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NEURAL NETWORKS APPLICATIONS IN CHEMISTRY AND CHEMICAL ENGINEERING

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ABSTRACT. The paper presents results of using artificial Neural Networks (NN) in chemistry and chemical engineering applications. Several incentives accomplished by NN are revealed and sustained by relevant examples: capability of predicting the behaviour of systems for which the first principle modelling is difficult or insufficiently known (Simulation of Electrochemical Impedance Diagrams), capacity of building reliable dynamic models subsequently used for model based control (Model Predictive Control of the Drying Process of Electric Insulators) and ability of ANN to perform classification tasks where the membership of individuals to specific groups is not obvious (Characterisation of Commercial Vinegar).

1. INTRODUCTION

The field of NN has a history of five decades but it has found solid application only in the last decade and the field is still developing rapidly. Founded on an idealised model of the biological neuron, the calculation paradigm of NN is able to represent information on complex systems. The main characteristics of the NN model are the inputting of information (signals) from exterior or other units of the network, feeding it to the given unit (neuron) that processes it and then sending it, as output, to other units or output of the network. The main benefits of the NN approach consist in its remarkable ability of learning, generalisation and robust behaviour in the presence of noise [1]. As a consequence, the NN may be successfully used for modelling systems in which detailed governing rules are unknown or are difficult to formalise, but the desired input-output set is known. The NN prediction capability of generating new values of outputs is usually highly appreciated.

The use of NN models for control purposes has gained considerable attention in the field of chemical process control, being the subject of several scientific reports, as they are increasingly applied for system identification and controller design [2]. The favourable opportunities offered by the NN also consist in important savings of computer resources, having direct effect over the on-line computation in control oriented applications. The results presented in the following, reveal the potential of NN for both simulation and control applications.

2.SIMULATION OF ELECTROCHEMICAL IMPEDANCE DIAGRAMS

The NN approach has been used to improve the electrochemical impedance spectroscopy (EIS) set of experimental data by NN simulation, for copper electro-deposition. The trained NN, with data obtained in different experimental conditions (electrode potential, and thiourea concentrations), were used: (i) to improve the estimation of two important electrochemical parameters (the double layer capacity of the interface and the electrolyte resistance) by generating supplementary output data for new frequency values, both inside and outside the investigated domain; (ii) to generate impedance spectra for new electrode potential values within the investigated range of the potential [3].

The investigated system has been considered as having three input variables: the continuous amplitude component of the sinusoidal applied voltage, the signal frequency and the thiourea concentration. First, the NN has been trained using the bulk set of experimental data (90% of the original experimental set). Second, in order to test the NN generalization capability, the remained subset of experimental data (10% of the original experimental set) was operated. Results showing the comparison between the experimental and NN simulated data are presented in figures 1 to 3, for for all the three NN output variables: current amplitude, real part and imaginary part of the electrochemical impedance.

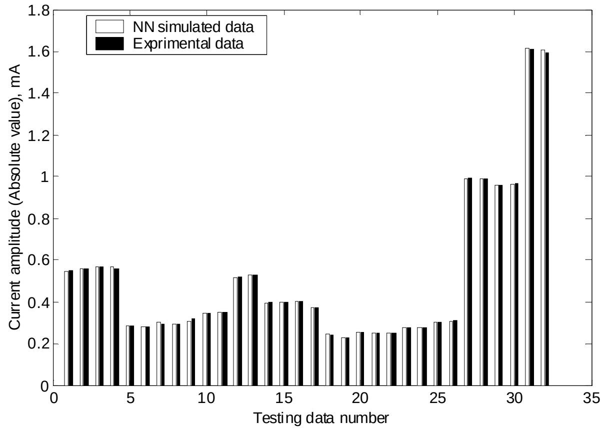

Figure 1. Comparison between NN simulated data and experimental data for current amplitude. Experimental conditions: sinusoidal frequency ranging from 2.2 Hz to 6.8 kHz; thiourea concentration c=10 mg/l.

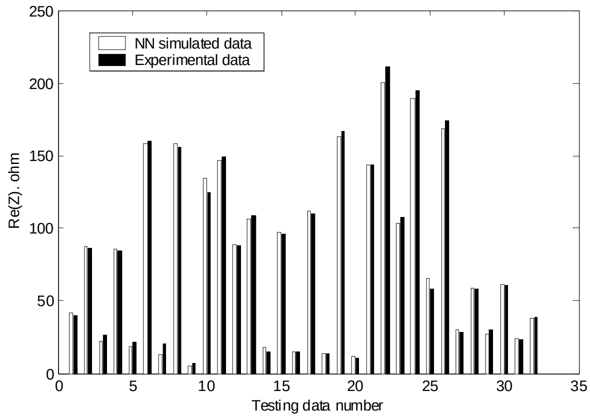

Figure 2. Comparison between NN simulated and experimental data for the real part Re(Z) of the electrochemical impedance. Experimental conditions: sinusoidal frequency ranging from 2.2 Hz to 6.8 kHz; thiourea concentration c=10 mg/l.

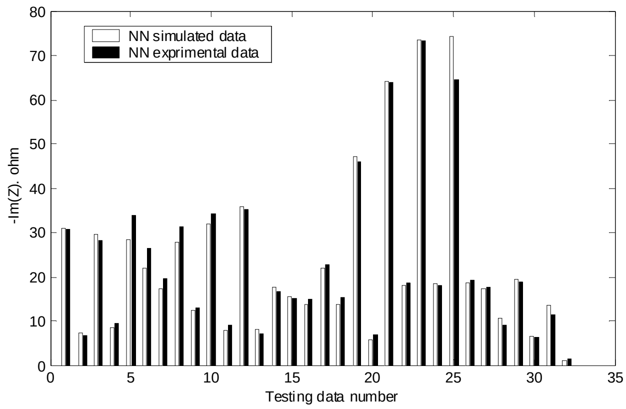

Figure 3. Comparison between NN simulated and experimental data, for the imaginary part Im(Z) of the electrochemical impedance. Experimental conditions: sinusoidal frequency ranging from 2.2 Hz to 6.8 kHz; thiourea concentration c=10 mg/l.

Results presented in figures 1 to 3 reveal a good prediction capability of the trained NN, with good results for the case of the presented frequency range: 2.2 Hz to 6.8 kHz.

Additional investigation has been performed to study the prediction capability of the trained NN for generating outputs of the electrochemical system for input variables having values situated between (different from) the experimental values used in the training or testing steps. Thus, different values for the amplitude and for the frequency of the input electrode potential have been supplied to the NN and the obtained results are presented in figure 4.

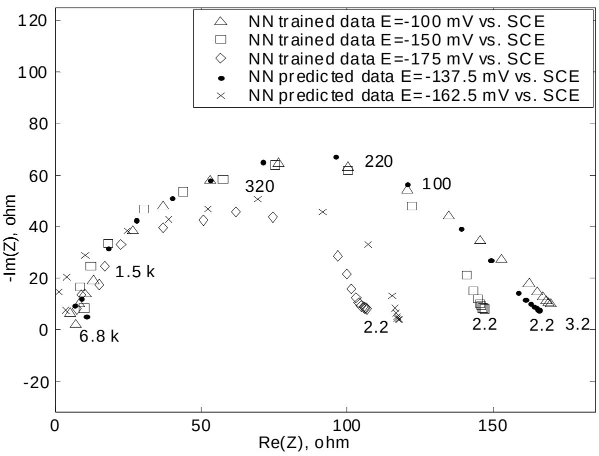

Figure 4. Nyquist plot presenting NN predicted values of the electrical impedance for different values of the input electrode potential. Experimental conditions: sinusoidal frequency ranging from 2.2 Hz to 6.8 kHz; thiourea concentration c=10 mg/l.

The obtained results fit to a shape that conforms with the expected form (qualitatively inferred by induction) for the electrochemical impedance. This agreement proves that the trained NN were able to predict well the electrochemical impedance of this complex system, otherwise difficult to be described using analytical models.

3. MODEL PREDICTIVE CONTROL OF THE DRYING PROCESS OF ELECTRIC INSULATORS

The high-voltage electric insulator production implies a first step batch drying process in order to reduce the moisture content of the drying product from 18-20% to 0.4%, performed in special gas-heated drying chambers. Gas and air flow rates are controlled according to a special program, during a period of about 100 hours, in order to obtain the desired moisture content and avoiding the risk of unsafe tensions in the drying products. Building a detailed first principle model, able to thoroughly describe the spatial and temporal evolution of properties inside the electric insulator body and needed in model based control, is complex and not yet reliable [4]. The NN-based control may overcome some of these problems.

The NN model of the dryer has been developed to serve two goals. The first one is to provide information on time evolution of target variables, inherently needed for prediction in the Model Predictive Control (MPC) algorithm. The second one is to infer the moisture content of the drying product, based on available measured variables, model that is later used for NN observer based MPC. The NN developed model has the complimentary property of requesting reduced computation effort, supplying the algorithm with speed necessary for real time implementation.

Taking into account that the target variable, i.e. the moisture content of the product, is not available for direct measurement, a NN based state observer is proposed for its estimation. The data provided by the NN state observer is used for feedback MPC of the product moisture content, as shown in figure 5, [5].

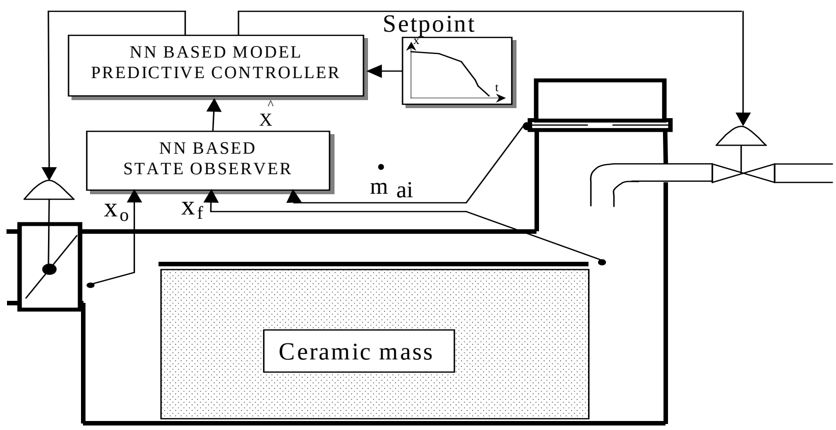

Figure 5. Structure of the control system for direct moisture content control using NN based state observer and NN based MPC controller.

The time dependent setpoint selection for the moisture content is based on practical and theoretical considerations concerning the time evolution of the product drying-rate. The conditions stated by the above mentioned considerations are best fulfilled by a seven-segment ramp function, which is actually used as setpoint. Simulations were conducted using this control structure and the results, for two *10%* heating power disturbances applied at both *t=3000 s* and *t=1250000 s*, are presented in figure 6.

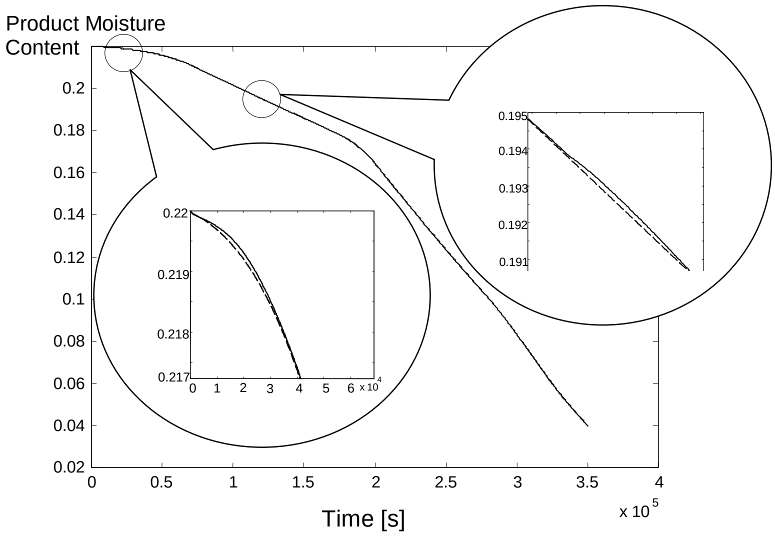

Figure 6. Setpoint (dashed line) following and disturbance rejection ability for direct moisture content control using NN based state observer and NN MPC (solid line).

The simulation results for this control structure show both good setpoint following and disturbance rejection capability. The applied model predictive algorithm has a few special features that makes it more effective: it operates with constraints on manipulated variables and controlled variables; in order to obtain the feasible and feasible control performance, a nonlinear form of the MPC algorithm was used; the MPC controller was tuned according to the dynamic sensitivity analysis. A significant reduction of the computation time (2:1 ratio) has been also observed as a consequence of NN model use that requires less computer power compared to the first principle model.

4. CHARACTERISATION OF COMMERCIAL VINEGAR BY NEURAL NETWORKS

The aim of this appication was to classify several vinegars using the Neural Networks approach [6]. The classification was based on the determination of eleven analytical parameters of commercial vinegars, divided into five categories according to the raw material: white wine vinegar (WWV), red wine vinegar (RWV), alcohol vinegar (ALV), apple vinegar (APV), and non-coloured industrial vinegar (NIV). The analytical parameters assessed were the usual employed for controlling and monitoring the quality of vinegar: total acidity ($g \cdot L^{-1}$) detached in volatile acidity ($g \cdot L^{-1}$) and fixed acidity ($g \cdot L^{-1}$), pH, dry matter ($g \cdot L^{-1}$), ash content ($g \cdot L^{-1}$), sulphates ($g \cdot L^{-1}$), chlorides ($g \cdot L^{-1}$), glycerol ($g \cdot L^{-1}$), total polyphenol index (TPI, dimensionless), Folin-Ciocalteu index (FCI, dimensionless) and modified colour intensity (MCI, dimensionless). Ninety-four commercial vinegars were acquired in six supermarkets in Tarragona (Catalonia, Spain) during the year 2000. The markets selected cover most of the sales to consumers of food and beverages. Samples were taken directly from every meter of displayed product in the stand (i.e. those brand names with more rack space are represented with more samples). Using this sampling method a representative mix, directly related to the market share of each vendor, was achieved. As a result, the number of vinegars per categories was not homogeneous (WWV: 54 samples; RWV, 32 samples; ALV, 3 samples; APV, 4 samples; and NIV, 2 samples).

First, the NN has been trained to learn the vinegar categories based on the original set of data from which a randomly selected set of data (testing set) has been extracted. Second, the trained NN was used for testing the prediction capability using the testing set of not yet encountered data. For the testing set of 13 vinegar samples the NN succeeded to predict the right category, without any mismatch. The obtained results showed a good capability of the NN to classify the vinegar category.

Results of this NN based category classification, for the testing set of data, are presented in Table1.

Table 1

NN based category classification

Sample no.	Actual category	Predicted category				
		WWV	RWV	NIV	APV	ALV
4	WWV	✓	-	-	-	-
10	APV	-	-	-	✓	-
15	WWV	✓	-	-	-	-
25	ALV	-	-	-	-	✓
30	RWV	-	✓	-	-	-
34	RWV	-	✓	-	-	
44	RWV	-	✓	-	-	-
47	WWV	✓	-	-	-	-
56	WWV	✓	-	-	-	-
64	WWV	✓	-	-	-	-
72	RWV	-	✓	-	-	-
78	WWV	✓	-	-	-	-
82	WWV	✓	-	-	-	-

Another NN was trained to classify the producer. For the same set of 13 vinegar samples, this special trained NN succeeded to predict the producer (among the set of 9 producers), with small errors. For this case the classification errors vary, when picking different sets of training/testing data, from perfect match to one mismatch out of the 13 samples. A perfect match case is presented in Table 2.

The results show a good capability of the NN to classify the vinegar, according to both investigated criteria: category and producer. Results also demonstrate that NN succeed to capture the intrinsic relationship between vinegar properties and producer, thus stating that product characteristics are strongly dependent on the production techniques.

Table 2

NN based producer classification

Sample no.	Actual Producer	Predicted Producer								
		P1	P2	P3	P4	P5	P6	P7	P8	P9
4	P1	✓	-	-	-	-	-	-	-	-
9	P1	✓	-	-	-	-	-	-	-	-
14	P2	-	✓	-	-	-	-	-	-	-
26	P2	-	✓	-	-	-	-	-	-	-
30	P3	-	-	✓	-	-				
34	P3	-	-	✓	-	-	-	-	-	-
44	P4	-	-	-	✓	-	-	-	-	-
47	P4	-	-	-	✓	-	-	-	-	-
56	P5	-	-	-	-	✓	-	-	-	
64	P6	-	-	-	-	-	✓	-	-	-
72	P7	-	-	-	-	-	-	✓	-	-
78	P8	-	-	-	-	-	-	-	✓	-
82	P9	-	-	-	-	-	-	-	-	✓

5. CONCLUSIONS

Important incentives of the ANN approach are explored, such as modelling process for which detailed governing rules may be difficult to formalize as first principle models and reducing the computation time in the case of nonlinear model based control. Simulation results also reveal benefits for the NN based MPC using the NN based observer approach (control of inferred variables), compared with traditional direct feedback control methods.

For the simulation of electrochemical impedance diagrams the employed NN architecture was of the multilayer feed-forward structure with the backpropagation training algorithm used for computing the network biases and weights. Two layers of neurons have been considered, having the tan-sigmoid transfer function for the hidden layer and the purelin transfer function for the output layer. The quasi-Newton Levenberg-Marquardt algorithm was used for training the NN and an early stopping method was applied for preventing the NN overfitting and for improving generalisation.

For the dynamic NN based model of the drying process the NN inputs are the states of the system at current moment of time t together with the manipulated variables, and the NN-outputs are the three state variables considered at the next sampling time $t+\Delta t$. The trained NN is designed to predict one step into the future the behaviour of the state variables. Applied repeatedly, the dynamic NN predicts the time evolution of the state variables over a desired time horizon.

For the classification purposes the NN architecture had a radial basis layer and a competitive layer. The first one computed the distance between the input vector and the training input vectors, generating a vector with elements measuring the deviation with the training input vector. The second layer processed the deviation vector for each class of inputs creating a vector of probabilities from which the competitive transfer function selects the maximum of the probabilities.

Designing the network architecture, i.e. the number of layers and the number of neurons in each layer of the NN, is not a trivial task as there are no simple rules for setting them. Trial and error techniques still play an important role for setting a minimal but successful NN structure. Another important aspect is the pre-processing of the data used for training the NN. Expunging the outliers in the process of training and repeating the training procedure with the remaining set of data may be a good choice to filter the noisy or non-confident process data.

Although NN do not prove to be a panacea for all modelling or control purposes they present incentives for a large set of applications showing continuous open improving potential for further development.

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