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Application of Artificial Neural Networks in an Experimental Batch Reactor of Styrene Polymerization for Predictive Model Development

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Batch reactors are widely used in the polymer industry, especially for multi-purpose processes where different types of polymers are produced on demand. Batch polymerization reactors impose rather great operational difficulties due to the complex reaction kinetics and inherent process nonlinearities. Thus it is a difficult task to develop mathematical models for polymerization processes. If required for process control purposes the model should be accurate but simple, so that it can be used in a control loop. The present work shows the application of the neural network approach in the development of a predictive model for a styrene polymerization pilot plant, located at the Laboratory of Chemical Systems Engineering, School of Chemical Engineering at UNICAMP. Artificial Neural Networks have become a usual application in many areas of engineering, and are well suited for chemical processes due to their ability to describe multi-variable non-linear models. However to control purposes, the consideration of the variation of the process variables in real time is required as input to the model, to ensure the representation at the dynamics of the process. The experimental prototype consists of a jacketed stirred reactor, using thermal fluid as a heat source. Reaction progress was measured by a density sensor situated in a external recycle loop. Temperature sensors were positioned both inside the reactor and in the inlet and outlet of the jacket. Traditional *feedforward* neural networks with back-step inputs and the *Elman* network were applied to obtain the best model to be employed in a control loop. A comparison between the networks was performed, showing that, for process dynamics modeling, both networks were able to create suitable polymerization models.

1. Introduction

One of the many advantages of batch processes is ability to allow for the change of operating conditions, both at start up and along the reaction, in order to conduct them to desired products and material properties. According to Hosen et al. (2011), it is common to manufacture different products in the same equipment, for instance the polymer industry, where batch reactors are used to produce materials aggregate to demands of all potential customers in a specific market.

Many batch processes present serious difficulties for control due, especially, to the inherent complexity of the involved reactions, like the nonlinear behaviour and with properties that vary whole batch period, namely, require the frequent change of operating parameters (Galván et al. 1997). Lastly, the results are products yield lower than expected.

Searching for performance improvements of process is usual to develop mathematical models for the dynamic simulation of the systems, so that studies to improve yield, cost minimization and test of new control strategies can be performed.

The development of phenomenological models requires much time and can lead to high costs due the range of operating conditions that have to be considered, and the large number of components and reactions involved. Also, the additional cost might not be compensated for, as many batch reactors are used for smaller scale production.

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In this context, a faster way of developing models for control purposes is the so-called system identification, which uses mathematical models to create relationship between process variables, using actual data from the process, but without resorting to phenomenological models. Through optimization techniques the model parameters are adjusted so that the prediction model performs close to the actual dynamic behaviour. In this field artificial neural networks (ANN) have become of widespread use. According Chen et al. (1992), the ANNs have the ability to be adjusted as nonlinear models, being well suited to be used as models in system identification.

2. Experimental Configuration

The styrene polymerization pilot plant, located at the Laboratory of Chemical Systems Engineering, School of Chemical Engineering at UNICAMP, was used for the process data acquisition. The experimental prototype consists of a stirred jacketed reactor (total volume of 1.2 L).Thermal oil was used as heat transfer medium in the jacket. An electrical heater, connected to a thyristor, provided heating to the thermal fluid inside a storage tank. The dissolved oxygen was purged by bubbling pure nitrogen gas through the reaction mixture. The monomer was obtained with 99% purity from *Sigma Aldrich*. Toluene was used as solvent and it was purchased from *Ecibra* with a purity of 99%. No further purification was used. The initiator agent for the reaction was benzoyl peroxide (BPO) from *Sigma Aldrich*, presenting 70% purity. There are temperature sensors inside the reactor and in the inlet and outlet of the jacket. A scheme layout of the prototype is shown in Figure 1.

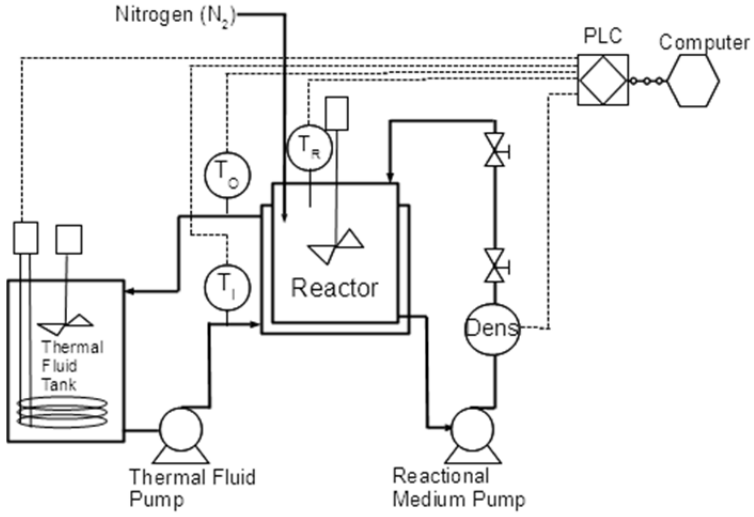


Figure 1: Scheme layout of the polymerization prototype.

Table 1: Experimental Conditions of the Styrene Solution Polymerization Reactor

Parameter	Value
Reaction volume	800 mL
Reactor stirring speed	200 rpm
Jacket flow rate	300 L/h
Heating power	3000 W
Initiator concentration	0.0185 mol/L

After loading the monomer and the solvent into the reactor, the system was heated to reach the desired operating temperature of 90 °C (Ghasem et al., 2007), when the initiator BPO was added to start the polymerization reaction. Typical experimental operating conditions are shown in Table 1. Experimental runs were conducted using several monomer/solvent ratios: 30, 50 and 70 V/V %.

For this kind of reaction the prediction of reactor temperature is important for product quality. Therefore, the aim of the sought model was to predict reaction temperature *TR* at time $k + 1$, given the following process variables at time *k*: actual reactor temperature *TR* [*k*], jacket inlet temperature *Ti* [*k*] and the power *P* [*k*] being sent for the oil heater. In the *feedforward* topology the same variables were inputted at time [k-1] and [k-2].

3. Neural Networks Models

Artificial Neural network is an artificial intelligence technique that has been widely used in the chemical industry for the development of nonlinear process models, due to its great ability to generalize and represent nonlinear systems satisfactorily. These models are of the black-box type as the model is generated by adjusting parameters (weights and bias) until the error between the dummy variable and real variable is small enough. In the case of ANN´s the building blocks of the models are the neurons, which generate an output signal by processing an input signal through an activation function.
The ANN is built by the union of a several neurons. Figure 2 illustrates the topology of a typical *feedforward* network with three inputs, one output and an intermediate layer.

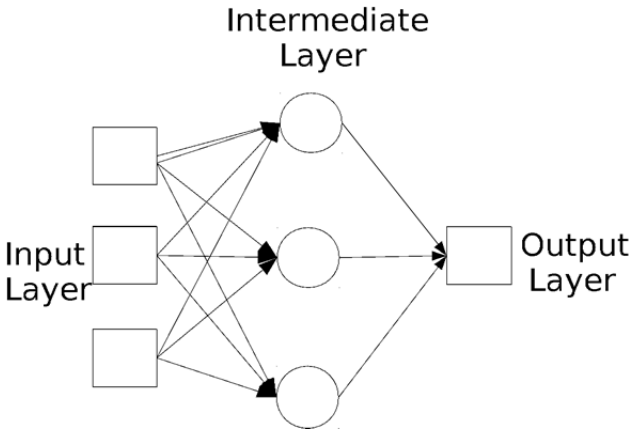


Figure 2: Topology layout of a feedforward ANN.

In the *feedforward*, the input signals are processed from left to right as shown in Figure 2. For a network with two neurons, one in each layer, see Figure 3, the output *Y* as a function of input *X* is given by:

$$Y = f_2(w_2 . f_1(w_1 . X + b_1) + b_2) \quad (1)$$

where f_1 and f_2 are the activation functions, w_1 and w_2 the weights, and b_1 and b_2 the bias of neurons 1 and 2, respectively.

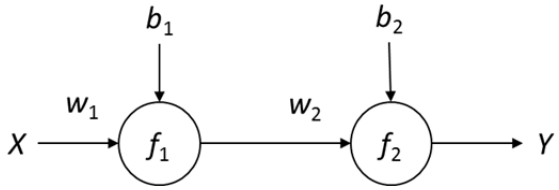


Figure 3: Feedforward ANN topology for 1 neuron in each layer

In an *Elman* network (recurrent network), there is a return signal from the output of the intermediate layer neuron to the input of the same neuron, see Figure 4. This signal is delayed by one time step and multiplied by a weight (w_2) thus allowing for the consideration of the dynamic behaviour of the process.

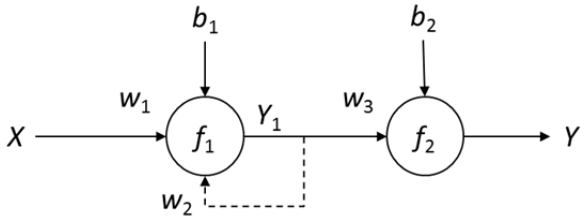


Figure 4: Elman ANN topology for 1 neuron in each layer

The equation representing the output of an *Elman* network is shown by Eq(2) and Eq(3).

$$Y[k] = f_2(w_3 . f_1(w_1 . X[k] + w_2 . C + b_1) + b_2) \quad (2)$$

where,

$$C = Y_1[k-1] \quad (3)$$

Compared to the *Elman* network the *feedforward* network is much simpler. For a two-neuron network the *feedforward* network has four adjustable parameters, two weights (*w*) and two bias (*b*). An *Elman* network of the same size requires five adjustable parameters, three weights (*w*) and the two bias value (*b*). With the growth in the number of inputs and neurons in the intermediate layer, it is expected for the *Elman* ANN to become harder to be trained, as the number of parameter to be fitted increases. For both cases the activation functions are of a non-linear type, usually of sigmoid shape.

4. Results and Discussion

The dynamic behaviour of the reaction system was measured in four different batches, resulting in 9068 samples. Due to the short period of time used to sample variables five seconds from each batch run the number of data points used for the ANN training had to be assessed. The training data was evaluated by a cross-covariance technique, as suggested by Aguirre (2007). This technique is a simple heuristic method to identify the most adequate time span between data points, so avoiding either redundancy or lack of suitable data points for training, and is based on the covariance of the sampled data:

$$Cov_{x,y}(\varepsilon) = \frac{1}{N} . \sum_{k=1}^{N} [(x(k) - \underline{x}) . (y(k-\varepsilon) - \underline{y})] \quad (4)$$

where:

N is the number of elements in vectors x and y.
x and *y* are the input and output vectors, respectively
$\underline{x}$ and $\underline{y}$ are the mean values of the respective vectors
ε is the number of delays

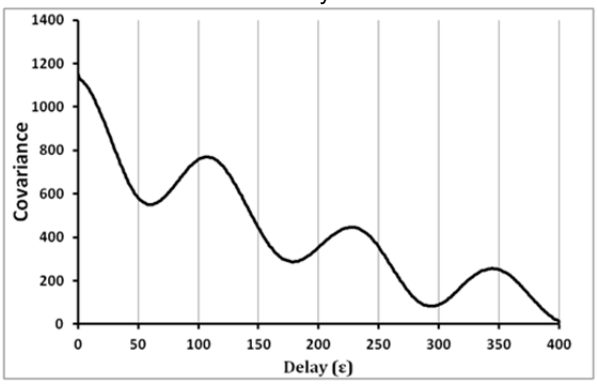
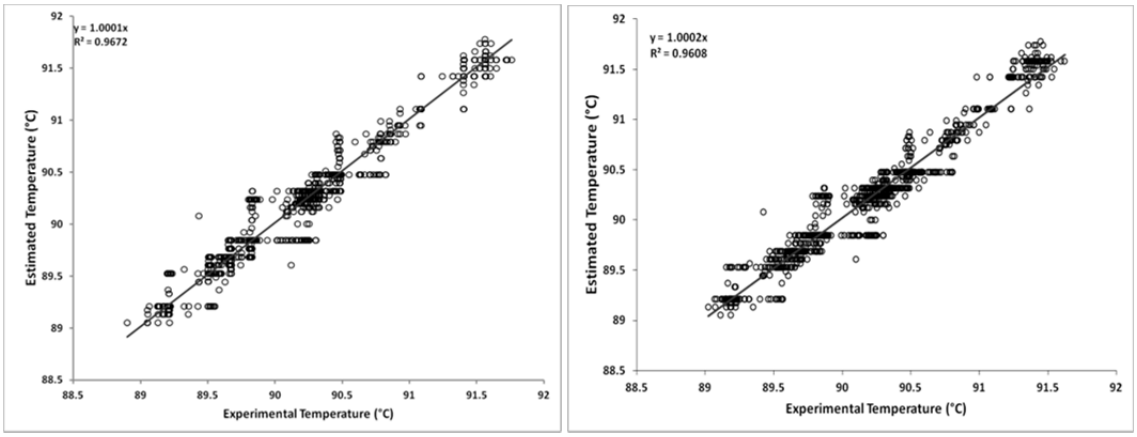


Figure 5: Covariance as a function of delay

Applying the technique to the sampled data leads to the identification of the first local minimum, in Figure 5, leading to an optimal sampling time of 20 s, and a reduction of the data set from 9068 to 453 points.
Once the training data set was obtained, the dynamic behaviour of the reaction systems was modelled with both the *Elman* and *feedforward* ANNs. The networks were developed and trained using *Matlab ® 7.0*, Neural Networks Toolbox.
Both networks showed a good performance with regard to minimizing the error for training and validation, obtaining correlation coefficient of 0.96. Figure 6 a comparison between both networks is presented, showing

a good agreement between actual and estimated data. In both cases a maximum variation of 0.5% for the reactor temperature was achieved.



a) b)

Figure 6: Comparison between performance on the validation of a) Elman ANN and b) Feedforward ANN.

Other parameters were also evaluated in order to point out differences between these two types of ANN. Table 2 shows additional information about time, training algorithms and number of adjustable parameters. Although the performance to predict reaction temperature is similar for both cases, the number of parameters and training time for *Elman* network is much greater than for the *feedforward* network of the same topology.
A problem found in the *Elman* network is the difficulty in training, as it was not possible to uses a training procedure with bayesian regularization, which avoids overfitting. The use of the Levenberg–Maquardt method increased the training time for the *Elman* network 28 fold, compared to the *feedforward* network. The best performance of the *Elman* network was only obtained when the hyperbolic tangent function for the intermediate and output layers was chosen, which demands greater complexity to the model.
In point of view of the model implementation in a control loop, the *Elman* network is also more complex as it requires more memory to store signal until the next loop.

Table 2: Neural Networks Performance Comparison

Parameters	Elman	Feedfoward
R^2	0.9672	0.9608
MSE (°C)	$1.87x10^{-3}$	$1.92x10^{-3}$
Topology[1]	3x3	9x20
Train Time(s)	60.19	2.18
Transfer Functions	Tansig/Linear	Tansig/Tansig
Train Algorithm	LM[2]	BRB[3]
Number of Weights	21	200
Number of Bias	4	21

1. Number of input neurons and neurons in the intermediate layer.
2. Levenberg-Maquardt algorithm
3. Bayesian regularization backpropagation algorithm.

5. Conclusions

In this work an application of artificial neural networks to the identification of a polymerization system was presented. With data obtained from four experiments, the *Elman* neural networks and traditional *feedforward* were trained to model the dynamic behaviour of the process and used to estimate the reactor temperature.
The comparison between the performances of both networks showed that although *Elman* networks intrinsically have the capacity to describe dynamic models, they suffer in aspects related to the computational effort and training time, and in implementation in a real system due to the additional complexity of requiring intermediate storage.

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