

Neutralize the pH of wastewater using intelligent controllers for industrial reuse

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Graphical abstract



Abstract

Water waste management is one of the significant hassles which is faced by the majority of industries. The industries let the wastewater into flowing streams or rivers near them, polluting the water resources in that locality. To prevent and safeguard the water resources, the government has given many regulations on the water quality before draining into the water sources. One of the primary problems is the amount of pH in the wastewater. The pH is a highly nonlinear quality of the liquids, as a single drop may change the quality to either acidic or alkaline. This paper proposes various intelligent controllers to maintain the wastewater's pH. The Nonlinear Autoregressive with External Controller (NARX) and Nonlinear Autoregressive (NAR) under deep neural network where the NARX model is synthesized based on two different algorithms which involve the Levenberg Marquardt (LM) and Scaled Conjugate Gradient (SCG) for maintaining pH. The controllers are being implemented on the pilot plant's process tank to determine and neutralize the pH. The pilot plant is interfaced MATLAB R2019b with the computer as a controller where the controller action for the neutralization is processed. The proposed NAR model did produce better results than the other models, whose settling times were 8.6 seconds and 0.5% overshoot. Furthermore, the NAR model has better results when compared with the other two neural models.

Keywords: Wastewater, pH, deep learning, intelligent controller, nonlinear

1. Introduction

Water is one of the most basic needs of a human being and is used to make most of the components in the industry. In recent years, water has become scarce because of improper usage and wastage; therefore, water conservation is significant. The water used for the processing is let out into the water reservoirs near the industries without properly treating/recycling the water, in turn causing the contamination of the reservoir water and the groundwater. The water quality is given through different forms, in which pH stands as the most crucial quality test of the water. The pH measures the water quality as three other liquids on a scale of 0-14. In most industries, different techniques are used to control the pH level based on the product manufactured. For example, in sugar industries, the pH level of sugarcane juice is around 4.5, which has to be at 7.

Process Information: Domestic water consists of a pH value of around 6.5 to 8.5, considered a neutral or consumable liquid by the World Health Organization. In industries, the pH level of the water used for the manufacturing process is to be maintained near seven based on the requirement of the process involved. The process liquid is to be reused based on the industry's standards, where the pH of the process liquid will be either acidic or alkaline based on the process involved. Neutralization of the liquid process is required to reuse such that the liquid does not affect the boilers/pipes through which the liquid flows. When water levels are dropping, regulations are becoming stricter, and the cost of wastewater treatment and supply is rising, water management, recycling, and reuse in industrial facilities is a hot topic in today's expanding economy. The concentration of hydroxyl ions is often higher than that of hydrogen ions at higher pH values and vice versa. Different solutions of pH range from 0 to 14. Acidic solutions have a pH under 7, while bases have a pH over 7, and a pH value of 7 is regarded as neutral. The pH scale is logarithmic, with each increase containing ten times as many hydrogen ions as the pH before it.

2. Related work

The following papers are surveyed to study the existing systems - based on various techniques applied for pH measurements.

Alessandro Lusci *et al.*, 2013 discussed the deep learning techniques for cheminformatics. The undirected Graph Recursive Neural Networks (UG_RNN) method is introduced to identify the molecules of the Aquasol solution (Lusci *et al.*, 2013). Ariane Silva Mota *et al.*, 2016 proposed the Adaptive Neuro-Fuzzy Inference System (ANFIS) model predict and maintain the pH value. The model was tested to predict the flow rates of the output variable (Mota *et al.*, 2016). Finally, Ashok Kumar *et al.*, 2018 investigated the NARX controller to forecast the closing index of the stock market (Kumar *et al.*, 2018).

Ethar H. Alkamil *et al.*, 2018 worked on a lab-scaled pH neutralization system while drilling hydrogen sulfide. A fuzzy neural network combined with the fuzzy logic controller to identify the pH in a drilling system and assess its harshness (Alkamil *et al.*, 2018). Febina C. *et al.*, 2020 implemented an optimized new generation Robustness, Tracking, Disturbance rejection, and overall Aggressiveness (RTDA) controller for a nonlinear conical tank system. However, it has one limitation it cannot handle input-output constraints effectively (Febina *et al.*, 2020).

Hernan Alvarez *et al.*, 2001 discussed nonlinear control on pH neutralization to define the pH region concerning Proportional Integral Derivative (PID) controller to get the performance of the nonlinear system (Alvarez *et al.*, 2001). Imam Sutrisno *et al.*, 2019 simulated the pH neutralization process with the Backpropagation Neural Network and Extreme Learning Machine to maintain the region at a pH of 7, which resulted in Backpropagation showing better results for the system (Sutrisno *et al.*, 2019).

Mohanad Hamad Eljack Elameen *et al.*, 2019 considered active disturbance rejection control (ADRC) for the pH neutralization process as the challenge to controlling the pH. The simulation of the system with the ADRC control was presented to the pH data, and it found that ADRC performance is better than the feedback linearization method (Elameen *et al.*). M.F. Zamil *et al.*, 2014 presented a model comprising neuro-fuzzy and the first principal models to identify the characteristics of pH. The model was implemented to check for the robust control for online pH control where the proposed model has given the best fit with the neutralization process (Zamil *et al.*, 2014).

P.K. Singh *et al.*, 2018 proposed a pH neutralization using strong acid/base of HCL and NaOH, respectively. An artificial neural network (ANN) model and a Fuzzy Logic Controller (FLC) are used to optimize the control specifications using the different scaling factors to incorporate dynamic pH variation between 6 and 9. Different techniques are involved in maintaining the pH between 6-7 or 8-7, where the DE has given better results than other controllers (Singh *et al.*, 2018). R. Babu *et al.*, 2017 conducted a study about the control of pH using a PID controller where the six different tuning techniques of PID were implemented, and the Chien, Hrones, and Reswick

Method of tuning PID has shown better output for the FOTD system (Babu *et al.*, 2017).

Biagiola S.I *et al.*, 2016 designed a controller and robust analysis of the Wiener system under various uncertainty. The nonlinear system is analyzed as a sector, where strength investigation is performed using μ -theory and checked with a pH neutralization process (Biagiola *et al.*, 2016). T. Pravin Rose *et al.*, 2020 proposed the control of pH in multiple tanks' which vary in size and quantity. In this paper, the ANFIS model has enhanced the prediction of the fractional-PI controller. The proposed system has presented the various parameters' offset & overshoot as null, and the settling time is less than 10 mins (Rose *et al.*, 2020).

X Chen *et al.*, 2012 presented a control for pH neutralization using the Fuzzy model, where the different rules were adjusted to maintain the pH of the liquid to be at a 7 neutral state (Chen *et al.*, 2012). Y. Dharshan *et al.*, 2020 have proposed a pH neutralization process using various controllers in the pilot plant where the results of the system have shown that the Model Predictive Controller has obtained better in terms of settling time, rise, and overshoot when compared with the P, PI, and PID controller (Dharshan *et al.*, 2020). Khatri *et al.*, 2018 outline a clever technique for regulating the pH of the water to treat municipal wastewater and reuse it for gardening and agriculture (Khatri *et al.*, 2018). Mushiri *et al.*, 2014 present a design for an automated control employing fuzzy logic to treat industrial wastewater (Mushiri *et al.*, 2014). G. Guyer *et al.*, 2016 explored the direct reuse and recycling ability of washing/bleaching effluent from reactive dyeing cotton fabric using advanced oxidation processes (AOPs) (Güyer *et al.*, 2016). C. Chen *et al.*, 2021 research shed light on the large-scale ZLD procedure for wastewater treatment (Chen *et al.*, 2021). In contrast to the poor performance and erratic system behavior caused by network latency, missing data and noise are critical red flags in the design of Networked Control Systems (NCS) (Dharshan *et al.*, 2017 - Srinivasan *et al.*, 2018). The standard PID controller with various tuning techniques, as well as sophisticated controllers like the fractional order PID controller (FOPID), neural controller, and internal model controller, have been used to address such a control problem (IMC). The addition of these reactants into the process tanks is done with the help of two dosing pumps which acts as the final control element for the process (Srinivasan *et al.*, 2018 – Srinivasan *et al.*, 2016).

3. Modeling of pH Process

The systematic approach for developing a model for the pH neutralization process involves the following general Eq. (1) and Eq. (2).

Conservation of mass:

$$\left(\begin{array}{c} \text{Rate of mass} \\ \text{Accumulation} \end{array} \right) = \left(\begin{array}{c} \text{Rate of } \square \\ \text{Mass } \square \text{ in} \end{array} \right) - \left(\begin{array}{c} \text{Rate of} \\ \text{Mass out} \end{array} \right) \quad (1)$$

Conservation of Energy:

$$\begin{aligned} \left(\begin{array}{l} \text{Rate of energy} \\ \text{Accumulation} \end{array} \right) &= \left(\begin{array}{l} \text{Rate of energy} \\ \text{in by Convention} \end{array} \right) \\ &- \left(\begin{array}{l} \text{Rate of energy} \\ \text{out by Convention} \end{array} \right) \\ \left(\begin{array}{l} \text{Net Rate of Heat} \\ \text{Addition to the system} \\ \text{from the surroundings} \end{array} \right) &+ \\ \left(\begin{array}{l} \text{Net Rate of Work} \\ \text{performed on the system} \\ \text{from the surroundings} \end{array} \right) & \end{aligned} \quad (2)$$

The overall energy of a thermodynamic system is given by Eq. (3).

$$U_{\text{tot}} = UKE_{PEint} \quad (3)$$

The process of pH neutralization is considered the process of the general continuous stirred tank reactor (CSTR), where the liquid undergoes chemical reactions based on the reactants added. The reactor is shown in Figure 1, the reactor system, where the process undergoes. The mass balance of the stirring system under unsteady conditions is given by Eq. (4) where w_1 , w_2 , and w are the mass flow rates into the reactor system.

$$\frac{d(V\rho)}{dt} = w_1 + w_2 - w \quad (4)$$

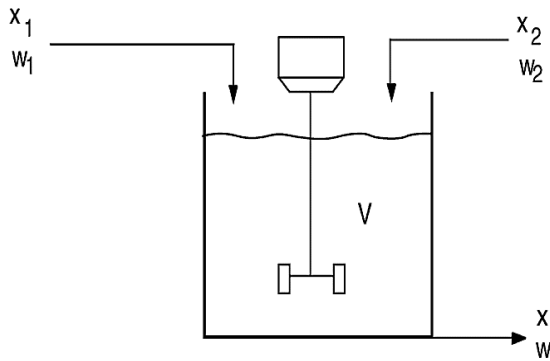


Figure 1. Process diagram of pH reactor

The unsteady balance is given by Eq. (5),

$$\frac{d(V\rho)}{dt} = w_1x_1 + w_2x_2 - wx \quad (5)$$

Eq. (6) and Eq. (7) give the convenient model for the stirred tank reactor.

$$\frac{d(V)}{dt} = \frac{1}{\rho}(w_1 + w_2 - w) \quad (6)$$

$$\frac{dx}{dt} = \frac{w_1}{V\rho}(x_1 - x) + \frac{w_2}{V\rho}(x_2 - x) \quad (7)$$

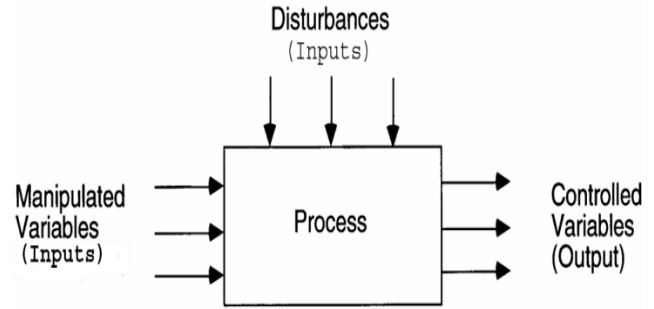


Figure 2. Block diagram of the process involved

During the stirred tank system processes, they are typically performed in a batch or semi-batch reactor. Most industries use the semi-batch reactor system to process their products' substrates. Based on the process involved shown in Figure 2 yields of the products are given by Eq. (8).

$$Y_{x/s} = \frac{\text{Mass of new components}}{\text{Mass of substrate consumed to form component}} \quad (8)$$

$$\text{Rate of accumulation} = \text{Rate input} + \text{rate of formation} \quad (9)$$

The general balanced form of the individual product formed is given by the general Eq. (9), which gives information about the compound, which is developed from base liquid with the addition of the reactants used to complete the process.

4. Hardware utilized

The above section has given the modeling process, which gives information about the process involved in developing the proposed output for the product. This section is about the development of the hardware setup involved in the process of determining the required output based on the model.

The system consists of the following hardware setup to process the liquid for neutralization. The system is highly nonlinear; therefore, the system processes in a batch manner where the liquid is stored in a storage tank. Then, the process liquid is passed into the Process tank, where a batch is taken for the process. In the hardware setup developed, 5 liters of the process liquid is accumulated.

Two reagent tanks hold the two reactants' acid and base solutions. The reactants are added to the process tank based on the pH value set by the user. For example, if the process liquid is acidic (pH 4), it is neutralized for the following process; the reactant base solution is added into the process tank such that the liquid reacts with the base solution to reduce the H^+ ions in the liquid to neutralize. The vice versa operation is done to neutralize a base solution in the process tank which would reduce the OH^- ions in the process liquid.

These dosing pumps work using the current input, which varies from 4-20mA, being 4mA in fully closed condition and 20mA in fully open condition. The dosing pumps allow the reactants to process the tank in drops or liters per hour.

The pump specifications are 5 liters/hour and 9 liters/hour for acid and base solutions, respectively.

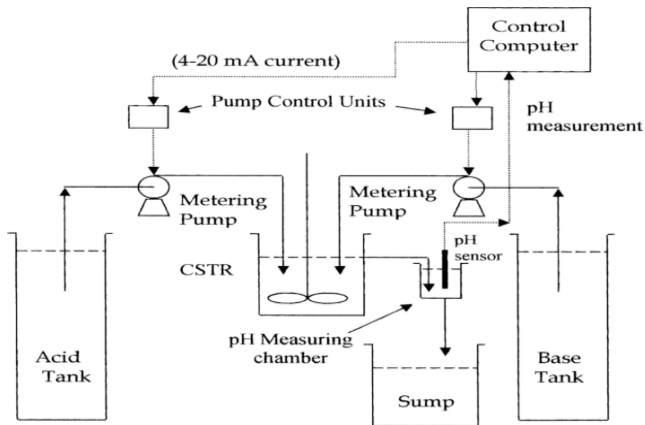


Figure 3. Process diagram of the pH neutralization

The pH of the liquids measured using the electrode involves the measurement of H^+ and OH^- ions in the liquid process. The pH electrode is manufactured on different materials based on usage. The electrode utilized for the measurement is of the glass electrode type, which measures and transmits the pH value in the form of current at 4-20mA.

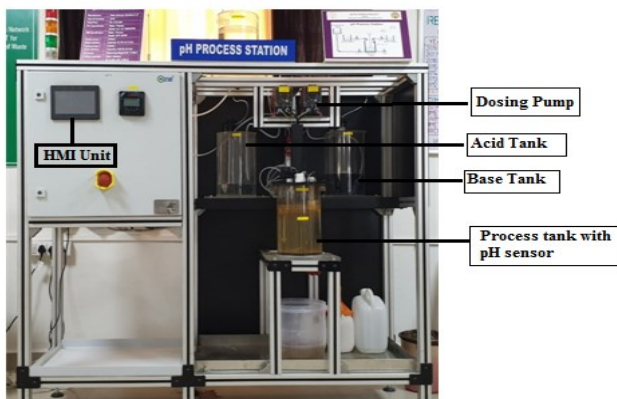


Figure 4. Experimental setup for pH process

The process involved in the pH neutralization diagrammatically represented in Figure 3 is similar to a continuous stirred tank reactor (CSTR). The process tank contains the solution that has to be processed, and two more tank structures are present to hold the acid solution and base solution, respectively. A pH sensor is placed in the process tank, where the liquid to be neutralized is present. Based on the pH value retrieved from the sensor, the controller connected to the system decides on an action to neutralize the liquid. The controller controls two dosing pumps, in which the acid solution and the base solution are fed to the process tank based on which solution to be added to make the process liquid neutral. The process of addition of the acid/ base solution is done by,

- i. If the pH sensor measures the process liquid in the tank to be acidic, the controller turns ON the

dosing pump, which is connected to the base solution tank.

- ii. If the pH sensor measures the process liquid in the tank to be alkaline, the controller turns ON the dosing pump, which is connected to the acid tank.

The user decides on the addition of acid and base, where the dosing pump can be varied by adjusting the amount of solution to be added to the process tank. This experimental setup is set to be at 1000 ml per hour for an acid solution and 2500 ml per hour for a base solution. The variation in the amount of addition is due to the property of those solutions. For example, a small amount of the acid would react with the liquid, in turn changing the pH value much quicker, as the base solution is less reactive; thus, a large amount of the solution is required for the change. The experimental setup for experimenting is shown in Figure 4.

5. Controllers for neutralization

In this chapter, we will discuss the different controllers utilized for the neutralization of liquid. Deep learning is an intelligent controller that uses neural networks as the base with several hidden layers, which are used to identify the minor changes in the values obtained from the input and output of the process based on the prediction. These models are used to build dynamic models of real-time physical systems to analyze, simulate, monitor, and control the physical system.

The deep learning in this pH neutralization process involves two different controlling techniques.

- i) Nonlinear Autoregressive with External (NARX) controller.
- ii) Nonlinear Autoregressive (NAR) controller.

5.1. Nonlinear autoregressive with external (exogenous) input (NARX) controller

The times series problem using the NARX controller uses the values of the given data set to predict the value to be adjusted and controlled to the final control element of the process. The pH value is given as the input to the controller to decide which dosing pump should be energized to add reactant to the process. The process repeats till the value of the pH is reached, which is given as the set point. The dataset can be used to train a neural network to predict a solution's pH in a tank from acid and base flow.

5.1.1. Dataset creation and training

pH Inputs - 1x8000 cell array of 2x1 vectors representing 8000-time steps.

pH Targets - 1 x 8000 cell array of scalar values which states the output pH values of the liquid.

This dataset can train a neural network to predict pH solutions in a tank from acid and base flow.

The data created is of a pH neutralization process in a constant tank of 7 liters. The acid solution Hydrochloric acid (HCL) concentration was 0.0032Mol/L. The base solution Sodium Hydroxide (NaOH) concentration was 0.05Mol/L.

The dataset created is used to design the neural network that predicts the target using the previous data given as

input to the NARX network. The algorithmic steps to develop the network involved are given,

Step 1: $[X, T] = \text{pH}$ dataset:

Step 2: $\text{Net} = \text{narxnet}(1:2, 1:2, 10)$

Step 3: $[X_s, X_i, A_i, T_s] = \text{preparets}(\text{net } X\{, T\})$:

Step 4: $\text{Net} = \text{train}(\text{net}, X_s, X_i, A_i, T_s)$

Step 5: $\text{view}(\text{net})$

Step 6: $\text{plot response}(T_s, Y)$

The above is the process involved in designing the neural network for the input and target given to the network as inputs and targets.

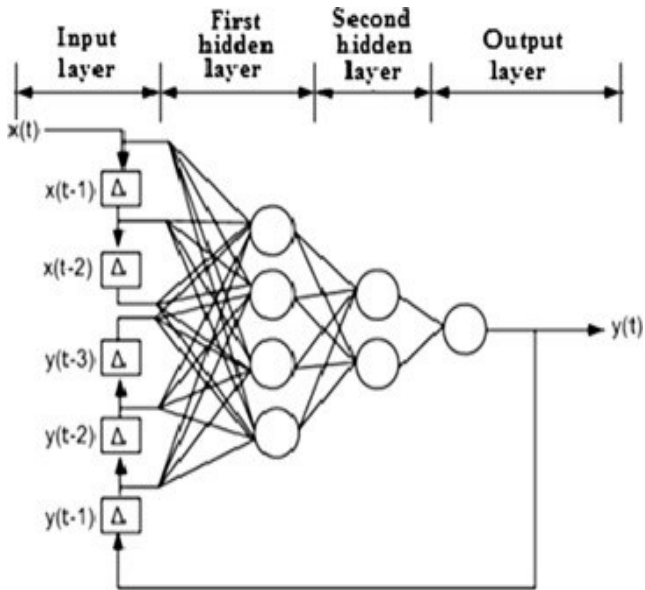


Figure 5. Structure of nonlinear autoregressive with external controller NARX neural network

The dataset developed is then divided into 60% - Training, 20% - Validation, and 20% - Testing, used to design and analyze the developed neural network. The NARX net, which was designed, consists of ten hidden layers and two network delays to determine the disturbance in the network. The flow diagram of the NARX net is given in Figure 5, and the schematic of the NARX net is given in Figure 6.

After the data alignment is done, the data given is to be trained into the network for the prediction. The NARX net consists of 3 training algorithms, of which two.

Levenberg – Marquardt: The algorithm, as mentioned earlier, requires more memory to process the data information given to it. However, the process takes less time than the other two algorithms. The training of the dataset stopped when a generalization of data concerning the mean square error increased with the validation data.

Scaled Conjugate Gradient: The algorithm takes less memory to complete the training of the dataset given, but the result is better for the systems that are noisy, difficult, and small. Data processing is adaptive, which adjusts the weight of the network on minimization.

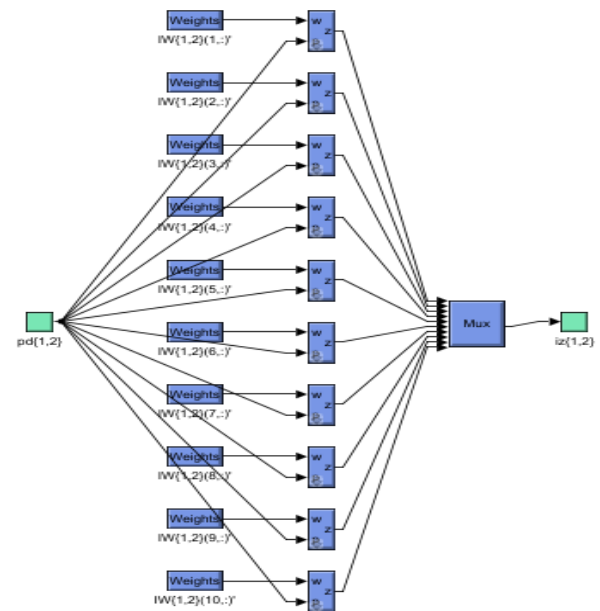


Figure 6. Nonlinear autoregressive with external controller NARX network for the pH model developed

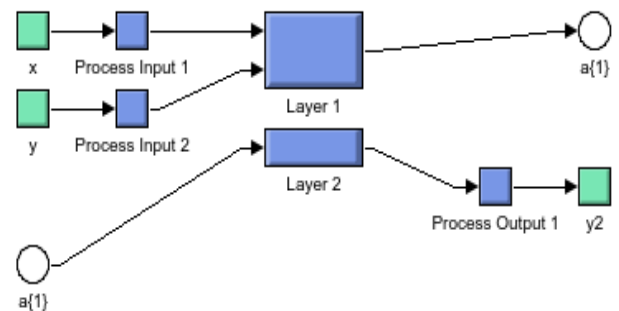


Figure 7. Nonlinear autoregressive with external controller NARX net base flow diagram

The NARX net, developed for the given data set, is shown in Figures 7 and 8. The trained network is introduced into the network to process the real-time data set for the validation and testing of the NARX net. Finally, the response of the test network is given in Figure 9.

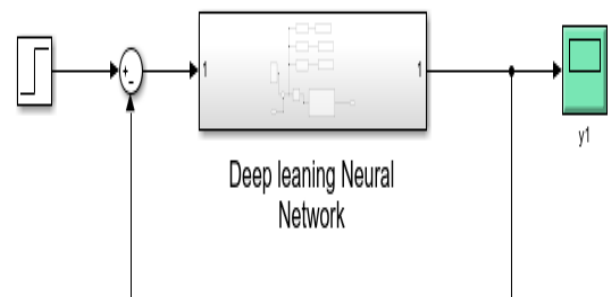


Figure 8. Schematic of nonlinear autoregressive with external controller NARX model developed

5.2. Nonlinear autoregressive (NAR) controller

The simple Nonlinear Autoregressive (NAR) controller is another type of neural net that insists on the deep learning

of data with an increase of the hidden layers to process and predict the data given to the system. The NAR net is a simple process net that predicts future data based on the dataset set trained into the network. The dataset given to it is the pH target values, which act as the input to the NAR net to depict the system's output.

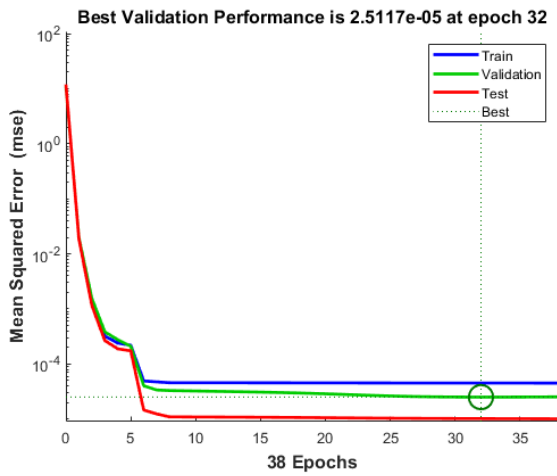


Figure 9. Validation performance of the nonlinear autoregressive with external controller NARX network

5.2.1. Dataset creation and training

pH Targets – 1 x 8000 cell array of scalar values, which states the output pH values of the liquid.

The data created is of a pH neutralization process in a constant tank of 7 liters. The acid solution Hydrochloric acid (HCL) concentration was 0.0032Mol/L. The base solution Sodium Hydroxide (NaOH) concentration was 0.05Mol/L.

The algorithm flow to design the NAR network,

- Step 1: T = simplenar_dataset;
- Step 2: net = narnet (1:2,10);
- Step 3: [Xs,Xi,Ai,Ts] = preparets(net,{}, {},T);
- Step 4: net = train (net, Xs, Ts, Xi, Ai);
- Step 5: view (net)
- Step 6: Y = net (Xs, Xi, Ai)
- Step 7: plotresponse(Ts,Y)

$$y(s) = f(y(s-1), \dots, y(s-d)) \tag{10}$$

The above equation 10 states the function of the NAR net, which predicts the data using the previous data.

The NAR network consists of ten hidden layers and two delay networks for the process weight data of the net to be adjusted. The NAR net schematic is shown in Figure 10. The NAR net is trained using the Levenberg-Marquardt algorithm, which uses 70% of the data set for the training, 15% for validation, and the final 15% for the testing of the network, which is formed.

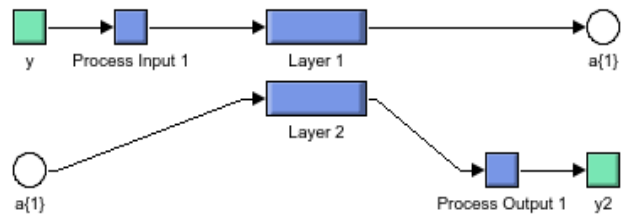


Figure 10. Schematic of nonlinear autoregressive (NAR) controller net

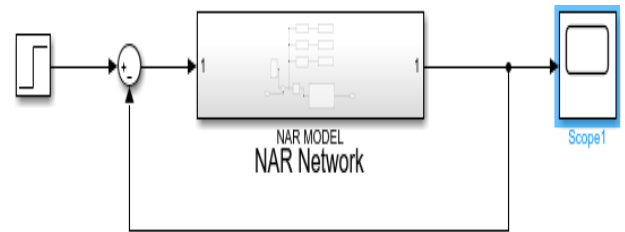


Figure 11. NAR net formed for the prediction of the pH neutralization process

The NAR network is developed for the pH process given in Figure 11. The network, which has been trained, is introduced into the real-time process to depict the output and control the neutralization process of the system. The response of the plot developed by the dataset given as the input to the NAR net is shown in Figure 12. The NAR net's regression points and the NAR network's validation performance for the dataset are given in Figures 13 and 14, respectively.

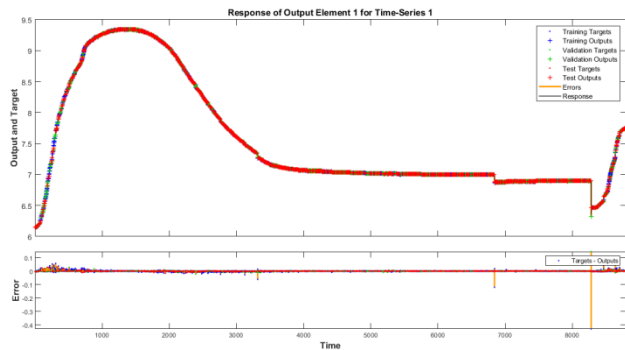


Figure 12. Plot response of the NAR network for the pH neutralization dataset

6. Results and discussions

The pH neutralization is a highly nonlinear process, and the output prediction has been very complicated for real-time data application. The development of deep learning neural networks has shown a way to process the information of the dataset, which is created by the pH process under the open-loop condition. The data is processed under two different neural networks, such as the NARX net and NAR net, which predict the output of the neutralization of wastewater or any liquid.

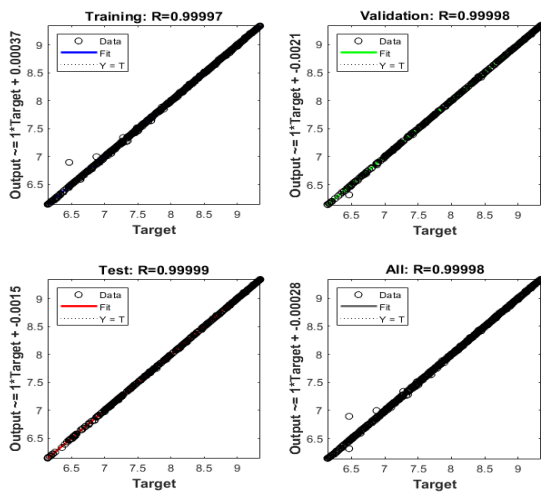


Figure 13. Regression values of the NAR net for the dataset of pH neutralization

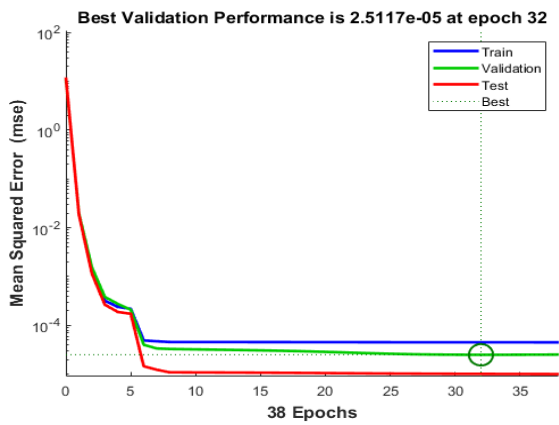


Figure 14. Validation performance of the NAR network for the pH dataset

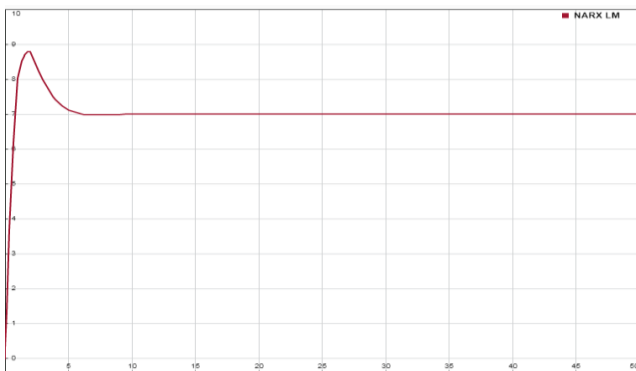


Figure 15. Output of NARX model using the Levenberg – Marquardt

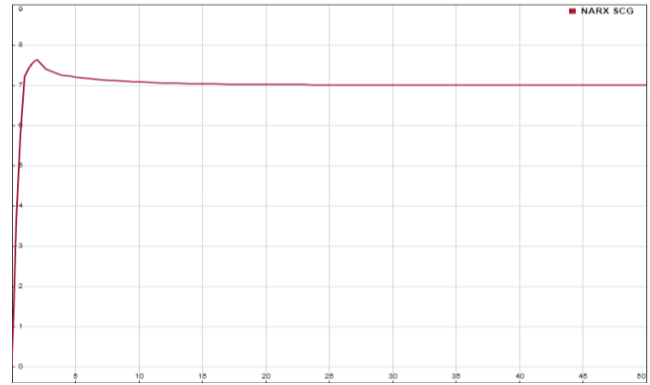


Figure 16. The output of the NARX model using the scaled conjugate gradient

The output of the NARX model for the pH neutralization is given in Figures 15 and 16. The NARX- based network system has shown results that are optimized for the neutralization process to obtain the required value for the processed system. The results show a 25% overshoot for the Levenberg – Marquardt algorithm for neutralization and a 9% overshoot for the Scaled Conjugate Gradient algorithm. The settling time for the NARX net system is 10.5 seconds, and 30 seconds for the respective algorithm (Table 1, Figures 17 and 18).



Figure 17. Output of NAR model using the Levenberg – Marquardt

Table 1. Comparison of NARX & NAR model

	NARX (L-M)	NARX (SCG)	NAR Model
Overshoot (%)	25.9	9.3	0.5
Settling time (in a sec)	10.5	30	8.6
Rise Time (sec)	6.1	7.2	3.6
High value (pH)	8.7	7.7	7.0
Low value (pH)	4.3	3.8	3.5
Slew rate (per sec)	9.0	7.7	1.5

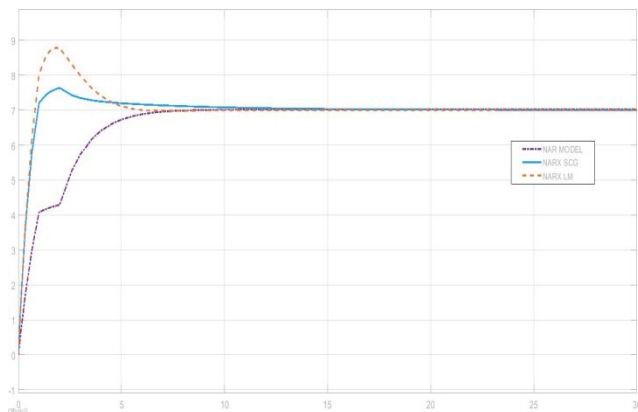


Figure 18. Output comparison of the NARX model and NAR model

7. Conclusion

The pH neutralization is highly nonlinear due to the chemical property of the liquids. Various controlling algorithms were implemented to identify better results to neutralize the wastewater pH. The deep learning algorithms were introduced to the pH process, where NARX net and NAR net were implemented to neutralize the pH. Based on the evaluation of the results of the system, NAR net algorithm has shown better results, such that the overshoot (%) is 0.5 and the settling time is 8.6 seconds which is better than the NARX algorithms. Thus, the NAR model shows better results for the pH neutralization process. In recent years, physiochemical treatments have offered several benefits, including cheap cost, the convenience of use, economic viability, and flexibility in modifying a chemical plant as needed. On the other hand, high operational costs, sludge output, energy usage, and metal selectivity are the techniques' drawbacks. The future development should consider operational costs while undergoing remediation, which would make it more beneficial for use in practical scenarios.

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Conflicts of Interest

The authors declare they have no conflicts of interest to report regarding the present study.

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