Application of Artificial Neural Networks to Combinatorial Catalysis: Modeling and Predicting ODHE Catalysts

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This paper shows how artificial neural networks are useful for modeling catalytic data from combinatorial catalysis and for predicting new potential catalyst compositions for the oxidative dehydrogenation of ethane (ODHE). The training and testing sets of data used for the neural network studies were obtained by means of a combinatorial approach search, which employs an evolutionary optimization strategy. Input and output variables of the neural network include the molar composition of thirteen different elements presented in the catalyst and five catalytic performances (C_2H_6 and O_2 conversion, C_2H_4 yield, and C_2H_4, CO_2, and CO selectivity). The fitting results indicate that neural networks can be useful in high-dimensional data management within combinatorial catalysis search procedures, since neural networks allow the ab inito evaluation of the reactivity of multicomponent catalysts.

KEYWORDS:

combinatorial catalysis · combinatorial chemistry · heterogeneous catalysis · neural networks · ODHE processes

The new development of high-throughput experimentation (HTE) techniques in the framework of heterogeneous catalysis is exponentially increasing the number of catalytic data, derived from the parallel synthesis, characterization, and catalytic testing. Nevertheless, the rapid progress of those accelerated experimental tools[1–3] requires the development of more powerful data management techniques adapted to this specific research methodology. These software techniques include a) the efficient administration and scheduling of large amounts of experimental data, b) the comprehension and modeling of the organized data, and c) a global search strategy for optimizing the catalytic performance.

Combinatorial catalysis can be defined as a methodology in which a large number of new materials are synthesized and tested for a specific catalytic performance in a parallel way. A generalized approach employs a genetic algorithm (GA) as a search strategy in order to optimize the catalyst composition. This optimizing strategy (GA) is quite appropriate for heterogeneous catalysis since a) GA tolerates noisy data (experimental data) with considerable error, b) GA uses a population of points to conduct the search, which fits quite well with the application of HTE techniques, and c) the goal is to find an approximate global maximum in a high-dimensional space, minimizing the number of trials.

In contrast with the conventional research method,[4] whereby scientists process all the experimental data and guide the research process, these novel accelerated methods are applying different data mining techniques in order to find relationships and patterns between the input (catalyst composition, preparation, and reaction conditions) and output data (characterization data and catalytic performance). Data mining does not need any starting hypotheses, since it automatically discovers hidden relationships and patterns as its ultimate goal is to discover knowledge from large quantities of raw data.

In this sense, data mining techniques[5] have been applied to combinatorial (bio-)chemistry[6–9] and could, in principle, be suitable for combinatorial heterogeneous catalysis. Hence, artificial intelligence (AI) techniques have an important potential for modeling and predicting complex high-dimensional data. Expert systems have been applied, but most of those methodologies would require much time to acquire the knowledge, which constitutes the expert system, and this knowledge may not be universal.[10]

Recently, artificial neural networks (NN) have been successfully applied to conventional catalytic modeling and the design of solid catalysts. These applications[11] include the design of catalysts for the ammoxidation of propylene[12] and methane oxidative decoupling,[13] and the analysis and prediction of the results of the composition of NO over zeolite Cu/ZSM-5.[14] In these studies (see, for instance, Huang et al.[13]), neural networks were applied to catalytic reactions, but the samples used for modeling with artificial NN were based on human knowledge

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and the number of catalyst variables was much reduced. In the present work, the catalysts and catalytic results were all provided by a combinatorial evolutionary method, in which thirteen elements have been combined for catalyst formulation (input) and six results (output) are modeled. The practical application of the methodology has been done on the oxidative dehydrogenation of ethane (ODHE) reaction. The necessary experimental data for the training and testing of neural networks were derived from a combinatorial approach search, which employs an evolutionary optimization strategy (GA).[15–17] The theoretical total number of combinations is very high and only a reduced number (about $2 \cdot 10^{-22}$ %) of samples were used to train and test the neural network.

A promising tool for the combinatorial approach applying search algorithms is the use of NN within the search procedure in order to understand the prior information generated during the experimental cycles. The use of NN predictions could be employed as a theoretical pre-screening, enabling us to skip the test of statistically poor active materials. However, it should be taken into account that data with singularities or discontinuous points could be more problematic for modeling by NN. This could be an important limitation of this approach, since different catalytic systems present this type of unexpected behavior.

Experimental Section

Catalyst Formulation: A pool of thirteen elements (Ga, Cu, P, Mn, Mo, W, Sn, Cr, Co, Zr, Ca, La, Au) were selected for the evolutionary[16, 17] approach from fundamental knowledge, including elements which are believed to play different roles, such as a) redox properties (removable lattice oxygen), b) activation by adsorbed oxygen, and c) activation by lattice oxygen. To discover a new class of active materials, elements with a known activity, such as V, Nb, and Mg, were replaced by elements and their mixtures, which have not been tested before.

The evolutionary approach (GA) was initialized by generating 64 catalyst compositions, random combinations of three to four elements from the pool of thirteen elements previously selected. Successive generations were set up by the genetic algorithm, taking into account the catalytic performances of the previous generation, applying crossover and mutation operators. Further details concerning the genetic algorithm employed (the objective function, mutation and crossover operators, the selected techniques, and encoding mode) can be found elsewhere.[16, 17]

The total data available for the present work were derived from seven successive generations.

The total combinatory to be explored by the evolutionary approach comes from combining thirteen elements in 1000 possible loadings, yielding[*] 10^{39} combinations. However, until now, during the evolving process the maximum number of elements in a single catalyst is seven. Therefore, the combinatory is reduced[*] to 1.72×10^{24}.

Catalytic Performance: Resulting data from catalytic testing is expressed in the form of a) conversion of ethane and oxygen, b) yield of ethylene, and c) selectivity of ethylene, CO_2, and CO.

Catalytic testing was carried out[16] by means of a 64-reactor assembly, operating under fixed reaction conditions: 500 °C, $C_2H_6/O_2/Ar = 20/10/70$, and $\tau = 0.4\ g\ s^{-1}\ mL^{-1}$.

Computer-Aided Catalyst Modeling

Neural Network Description

Artificial neural networks consist of a number of simple interconnected processing units, also called neurons, which are analogous to biological neurons. Thus, artificial neural networks are composed of several neurons, which can be organized in different ways, defining what is termed topology, or neural architecture. Each different structure could be appropriate in a different way for the problem under consideration.[18]

The basic unit of a neural network is the neuron, or node, composed of:

- a *set of connections or inputs*, $x_j(t)$, each of which is characterized by a *synaptic weight* w_{ij}, that represents the intensity of interactions between each neuron j of a previous layer and the actual neuron i;
- a *propagation rule*, which determines the effective input of neuron k from all individual inputs to this neuron;
- an *activation function* F_k, that determines the output y_k of neuron k by means of its level of excitation.[19]

Two important features of neural networks are the ability to supply fast answers to a problem and the capability of generalizing their answers, providing acceptable results for unknown patterns. In this way, they need to learn about the problem under study and this learning is commonly named the *training*

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[*] *Members of the Editorial Advisory Board will be introduced to the readers with their first manuscript.*

[*] *Calculated from* $N = \frac{n_E!}{n_R!(n_E - n_R)!} n_\Delta^{n_R}$, *where* n_E *is the total number of elements,* n_R *is the number of elements per sample, and* n_Δ *is the number of intervals of possible compositions of each element over a support.*

process. During this training process, neural networks are supplied with a set of samples belonging to the problem domain. Then they establish mathematical correlations between the samples.[20] A large quantity of information and time are required for analysis and processing.

One of the most well-known structures of neuronal networks for supervised learning is the *multilayer perceptron* (Figure 1), which is generally used for classification and prediction problems. In the multilayer perceptron, neurons are grouped into

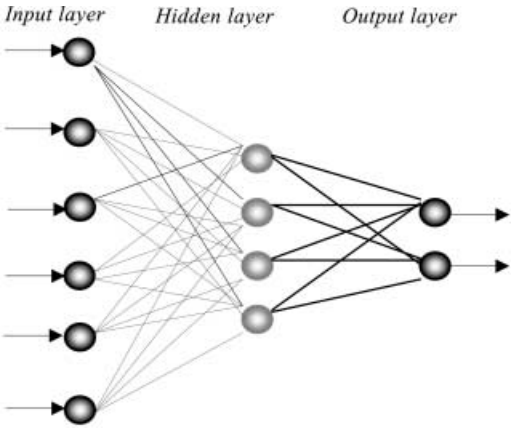

Figure 1. *Example of a multilayer perceptron.*

layers, so that each input of a neuron is composed of the outputs from the neurons of the previous layer. However, the neurons in the input layer have as input all the input data from the problem. Therefore, the number of nodes at the input and output layers are determined by the problem's features.

Neural Networks Applied to ODHE Catalyst Modeling

In the evolutionary approach,[16, 17] all catalysts must be tested empirically in order to obtain their catalytic performances. Then, the fitness function of the genetic algorithm can be calculated. The evolutionary approach designs the composition of the next catalysts set to be synthesized and tested, taking into account the composition and activity of the material already processed.

By using neural networks for predicting catalytic activities, it would be possible to make a statistical prescreening of the new GA-designed catalysts set, thus reducing the number of catalysts to be empirically analyzed.

As more samples (catalysts) could be passed to the neural network, better predictions could be obtained. However, in the problem under study, the number of available samples is small, as their generation implies a long and expensive process. Therefore, it would be interesting to analyze in the first term, if neural networks could really be used to predict catalyst behavior.

3.2.1. Network Topology Optimization and Training Process

A general rule for selecting the most suitable topology of a neural network for a given problem does not exist. Therefore, it is useful to analyze different topologies and different learning algorithms in order to find the best adapted to the problem under study.

A study of the art was carried out to determine which neural network topologies, such as the Kohonen, radial basis function, or multilayer perceptron networks, had been employed in similar prediction problems. Kohonen and other radial basis functions seem to be inappropriate for the problem under study, so we decided to adopt a multilayer perceptron, which was also used by Huang et al. and Sasaki et al.[13, 14] This type of topology requires supervised learning.

Supervised learning implies providing the neural network with training samples (input data) and desired results (output data).[21, 22] In each training step, the neural network adjusts its neuron weights, minimizing the error between the output calculated by the neuron and the desired values. The application of neural networks could be considered as a black box process (Figure 2), having all the relevant attributes of the catalyst as inputs (catalyst composition) and experimental testing results as outputs (catalytic performances).[23]

A set of 329 catalysts for the ODHE reaction was available for the neural network study, obtained from seven successive generations. Input data were molar percentages of each element in the active phase of each catalyst, whereas output data were the conversions X [%], yields Y [%], and selectivity S [%]: X(Ethane), $X(O_2)$, S(Ethylene), $S(CO_2)$, S(CO), and Y(Ethylene). All the calculations were made on a 500 MHz Pentium processor, using the SNNS neural networks simulator, developed by the Institute for Parallel and Distributed High Performance Systems at the University of Stuttgart.[24]

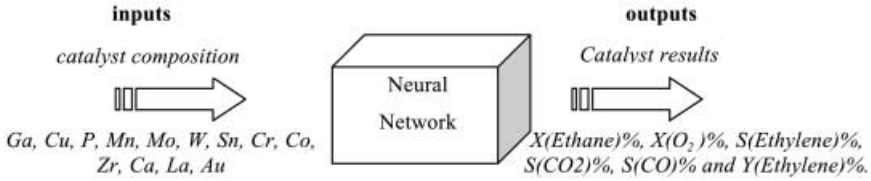

Figure 2. *Scheme of the use of a neural network for the problem under study.*

Concerning the data required for the study of the behavior of different neural network topologies, a set of 50 catalysts corresponding to the ten best catalysts from the first five generations of the evolutionary method[17] was chosen. It was decided to employ this small quantity of catalysts because the combination of neural networks and the evolutionary approach starts from a small set of samples.

Data were divided into training samples and testing samples. Usually in research, 80 – 90 % of the available samples is used for training and the remaining 20 – 10 % is used for testing or validating the proper prediction behavior of the neural network. Therefore, neural networks were trained using 40 catalysts and tested with the 10 remaining catalysts.

Using supervised learning, an incremental method was applied, testing different neural network topologies based on the multilayer perceptron. Starting with one single layer and few neurons, the topology was modified by increasing the number of neurons and the number of hidden layers. The inputs of the network were the catalyst composition variables and the outputs were the catalytic results. Different experiments were also carried out with those algorithms that turn out to be more suitable for the multilayer perceptron according to the literature.[20, 24, 25] Specifically, neural networks were trained with backpropagation and backpropagation with momentum, with different parameters (the learning factor α and the momentum μ terms).

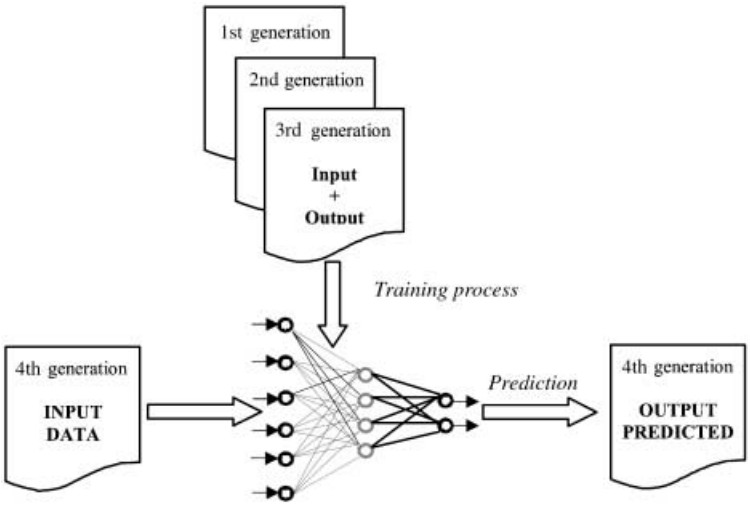

Figure 3. *Methodology applied to the study of training sample influence: Neural network training with previous generations for predicting actual generation.*

Combination of Neural Networks and Genetic Algorithms

Once a suitable neural network for the prediction of catalytic results was established, its combination with genetic algorithms was analyzed. This study also allowed the analysis of the influence of the amount of training data in the prediction performance. In this way, neural networks were trained using sets of catalysts from the previous generations given by the evolutionary approach and tested with some catalysts from the present generation. This methodology emulates the run of a combinatorial search procedure but, in this case, the experimental process (preparation and reaction assay) was replaced by the neural network.

For example, in one of the training processes, neural networks were trained with the first, second, and third generations and tested with some catalysts from the fourth generation (Figure 3). In the testing, the composition of the fourth generation catalysts was passed to the neural network, which predicted the catalytic performances of this fourth generation, based on the learned knowledge of first, second, and third generations. Next, the neural network was retrained with the catalyst compositions (input data) and experimental catalytic activities (output data) of this fourth generation.

Results and Discussion

As a result of the first study with 50 catalysts, a suitable neural network from those analyzed turned out to be a multilayer perceptron with 13 nodes in the input layer (corresponding to the percentage molar concentrations of each element of the catalyst active phase), 26 nodes in the first hidden layer, 12 nodes in the second hidden layer, and 6 nodes in the output layer (corresponding to X(Ethane), $X(O_2)$, S(Ethylene), $S(CO_2)$, S(CO), and Y(Ethylene)). In Tables 1 and 2, the mean error of the six

Table 1. *Mean square prediction error [%] and mean absolute error [%] for different neural network topologies and different training algorithms for training samples.*

					Training data (40 samples) Learning process											
					Backpropagation			Backpropagation Momentum								
				α:	0.2	0.5	0.8	0.2	0.2	0.2	0.5	0.5	0.5	0.8	0.8	0.8
				μ:				0.2	0.5	0.8	0.2	0.5	0.8	0.2	0.5	0.8
Neural network topology																
input layer	1st hidden layer	2nd hidden layer	output layer	weights												
					Mean square error											
13	8		6	152	98.43	59.20	48.54	88.30	65.65	48.25	54.16	43.80	27.85	44.99	34.43	26.80
13	20		6	380	104.81	64.92	44.23	99.51	71.66	34.89	54.54	33.82	11.20	35.63	21.87	8.09
13	40		6	760	101.14	63.27	38.07	96.06	70.24	26.86	50.29	27.08	11.40	27.62	14.25	9.01
13	8	6	6	188	227.88	125.87	115.83	144.71	131.26	123.74	122.10	117.97	117.04	114.58	112.20	113.03
13	26	12	6	722	87.56	38.87	22.00	78.13	51.64	15.64	28.60	15.74	12.13	15.32	10.72	8.10
13	40	20	6	1440	74.59	33.01	22.52	62.55	39.90	14.52	25.31	16.29	7.94	16.12	8.81	8.64
					Mean absolute error											
13	8		6	152	7.14	5.48	4.88	6.82	5.90	4.79	5.17	4.58	3.57	4.64	4.08	3.43
13	20		6	380	7.23	5.80	4.64	7.02	6.16	4.07	5.25	4.00	2.45	4.19	3.30	2.03
13	40		6	760	7.03	5.78	4.42	6.82	6.03	3.78	5.12	3.79	2.49	3.89	2.79	2.16
13	8	6	6	188	10.43	7.89	7.68	8.33	7.98	7.85	7.82	7.70	7.71	7.65	7.60	7.66
13	26	12	6	722	6.76	4.43	3.34	6.29	5.07	2.87	3.75	2.90	2.59	2.83	2.35	2.08
13	40	20	6	1440	6.24	4.18	3.69	5.61	4.47	2.90	3.80	3.05	1.99	3.03	2.07	2.04

Table 2. *Mean square prediction error [%] and mean absolute error [%] for different neural network topologies and different training algorithms for test samples.*

					Testing data (40 samples) Learning process											
					Backpropagation			Backpropagation Momentum								
				α:	0.2	0.5	0.8	0.2	0.2	0.2	0.5	0.5	0.5	0.8	0.8	0.8
				μ:				0.2	0.5	0.8	0.2	0.5	0.8	0.2	0.5	0.8
Neural network topology																
input layer	1st hidden layer	2nd hidden layer	output layer	weights												
					Mean square error											
13	8		6	152	177.2	252.9	224.0	184.0	224.7	237.1	255.4	216.1	218.7	216.1	241.3	180.2
13	20		6	380	178.9	208.1	225.2	183.7	188.3	249.6	226.8	233.5	297.2	230.8	260.8	310.3
13	40		6	760	188.0	219.1	260.1	193.8	202.7	285.3	237.6	286.0	302.1	287.5	301.3	313.2
13	8	6	6	188	238.9	189.2	198.8	163.3	185.3	216.3	198.9	211.6	213.0	205.8	209.2	213.9
13	26	12	6	722	154.4	211.2	334.6	153.4	172.5	328.2	304.2	321.7	387.9	316.3	330.1	401.0
13	40	20	6	1440	184.8	235.7	300.2	213.4	234.4	361.8	281.8	334.6	328.3	344.6	308.7	338.4
					Mean absolute error											
13	8		6	152	8.71	10.53	10.46	9.00	10.02	10.61	10.79	10.38	10.03	10.32	10.61	8.51
13	20		6	380	8.75	9.77	9.90	8.86	9.45	10.37	9.87	10.23	11.75	10.16	11.20	11.82
13	40		6	760	9.05	10.07	10.81	9.22	9.80	11.47	10.32	11.46	11.50	11.39	11.36	12.05
13	8	6	6	188	11.31	9.05	9.60	8.39	8.89	9.63	9.31	9.80	9.76	9.73	9.92	9.87
13	26	12	6	722	8.48	10.08	11.13	8.53	9.48	11.34	11.19	11.05	13.07	10.98	11.91	13.56
13	40	20	6	1440	9.40	10.91	11.13	10.26	11.04	12.09	11.33	11.93	12.22	11.92	11.28	12.73

outputs of some studied NN topologies with different training algorithms is shown. Table 1 displays the mean square errors (MSE) and mean absolute errors of the training data (40 samples) for different NN trained with backpropagation with and without the momentum term. These errors indicate the fitting rate of the NN to the model. Table 2 shows the MSE and mean absolute errors of the test data (ten samples), which indicates the generalization rate of the NN (capability to predict unknown catalysts).

The number of data used for training an NN should be at least as large as the number of weights, although this rule is not always followed.[26] In the present study, those NNs with a higher number of nodes (such as the NN selected above) provide lower errors in both the training and test samples. Moreover, backpropagation without momentum fits the test samples better then when using the momentum term. The differences are not very significant, though. The selection of the most suitable NN topology for this application in catalysis was also influenced by the way in which predicted values follow the experimental trends. The NN selected above provided in both studies predicted values that are more adjusted to the graphical curves of the experimental ones.

Figure 4 represents the prediction results of the ten catalysts tested using the neural network topology explained above. The experimental values of oxygen conversion and ethylene yield obtained for each catalyst, the values predicted by the neural network, and a *predicted area* established are shown. As can be seen in Figure 4, the predicted values are close to experimental ones (taking into account the methodology in catalysis research and experimental errors) and several experimental results are inside the *predicted area* established for the neural network.

This *predicted area* determines the region for the NN in which the probability of finding the experimental values of the catalyst performances is higher. The lower limit (LL) and the upper limit (UL) of the above interval were calculated as given in Equations (1) and (2), where $X(i)$ is the prediction made by the neural network for sample i and ε is the average of the absolute error made by the neural network for each one of the forty catalysts used in the training process.

$$\mathrm{LL}(i) = X(i) - \varepsilon \quad (1)$$

$$\mathrm{UL}(i) = X(i) + \varepsilon \quad (2)$$

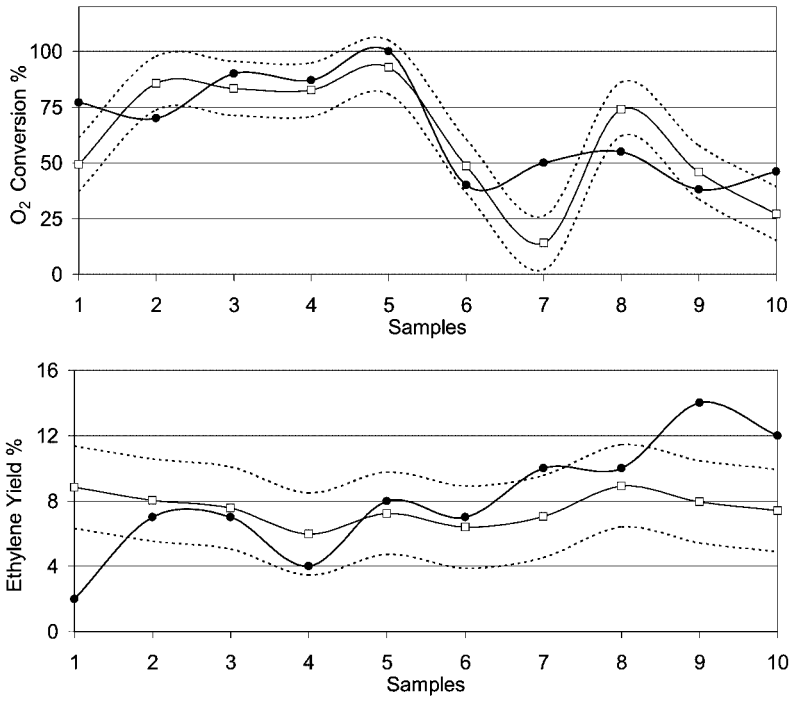

Figure 4. *Experimental (●) and predicted (□) results of O_2 conversion and ethylene yield (40 samples for training data, 10 samples for testing data). Dotted lines represent the predicted area.*

From these results, it is noticeable that the prediction performance is quite good, since the neural network is able to follow the experimental trends. Here, the tiny percentage of available training samples with respect to the huge combinations space should be pointed out.

The results from the second study (combination of the evolutionary approach and the neural network) can be observed in Figure 5. This Figure represents the experimental and predicted data (oxygen and ethane conversion) for each generation when the neural network was trained (and retrained) with all the previous successive generations. In Table 3, the mean absolute errors of predictions for each generation are shown.

As a first relevant aspect, an improvement on the quality of the predictions can be noticed, namely the neural network gains more knowledge as more steps in the search combinatorial process are carried out. This knowledge makes possible the

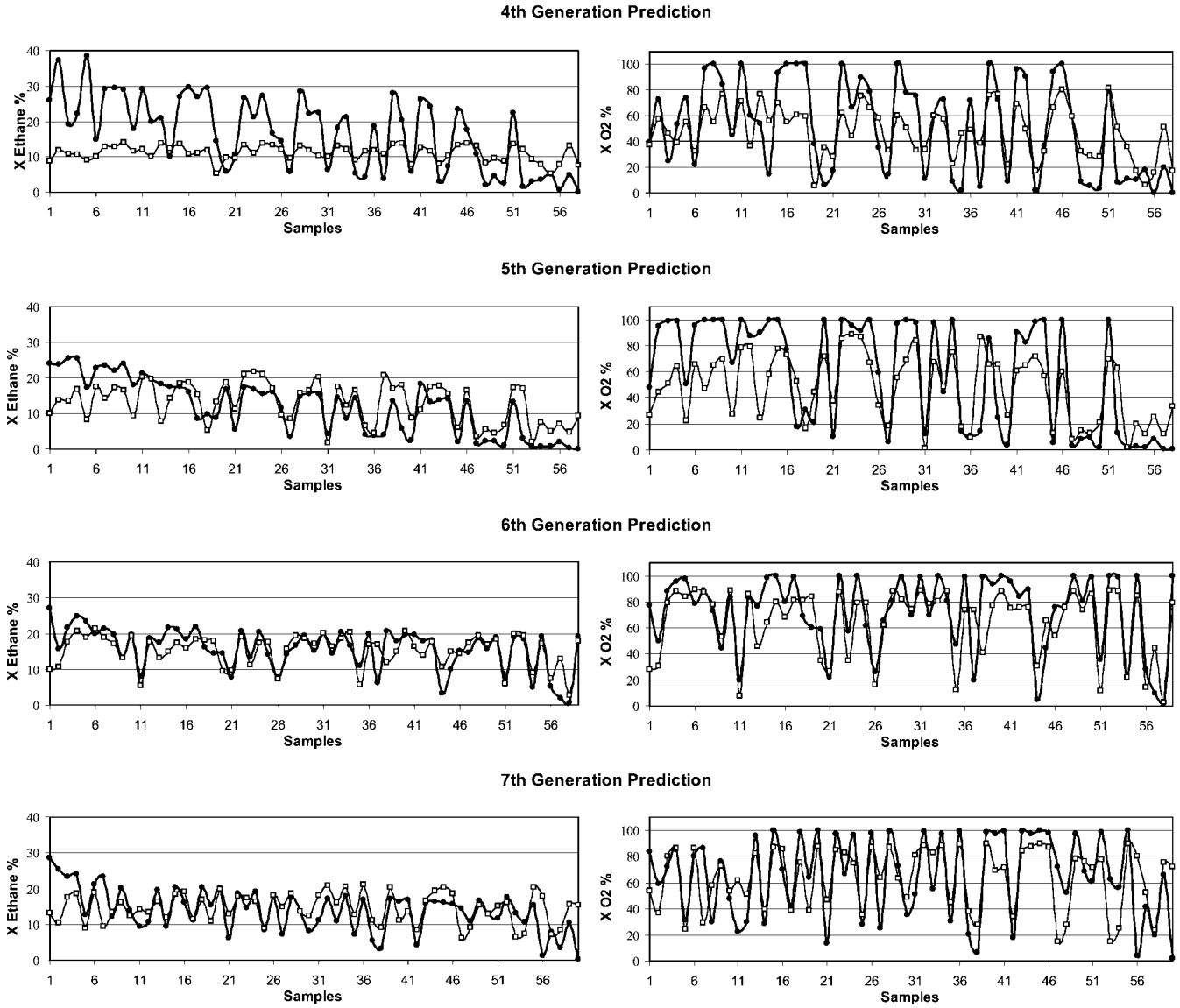

Figure 5. *Evolution of prediction performances with the number of training data within a combinatorial procedure: Experimental (●) and predicted (□) results of O_2 and ethane conversion for the different generations. (See Figure 3 for the methodology followed).*

Table 3. *Mean absolute errors of predictions made by the neural network for each genetic algorithm generation.*

	Absolute error						
Predictions	*X*(Ethane)	*X*(O_2)	*S*(Ethylene)	*S*(CO_2)	*S*(CO)	*Y*(Ethylene)	Mean
4th generation	8.54	22.52	16.48	20.85	8.07	2.51	13.16
5th generation	6.47	21.14	16.30	23.78	12.28	2.46	13.74
6th generation	3.47	17.78	9.43	14.35	5.44	1.71	8.70
7th generation	4.58	21.15	14.60	16.45	8.31	2.52	11.27

enhancement of modeling the experimental trends in each step, as can be seen in Figure 5, where the topologies of the predicted and experimental trends are similar. Although even in the prediction of the last generation some poor predictions can be seen, this NN-prediction routine could be good enough for its application in a pre-screening process in the frame of combinatorial catalysis. In fact, these predictions are better in this case than those that one could make by using a fundamental knowledge. Moreover, predicted values could be compared between them, as the absolute error of those predicted values with the corresponding experimental ones might probably be very similar.

Second, data singularities are quite difficult to predict by a neural network, because they do not present the general tendency. In the present problem, although the NN does not properly predict singularities, in many cases it can provide an initial idea of possible points with promising activities.

Finally, it is important to consider that the combinatorial search approach looks for combinations of elements and concentrations which would yield better experimental results. Through the searching process, the combinatorial procedure focuses on specific regions (exploitation) of the whole combinatorial space in which catalyst results are supposed to be more promising. On the other hand, the combinatorial procedure tries to conserve the diversity up to a point, exploring different regions (exploration). As seen up to now, neural networks can be used together with the combinatorial search approach. Neural networks can establish a better correlation between the catalyst composition and catalytic results in the regions where the combinatorial process is focused (exploitation). However, neural networks are limited when generalizing from the samples obtained by exploration.

Conclusions

In this work, the viability of artificial neural networks in the analysis and prediction of catalytic results within a population of catalysts produced by combinatorial techniques, depending on the catalyst elemental composition, has been proven.

The application of artificial neural networks to modeling and prediction in the field of combinatorial catalysis is a new and powerful tool that could accelerate the development and optimization of new catalysts. An important characteristic of these models is that they do not require any theoretical knowledge or human experience during their training process. Fundamental knowledge has only been used in the determination of the data structure (parameters, input, and output data). However, once those parameters are established, neural networks can be trained and used without further previous knowledge.

Furthermore, these artificial intelligence techniques could be conveniently applied to extract general principles from high-dimensional data resulting from catalysis experimentation.

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