Optimal Design of Experiments for Artificial Neural Network-based Kinetic Model Recognition

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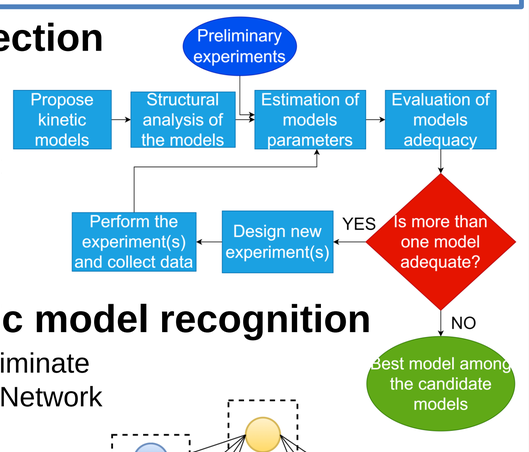
INTRODUCTION

The problem of kinetic model selection

The classical model discrimination approach [1] is characterised by several steps.
The modelling process may require a significant amount of time and the quantity of experimental data may be limited due to cost and time limitations.



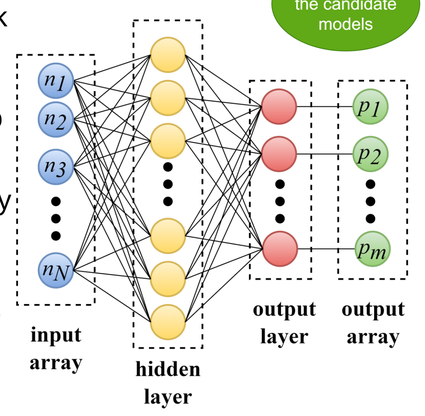
Artificial neural network for kinetic model recognition

A different approach may be considered to discriminate among the models by means of Artificial Neural Network classifiers. [2]
The advantages are

- ▲ Small amount of experimental data required (in silico dataset generation)
- ▲ No structural identifiability test ⟶ no rejection of any model a priori

The limitations are:

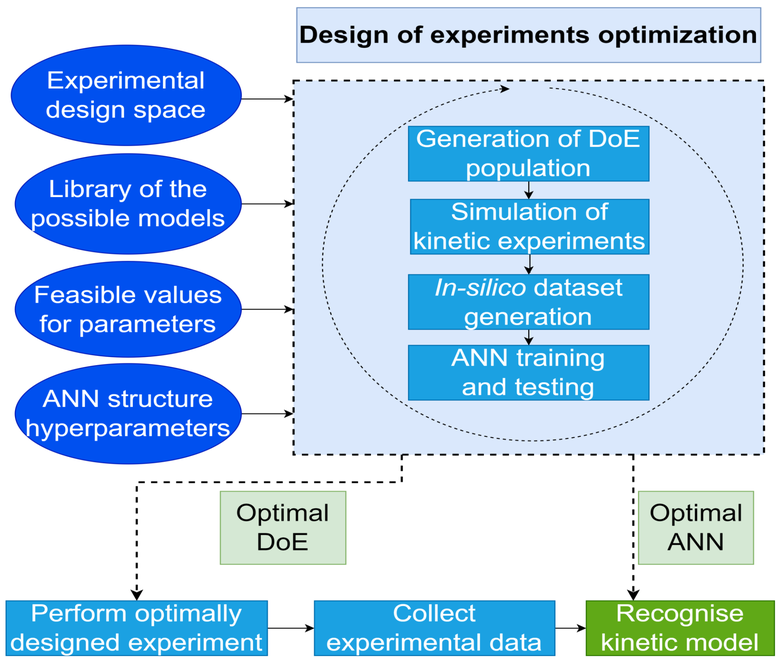
- ▼ No information about the model parameters from the discrimination
- ▼ Experimental design is fixed



APPROACH TO OPTIMIZE THE DESIGN OF EXPERIMENTS

We propose to couple the ANN application to model recognition with a **population algorithm** for optimizing the experimental conditions, **aiming to improve the ANN performance**.

- ➢ The **number of experiments** is provided as an input to the algorithm
- ➢ The ANN training is performed with **simulated experimental data**, which include parameter uncertainty within feasibility regions.
- ➢ **Physical experiments** are only required to be conducted **at the identified optimal conditions**, then the acquired experimental data is fed to the trained ANN.



Objective function to be maximised: Artificial Neural Network **accuracy on unseen data**

$$\mathrm{Acc}_{\Psi_{\mathrm{test}}} = \frac{\left|\left\{i \in \{1,\dots,N_{\Psi_{\mathrm{test}}}\} \text{ s.t. } (\mathrm{n}_i, l_i) \in \Psi_{\mathrm{test}} \wedge \hat{l}_i = l_i\right\}\right|}{\left|\left\{i \in \{1,\dots,N_{\Psi_{\mathrm{test}}}\} \text{ s.t. } (\mathrm{n}_i, l_i) \in \Psi_{\mathrm{test}}\right\}\right|} \cdot 100\%$$

where Ψ_{test} is the testing-set of data (size $N_{\Psi_{\mathrm{test}}}$), n_i experimental data and l_i the label of the model used to generate the data and $\hat{l}_i$ the label predicted by the ANN.

CASE STUDY DESCRIPTION

The proposed approach has been tested on a case study with in silico generated experimental data. The system considered was an **isothermal batch reactor**, where the experimental conditions were defined by:

- ➢ **Initial concentration of reactant A**
- ➢ **Temperature**
- ➢ **Sampling times**

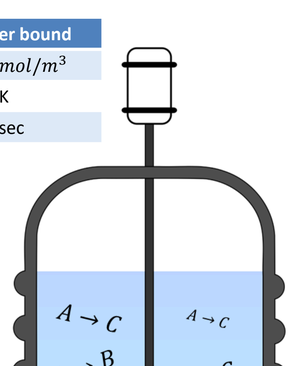
Symbol	Lower bound	Upper bound
C_{A0}	0 mol/m^3	250 mol/m^3
T	520 K	720 K
t	50 sec	350 sec

The reacting system was characterised by three species: A, B, C

We aimed at discriminating among **8 candidate model structures**, assuming the reactions could take place either in series or parallel, and being described either by 1st or 2nd order power law kinetic equations.

Series mechanism: $A \xrightarrow{k_1} B, \quad B \xrightarrow{k_3} C$

Parallel mechanism: $A \xrightarrow{k_1} \mathrm{B}, \quad A \xrightarrow{k_2} C$



Kinetic models formulation

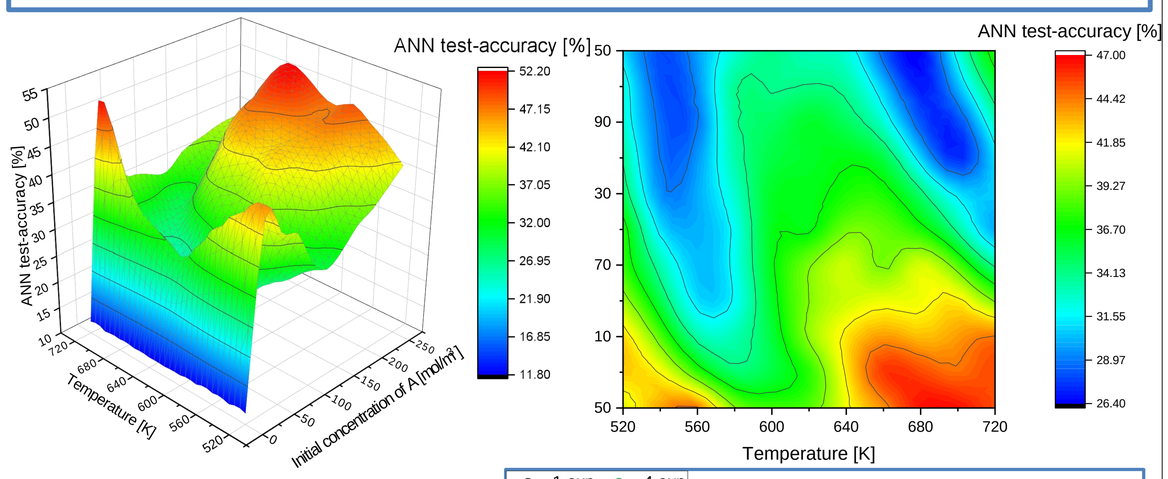
Material balances
$\frac{\mathrm{d}c_i}{\mathrm{d}t} = \sum_{j=1}^{3} \nu_{ij} r_j \quad \forall i = A, B, C$

Arrhenius-type kinetic factors
$k_j = A_j e^{-E_{a,j}/RT} \quad \forall j$=1, 2, 3

Experimental error
$\varepsilon \sim \mathcal{N}(0, \sigma^2)$

	Series mechanism				Parallel mechanism			
Label:	1	2	3	4	5	6	7	8
$\boldsymbol{r_1}$	$k_1 \cdot C_A$	$k_1 \cdot C_A$	$k_1 \cdot C_A^2$	$k_1 \cdot C_A^2$	$k_1 \cdot C_A$	$k_1 \cdot C_A$	$k_1 \cdot C_A^2$	$k_1 \cdot C_A^2$
$\boldsymbol{r_2}$	$k_2 \cdot 0$	$k_2 \cdot 0$	$k_2 \cdot 0$	$k_2 \cdot 0$	$k_2 \cdot C_A$	$k_2 \cdot C_A^2$	$k_2 \cdot C_A$	$k_2 \cdot C_A^2$
$\boldsymbol{r_3}$	$k_3 \cdot C_B$	$k_3 \cdot C_B^2$	$k_3 \cdot C_B$	$k_3 \cdot C_B^2$	$k_3 \cdot 0$	$k_3 \cdot 0$	$k_3 \cdot 0$	$k_3 \cdot 0$

RESULTS

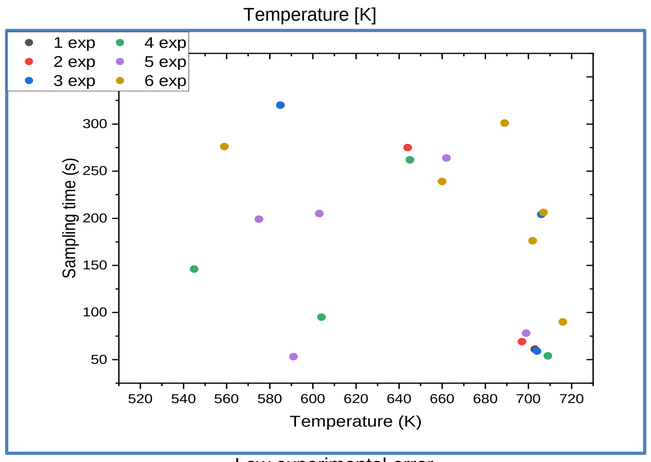


SENSITIVITY ANALYSIS

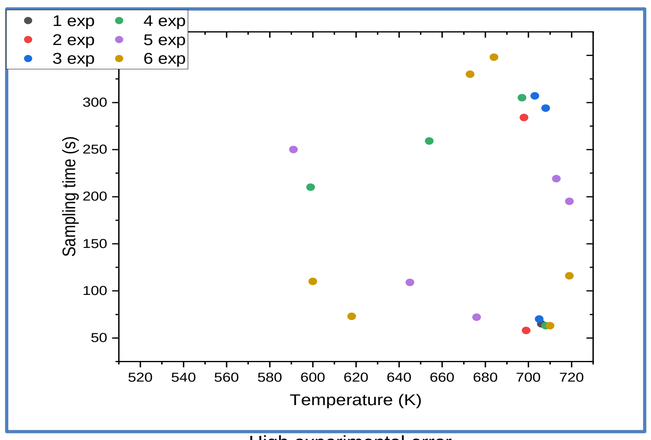
- ➢ The choice of the experimental conditions has a strong impact on the ANN performance.
- ➢ Using a fixed experimental design may lead to suboptimal performance or to conduct unnecessary experiments.

OPTIMAL DESIGN OF EXPERIMENTS

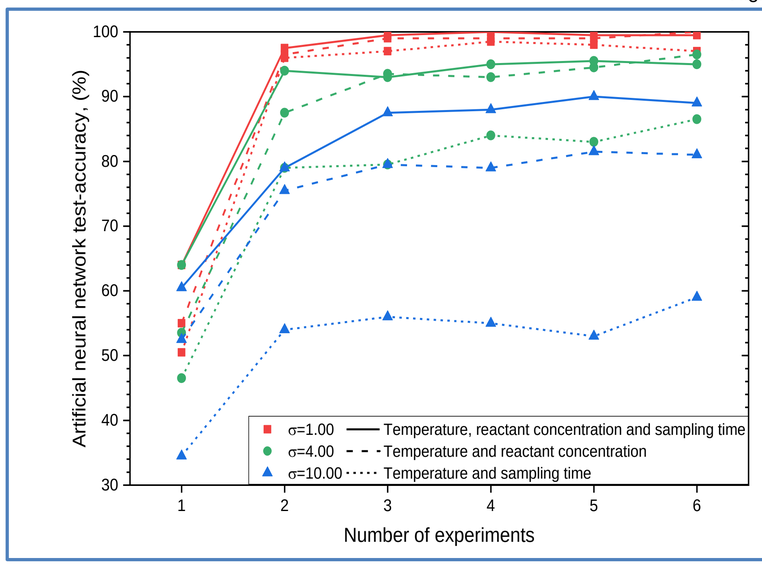
- ➢ The optimization algorithm performance was satisfactory both in the case of low and high noise on experimental data.
- ➢ High levels of accuracy were obtained with just a few experiments required even in the high experimental error scenario.
- ➢ The choice of which experimental variables to manipulate affected the outcome in terms of ANN accuracy.
- ➢ When the optimal ANN failed, the correct model was always among the top 3 ranked models.



Low experimental error



High experimental error



Noise σ	Max ANN accuracy	N exp.
1.00	100 %	4
4.00	97 %	5
10.00	92 %	5

SCAN ME

GitHub repository for the Python code

CONCLUSIONS

- ➢ The identification of kinetic model structures using ANNs was improved by optimizing the experimental design.
- ➢ Steep increase in ANN accuracy with respect to the number of experiments.
- ➢ High levels of accuracy were obtained with a minimal number of experiments to be conducted.
- ➢ The proposed approach is promising for conducting a first discrimination among many candidate kinetic models.

REFERENCES

[1] S.P. Asprey and S. Macchietto, 2000, Statistical Tools for Optimal Dynamic Model Building, Computers & Chemical Engineering, 24, 1261-1267
[2] M. Quaglio, L. Roberts, M.S. Bin Japaar, E.S. Fraga, V. Dua, F. Galvanin, 2020, An Artificial Neural Network Approach to Recognise Kinetic Models from Experimental Data, Computers & Chemical Engineering, 135

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