Master thesis proposal

# Development of a Bayesian optimisation pipeline integrated into a catalytic multi-reactor system for self-optimisation of the catalytic reduction of $CO_2$ into methane

**Keywords:** $CO_2$-to-methane, Bayesian optimization, self-optimizing catalytic process, heterogeneous catalysis

## SCIENTIFIC DESCRIPTION:

In the race for using artificial intelligence-based methods to improve and accelerate the development of new materials involved in catalytic processes many strategies are competing.[1,2,3] Machine learning approaches are booming, taking advantage of the rapid creation of unified databases and new algorithms that take into account more and more aspects of catalytic material development, from predictive DFT to algorithms that capture the properties of catalysts (type of support, particle size, solid phases present, number and types of catalytic sites, etc.). Although these models are increasingly effective at predicting which materials to use, they still struggle to predict the best set of operating conditions under which they should be used in a reactor.

One solution is, of course, to use DOE, as has long been done. Recently, however, we have seen the return of Bayesian optimisation algorithms, this time with a direct application in chemical catalysis.[4] By proposing an iterative process, unlike many DOE models, and above all by focusing on reducing the uncertainty of the modelling function rather than on a specific optimisation parameter, this type of approach offers the possibility of greatly reducing the number of experiments required to obtain a similar mapping of the parameter space to that offered by the best DOE models. However, one of the main limitations of Bayesian optimisation systems lies precisely in their iterative nature. Still requiring a significant but also indeterminate number of measurements, this approach is hence more efficient using high-throughput testing platforms. Unlike DOE, which can be performed in batches of measurements, Bayesian optimisation, which requires only a limited number of points per cycle, ideally requires a minimum time between two consecutive measurements and, therefore, in the best cases, a continuous measurement system whose parameters can be modified on the fly.

We have typically demonstrated the crucial importance of this point through the recent thesis of Dr. Markus Grimm, who developed a new Bayesian optimisation algorithm applied to the optimisation of propane conversion to propylene. Indeed, while the tests were carried out on a Flowrence unit, which has 16 reactors in parallel, the steps involved in defining the reaction parameters were still carried out manually, preventing 1) the modification of these parameters during the reaction in order to adjust the catalytic processes in real time according to the prediction made by the Bayesian optimisation algorithm, and also 2) the closing of the loop without human intervention to allow the system to converge on its own towards the best catalytic conditions in a defined parameter space.

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That is why we would now like to continue this work, this time focusing on setting up a fully circular system with the aim of optimising the hydrogenation of $CO_2$ to $CH_4$. We have recently developed several processes using new catalytic materials tested at high throughput on our Flowrence unit on the REALCAT platform, and we believe that we could quickly optimise the reaction conditions for these materials by deploying this new Bayesian optimisation strategy. The objective of this internship will therefore be to develop an autonomous optimisation system based on a complete feedback closed loop that does not require human intervention, in order to achieve a proof of concept of a self-driving catalytic reactor.

1. Toyao et al., ACS Catalalysis, 2020, 10 (3), 2260-2297
2. Yang et. Al, ACS Omega, 2020, 5 (1), 83-88.
3. McCullough et al., Physical Chemistry Chemical Physics, 2020, 22 (20), 11174-11196
4. Wu et al., Digital Discovery, 2024, 3, 1086-1100
5. Grimm et al., Computers & Chemical Engineering, 2024, 189, 108779
6. HAL: tel-04870317

**Techniques/methods in use:** Gas phase catalytic tests; gas chromatography analysis; use of Bayesian optimisation algorithms; programming of a computer and robotics pipeline.

**Applicant skills:** The person applying for this position must have a good understanding of chemical catalytic systems, without necessarily having significant experience in heterogeneous catalysis, although this would be an advantage. They must have solid knowledge of analytical methods, ideally gas chromatography, and be able to interpret a chromatogram. They must have a minimum background in programming, ideally in Python.

**Industrial partnership:** No

**Internship supervisor(s):** PAUL Sébastien & HEUSON Egon

**Internship location:** This internship will take place at the REALCAT platform at the Ecole Centrale de Lille in Villeneuve D'Ascq.

**Possibility for a Doctoral thesis:** Yes, but no grant is yet secured