Active Learning-based Experimental Optimization of CO_2 Hydrogenation to Methanol

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Highlights

- Process conditions to be optimized for single-pass methanol yield.
- Active learning-based design of experiments will be employed.
- Expected results applicable to larger-scale, complex systems.

1. Introduction

The catalytic hydrogenation of CO_2 to methanol is one of the most important reactions in the defossilization toolkit, as it converts a greenhouse gas into a widely used platform chemical and potential future marine fuel. In a previous study it had been shown that increasing the single-pass yield of this reaction could significantly reduce the levelized cost of green methanol, bringing it closer to economic viability[1]. Single pass yield can be increased by improving the catalyst or finding the optimal operating conditions. This study focuses on the latter by applying Bayesian Optimization Structure Search (BOSS), an active learning-guided Design of Experiment (DoE) framework [2]. In contrast to traditional DoE methods (space-filling design, factorial design, etc.), BOSS takes advantage of dynamic data acquisition and can be efficient in high dimensions. By demonstrating the applicability of BOSS to maximizing single-pass methanol yield on the lab scale, this study is the first step in developing experimental design frameworks for larger, more complex cases (such as pilot- or demonstration plants).

2. Methods

The overall workflow of this study is presented in Figure 1. Based on the results of a few initial baseline experiments, the BOSS methodology will suggest new experimental conditions (pressure, temperature, and gas hourly space velocity). Data obtained at these new points in the design space will, in turn, be fed back to BOSS until the optimum (highest single-pass methanol yield) is found.

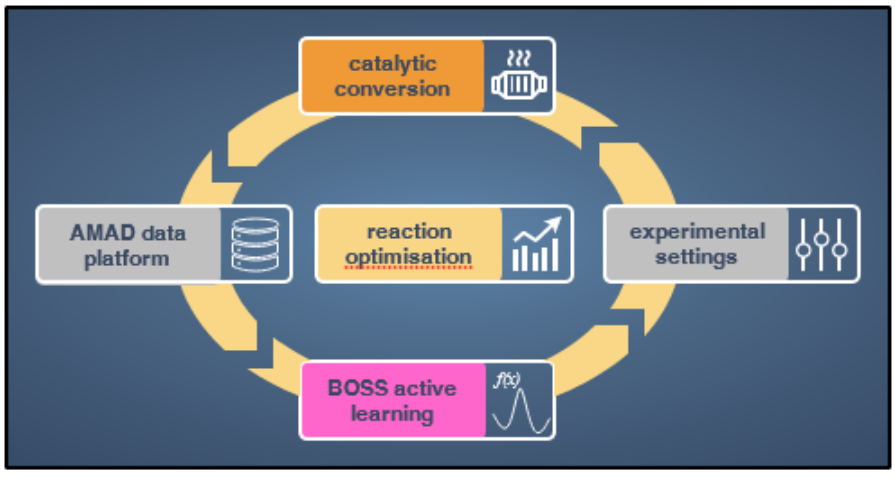

Figure 1: schematic of the active learning-based experimental design workflow used for this study.

Experiments will be carried out with a hydrogenation millireactor (ThalesNano Phoenix with backpressure controller, and a Bronkhorst EL-FLOW mass flow controller) supplied with a CO_2:H_2:Ar gas mixture in the respective molar ratios of 23.5:70.5:6. Analysis will be carried out with a gas chromatographer equipped with a thermal conductivity detector (ThermoFisher 1300 series). Argon will be used as the internal standard for quantification, and the carrier gas is helium. A commercial copper-zinc-alumina catalyst will be used for this study.

Active learning-based experimental design using the BOSS methodology will be implemented in the Aalto Materials Digitalization Platform (AMAD) and connected Jupyter notebooks.

3. Expected Results

Process conditions with the highest single pass methanol yield will be found using BOSS-based iterative experimental design. It is expected that these optimized process conditions could result in improvements in most technical key performance indicators (KPIs) in a large-scale CO2-based methanol plant. Therefore optimized conditions will be used as an input for a techno-economic simulation based on Nyári et al. [1], and the resulting levelized cost of methanol will be compared to literature data and market prices.

References

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Keywords

"methanol" "design of experiments" "Bayesian optimization" "CO_2 hydrogenation".