory behavior of the control result is shown in Fig. 3. The same type of control is also realized by adding 30 g  $\text{CaCO}_3$  as a disturbance. The control result under this load effect is shown in Fig. 4. STPID control result is very good in the face of instant  $\text{CaCO}_3$  addition as a load affect.

PRBS signal was used to found model parameters. The model parameters of the system is given in Table 1. The sampling inter-

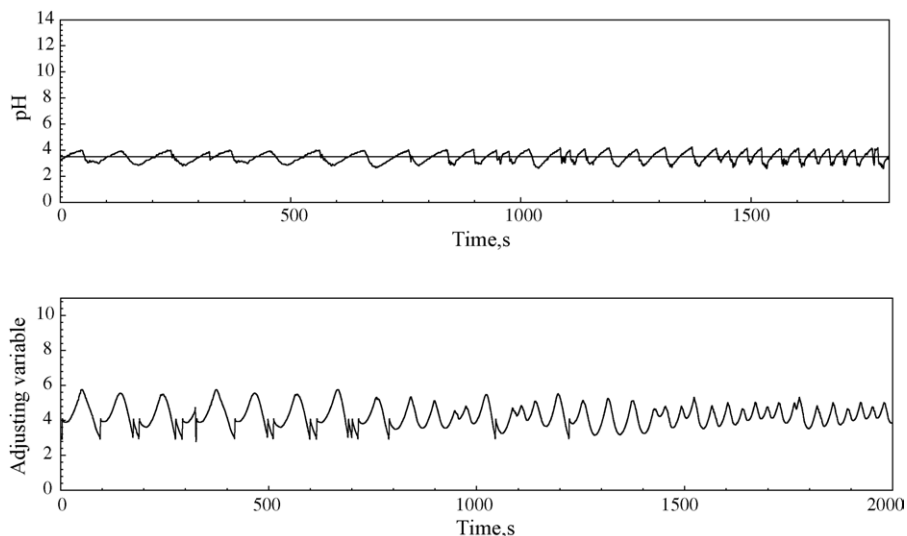


Fig. 3. pH control of a neutralization process with STPID ( $m_{\text{CaCO}_3} = 0$ ,  $t_1 = 0.00005$ ).

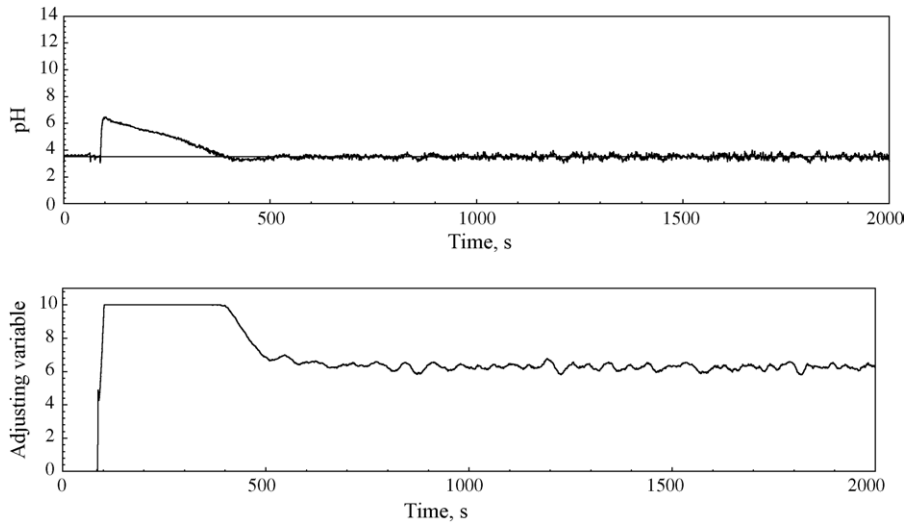


Fig. 4. pH control of a neutralization process with STPID ( $m_{CaCO_3} = 30$  g vs.  $t_1 = 0.00005$ ).

Table 1  
Model parameter values

Model parameters	Parameter value
$a_1$	-0.5227851
$a_2$	0.1826556
$b_0$	0.0046153

Table 2  
ISE vs. IAE values for linear model (not under charge force)

$t_1$	ISE	IAE
0.00005	1355.656	1323.823
0.00001	1313.369	1243.175
0.000005	895.028	564.603
0.000001	851.081	599.151

Table 3  
ISE vs. IAE values for linear model (under charge force)

$t_1$	ISE	IAE
0.00005	1034.929	721.345
0.00001	1369.289	785.433
0.000005	914.811	733.949
0.000001	677.779	541.524
0.0000005	1146.241	723.008
0.0000001	2248.228	976.931
0.00000005	2467.379	1059.256
0.00000001	2563.584	1623.250

val is chosen experimentally in our system. 1.0 s is found as the best sample time. This is a feedback strategy in control. Controller handles any disturbances between sample times by measuring the error in the next sample time. In Eq. (16) the valve of  $t_1$  is used as a tuning parameters of STPID. The effect of this value is shown in Tables 2 and 3. The performance is given by using ISE and IAE criterias in the same table.

To find the control performance, ISE and IAE criterias were calculating by using the following Eqs. (21) and (22):

$$ISE = \sum_{t=0}^{t_1} [y(t) - r(t)]^2 \quad (21)$$

$$IAE = \sum_{t=0}^{t_1} [y(t) - r(t)] \quad (22)$$

For one of  $t_1$  value, control result (not under charge force) is given in Fig. 3, and control result (under charge result) is given in Fig. 4. A standart PID application is given in Fig. 5. The

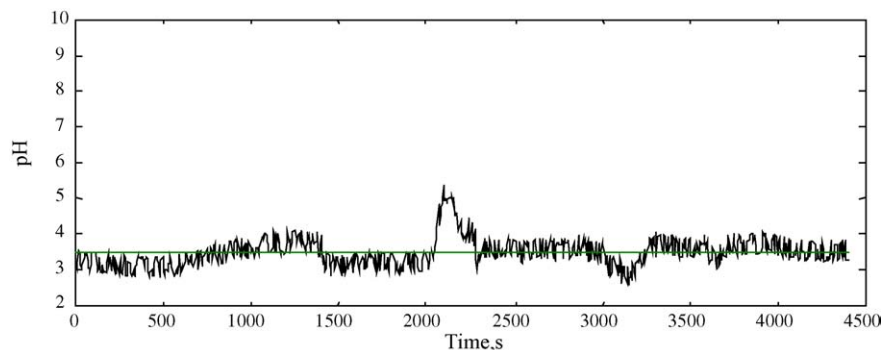


Fig. 5. pH control of a neutralization process with PID ( $m_{CaCO_3} = 30$  g,  $K_c = 31.17$ ,  $T_1 = 118.9$ ,  $T_d = 17.9$ ).

comparison of control performances between STPID and PID is done. The STPID shows very satisfactory control than PID. The reactions for  $\text{SO}_2$  and the ones given for this study in reaction of disulfurization equations shows that  $\text{H}_2\text{SO}_4$  used in the control algorithm is replaced into  $\text{H}_2\text{SO}_3$  in reaction for  $\text{SO}_2$ . The solution of  $\text{H}_2\text{SO}_3$  does not behave in exactly the same way as sulfuric acid. However, from a process control point of view sulfuric acid can be used as manipulating variable [12]. In the wet limestone flue gas desulfurization process, powdered limestone dissolves and neutralizes acidity produced by  $\text{SO}_2$  absorption in the liquid phase.

## References

- [1] A.V. Slack, G.A. Halliden, Noyes Data Corp, Dark Ridge.
- [2] V. Boscak, A. Saleem, Proceedings of the Third Annual Symposium on Industrial Coal Utilization, Tennessee, 1981.
- [3] U. Naohiko, O. Susumu, O. Michio, O. Tsuyoshi, J. Chem. Eng. Jpn. 26 (1993) 112–113.
- [4] T.E. Wentzler, F.F. Aplan, Proceedings of the Environmental Control Prac. Symposium, AIME, 1972; A.A. Metry, The Handbook of Hazardous Waste Management, Technomic, Westport, CT, 1980.
- [5] E. Katende, A. Jutan, A new constrained self-tuning PID controller, Can. J. Chem. Eng. 71 (1993) 625–633.
- [6] A. Loh, D. Sankar De, P.R. Krishnaswamy, pH and level controller for a pH neutralization process, Ind. Eng. Chem. Res. 40 (2001) 3579–3584.
- [7] T.K. Gustafsson, B.O. Skrifvars, K.V. Sandström, K.V. Waller, Modeling of pH for control, Ind. Eng. Chem. Res. 34 (1995) 82–827.
- [8] D.E. Seborg, T.F. Edgar, S.L. Shah, AIChE J. 32 (1986) 881.
- [9] R.B. Newell, P.L. Lee, Applied Process Control, Practical Hall, Newyork, 1989.
- [10] P.E. Wellstead, M.B. Zarrop, Self-Tuning Systems Control and Signal Processing, Wiley, Great Britain, 1991.
- [11] G.J. Bierman, Measurement updating using the U–D factorization, Automatica 12 (1976) 375–382.
- [12] N. Ukawa, S. Okino, M. Oshima, T. Oishi, Effects of salts on limestone dissolution rate in wet limestone flue gas desulfurization, J. Chem. Eng. Jpn. 26 (1) (1993) 112–113.