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An Optimisation Strategy for the Catalytic Transformation of Bioethanol into Olefins Using Computational Intelligence

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Abstract: This paper presents a strategy for the optimisation of the operational conditions of the catalytic transformation of Bioethanol into Olefins (BTO) process. The variables to optimise are the main operating variables of the process (temperature, space-time and water content in the feed), and the objective function is to maximise the total production of olefins. The proposed strategy is based on evolutionary algorithms guided by surrogate models used to simulate the process behaviour under different experimental conditions. This paper compares the optimisation results of the BTO process obtained using an existing mechanistic model with those obtained with a surrogate model. The results suggest that the proposed methodology achieves similar results than those using mechanistic models but 43 times faster. This is a preliminary study where only constant set points have been tested; further research will include dynamic optimisation of the operational conditions by testing expected dynamic trajectories for each operating variable.

1 Introduction

Nowadays, we are becoming aware that crude oil is a finite source of energy and raw material. Therefore, our society begins to impulse the sustainable development using alternative sources of energy and raw materials, such as coal or biomass. In nature, being 170 billions tones of biomass annually produced, only the 3-4 % is exploited. Thus, there is a huge quantity of biomass available for its valorization as raw material to obtain biofuels and other chemical products [6]. Consequently, the scientific developments in this field are very important in order to advance in a future post-petroleum society and to reduce our dependency from the petroleum and its derivatives. The development of new tools to study the optimal operation of biomass transformation processes for a future scaling up to industrial level is a new interesting research line.

An important biomass transformation process is the Bioethanol-To-Olefins (BTO) process. The use of biomass as raw material has a great interest as an alternative to the petrochemistry for the production of light olefins like ethylene and propylene. The use of the computational intelligence in this research field can improve the optimisation procedures and allow faster developments in the design and optimal operation of the production processes.

The optimal control laws of biorefinery production processes are mainly unknown. Therefore, it is necessary to test several operational conditions over the whole operational range to study the influence of each manipulated variable on the final production objectives.

One of the key points for the implementation of the BTO process is to perform an advanced control strategy by adjusting the operating variables to maintain product quality while extending the lifespan of the catalyst. Due to the influence of multiple variables simultaneously over the reaction kinetics and the catalyst deactivation, it is necessary to develop advanced optimisation strategies of the operational conditions that guarantee specific production objectives without exceeding the operation limits to avoid an irreversible deactivation of the catalyst.

Therefore, the search of the parameters and operational conditions that allow to reach those production objectives give rise to optimisation problems in which the calculation of an analytic solution can present some difficulties with conventional search techniques [19]. While these techniques require characteristics of the process or from the optimisation problem, such as gradients, Hessians or linearities, to calculate the next points; there are stochastic search techniques as the Evolutionary Algorithms (EA) that solve the optimisation problems only with stochastic rules.

In the field of chemical engineering, there are several applications where these algorithms have been employed for the design, optimisation and optimal control of chemical reactors and plants [8, 1], such as in fermentation processes with fed-batch reactors [19] or to optimise the operational conditions of industrial scale reactor [18] or chemical plants [16].

To perform a dynamic optimisation of a process or reactor, one of the problems is that each cost function evaluation may require from minutes to hours of calculation time, and when using EA hundreds of evaluations are generally needed [14]. Therefore, as knowledge models use to be non linear (and hence difficult to be solved, even numerically), surrogate models are commonly used to simulate the real process during its optimisation [13, 10]. Thanks to their characteristics, Artificial Neural Networks (ANN) are being increasingly used as a modelling technique for process simulation using evolutionary optimisation techniques [2, 5].

ANN attempt to mimic the structural principles of the

biologic brains to learn the existing relations inside input-output datasets. They have been able to successfully model any type of complex models and chemical reactors, such as batch reactors [7, 12], laboratory or industrial scale reactors [15] or even catalytic reactions as in the case of the BTO process [11].

The present work has the objective of performing an optimisation of the Bioethanol-To-Olefins (BTO) process in order to maximise the olefins total production while extending the catalyst lifespan. The BTO process, as other biomass transformation processes, uses a specific catalyst to stimulate the formation of a specific product at the reactor output. This catalyst is deactivated with time depending on the operational conditions. Therefore, in the design and optimisation of new catalytic transformation processes, such as the BTO process, there are two aspects to take into account in order to maximise the production of the desired product: the composition of the catalyst and the operational conditions. In the first case, different approaches based on soft computing techniques have been proposed to optimise the catalyst composition [17]. For the operational conditions, we can find some studies for different processes but without catalyst deactivation [16] or only in a discrete way [9]. In this work, we proposed a strategy to study the optimal dynamic operational conditions (with a specific catalyst composition) to achieve the optimal production results, taking into account the catalyst deactivation to design an optimal operation policy that not only maximise the production objectives but it is also able to counteract the catalyst deactivation.

This paper presents the preliminary results for the optimisation of constant operation set points. The optimisation objectives are the operating conditions that govern the BTO process. The optimisation process has been implemented using computational intelligence algorithms. An EA explores the possible optimal solutions guided by a surrogate model of the process based on ANN that is integrated in its evaluation function.

The paper is organized as follows: Section 2 presents the BTO process. Next, Section 3 describes the proposed methodology for the optimisation of the BTO process. Section 4 presents the obtained results. The conclusions and future works are finally summarized in Section 5.

2 BTO Process

The BTO process consists in the catalytic transformation of bioethanol into olefins over an acid catalyst. This is a key process in the concept of sustainable refinery, incorporating biomass or derivatives as an alternative feedstock to petroleum. This process uses a very selective catalyst to maximise the olefins conversion rate (X_O). However, this catalyst is deactivated due to the accumulation of coke. Therefore, when the catalyst reaches a minimum activity value, it is necessary to stop the production step and carry out the catalyst regeneration phase. In addition, the regeneration does not achieve the total catalyst recovery, so the number of possible cycles of production-regeneration is also limited. Depending on the operational conditions, both the production and the catalyst deactivation reversibility will be affected.

Therefore it is necessary to explore experimentally those operational conditions that fulfill the desired objectives, which would be very costly and complicated. The use of EA and surrogate models to explore those operational conditions is much more practical and reasonable.

The following variables are the main operating variables that govern the behaviour of the BTO process:

- **Operating variables**:

  T: reaction temperature (K).

  X_w: mass fraction of water based on the equivalent mass of ethylene in the reactor feed $\left(g_{water}g^{-1}\right)$.

  WF_{EO}^{-1}: space-time $\left(g_{catalyst}h^{-1}(g_{ethanol})\right)$.

- **Activity level**:

  a: catalyst activity. The activity has been considered as a disturbance that quantifies the rate of catalyst deactivation by coke.

In previous works, experimental runs of this process were carried out in an automated device equipped with an isothermal laboratory scale fixed bed reactor connected online to a gas chromatograph and a micro-GC for the analysis of the reaction product (see Figure 1). Details about the reaction equipment, catalyst preparation and experimental methodology can be found in previous works [3, 4].

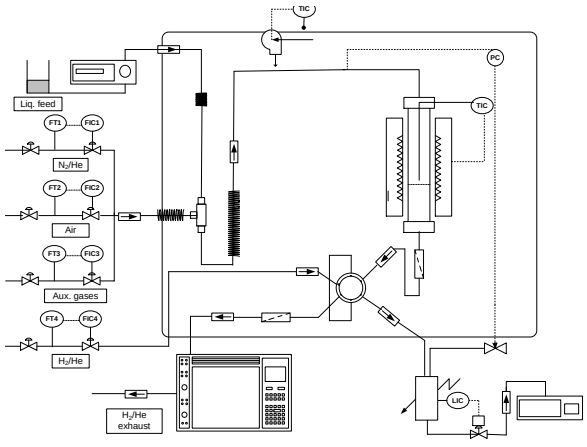

Figure 1: Diagram of the reaction equipment used to obtain the experimental data.

3 Methodology

This section presents the proposed dynamic optimisation strategy using Soft Computing techniques. An EA guided

by an ANN based surrogate model of the process is proposed. The surrogate models are used in the evaluation operator to simulate the process behaviour under each operational condition proposed by the EA. The results will be validated with an existing mechanistic model (MECH) [3, 4], since the experimental validation is not technical or economically viable.

The EA and the BTO process models (both the MECH and the surrogate) have been implemented using the programming package MATLAB™ (version 8.0, 2012b, Mathworks Company). Simulation and optimisation have been carried out in an PC with an Intel® Core™ i5-2467M CPU at 1.6 GHz and 4.0 GB of physical memory (RAM).

3.1 Surrogate Model

Chemical knowledge models are generally computationally very demanding to be used in evolutionary approaches [14]. Thus, in this work, an ANN based surrogate model is proposed to dynamically simulate the process behaviour.

A nonlinear autoregressive with exogeneous inputs (NARX) neural network topology has been selected to model the BTO process. The developed ANN model estimates the olefins conversion rate at the reactor output (X_O) using as inputs the previously mentioned process operating variables (T, X_W,WF_{EO}^{-1}), the catalyst activity level (a) and the previously estimated output values in a recursive loop ($\hat{X}_O$).

An iterative methodology modifying the number of layers and neurons in each layer has been carried out in order to select the neural model structure that better fits the process using the Leave-One-Out Cross-Validation (LOOCV) technique. This technique consists of setting aside a set of experiments (representing unique operational conditions) from the model training phase and only using them for the validation phase. The process is repeated until every single set of experiments is used in the validation stage. These type of techniques test the generalization capability of a model structure.

Consequently, the available data are divided into training, validation and test datasets. The training and validation datasets are used in the model structure selection. The first ones are used to train several models with different structures. Aspects such as the number of hidden layers, the neurons in each layer or the connections between neurons are iteratively modified. The performance of each model is tested and compared following the LOOCV procedure.

Once the neural model structure is selected, the final model is trained with all the available data except from the test dataset which is used to validate the fitted model. The Levenberg-Marquardt Algorithm has been used for the training and the main comparison criterion has been the Root Mean Squared Error (RMSE) of the model (Equation (1)).

$$RMSE := \sqrt{\frac{1}{n}\sum_{i=1}^{n}(X_O - \hat{X}_O)^2}. \tag{1}$$

A first ANN model, trained only with experimental data, was able to describe correctly the process behaviour for the experimentally tested operating conditions (see Section 4). However, for optimisation purposes, it is necessary to test several operating conditions where limited experimental data are available. In particular, the dataset does not contain any experiment describing the dynamic behaviour of X_W and WF_{EO}^{-1}. To provide the ANN with the required information about the process behaviour in the whole operating range, some experiments were simulated with the MECH model and introduced in the training dataset.

3.2 Optimisation Problem

The aim of this optimisation problem is to obtain the operational conditions that achieve the best production results per amount of catalyst needed. Due to the catalyst deactivation, is important to bear in mind the deactivation rate, keeping in the whole simulation time the catalyst activity and the olefins production rate over the established minimum value of 0.10. Therefore, the Equation (2) defines the objective function in order to maximise the total production of olefins.

$$\max_{T,X_w,WF_{EO}^{-1}} \frac{\int_0^{\tau} X_O\left(T, X_w, WF_{EO}^{-1}\right) dt}{WF_{EO}^{-1}}. \tag{2}$$

Please note that there are two stopping criteria (τ constant in the upper bound of the integral):

- The catalyst involves a major process cost. Therefore, to maximise the production per amount of catalyst we have set a lower bound on the activity in order to reduce the total production costs. This bound has been set to 0.10 ($a < 0.10$).
- In order to maintain the production, it is needed that the olefins conversion rate does not decrease below the 0.10 ($X_O < 0.10$).

If any of the above criteria are passed over, we consider that the production has reached its maximum span and should be stopped to proceed with a catalyst regeneration phase.

The operating variables to optimise are bounded based on the physical-chemical properties of the process. In fact, for the temperature (T), $573K$ is the inferior bound where the complete dehydration of the ethanol happens and $673K$ is the upper bound to avoid the irreversible deactivation of the catalyst. The variable X_W will range between $[0.0821, 4.8889]$ and WF_{EO}^{-1} will range between $[0.068, 1.525]$ respectively. Please note that those intervals have been chosen as are the ones used to adjust the mechanistic model [3, 4].

As previously stated, in order to solve the optimisation problem an EA has been used. In particular we have implemented a Genetic Algorithm (GA). Table 1 summarizes the principal parameters of the EA used. The surrogate

Parameter	Value
Individuals	Vector of real numbers
Population	50 individuals
Generations	200 generations
Fitness Operator	Olefins Production
Genesis Operator	Random (uniform) initialization
Selection Operator	4-Tournament
Crossover Operator	Arithmetic crossover
Mutation Operator	Adaptive mutation
Elitism	True

Table 1: Main parameters of the GA used to optimise the operational conditions.

model previously developed will be used now to simulate the temporal behaviour of the process under the operating constant set points provided by the GA.

4 Results

In this section the results obtained for the modelling and optimisation of the BTO process are shown. Following the modelling procedure mentioned above, an ANN based surrogate model, with the topology showed in the Figure 2, has been implemented.

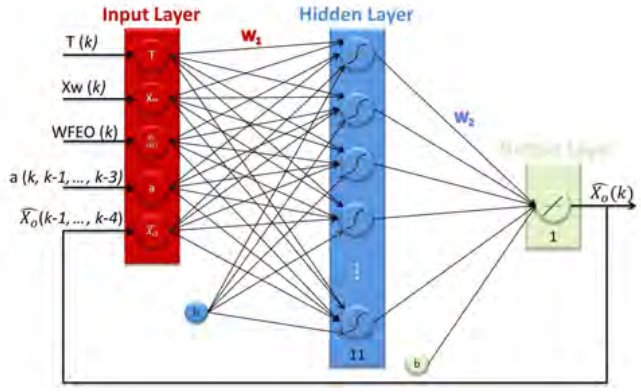

Figure 2: Feed-Forward ANN topology for the BTO process model.

Table 2 shows the mean estimation errors of the mechanistic and surrogate models for the experimental data. The discrepancy between both models is rather low, but the surrogate model simulates the process behaviour 43 times faster.

Model	RMSE ($g_o g^{-1}$)
MECH	0.0323
ANN	0.0387

Table 2: Root mean square error of the mechanistic (MECH) and surrogate (ANN) model to experimental data.

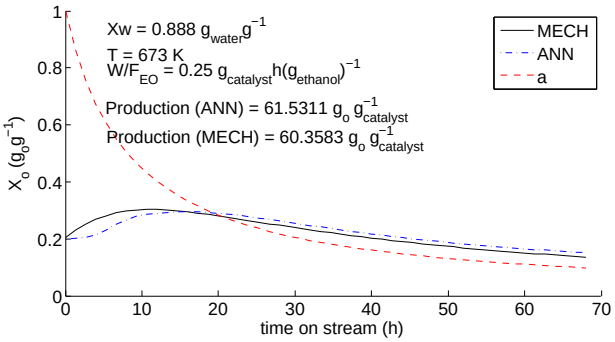

Figure 3: Comparison of the estimation generated by the MECH and ANN models for a test experiment.

MECH ($g_o g^{-1}_{catalyst}$)		ANN ($g_o g^{-1}_{catalyst}$)	
$\mu_{Production}$	$\sigma_{Production}$	$\mu_{Production}$	$\sigma_{Production}$
70.2204	0.6385	63.7376	1.6946

Table 3: Comparison of the mean production results for the "optimal" operational conditions when repeating several times the evolutionary optimisation using the mechanistic (MECH) and surrogate (ANN) models.

Figure 3 represents the estimates of the process behaviour calculated with both models for a test operational conditions that were excluded for the training procedure. The ANN model has been validated using the testing dataset (see Section 3), obtaining a root mean squared error of 0.0323 $g_o g^{-1}$ for the whole test experiments. These results show the capacity of the ANN to properly assimilate and reproduce the BTO process dynamics in the same way as the mechanistic model.

Once the surrogate model has been validated, the evolutionary optimisation has been carried out. Table 3 shows the mean production results obtained with the "optimal" operational conditions generated by the EA. Being the optimisation procedure stochastic, it has been launched several times to guarantee its convergence to a local optimum. Notice the small deviations repeating all the procedure 50 and 100 times for the mechanistic and surrogate approaches respectively. Although the standard deviation using the surrogate model doubles the deviation that results from using the mechanistic model, in both cases they are still very small. So we can conclude that the proposed approach is converging to a local optimum.

Finally, Figure 4 shows the behaviour of the process under the optimum solution provided by the evolutionary optimisation using the surrogate model. The maximum production has been 68.05 $g_o g^{-1}_{catalyst}$. Please note that the maximum production using the surrogate model only differs a 5.67 % over the optimisation carried out using the mechanistic model, but with much less computation cost. This maximum is reached by operating the reactor at $645K$ with a high content of water in the feed ($X_W = 4.875 g_{water} g^{-1}$)

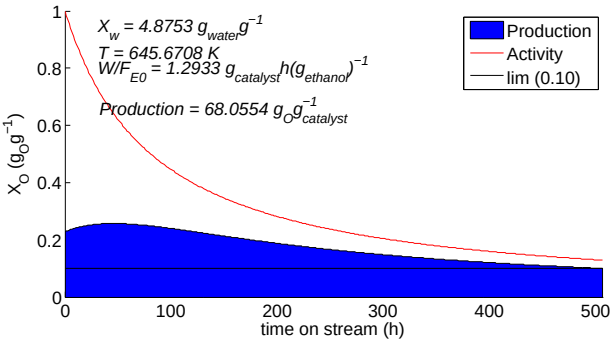

Figure 4: BTO process performance under the best solution provided by the evolutionary optimisation guided by the surrogate model.

and a space-time of $1.293 g_o h(g_{ethanol})^{-1}$. This water quantity attenuates the catalyst deactivation, which allows the extension of the production phase.

5 Conclusions

An optimisation strategy for the catalytic transformation of Bioethanol-To-Olefins (BTO) process based on computational intelligence has been presented. The proposed evolutionary optimisation guided by an Artificial Neural Networks based surrogate models has been able to optimise the process obtaining similar results than those obtained when using a mechanistic model but 43 times faster. A clear optimum has been defined. The optimum operational conditions have shown to be, temperature: $645K$; water quantity in the feed: $4.875 g_{water} g^{-1}$; space-time: $1.293 g_o h(g_{ethanol})^{-1}$. These operational conditions allow to extend the catalyst lifespan maximising the production phase and hence the total production.

The presented results are a preliminary study on the optimisation of the BTO process looking only for constant set points that maximise the production objectives. This has been the first step for a more complex dynamic optimisation of the process, in which the optimal dynamic trajectory for each operating variable will be described. In this line, several trajectory types will be defined for each of the operating conditions to study the process dynamic behaviour.

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