MIMO control application for pulp and paper mill wastewater treatment by electrocoagulation

Sule Camcioglu*, Baran Ozyurt, Hale Hapoglu

Ankara University, Faculty of Engineering, Department of Chemical Engineering, Tandogan Ankara, Turkey, Tel. +90 3122033465, email: camcioglu@eng.ankara.edu.tr (S. Camcioglu), bozyurt@ankara.edu.tr (B. Ozyurt), hapoglu@eng.ankara.edu.tr (H. Hapoglu)

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ABSTRACT

In this work batch treatment of pulp and paper mill wastewater using electrocoagulation (EC) has been investigated. Electrical conductivity, temperature and pH were selected as controlled variables; supporting electrolyte, cooling water, coordinated acid and base solution flow rates were selected as manipulated variables respectively. Dynamic analyses were carried out to statistically model the variations of controlled variables with time. Second order auto regressive moving average with external input (ARMAX) was used as model and obtained dynamic data were used to fit model parameters with recursive least squares (RLS) method. Pole placement approach was utilized to design robust proportional integral derivative (PID) controllers. PID parameters were found as 5, 0.05 and 0.01 for conductivity, 48, 0.5 and 0.01 for temperature, 1000, 0.05, 0.01 and 1, 0.06, 0.4 for coordinated pH acid and base controllers respectively. Controllers were tested in simulation environments depending on ARMAX models. Experimental multi input-multi output (MIMO) control of variables was achieved using real time PID control algorithms coded in Simulink®. MIMO conductivity, temperature and pH control increased color, turbidity and chemical oxygen demand (COD) removal efficiency as 8.19%, 13.29% and 10.81% respectively, when compared with uncontrolled treatment. Energy consumption of the EC process was reduced by 31.65% and 21.72% compared to the energy consumption of the uncontrolled process using MIMO conductivity and temperature control and MIMO control of three parameters, respectively. It is observed that MIMO control increased pollutant removal and decreased energy consumption simultaneously, which makes the process more economical.

Keywords: Multi input–multi output control; Coordinated control; Electrocoagulation; Pulp and paper mill wastewater

1. Introduction

It is reported that the most commonly used treatment methods of pulp and paper effluent are physical adsorption [1,2], chemical oxidation [3,4] and biochemical [5,6] methods. The constituents of wastewater generated from pulp and paper industry is very complex for component characterization as for other industrial wastewaters [7]. A comparison of some treatment methods of industrial wastewaters is noted in Table 1. One of the environmental impacts related to the wastewater is the increase of high organic matter in receiving waters which leads to ecosystem disturbance [8,9]. Moreover, the discharge of high turbidity wastewater may have been one of the major threats for the organisms' activity in the receiving environments like rivers or sea [10].

The low biodegradability index of pulp and paper wastewater indicates that this kind of effluent cannot be treated effectively through biochemical methods. On the other hand, the chemical methods generate considerable amount of sludge which needs further treatment. Due to these disadvantages there has been an increasing interest in the use of electrochemical technologies for the treatment of pulp and paper effluent. High removal yield is achieved using electrocoagulation (EC) method without adding any chemical coagulant or flocculants, thus reducing the amount of sludge, which must be disposed [11].

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^{*}Corresponding author.

Table 1 Comparison of some recent industrial wastewater treatment processes

Industrial wastewaters	Treatment process	COD removal (%)	Turbidity removal (%)	Reaction time (min)	Ref. No.
Pulp and paper	Combined EC and UV-based sulphate radical oxidation	61	-	33.7	[7]
Olive mill	Peroxi-EC/electroxidation- electroflotation	96	88.7	30	[8]
Currant	Anaerobic/aerobic -bio	98.1	-	2880	[9]
Water-based paint	Coagulation	34.4	90.6	20	[10]
Pulp and paper	EC	55.3	98.9	45	The present work

Some operational parameters such as temperature and pH have an effect on system behavior during wastewater recycling or treatment process. Specially, the pH control has of importance to get adequate conditions for most of the processes [12]. Treatment of pulp and paper mill wastewater by EC is a very complex process and is greatly influenced by many factors, including electrode configuration and material, current density, electrolysis time, pH, temperature and electrical conductivity [13–15]. Among these operational parameters, electrical conductivity, temperature and pH have dynamic behavior during process, which must be controlled. The electrical conductivity depends on the temperature and ion concentrations in a solution [16]. Control of electrical conductivity is crucial because due to Ohm's law of resistance, high values of electrical conductivity reduces the resistance, the potential difference and power consumption of the EC process under constant current conditions [17]. Temperature also affects the efficiency of pollutant removal in many ways such as rate of reactions, liquid conductivity and kinetics of gas bubbles or small colloidal particles[18]. Uncontrolled rise of temperature during EC process causes variation of pH and electrical conductivity and poses unsafe process operation. Chemical dissolution of metal electrodes is related with the pH value of the wastewater. At an appropriate pH range, the metal ions in the wastewater can form various coagulated species and metal hydroxides which are able to adsorb dissolved contaminants and destabilize suspended particles [19,20]. Efficient control of these parameters in their optimum values allows not only maintenance of high removal efficiency, but also reduces costs and provides safe process operations. Despite these advantages, few studies have been reported for process control applications on EC process.

In this work batch treatment of pulp and paper mill wastewater using EC has been investigated. Conductivity, temperature and pH were selected as controlled variables; supporting electrolyte flow rate, cooling water flow rate, coordinated acid and base solution flow rates were selected as manipulated variables respectively. Dynamic analyses of conductivity, temperature and pH were performed in the face of step changes given to manipulated variables. Auto regressive moving average with external input (ARMAX) models' parameters were evaluated using experimental data and recursive least squares (RLS) estimation method. Proportional integral derivative (PID) control is the most generally used feedback controller in industry. Although tuning of PID controller has always been very important, Table 2

Characteristics of pulp and paper mill wastewater

COD (mg/L)	420
Turbidity (NTU)	173
Color (CU)	923
Conductivity (mS/cm)	2.60
pH	6.45

it has often poorly tuned [21]. Pole placement approach is a simple and reliable method for PID tuning and was used in this study [22]. Real time experimental multi input-multi output (MIMO) control of conductivity, temperature and pH under constant current conditions were achieved using MIMO PID control algorithms coded in MATLAB™. Algorithms were adapted to a designed real time Simulink® model which is capable of transferring real time data of input measurement signals from conductivity, temperature and pH sensors to designed controller for required calculations of manipulated variables actions and calculated controller output signals to supporting electrolyte pump, cooling water valve and acid and base solution pumps for simultaneous manipulation. All the experimental results, throughout the present study have been evaluated in terms of pollutant removal efficiency and energy consumption. Although most of the wastewater treatments in the previous studies are executed only by adjusting the initial value of the operational parameters, this work has the novelty of maintaining conductivity, temperature and pH parameters at their desired value during the operation.

2. Experimental procedure

Experiments were conducted in batch process using a corrosion resistant 2 LEC reactor made of plexiglass. In each run, 1 L of pulp and paper mill wastewater, obtained from a craft paper mill with production capacity of 100.000 tons/ year in Turkey, was fed into the electrochemical reactor. The characteristics of the wastewater are shown in Table 2. Electrodes made of aluminum with dimensions of 60 mm \times 60 mm \times 2 mm were used. Distance among electrodes in mono-polar parallel connection was arranged as 10 mm. In order to carry out the experiments under constant current conditions, the electrodes were connected to a DC power

supply (MAY 11-PS Constant Current Power Supply) operating in the range of 0–2 A. Supporting electrolyte (0.04 M NaCl), acid (0.1 M HCl) and base (0.1 M NaOH) solutions were added to the wastewater with three peristaltic pumps (Longer Pump LEAD-2). A stirrer (MTOPS MS-3020) was used to maintain uniform concentration and temperature dispersion in the reactor. During experiments a heating/ cooling water circulator (Hoefer RCB 20-PLUS) was used in order to avoid possible temperature increase and keep temperature constant at a desired value. Cooling water was passed through a jacket surrounding the reactor with the aid of an on-off control valve.

A pH meter, conductivity meter and a thermocouple (Mettler Toledo M200 easy) were used for on-line measurements of pH, conductivity and temperature during wastewater treatment. These probes were placed into a separate compartment inside the reactor to avoid the measurements to be affected from charge distribution occurring in the reactor. Sample circulation between the compartment and reactor was carried out using a peristaltic pump (Aspen Standard Pump).

On-line signals of conductivity, temperature and pH from measurement devices were sampled and transferred towards controller using data acquisition device (MAY 11-ESA Electrophoresis Control Unit) and calculated signals from controller was transferred to related manipulated variable via the data acquisition device and input variables were regulated. In conductivity control studies, supporting electrolyte flow rate was the manipulated variable and its value was adjusted by the conductivity controller signals transferred to the supporting electrolyte peristaltic pump. In temperature control studies, on/off position of cooling water valve was the manipulated variable and its position was adjusted by the temperature controller. In pH control studies, acid and base solution flow rates were manipulated variables and their values were adjusted by acid and base peristaltic pumps. Flow rate adjustment signals were calculated by pH controller algorithm which depends on coordinated control strategy. Experimental set-up is given in Fig. 1.

A multi-purpose real-time MATLABTM/Simulink® model and a PID controller program were designed for performing EC, monitoring input and output variables, carrying out dynamic analyses and control experiments in electrochemical reactor. The model acquires pH, conductivity and temperature readings and uses PID controller algorithms in order to calculate flow rate signals. Although the algorithm designs of each controller are similar, coordinated pH control strategy and algorithm is more complex than that of conductivity and temperature algorithms. Coordinated pH PID controller algorithm written as a Simulink S-Function is given in Fig. 2. Real-time MATLABTM / Simulink® model is given in Fig. 3.

Initial conductivity of wastewater was adjusted to 2.6 mS/cm in order to reduce energy consumption and avoid excessive addition of supporting electrolyte solution more than 10% of wastewater volume. Initial pH and temperature of wastewater was adjusted to 8 and 20°C respectively. All experiments were carried out under 1 A constant current conditions. 50 ml samples were taken after treatment processes and kept at 20°C for 3 h for sedimentation. Supernatants were collected for analyses. COD analyses were performed according to SM 5220 D [23]. 2.5 ml samples were treated with 1.5 ml high range digestion solution and 3.5 ml sulfuric acid reagent in 16×100 mm culture tubes. Treated samples were digested at 150°C for 2 h using a thermo-reactor (Velp ECO-16). After digestion process, tubes were cooled down to room temperature and absorbance of the samples were read at 600 nm using a spectrophotometer (PG Instruments T60V). COD of the samples were calculated with a calibration curve prepared using potassium hydrogen phthalate standards. Color analyses were performed in accordance with SM 2120 C [23].Sample absorbances were read at 456 nm using the spectrophotometer. Color values were calculated by a calibration curve prepared using 500



Fig. 1. Experimental set-up (1: EC reactor, 2: EC reactor heating/cooling jacket, 3: electrodes, 4: pH meter, conductivity meter, thermocouple, 5: sample circulation pump, 6: stirrer, 7: water circulator, 8: acid pump, 9: base pump, 10: supporting electrolyte pump, 11: pH and temperature display, 12: conductivity display, 13: control unit, 14: power supply, 15: computer).



Fig. 2. Coordinated pH PID controller algorithm.

CU Pt-Co stock solution. Sample turbidities were measured using a turbidity meter (Aqualytic AL250T-IR). Energy consumption of treatment processes per kg COD_r were calculated using Eq. (1).

$$Energy\ consumption = \frac{IVt}{(COD_i - COD_t)V_R}$$
(1)

Here, I refers the applied current intensity in A, V represents the mean potential difference applied in V,

 COD_i and COD_t are the initial and final COD values of the wastewater in mg/L, *t* is the treatment time in h, V_R is the volume of effluent in L and unit of energy consumption is kWh/kg COD_r.

3. Results and discussion

Four studies were carried out to investigate the effect of MIMO control of conductivity, temperature and pH on pulp



Fig. 3. Real-time MATLAB[™] /Simulink® model.

and paper mill wastewater treatment with EC. In the first study variation of conductivity, temperature and pH was observed without any control action. Results are shown in Fig. 4.

As can be seen in the figure conductivity was decreased approximately 0.2 mS/cm. $Cl_{2(g)}$ formation causes electrical conductivity reduction during EC without control. Serikawa et al. [24] observed strong catalytic effect of chloride ion on oxidation of organic pollutants. Raju et al. [25] indicated that indirect electro-oxidation involving active chlorine was a predominant process in removing organic pollutants from chloride medium and that at 25°C and normal atmospheric pressure $Cl_{2(g)}$ that revealed from anode dissolved in water to the extent of 6.413 g/L. If its solubility is exceeded locally at the electrode surface, then Cl, bubbles may form [25].In parallel with the drop of conductivity, operational cost of treatment, energy consumption and required potential difference for constant current condition were increased. Total energy consumption of this experiment was calculated as 45.81 kWh/kg COD, Also, temperature was increased approximately 4°C and pH was increased approximately 1.1 unit during treatment.

Generation of coagulated species and metal hydroxides during treatment were expected to be affected by the increase in temperature and pH. Therefore, need for efficient process control of conductivity, temperature and pH parameters were emerged. Dynamic analyses were carried out to statistically model the variations of conductivity, temperature and pH with time to design robust controllers for these parameters. The variation of conductivity with time was observed by applying a step change of 4.4 ml/ min to the flow rate of supporting electrolyte solution in an experimental treatment with EC. In another experimental treatment, cooling water valve in turn off position was switched to on position without any feed of supporting



Fig. 4. Treatment without any control action.

electrolyte solution to the system and the variation in temperature was observed by passing cooling water at 12°C through the jacket surrounding the reactor. In our previous study [26], a pulse change having a magnitude of 5 ml/min was applied to acid solution flow rate for 180 s. After the pulse input, a step change with a magnitude of 5 ml/min was given to base solution flow rate in order to



Fig. 5. Variation of conductivity with step input and calculated model parameters with RLS.



Fig. 6. Variation of temperature with step input and calculated model parameters with RLS.



Fig. 7. Variation of pH with pulse and step inputs and calculated model parameters with RLS.

observe the dynamic behavior of pH. The data obtained from these experiments was used separately in RLS algorithm coded in MATLAB[™] and second order ARMAX model parameters were evaluated. Variation of conductivity,temperature, pH and calculated model parameters are shown in Figs. 5–7.

PID parameters of controllers for MIMO control of conductivity, temperature and pH were determined with pole placement approach. Model parameters used in pole placement approach are shown in Table 3.

Table 3 Second order model parameters for conductivity, temperature and pH processes

	<i>a</i> ₁	<i>a</i> ₂	b _o
Conductivity	0.783	0.072	0.01152
Temperature	-0.6424	0.1	0.03325
pH (acid)	0.8297	-0.03538	0.0000293
pH (base)	-0.9367	0.0367	0.0002312

Poles of the system obtained by using the model and estimated optimum controller parameters for conductivity, temperature and pH processes are given in Fig. 8.

Results show that the roots of conductivity, temperature and pH processes are all real and the closed loop responses should be non-oscillatory. Poles of the system and optimum controller parameters for these three processes are given in Table 4.

On the first step of control studies, SISO conductivity, SISO temperature and SISO pH control simulations were performed with an algorithm coded in MATLAB using ARMAX model parameters and determined optimum controller parameters. A sample simulation algorithm is given below in Fig. 9.

SISO PID control simulations were carried out for conductivity, temperature and pH using optimum controller parameters given in Table 4 to investigate designed controllers' performances and process response. Set point of conductivity, temperature and pH were selected as 2.6 mS/cm, 18°C and 8 respectively. Integral of the square of the error (ISE) and the integral of the absolute value of the error (IAE) were used as controller performance criteria. ARMAX model based simulation results given in Table 5 show that tuning of PID parameters with pole placement approach for conductivity, temperature and pH controllers were achieved successfully and that they are well suited to be applied in experimental studies.

On the second step, a SISO PID control experiment was carried out for conductivity process using optimum parameters given in Table 4. Set point of conductivity was selected as 2.6 mS/cm for maintaining the adjusted initial condition during process. Results are shown in Fig. 10.

ISE and IAE values of control performance were calculated as 2.4916 and 93.8929 respectively. Total energy consumption was calculated as 37.10 kWh/kg COD_r in this experiment. Results show that control of conductivity at 2.6 mS/cm reduces 19 % of the energy consumption for the process compared with uncontrolled treatment. Removal efficiency of color, turbidity and COD were found as 93.17%, 91.47%, and 47.38% respectively in this experimental case. Temperature variations during the experimental cases with conductivity control and without control were compared and no significant difference is observed between two experiments due to interaction of conductivity and temperature.

A SISO PID control experiment was carried out for temperature process using optimum parameters given in Table 4. Set point of temperature was selected as 18°C. Results are shown in Fig. 11. ISE and IAE values of control performance were calculated as 2892.687 and 3135.218



Fig. 8. Poles of systems for (a) conductivity (b) temperature (c) pH (acid) (d) pH (base).

Table 4 Poles and optimum controller parameters for conductivity, temperature and pH processes

	K _c	$\tau_{I}(s)$	$\tau_{\rm D}({\rm s})$	Poles		
Conductivity	5	0.05	0.01	0.701963	-0.684202	-0.147513
Temperature	48	0.5	0.01	-0.828502	0.310029	-0.265047
pH (acid)	1000	0.05	0.01	-0.8915	0.8373	0.0482
pH (base)	1	0.06	0.4	0.9750	0.9196	0.0407

respectively. Total energy consumption was calculated as $44.78 \text{ kWh/kg COD}_r$ and removal efficiency of color, turbidity and COD were found as 93.01%, 90.17%, 46.19% respectively in this experiment.

Since pH is one of the most effective parameters in EC, an experimental study on variable set point was performed to observe the robustness of PID controller for pH process. The results of the study in which 0.5 units of positive and negative step changes were applied to the pH set point are given in Fig. 12.

ISE and IAE values were calculated as 425.63 and 1860 for servo problem of pH. Satisfactory control was obtained because it had a good experimental set point trajection. Performance of pH controller was found acceptable when ISE and IAE values were considered with various set points and electrolysis time.

A MIMO PID control was carried out experimentally for conductivity and temperature processes using optimum parameters given in Table 4. Set points of conductivity and temperature were selected as 2.6 mS/cm and 18°C respectively. Results are shown in Fig. 13.

Conductivity control performance criteria ISE and IAE were calculated as 3.0287 and 111.124 respectively. Accordingly, ISE and IAE values of temperature control performance were calculated as 324.5943 and 953.6196 respectively. Total energy consumption was calculated as



Fig. 9. Sample simulation algorithm.

Table 5

Performance criteria of conductivity, temperature and pH processes for simulation studies

Performance criteria	Conductivity	Temperature	pH (acid)	pH (base)
ISE	0.0361	282.8887	2.5510	8.6206
IAE	7.7416	346.9500	93.8273	171.6856



Fig. 10. Experimental SISO PID control of conductivity.

31.31 kWh/kg COD_r and removal efficiency of color, turbidity and COD were found as 94.72%, 92.01%, and 50.26% respectively in this experiment.

A MIMO PID control was carried out experimentally for conductivity, temperature and pH processes using opti-



Fig. 11. Experimental SISO PID control of temperature.

mum parameters given in Table 4 to determine the effect of constant pH on pollutant removal efficiency and energy consumption. In this control study, conductivity and temperature were also kept constant by means of PID controllers to avoid the interaction among these parameters. The aim of conductivity control was to reduce the potential difference and therefore energy consumption. Uncontrolled decrease in conductivity increases the resistance which also causes an increase in the amount of heat generated and temperature of the process. pH can also be affected by temperature rise. For this reason maintaining temperature at a constant value during treatment was necessary. In this study, set point of conductivity, temperature and pH was selected as 2.6 mS/cm,20°C and 8 respectively. Results are



Fig. 12. PID control of pH under servo problem.



Fig. 13. Experimental conductivity and temperature control with MIMO PID algorithms.



Fig. 14. Experimental conductivity, temperature and pH control with MIMO PID algorithms.

shown in Fig. 14. In this study, total energy consumption was calculated as 35.86 kWh/kg COD.

Removal efficiencies, energy consumption and operational cost values of five studies mentioned above are given in Table 6 for comparison. Both simulation and experimental control performance results are given for comparison in Table 7.

The 3 units pH increments were observed by Mouedhen et al. [27] for 30 min EC treatment using Al electrodes at 0.5 A/dm^2 current density. Oncel et al. [28] mentioned that the optimum pH for maximum Al precipitation was 8 and for higher pH values, due to formation of soluble species, EC efficiency reduction occurred. Suitably, the present study showed that at an initial pH of 8, 1.1 units pH increments were observed for 45 min EC treatment using Al electrodes at 1 A current intensity, and the control action prevented the pH increase due to the nature of the process, also reduced the amount of soluble species, which increased pollutant removal.

Table 6

Removal efficiencies, energy consumption and operational cost values of wastewater treatment with different control approaches

Control action	Color	Turbidity	COD	Energy consumption		Operational
	removal (%)	removal (%)	removal (%)	(kWh/kg COD _r)	(kWh/m ³)	$-\cos(({m^3}))$
Treatment without control	89.52	85.88	44.52	45.81	8.58	1.01
SISO conductivity control	93.17	91.47	47.38	37.10	7.38	0.87
SISO temperature control	93.01	90.17	46.19	44.78	8.69	1.03
MIMO conductivity, temperature control	94.72	92.01	50.26	31.31	6.95	0.82
MIMO conductivity, temperature, pH control	97.01	98.92	55.33	35.86	8.33	0.98

Table 7

Controller performances

Control action		ISE	IAE
SISO conductivity control		2.4916	93.8929
SISO temperature control		2892.6870	3135.2180
SISO pH servo control		425.6300	1860.0000
MIMO conductivity, temperature control	Conductivity	3.0287	111.1240
	Temperature	324.5943	953.6196
MIMO conductivity, temperature, pH control	Conductivity	2.1372	86.6256
	Temperature	313.4573	1040.2534
	pH	92.0591	586.2427

Table 8

Comparison of results with the previously published studies

Operating conditions	Energy consumption for EC	Maximum COD romoval officion cy (%)	Rof No
operating conditions	treatment	reached for EC treatment	Ref. INO.
Treatment time: 45 min	35.86 kWh/kg COD	55.33	The present
Current density: 55.56 A/m ²	(8.33 kWh/m ³)		work
The controlled pH: 8			
Condition choice: arbitrary			
Treatment time: 6.9 min	1.19 kWh/kg COD	55	[29]
Current density: 112.9 A/m ²			
The initial pH: 7.3			
Condition choice: adequate			
Treatment time: 20 min	11.055 kWh/m ³	90	[15]
Current density: 150 A/m ²			
The initial pH: 7			
Condition choice: adequate			
Treatment time: 120 min	5.16 kWh/m ³	85	[30]
Current density: 40 A/m ²			
The initial pH: 7			
Condition choice: adequate			
Treatment time: 15 min	11.5 kWh/m ³	75	[31]
Current density: 300 A/m ²			
The initial pH: 3			
Condition choice: adequate			

4. Conclusion

The comparison of constant and time varying operating conditions were investigated in this study to express the effect of process control on treatment of pulp and paper mill wastewater treatment by means of EC. After determining the best PID controller parameters for conductivity, temperature and pH processes using pole placement approach, theoretical and experimental studies were performed. For SISO conductivity and SISO temperature controllers, performance criteria of the experimental cases were higher than that of the theoretical cases due to high level of measurement noise of the real system.

Controller performance results of this study show that MIMO control of conductivity and temperature has a positive effect on temperature control performance due to reduced energy consumption and heat accumulation and reduced ISE by 88.8% compared to SISO temperature control.pH controller addition to the MIMO conductivity and temperature control increased conductivity and temperature control performance by 29.4% and 3.4% respectively in terms of ISE compared with MIMO control of conductivity and temperature.

MIMO conductivity, temperature and pH control increased the color, turbidity and COD removal efficiency as 8.19%, 13.29%, and 10.81%, respectively, when compared with uncontrolled treatment. Although color, turbidity and COD removal efficiencies of SISO conductivity, SISO temperature and MIMO conductivity and temperature control studies are similar, pH control addition to the MIMO conductivity and temperature control increased pollutant removal efficiencies by about 5%. This result indicates that pH control is the most important action in terms of pollutant removal compared with other control actions. The energy consumption of the EC process was reduced by 31.65% and 21.72% compared to the energy consumption of the uncontrolled process using MIMO conductivity and temperature control and MIMO control of three parameters, respectively. This result shows that conductivity control plays the most important role in reducing energy consumption and temperature control is also helpful.

The EC treatment results of pulp and paper mill wastewaters obtained from different sources in the previously published work were roughly compared with the present treatment in Table 8. The energy consumptions in the literature were 11.055 kWh/m3 for 20 min treatment with 150 A/m^2 , 5.16 kWh/m³ for 120 min treatment with 40 A/ m^2 , 11.5 kWh/m³ for 15 min treatment with 300 A/m². It was observed that the present operational conditions were quite different from the ones in literature. Although the conditions of the present work might not be the adequate ones, the comparison still revealed the relative potentials of their energy consumptions. It is noted that the present treatment with 8.33kwh/m³ energy consumption in 45 min was highly competitive.

It is concluded that SISO and MIMO PID control of conductivity, temperature and pH are achieved successfully for pulp and paper mill wastewater treatment with EC. Controller performances were found similar for both simulation and experimental studies which show that ARMAX process models were successfully obtained and controller parameters were determined appropriately. It is observed that MIMO conductivity, temperature and pH control increase

pollutant removal and decrease energy consumption simultaneously, which makes the process more economical. The results are considered promising in terms of applicability of the EC process to the pulp and paper industry.

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Symbols

- Α Monic polynomial in the z-domain representing the poles of the discrete time system В Polynomial in the z-domain representing the
- zeros of the discrete time system С Monic polynomial in the z-domain representing
 - the zeros of the process noise
- COD Initial COD
- COD removed COD COD, COD after treatment
- ____ *e*(*k*) kth error
- *e*(*n*)
- Error at the nth sampling instant e(t)Error at time t
- Current intensity I
- K Controller gain
 - Hold element
- Ŕ Set point at time t r(t)
- S
- Self-tuning polynomial
- Reaction time
- Tailoring polynomial Δt
 - The sampling period
- Initial value of manipulated variable u_0
- *u*(*n*) Controller output at the nth sampling instant $(n = 1, 2, \ldots)$
- u(t)Input variable at time t
 - Cell voltage
- V_{i} Reaction volume
- y(t)Output variable at time t
- Z^{-1} Backward shift operator
- Integral time constant τ,
- Derivative time constant $\tau_{\rm D}$

References

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- A.R. Shawwa, D.W. Smith, D.C. Sego, Color and chlorinated [1] organics removal from pulp mills wastewater using activated petroleum coke, Water Res., 35(3) (2001) 745–749.
- [2] C.P. Das, L.N. Patnaik, Removal of lignin by industrial solid wastes, Practice Periodic. Hazard. Toxic Radioact. Waste Manage., 4(4) (2000) 156–161.
- S. Verenich, A. Laari, J. Kallas, Wet oxidation of concentrated [3] wastewaters of paper mills for water cycle closing, Waste Manage., 20(4) (2000) 287–293.
- M.M. Hassan, C.J. Hawkyard, Decolourisation of aqueous dyes [4] by sequential oxidation treatment with ozone and Fenton's reagent, J. Chem. Technol. Biotechnol., 77(7) (2002) 834-841.
- [5] A. Raj, M.M.K. Reddy, R. Chandra, Decolourisation and treatment of pulp and paper mill effluent by lignin degrading Bacillus sp., J. Chem. Technol. Biotechnol., 82(4) (2007) 399– 406

- [6] K.V. Rajeshwari, M. Balakrishnan, A. Kansal, K. Lata, V.V.N. Kishore, State-of-the-art of anaerobic digestion technology for industrial wastewater treatment, Renew. Sust. Energ. Rev., 4(2) (2000) 135–156.
- [7] N. Jaafarzadeh, M. Omidinasab, F. Ghanbari, Combined electrocoagulation and UV-based sulfate radical oxidation processes for treatment of pulp and paper wastewater, Process Saf. Environ. Prot., 102 (2016) 462–472.
- [8] Y. Esfandyari, Y. Mahdavi, M. Seyedsalehi, M. Hoseini, G.H. Safari, M.G. Ghozikali, H. Kamani, J. Jaafari, Degradation and biodegradability improvement of the olive mill wastewater by peroxi-electrocoagulation/electrooxidation-electroflotation process with bipolar aluminum electrodes, Environ. Sci. Pollut. Res., 22 (2015) 6288–6297.
- [9] J. Jafari, A. Mesdaghinia, R. Nabizadeh, M. Farrokhi, A.H. Mahvi, Investigation of anaerobic fluidized bed reactor/aerobic moving bed bio reactor (AFBR/MMBR) system for treatment of currant wastewater, Iran. J. Public Health, 42(8) (2013) 860.
- [10] Ş. Camcıoğlu, B. Özyurt, Z. Zeybek, H. Hapoğlu, Experimental application of one step ahead advanced pH control to waterbased paint wastewater treatment, J. Fac. Eng. Archit. Gazi., 31(3) (2016) 655–664.
- [11] K.S.P. Kalyani, N. Balasubramanian, C. Srinivasakannan, Decolorization and COD reduction of paper industrial effluent using electro-coagulation, Chem. Eng. I., 151(1) (2009) 97–104.
- using electro-coagulation, Chem. Eng. J., 151(1) (2009) 97–104.
 [12] S. Altuntaş, H. Hapoğlu, S. Ertunç, M. Alpbaz, Experimental pH advance control of the cheese whey bacthkefir-type drink production with manipulated base flowrate, J. Fac. Eng. Archit. Gazi, 31(3) (2016) 709–716.
- [13] E.-S.Z. El-Ashtouky, N.K. Amin, O. Abdelwahab, Treatment of paper mill effluents in a batch-stirred electrochemical tank reactor, Chem. Eng. J., 146(2) (2009) 205–210.
- [14] M. Zaied, N. Bellakhal, Electrocoagulation treatment of black liquor from paper industry, J. Hazard. Mater., 163(2) (2009) 995–1000.
- [15] R. Sridhar, V. Sivakumar, V. P. Immanuel, J.P. Maran, Treatment of pulp and paper industry bleaching effluent by electro-coagulant process, J. Hazard. Mater., 186(2) (2011) 1495–1502.
 [16] R.B. McCleskey, D.K. Nordstrom, J.N. Ryan, J.W. Ball, A new
- [16] R.B. McCleskey, D.K. Nordstrom, J.N. Ryan, J.W. Ball, A new method of calculating electrical conductivity with applications to natural waters, Geochim. Cosmochim. Acta, 77 (2012) 369– 382.
- [17] E. Terrazas, A. Vázquez, R. Briones, I. Lázaro, I. Rodríguez, EC treatment for reuse of tissue paper wastewater: Aspects that affect energy consumption, J. Hazard. Mater., 181(1) (2010) 809–816.
- [18] C. Phalakornkule, J. Mangmeemak, K. Intrachod, B. Nuntakumjorn, Pretreatment of palm oil mill effluent by electrocoagulation and coagulation, Science Asia, 36(2) (2010) 142–149.
- [19] B. Merzouk, B. Gourich, A. Sekki, K. Madani, C. Vial, M. Barkaoui, Studies on the decolorization of textile dye waste-water by continuous electrocoagulation process, Chem. Eng. J., 149(1–3) (2009) 207–214.
- [20] P. Cañizares, M. Carmona, J. Lobato, F. Martínez, M. Rodrigo, Electrodissolution of aluminum electrodes in electrocoagulation processes, Ind. Eng. Chem. Res., 44(12) (2005) 4178–4185.
- [21] P. Cominos, N. Munro, PID controllers: recent tuning methods and design to specification, IEE Proceedings-Control Theory Appl., 149(1) (2002) 46–53.

- [22] S. Karacan, H. Hapoglu, Y. Cabbar, M. Alpbaz, Pole placement self tuning control for packed distillation column, Chem. Eng. Process. Process Intensif., 36(4) (1997) 309–315.
- [23] American Public Health Association (APHA), Standard Methods for the Examination of Water and Wastewater, Maryland, 2005.
- [24. R.M. Serikawa, M. Isaka, Q. Su, T. Usui, T. Nishimura, H. Sato, S. Hamada, Wet electrolytic oxidation of organic pollutants in wastewater treatment, J. Appl. Electrochem., 30(7) (2000) 875– 883.
- [25] G.B. Raju, M.T. Karuppiah, S.S. Latha, S. Parvathy, S. Prabhakar, Treatment of wastewater from synthetic textile industry by electrocoagulation–electrooxidation, Chem. Eng. J., 144(1) (2008) 51–58.
- [26] S. Camcioğlu, B. Ozyurt, H. Hapoglu, Elektrokoagulasyonyontemiylekagitatiksuyuaritiminda pH kontrolu, Anadolu University J. Sci.Technol.-A Appl. Sci.Eng., 16(2) (2015) 107–115.
- [27] G. Mouedhen, M. Feki, M.D.P. Wery, H.F. Ayedi, Behavior of aluminum electrodes in electrocoagulation process, J. Hazard. Mater., 150(1) (2008) 124–135.
- [28] M.S. Oncel, A. Muhcu, E. Demirbas, M. Kobya, A comparative study of chemical precipitation and electrocoagulation for treatment of coal acid drainage wastewater, J. Environ. Chem. Eng., 1(4) (2013) 989–995.
- [29] P.A. Soloman, C. Ahmed Basha, M. Velan, N. Balasubramanian, P. Marimuthu, Augmentation of biodegradability of pulp and paper industry wastewater by electrochemical pre-treatment and optimization by RSM, Sep. Purif. Technol., 69 (2009) 109–117.
- [30] P. Asaithambi, Studies on various operating parameters for the removal of COD from pulp and paper industry using electrocoagulation process, Desalin. Water Treat., 57 (2016) 11746– 11755.
- [31] U. Tezcan Un, S. Topal, F. Ates, Electrocoagulation of tissue paper wastewater and an evaluation of sludge for pyrolysis, Desal. Water Treat., 57 (2016) 28724–28733.
- [32] K.J. Aström, R.M. Murray, Feedback systems: an introduction for scientists and engineers, Princeton University Press, 2010.
- [33] D.E. Seborg, T.F. Edgar, D.A. Mellichamp, Process Dynamics and Control, John Wiley & Sons, New York, 1989.
- [34] A.R. Karagoz, H. Hapoglu, M. Alpbaz, Generalized minimum variance control of optimal temperature profiles in a polystyrene polymerization reactor, Chem. Eng. Process. Process Intensif., 39(3) (2000) 253–262.
- [35] G. Ozkan, H. Hapoglu, M. Alpbaz, Identification and nonlinear pole placement self-tuning PID control of a jacketed well mixed reactor, Chem. Eng. Commun., 170(1) (1998) 199–215.
- [36] S. Ertunc, B. Akay, H. Boyacioglu, H. Hapoglu, Self-tuning control of dissolved oxygen concentration in a batch bioreactor, Food Bioprod. Process., 87(1) (2009) 46–55.
- [37] B. Akay, S. Ertunc, H. Boyacioglu, H. Hapoglu, M. Alpbaz, Discrete system identification and self-tuning control of dissolved oxygen concentration in a stirred reactor, Korean J. Chem. Eng., 28(3) (2011) 837–847.
- [38] D.R. Coughanowr, L.B. Koppel, Process System Analysis and Control, McGraw-Hill, New York, 1965.

APPENDIX A Digital PID Controllers

When a feedback control strategy implemented digitally, the controller input and output must be digital signals rather than continuous signals. Thus, the continuous signal from the transmitter is sampled or converted periodically to a digital signal by an analogue-to-digital converter (ADC). A digital control algorithm is then used to calculate the controller output and a digital signal is sent to a final control element before the system. This digital signal is converted to a corresponding continuous signal by a digital-to-analogue converter (DAC). Alternatively, the digital signal can be converted to a sequence of pulses representing the change in controller output. The pulses are then sent directly to a final control element that utilizes pulse inputs to change its position.

$$u(t) = u_0 + K_c \left[e(t) + \frac{1}{\tau_I} \int_0^t e(t) dt + \tau_D \frac{de}{dt} \right]$$
(1)

A straightforward way of deriving a digital version of the ideal PID control law in Eq. (1) is to replace the integral and derivative terms by their discrete equivalents. Thus, approximating the integral by a summation and the derivative by a first order backward difference gives Eq. (2).

$$u(n) = u_0 + K_c \left[e(n) + \frac{\Delta t}{\tau_I} \sum_{k=1}^n e(k) + \frac{\tau_D}{\Delta t} (e(n) - e(n-1)) \right]$$
(2)

Eq. (2) is referred to as the position form of the PID control algorithm since the actual controller output is calculated. If a controller with integrating action is used, the error will continue to be integrated. This means that the integral term may become very large or, colloquially, it "winds up". It is then required that the error has opposite sign for a long period before things return to normal. The consequence is that any controller with integral action may give large transients when the actuator saturates [32].

An alternative approach is to use a velocity form of the algorithm in which the change in controller output is calculated. Eq. (3) can be derived by writing the position form of the algorithm, Eq. (2), for the (n-1)th sampling instant.

$$u(n-1) = u_0 + K_c \left[e(n-1) + \frac{\Delta t}{\tau_I} \sum_{k=1}^{n-1} e(k) + \frac{\tau_D}{\Delta t} (e(n-1) - e(n-2)) \right] (3)$$

Subtracting Eq. (3) from Eq. (2) gives the velocity form of the PID algorithm. In velocity form the incremental change in controller output is calculated [33].

$$\Delta u(n) = u(n) - u(n-1) = K_c \bigg[(e(n) - e(n-1)) + \frac{\Delta t}{\tau_1} e(n) + \frac{\tau_D}{\Delta t} (e(n) - 2e(n-1) + e(n-2)) \bigg]$$
(4)

A velocity algorithm first computes the rate of change of the control signal which is then fed to an integrator. With this approach it is easy to avoid windup by inhibiting integration whenever the output saturates [32].

APPENDIX B Pole Placement

In this study, a computer algorithm was coded in MAT-LABTM in order to convert the velocity form of the PID algorithm into a self-tuning equivalent. The discrete time PID control algorithm was considered as shown in Eq. (5).

$$u(t) = \frac{S}{R} \left(r(t) - y(t) \right) \tag{5}$$

S polynomial and its coefficients are given in Eqs. (6) – (9). Here R is defined as $(1 - z^{-1})$.

$$S = s_0 + s_1 z^{-1} + s_2 z^{-2} \tag{6}$$

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

$$s_1 = K_c \left(\frac{\Delta t}{2\tau_1} - 1 - \frac{2\tau_D}{\Delta t} \right) \tag{8}$$

$$s_2 = \frac{K_C \tau_D}{\Delta t} \tag{9}$$

Hence

 t_0

$$u(t) = \frac{\left(r(t) - y(t)\right)\left(s_0 + s_1 z^{-1} + s_2 z^{-2}\right)}{\left(1 - z^{-1}\right)}$$
(10)

 $K_{\rm C'} \ \tau_{\rm I}$ and $\tau_{\rm D}$ are chosen by the system designer and placed in Eqs. (6)–(9).

$$y(t) = \frac{B}{A}u(t-1) + \frac{C}{A}e(t)$$
(11)

Eq. (11) represents an application of the auto regressive moving average (ARMA) type with an added control (or exogenous input) and therefore this equation is thus said to be a controlled auto regressive moving average (CARMA) or an auto regressive moving average with external input (ARMAX) model of the system [22,34,35]. Closed-loop equation of the system is given in Eq. (12).

$$y(t) = \frac{z^{-1}BS}{AR + z^{-1}BS}r(t) + \frac{RC}{AR + z^{-1}BS}e(t)$$
(12)

The properties of this closed-loop can be varied by placing the poles of the characteristic equation i.e. the denominator of Eq. (12) utilizing a 'tailoring polynomial' T, where the poles of T are determined by the computer program. Thus, the characteristic equation is shown in Eq. (13).

$$T = AR + z^{-1}BS \tag{13}$$

Coefficients of the T polynomial are given in Eqs. (14)–(17).

$$= 1$$
 (14)

$$t_0 = (s_0 b_0) + \alpha_1 \tag{15}$$

$$t_1 = (s_1 b_0) + \alpha_2 - \alpha_1 \tag{16}$$

$$t_3 = (s_2 b_0) + \alpha_2 \tag{17}$$

The coefficients of the A and B polynomials were estimated from the RLS algorithm. RLS updates the parameters as it receives each new data point. This allows quick calculation of the best estimate as time progresses. It also requires a minimum of storage space because all the estimation information is stored in a relatively small number of recursive variables. The ability of recursive algorithms to constantly update parameter estimate, while requiring minimal storage, makes them well suited for on-line parameter estimation [36,37]. The characteristic closed loop system behavior will only depend on the location of roots of T polynomial in the z-domain unit cycle. If the roots are all real, the response will be non-oscillatory. If two of the roots are complex and have negative real parts, the response will include damped sinusoidal terms which will produce an oscillatory response. If two of the roots are complex and have positive real parts, the response is a growing sinusoid [38].

Before a satisfactory response of a control system for a choice of control parameters, a concept of an ideal response must be defined. Two criteria that are often used to evaluate a response of a control system are ISE and IAE with respect to time. The objective is to obtain the minimum value of ISE and IAE by proper choice of control parameters. The summation of squared residuals will cause reduction of large errors that initially exists during the operation. The sum of the absolute values of the discrepancies takes all magnitude of errors into consideration in a uniform manner [38]. The definition of ISE and IAE is given below in Eqs. (18), (19) respectively.

$$ISE = \int_{0}^{0} e^{2}(t)dt \tag{18}$$

$$IAE = \int_{0}^{\infty} |e(t)| dt \tag{19}$$

In present work, the steps in the operation of the pole placement algorithm may be given as:

- (a) Apply a step input to the system as a forcing function and attain the plant output.
- (b) Estimate *A* and *B* from the CARMA model using the RLS algorithm.
- (c) Assign $K_{C'} \tau_1$ and τ_D and calculate $s_{0'} s_1$ and s_2 from Eqs. (7)–(9).
- (d)Calculate t_1 , t_2 and t_3 from Eqs. (15)–(17).
- (e) Determine and plot the roots of the T polynomial.
- (f) If the roots are complex, return to (c).