Artificial Intelligence Modelling Methodologies Applied to a Polymerization Process

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Abstract: A series of modelling methodologies based on artificial intelligence tools are applied to solve a complex real-world problem. Neural networks and support vector machines are used as models and differential evolution and clonal selection algorithms as optimizers for structural and parametric optimization of the models. The goal is to make a comparative analysis of these methods for the case study of the free radical polymerization of styrene, a complex, difficult to model process, where the monomer conversion and molecular masses are predicted as a function of reaction conditions, i.e. temperature, amount of initiator and time. Four modelling methodologies are developed and evaluated in terms of accuracy.

1 INTRODUCTION

Artificial neural networks (ANNs) are recommended tools for modelling complex nonlinear processes because they require only input-output data, with no need for in-depth knowledge of the rules governing the system. They often lead to accurate results and can be integrated into optimal control procedures.

Beside ANNs, support vector machines (SVMs) are gaining popularity over other learning methods, mainly due to their good generalization capability (Burges, 1998). Another important advantage is that SVMs perform well on high dimensional problems, and there is ongoing research on improving their scalability (Wang et al, 2011; Zhang et al., 2012).

Developing optimal ANN or SVM models with the adequate parameters is not an easy task. In the trial-and-error method (frequently applied by the majority of researchers, especially from engineering domains), the architecture is repeatedly modified by hand and evaluated with the goal of lowering the error. These repeated actions increase the computational overhead and the search is usually based on gradient descent, whose result is prone to being trapped in local minima (Cartwright and Curteanu, 2013). In the polymerization field, the use of ANN and SVM is increasing. Different types of processes are modelled with these techniques, as proven by the different review works (Noor et al., 2010; Cartwright and Curteanu, 2013).

Evolutionary algorithms (EAs) are promising methods for optimizing both the architecture and the internal ANN parameters (Almeida and Ludermir, 2008a; Almeida and Ludermir, 2008b). Among all EAs, differential evolution (DE) is an especially powerful approach. Its efficiency lies in a simple, compact structure that uses stochastic direct search (Subudhi and Jena, 2009). A series of applications recommend it as an efficient tool, particularly for highly non-linear objective functions. For instance, Lahiri and Ganta (2009) developed a method which incorporates a hybrid ANN and DE technique for the ANN parameter tuning. The algorithm was applied for the prediction of the hold up of the solid liquid slurry flow. The oxygen mass transfer in the presence of oxygen vectors was modelled using a feed forward multilayer perceptron neural network with parameters optimized using two DE-based versions: classical and self-adaptive (Dragoi et al., 2011). In combination with neural networks, a modified DE version, including two initialization strategies (normal distribution and normal distribution combined with the opposition-based principle) and a modified mutation, was applied for modelling the oxygen transfer when n-dodecane is added in aerobic fermentation systems of bacteria (Dragoi et al., 2013a). The pharmaceutical freeze drying process was studied from multiple points of

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view (modelling and system identification) using a hybrid combination of DE with ANNs and back-propagation as a local search procedure (Dragoi et al., 2012a; Dragoi et al., 2013b). The classification of some organic compounds based on their liquid crystalline property was performed using ANNs optimized with two different self-adaptive versions of the DE algorithm (Dragoi et al., 2012b).

Another optimization tool used in this work is the clonal selection (CS) algorithm, which belongs to the artificial immune system (AIS) class. AIS is a group of computational methods represented by highly abstract models of biological immune systems (Castro and Timmis, 2003). The main motivation of using immune systems as a source of inspiration for computational systems resides in their capabilities related to self-evolution, self-organization and self-sustainability (Ahmad and Narayanan 2011). In addition, unlike other biological systems such as the nervous system, the immune system is not centrally controlled and therefore detection and response can be locally executed (Dasgupta and Nino, 2009). Related to the combination of AISs with ANNs for chemical engineering processes, to the authors' knowledge, only a few studies can be found (Tao et al., 2012). For instance, different variations CS-ANN were applied by our group for the removal of heavy metals from residual water (Dragoi et al., 2012) and for the optimization of CO_2 absorption in pneumatic contractors (Cozma et al., 2013).

In this paper, ANNs and SVMs are employed as acceptable alternatives to the phenomenological models, which are difficult to develop and solve with satisfactory accuracy for a complex polymerization process. The goal is to perform a comparative analysis of different approaches and to identify the best method that generates simple but highly efficient models. The modelling methodologies include ANNs and SVMs, optimized with techniques such as DE, CS or grid search. One must emphasize the benefits of the hybrid modelling techniques in terms of accuracy and in connection with the particularities of the process. The novelty of this approach lies in several aspects: the application of different modelling techniques for a complex chemical process, new elements introduced in the DE optimizer and a novel combination DE-SVM.

2 DATABASE

The case study of our research work is the free radical polymerization of styrene performed through batch suspension technique. A complete mathematical model previously elaborated and solved (Curteanu, 2003) is used here as a simulator for producing the working database. The model contains the balance equations for the monomer conversion, initiator concentration, distribution moments of radicals and dead polymer and also, equations that take into account diffusion constraints (gel and glass effects). This last part is difficult to model with satisfactory accuracy; therefore, input-output data models are recommended alternatives to be applied.

Data quality and quantity are essential for modelling with machine learning techniques. In this case, the collected data was chosen to cover the whole domain of interest for the studied process and to be uniformly distributed within this domain. Thus, for the initiator concentration and temperature, the ranges specific to the suspension polymerization of styrene were 10-55 mol/l (variation step 5) and 60-90 °C (variation step 10), respectively. Regarding the reaction time, the interval was 0 to 2000 minutes, because for lower concentrations of initiator and lower temperature, the reaction time is longer (Curteanu et al., 2010).

After the data was generated, an internal step of data pre-processing was applied. This included normalization, randomization and splitting the data into training and testing subsets. The normalization was achieved using the 0-1 method:

$$x_{norm} = \frac{x - x_{min}}{x_{max} - x_{min}} \quad (1)$$

Concerning data randomization, it was applied in such a manner that all points from a single initiator-temperature combination belong to either the training or the testing set. In this way a separation between experiments is maintained and the testing of the model is not based on individual points but on an entire experiment. The amount of training data is 80%, with the remaining 20% used for testing.

For the styrene polymerization process considered here, the model input variables were chosen as: initiator concentration, I_0, temperature, T and reaction time, t. The other two variables, monomer conversion, x, and numerical average molecular weight, Mn, represent the outputs of the models. The modelling techniques aim to provide predictions about the main properties (molecular mass) and reaction characteristics (conversion) as a function of the working conditions.

3 MODELLING METHODOLOGY

Two modelling approaches, ANN and SVM, were applied to solve this real-world chemical engineering problem. Since both ANN and SVM have some parameters that need to be tuned in order to obtain optimal results, two bio-inspired algorithms, DE and CS, were applied and compared for model optimization.

3.1 Optimizing Neural Networks with Differential Evolution

DE, an algorithm based on the evolutionary paradigm, is used to simultaneously perform parametric and structural optimization of the neural network model for the styrene polymerization. The variant used in this work, called SADE-NN-2 (Dragoi et al., 2012a) is a combination of a self-adaptive DE with ANNs and back-propagation (BP). DE has the role of performing a global search, while BP locally improves the best solution found in each generation. This intertwinement of the two algorithms is possible because all DE individuals are in fact ANNs.

As in the case of all evolutionary algorithms, the evolution of the population occurs by applying mutation, recombination and selection steps until a stopping criterion is met. Initially, a set of potential solutions are generated using a random approach. In this work, a Gaussian distribution is used. After that, mutation has the role to add diversity and the population of mutants is combined in the crossover step with the current one to create a trial population. In the DE case, two types of crossover can be encountered: binomial (each characteristic of the trial individual is randomly copied from one of the two parents) or exponential (blocks of characteristics are inherited alternatively from the parents). In SADE-NN-2, the binomial version is used, as it was observed that efficiency is increased only for a small number of case studies when the exponential version is employed.

Concerning the selection step, where the trial individual competes with the current individual for the right to participate in the next generation, a tournament version with a "one-to-one" survival criterion is used.

The characteristic feature of DE when compared with other EAs is the mutation step in which a trial vector is generated by adding to a base individual a scaled differential term (Price et al., 2005). In SADE-NN-2, this mutation principle is modified, such that the individuals participating to the differential terms are sorted based on their fitness. Since it was observed that in various situations the DE version called DE/Best/2/Bin (where the base vector is represented by the best individual in the population, 2 differential terms are employed and the crossover type is binomial) obtains acceptable results, this version was considered as a base for the current study.

As the SADE-NN-2 is a self-adaptive method (in which the F and Cr control parameters are included into the optimization procedure, i.e. they evolve simultaneously with the individuals) a separate procedure for parameter optimization was not required.

A direct encoding with real values was chosen for the ANNs, because it is the least expensive computationally. For each position in the population, at each generation, at least one decoding procedure is required. The ANN parameters chosen for encoding are the number of hidden layers, the number of neurons in each hidden layer, the weights, the biases and the activation functions. Unlike the majority of applications where the variation of the activation functions is performed at the layer level, in the SADE-NN-2, the variation is applied at the neuron level.

3.2 Optimizing Neural Networks with Clonal Selection

The second algorithm employed for neural network optimization is clonal selection. It describes the basic characteristics of the immune response when an antigenic stimulus is applied to a vertebrate (Abdul Hamid and Abdul Rahman, 2010).

The main immunological principles used are: a specific memory set, selection and cloning of the best antibodies, removal of the worst antibodies, affinity maturation of the best immune cells and the generation of a diverse set of antibodies (Dasgupta and Nino, 2009). The main steps of the algorithm are initialization, selection, cloning, affinity maturation (the process of variation and selection achieved through hyper-mutation) and receptor editing. The last four steps are repeated until a stopping criterion is met.

As in the DE case, the initialization is based on the Gaussian distribution. In the selection step, the best 30% of the population is cloned 10 times. After that, each clone is hyper-mutated and its affinity (computed using an affinity function similar to the fitness function used in EAs) is determined. The mutated clones with the highest affinity are selected for introduction into the population. In the last step,

5% of the population with the worst affinity is replaced with newly generated individuals.

The CS-NN algorithm (Dragoi et al., 2012c) uses the same type of ANN (feed-forward multilayer perceptron) and the same encoding procedure as in SADE-NN-2. In this manner, the differences obtained between the two approaches are driven only by the optimization procedures (DE or CS) and their effectiveness can be assessed in a meaningful way.

The characteristics of CS-NN which distinguishes it among other CS variants (except its combination with neural networks) are: the introduction of the opposition-based principle in the initialization phase and the introduction of a hyper-mutation combining 3 types of hyper-mutation (Gaussian, non-uniform and pair-wise interchange), based on a random procedure.

3.3 Modelling with Support Vector Machines

One of the main advantage of the SVM is the small number of parameters that the user has to choose: the type of kernel with its parameters and a cost parameter which defines the balance between tolerance for training errors and generalization capability. For our case study, two support vector regression (SVR) models were designed, for each parameters of interest: x and Mn.

The experiments were performed using the implementation provided by the LIBSVM library (Chang and Lin, 2011), using the ε-SVR or the μ-SVR variants. In ε-SVR, ε is a parameter of the loss function with values in the $[0, \infty)$ domain. Also, the radial basis function (RBF) kernel was selected.

3.4 Support Vector Machines and Differential Evolution

The fourth approach for modelling the styrene polymerization process is a novel algorithm combining DE with SVM (DE-SVM). DE acts as an optimizer, while the SVM models the process. Distinctively from the SADE-NN-2, in DE-SVM, DE only performs parameter optimization. The training procedure is the classic one used for SVMs.

The same self-adaptive DE version used in SADE-NN-2 was also employed in DE-SVM. Thus, the performance differences obtained are solely determined by the performance of the model and not by the ability of the optimizer to determine the best solution. If the population was formed of neural models in the case of SADE-NN-2, in the case of DE-SVM, the individuals forming the population are lists of SVM parameters such as: SVM type (μ-SVR, ε-SVR), kernel type (linear, polynomial, RBF and sigmoid), degree (applicable only for polynomial kernel type), γ (a coefficient of polynomial and sigmoid kernels) and C, the cost parameter.

4 RESULTS AND DISCUSSION

After gathering the data describing the process, a series of simulations with the four considered algorithms were performed. In the case of the modelling approaches based on ANNs (SADE-NN-2 and CS-NN), some limitations to the structure of the network were imposed, in order to reduce the complexity of the encoded individuals and, therefore, to reduce the computational effort.

Consequently, for the hidden layers, it was considered that a network with one hidden layer can efficiently model the polymerization process. This restriction is based on the authors' experience: in the majority of our studies a network with one hidden layer provided satisfactory results. Also, it was considered that 30 neurons in the hidden layer are sufficient. A lower limit was imposed as well: the algorithms can generate networks with no hidden layers or with one hidden layer, with a number of neurons between 4 and 30.

Initially, with both SADE-NN-2 and CS-NN, a set of models with two outputs corresponding to the parameters of the process, x and Mn, were generated. Although the mean squared error (MSE) computed on the normalized data had acceptable values: 0.081 and 0.125 in the training phase for SADE-NN-2 and CS-NN, respectively, and 0.122 and 0.158 in the testing phase, the average relative errors (AREs) were not acceptable, exceeding 40% for some of the outputs. Therefore, for each output a separate neural model was created. A set of five best results are listed in Table 1 for SADE-NN-2 and in Table 2 for CS-NN.

As it can be observed from Tables 1 and 2, for the x and Mn parameters, the best error in the testing phase is obtained with CS-NN (CN1 model and CN6 respectively). These observations are also in trend with the average values.

Concerning the SVM models, Table 3 presents the best results obtained with ε-SVR. A grid-search approach was used to find the best values for the model parameters. It is recommended to use exponentially growing sequences of C and γ in order to identify good parameters (Hsu et a1., 2010). The

Table 1: Best results obtained with SADE-NN-2 for each process parameter.

Output variable	Topo-logy	MSE training	MSE testing	Model id
x	3:19:01	0.0128	0.0094	DN1
	3:19:01	0.0105	0.0104	DN2
	3:11:01	0.0147	0.0132	DN3
	3:04:01	0.0136	0.014	DN4
	3:19:01	0.0109	0.077	DN5
Average		0.0125	0.0248	
Mn	3:19:01	0.0039	0.0036	DN6
	3:11:01	0.0058	0.0048	DN7
	3:11:01	0.0045	0.0051	DN8
	3:14:01	0.0047	0.0073	DN9
	3:11:01	0.0118	0.0125	DN10
Average		0.0047	0.0045	

Table 2: Best results obtained with CS-NN for each process parameter.

Output variable	Topo-logy	MSE training	MSE testing	Model id
x	3:20:01	0.0069	0.0083	CN1
	3:19:01	0.0094	0.0103	CN2
	3:17:01	0.0091	0.0115	CN3
	3:11:01	0.0127	0.0128	CN4
	3:16:01	0.0099	0.0159	CN5
Average		0.0096	0.0117	
Mn	3:08:01	0.0009	0.0016	CN6
	3:10:01	0.0012	0.0017	CN7
	3:12:01	0.0327	0.0031	CN8
	3:16:01	0.003	0.0041	CN9
	3:20:01	0.0051	0.0064	CN10
Average		0.0085	0.0033	

chosen values for the experiments are (2^{-3}, 2^{10}) for C, and (2^{-3}, 2^{3}) for γ, with step 0.5. Low values for MSE show the good performance obtained with RBF kernel for both output variables, while the polynomial kernel performance is not suited for modelling the *Mn* variable.

For the combination of DE with SVM, the best models obtained for the process parameters are presented in Table 4.

In order to determine the efficiency of the best models, the coefficient of determination (r^2) was also computed (Table 5).

By analyzing the results it can be observed that in some cases, the coefficient of determination is not closely correlated to the MSE. This fact can be explained by the data distribution, since the system has a different dynamic for each combination of temperature-initiator.

In order to visualize the differences between the predicted and expected data, for the modelling of the

Table 3: Results obtained with ε-SVR for the output variables.

Output variable	Method parameters	MSE training	MSE testing	Model id
x	C = 256; γ = 2; RBF kernel; ε = 0.1	0.004	0.004	S1
	C=0.707; γ = 0.125; Polynomial kernel degree 2; ε = 0.25	0.021	0.02	S2
	C=0.353; γ = 0.125; Polynomial kernel degree 3; ε = 0.25	0.022	0.02	S3
Mn	C = 512; γ = 2; RBF kernel, ε = 0.1	0.09	0.27	S4
	C=2.828; γ = 0.125; Polynomial kernel degree 2; ε = 3	7.5	2.4	S5
	C=2.828; γ = 0.125; Polynomial kernel degree 3; ε = 3	7.5	2.4	S6

Table 4: Results obtained with DE-SVM for the output variables.

Para meter	Method parameters	MSE training	MSE testing
x	μ-SVM, RBF kernel C = 0.28;γ = 3.341	0.0085	0.0075
Mn	μ-SVM, RBF kernel C = 5.96; γ = 2.212	0.0010	0.0014

Table 5: The performance of the best models obtained with the four methods.

Para-meter	Model	r^2 training	r^2 testing
x	DN1	0.9617	0.9580
	CN1	0.9768	0.9581
	S1	0.96	0.9304
	DE-SVM	0.9714	0.9656
Mn	DN6	0.9871	0.9800
	CN6	0.9915	0.9731
	S4	0.99	0.9813
	DE-SVM	0.9936	0.9767

x parameter, a set of figures with testing data were generated. Since different combinations of temperature and initial value of the initiator were tested, two significant examples are given below: temperature of 368K and 10 mol/l of BPO initiator (Figure 1) and temperature of 338K and 50 mol/l of initiator (Figure 2).

Concerning the *Mn* modelling, the DE-SVM approach is the best in terms of MSE testing. Similar to the *x* parameter, a series of figures for two temperature-initiator value combinations were generated for it (Figures 3 and 4).

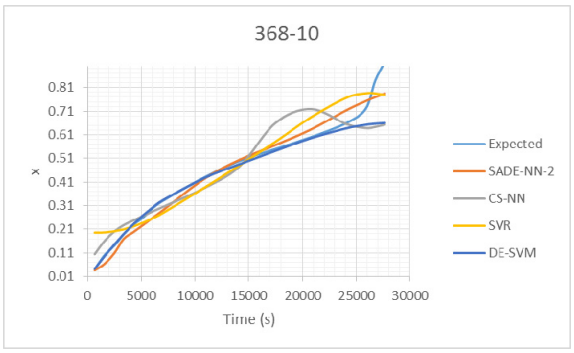

Figure 1: Comparison between the predictions of *x* obtained with the four methods and the expected data when the process parameters are 368K (temperature) and 10 mol/l (initial value of initiator).

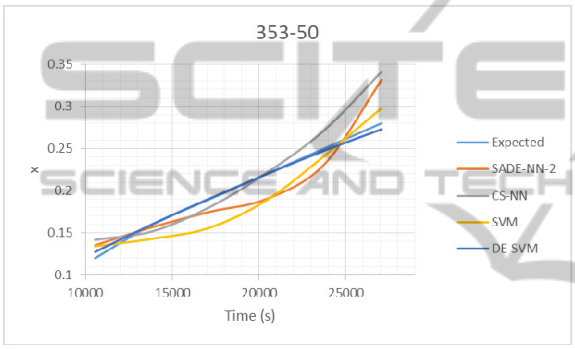

Figure 2: Comparison between the predictions of *x* obtained with the four methods and the expected data when the process parameters are 353K (temperature) and 50 mol/l (initial value of initiator).

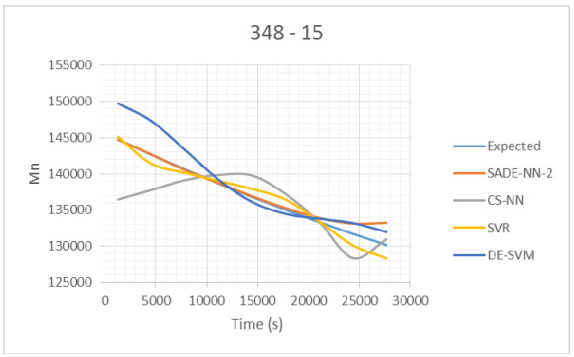

Figure 3: Comparison between the predictions of *Mn* obtained with the four methods and the expected data when the process parameters are 348K (temperature) and 15mol/l (initial value of initiator).

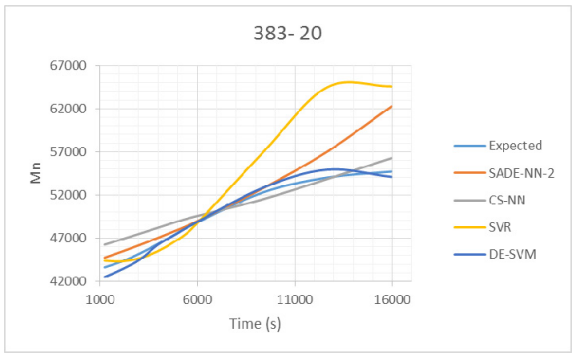

Figure 4: Comparison between the predictions of *Mn* obtained with the four methods and the expected data when the process parameters are 383K (temperature) and 20 mol/l (initial value of initiator).

4 CONCLUSIONS

Four modelling methodologies were developed and tested on a complex chemical process, i.e. free radical polymerization of styrene. They include ANN and SVM as models, structurally and parametrically optimized with DE and CS. Although both neural network and support vector machine models are found suitable for the polymerization process, selecting one of the techniques rely on the user experience. However, it must be mentioned that the combination SVM-DE deserves special attention due to its accessibility and accuracy observed in the results.

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