

Dynamic Modelling of Electrodialysis with Bipolar Membranes using NARX Recurrent Neural Networks

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Abstract

Electrodialysis with bipolar membranes (EDBM) is an innovative and effective process for the simultaneous production of acid and base solutions from salty streams. It has been proven to play a key role in several circular economy approaches to valorize waste industrial brines, but it can also be used for in situ generation of chemicals, especially in remote areas. The adoption of such technology at industrial scale requires reliable modelling tools capable of predicting both dynamic and stationary operations as process conditions vary, such as energy supplied to the system and the target concentration of chemicals. In this study, nonlinear autoregressive models with exogenous inputs (NARX) were applied for the first time to EDBM to predict the behaviour of this complex and non-linear process. Thus, an effective and low computational demanding neural-based modelling tool was developed. As a preliminary step, the network was trained with three different datasets, generated by a fully validated model. The best architecture was chosen to give good performance, testing the network with a new dataset. The NARX network accurately predicts the different behaviour of EDBM outputs (i.e. voltage and solutions conductivities) showing low average discrepancies between predicted and true values (lower than 0.5 %). These results suggest the possibility of using neural network-based models to effectively optimize and control EDBM process. Next step will focus on the training and validation of a network obtained with a set of data from a real EDBM plant.

Keywords: EDBM, BMED, Circular economy, Brine valorization, Artificial neural networks.

1. Introduction

Industrial processes are responsible for generation of waste streams which need to be treated to reduce their impact and reach environmental standards. In several cases, especially in the field of water treatment (Badruzzaman et al., 2009) and salt production (Turek et al., 2005), mixed salt solutions are produced. These streams represent a valuable unconventional source of minerals which can be recovered through circular economy approaches, improving the sustainability of industrial processes. Recently, the Horizon 2020 Water Mining project (*Grant Agreement*, 2020) has proved a minimum liquid discharge approach to valorize desalination brine, recovering chemicals, high value salts

and fresh water. This is accomplished through the adoption of different units based on membrane, thermal and reactive technologies. A crucial role in the treatment chain is played by ElectroDialysis with Bipolar Membranes (EDBM) unit, a membrane-based technology capable of producing acid and base solutions (Herrero-Gonzalez et al., 2023). These chemicals can be reused inside the chain as reagents for the reactive unit, but also to perform pH adjustment, regeneration of ion-exchange resins and cleaning. EDBM can also be suitable for *in-situ* generation of chemicals ready to use in all the industrial fields where medium-low concentration of acid and base are need (i.e., 1-5 wt.%). This motivates the importance of studying and optimizing this technology to promote its adoption on an industrial scale.

EDBM employs a salt solution and water as feed streams, and through the use of electrical energy is able to simultaneously produce acid and base streams and reduce the salinity of the salt solution. In this process ion-exchange membranes are employed, both monopolar (i.e., anionic and cationic) and bipolar. These membranes allow the selective passage of ions depending on their electrical charge. The bipolar system also promotes water dissociation into protons and hydroxide ions. The membranes are stacked together placing polymeric spacers between them to form the solution channels: acid, base and salt channels. The three solution channels and the ion-exchange membranes constitute the repetitive unit of an EDBM stack, also known as a triplet. Placing several triplets between a couple of electrodes and supplying a direct current allows the process to generate the desired amount of chemicals.

The EDBM process has been modelled adopting a first principles approach based on mass balances and on electro-neutrality requirements (Mier et al., 2008). These models show good performance compared to experimental results, although they require some calibration parameters to be estimated (Culcasi et al., 2022). A major drawback is that they require a significant computational effort, especially when dynamic behaviors are simulated, reducing the attractiveness of these modeling tools, especially for advanced control strategies.

To overcome these limits, AI-based models can be used to speed up predictive analysis and optimization (Ashraf & Dua, 2023). Among them, artificial neural networks have shown good performance in predicting the behavior of membrane-based processes (Asghari et al., 2020). Neural Networks (NN) are black box models capable of reproducing nonlinear relationships between inputs and outputs. The NN structure is composed of processing elements (called nodes or neurons) interconnected using weights and grouped into layers (Jing et al., 2012). The information flow between neurons of different layers can be directed from input to output (feedforward network), or from the output of a layer to the input of a preceding layer (recurrent or feedback network) or also between neurons of the same layer (lateral connections) (Porrizzo et al., 2013). Nonlinear autoregressive models with exogenous inputs (NARX) neural networks have been successfully implemented in many dynamic systems with complex nonlinearities, such as hysteresis phenomena (Wang & Song, 2014).

This study applies for the first time a NN to the EDBM process. Specifically, NARX neural networks were used to develop an effective modelling tool of a commercial EDBM unit. Data to train the network were obtained through a modified version of a first-principles model (Virruso et al., 2024). A semi-industrial scale EDBM unit (Cassaro et al., 2023) was simulated under transient operating conditions, investigating the effect of varying current density and solutions flowrates. The neural network model was tested using an additional dataset (not used during the training phase) showing an average error lower than 0.5 % for multi-step predictions.

2. Methodology

2.1. Data generation

The data used to develop the NARX model were obtained from a dynamic version of a multi-scale model (Virusso et al., 2024), used to simulate a commercial EDBM unit, FT-ED 1600-3 (FuMA-Tech GmbH). The EDBM unit operates adopting the feed and bleed process configuration for all the lines (i.e., acid, base, and salt), Figure 1. Four different datasets were generated by varying the operating conditions as a function of time and observing the dynamic response of the system. Fixed inlet concentrations of the solutions were used in all the datasets. A concentration of 1.5 mol L^{-1} of NaCl was employed for the salt steam, while, for acid and base line, water with a low salt content (1 mmol L^{-1} of NaCl) was utilized. Step changes were given to the outlet flowrates of the solutions and to the current density to investigate several operating conditions of interest for industrial applications. For each dataset consecutive changes were simulated with alternating periods of stable conditions (about 0.9 h) to observe the system's behavior, adopting a discretization time of 100 s. Details about datasets are reported in Table 1.

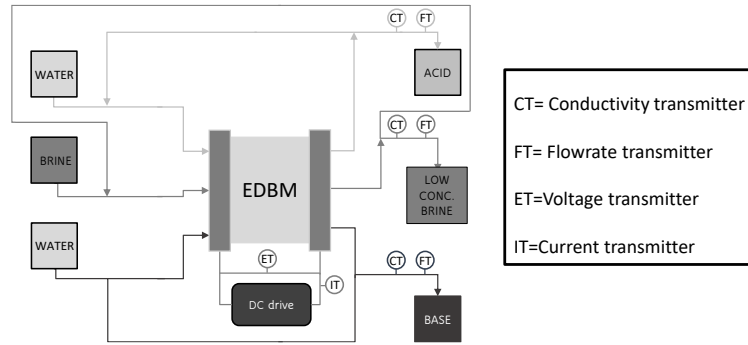


Figure 1. Schematic representation of an EDBM unit operated in the feed and bleed mode. Transmitters are depicted to show the variables used as inputs and outputs in the NARX model.

The variables subjected to step changes represent the inputs of the network and they were chosen to include all the major operating variables that are commonly manipulated in EDBM applications (Virusso et al., 2023). As output of the neural network model, physically meaningful and easy to measure online variables were selected. In particular, the outlet conductivity of the three solutions and the voltage applied to the stack were chosen. The former provides a partial indication of the outlet concentration reached (more detailed analysis must be performed offline), while the latter provides an indication of the energy consumed by the EDBM.

Table 1. Datasets obtained with the mechanistic model. The variation range for each variable, the operation time simulated and the number of step changes are reported for each dataset.

Dataset	$Q_{\text{acid,out}}$ (L min^{-1})	$Q_{\text{base,out}}$ (L min^{-1})	$Q_{\text{salt,out}}$ (L min^{-1})	I (A m^{-2})	$\frac{Q_{\text{base,out}}}{Q_{\text{base,out}}}$	Duration (h)	N. step changes
1	0.5-1.5	0.5-1.5	1-2.25	200-500	1.5-2	10.8	12
2	1-3	1-3	1.5-4.5	200-500	1.5	16.7	19
3	1.8-2.8	1.8-2.8	2.6-4.1	200-400	1.5	10.6	10
4	1.3-2.3	1.3-2.3	1.9-3.4	200-500	1.5	5.5	6

2.2. NARX neural network structure

The network structure presents four inputs (i.e., solutions outlet flowrates and current density) and four outputs (i.e., voltage applied to the stack and solutions outlet conductivities). A single hidden layer was used as this has been demonstrated to provide satisfactory results in different cases, both for feedforward (Jing et al., 2012) and recurrent networks (Cadenas et al., 2016). In addition to the number of nodes in the hidden layer, the order of the Tapped Delay Line (TDL) of input and output must be selected for NARX neural network. The value of TDL represents how many past values of inputs and outputs are needed by the network to estimate the current output (Wang & Song, 2014). The Mean Squared Error (MSE), as shown in Eq (1), was used as cost function to train the network.

$$MSE = \frac{1}{N} \sum_{k=1}^N (y(k) - d(k))^2 \quad (1)$$

Where $y(k)$ represents the predicted value at time step k , while $d(k)$ is the desired value. As activation functions, the Elliot symmetric sigmoid and the linear function were adopted for the hidden layer and the output layer, respectively.

3. Results and discussion

To train the NARX network, *MATLAB's Neural net time series* toolbox was employed. The structure described above was implemented and the Levenberg-Marquardt training algorithm was used. Three different datasets (1 to 3 in Table 1) were employed for the training phase, while dataset 4 was adopted for subsequent testing. The NARX network was trained in series-parallel configuration (also known as open loop), while its performance was evaluated using the parallel configuration (also known as closed loop) for multi-step predictions. Initialization of the network weight was adopted to avoid instability issues in closed loop. The training data were randomly divided into three subsets: training, validation and testing subsets. In particular, 80 % of the data was used for training, 10% for validation and the remaining 10% for verification. It is important to note that this division of results is useful to stop the training phase. The training phase stops at the number of epochs at which the error for the validation set starts to increase and uses the weights and biases determined at that point (Porrizzo et al., 2013).

To define the number of nodes in the hidden layer and the order of TDL of inputs and outputs, different networks were trained by changing the number of nodes between 1 and 15 as well as the number of TDL of the outputs between 1 and 5. The MSE obtained in dataset 4 was calculated using the network in parallel configuration and predicting the entire dataset. The average value of MSE between all the outputs was adopted as the evaluation criterion of the network performance. The best performance was obtained for a NARX network with 12 nodes and a TDL of 3 for the output. Finally, it was checked whether an increase in performance could be obtained using a non-zero value for the TDL of the input, and values between 1 and 3 were investigated. No improvements were observed, consequently a zero order TDL was used for the input.

The generalization performance of the NARX network in predicting dataset 4 are shown in Figure 2. As it can be observed the NARX network, in parallel configuration and doing multi-step predictions, is able to reliably predict the entire dataset 4. The dynamics of all the EDBM outputs were accurately predicted despite the much faster behavior of the voltage compared to the conductivities. The local error between the predicted value and

the true value was evaluated. Average values lower than 0.5 % were found for all the outputs demonstrating the high prediction capability of the NARX networks.

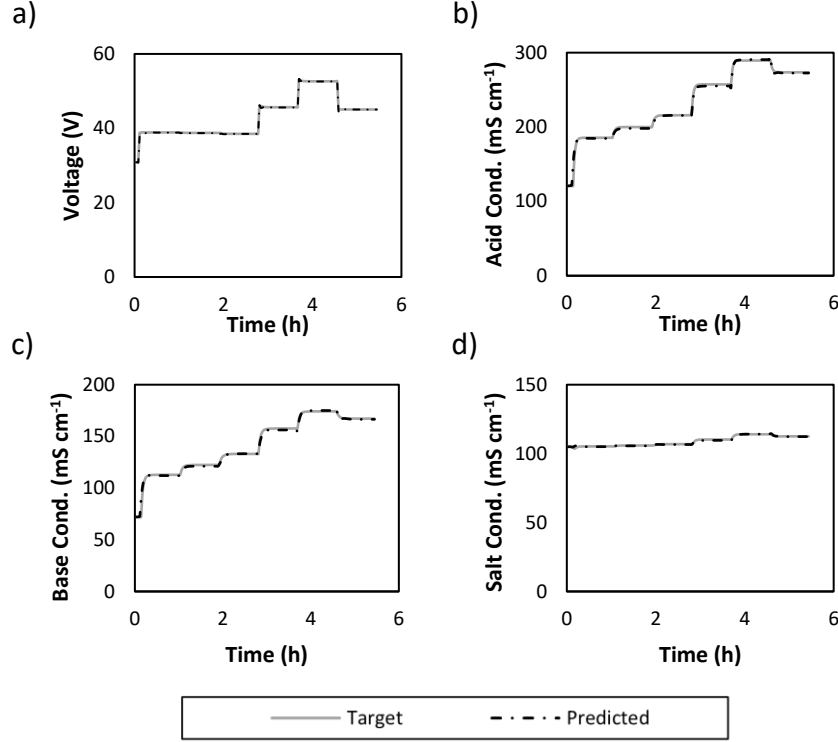


Figure 2. Comparison between the NARX results and the true value for the fourth dataset. The NARX neural network was employed in closed loop mode for multi-step prediction. The time-dependent profiles are shown of: a) voltage applied to the stack, b) outlet conductivity of the acid line, c) outlet conductivity of the base line and d) outlet conductivity of the salt line.

4. Conclusions

This work applies a neural network to electrodialysis with bipolar membranes for the first time. A recurrent dynamic network, NARX, was used to study the EDBM unit dynamic behaviour in feed and bleed operation mode. A NARX neural network with four inputs and four outputs and one hidden layer was employed. The best number of nodes and the order of tapped delay line (TDL) of input and output was determined evaluating different architectures. As criterion to select the best architecture the MSE obtained on the entire dataset 4 was used, adopting a parallel configuration and doing multi-step predictions. The selected network presents 12 nodes in a hidden layer as well as a 3rd order TDS for the output and no TDL for the input. The generalization performance was shown demonstrating that the different dynamic responses of the EDBM outputs can be predicted with high precision (average error <0.5 %). These results show the effectiveness of NARX networks in describing complex nonlinear electro-membrane systems, suggesting that they can be used for advanced control systems and to optimize EDBM process. Future research will also investigate the use of real data, obtained from an operating pilot scale

EDBM unit, to assess whether the effect of non-ideal phenomena and noisy signals can affect the network performance.

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