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# Optimizing Methane Conversion in a Flow Reactor System Using Bayesian Optimization and Model-Based Design of Experiments Approaches: A Comparative Study

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## ABSTRACT

Reaction processes require optimization to enhance key performance indicators (KPIs) such as yield, conversion, and selectivity. Techniques like Bayesian Optimization (BO), Model-Based Design of Experiments (MBDoE), and Goal-Oriented Optimal Experimental Design (GOOED) play pivotal roles in achieving these objectives. BO efficiently explores the design space to identify optimal conditions, while MBDoE maximizes the information gain by reducing kinetic model uncertainty. In contrast, GOOED focuses solely on maximizing the KPIs without considering the system uncertainty, identifying reactor conditions in the design space guaranteeing optimal performance. This study compares BO, MBDoE, and GOOED in optimizing methane oxidation in an automated flow reactor. Performance is assessed based on optimal methane conversion, reduced system uncertainty and minimal experimental efforts to achieve maximum conversion. BO quickly identifies high-conversion conditions, MBDoE minimizes experimental runs while providing insights into parameter sensitivities, and GOOED prioritizes conversion efficiency. The findings highlight trade-offs between convergence speed, robustness, and information gain, providing valuable insights for designing data-driven, physics-informed experiments..

**Keywords**: Bayesian Optimization, Model-Based Design of Experiments, Methane Conversion

## 1 INTRODUCTION

Reaction processes play a significant role in scientific and industrial applications in the production of chemicals, energy, and materials [1]. These processes are driven by interconnected factors such as reaction kinetics, thermodynamics, and transport phenomena [2], and their optimal performance in terms of key performance indicators (KPIs) such as yield, conversion, and selectivity depend on the precise control and fine-tuning of process parameters. Addressing these challenges requires advanced optimization techniques capable of navigating this intricate landscape.

Traditional methods like trial-and-error experimentation and design of experiments (DoE) [3] have proven effective in several applications. These techniques have been instrumental in refining reaction conditions, balancing exploration and exploitation of the design space, and offering insights into reactor behaviour. However, their limitations, particularly in high-dimensional, dynamic, or noisy systems, have spurred the development of novel approaches.

Emerging techniques such as Bayesian Optimization (BO) [4], Model-Based Design of Experiments (MBDOE) [5], and Goal-Oriented Optimal Experimental Design (GOOED) [6] provide more efficient strategies for optimizing reaction processes.

BO excels at identifying optimal conditions with a minimal number of experiments by balancing exploration and exploitation in black-box systems [7]. MBDOE emphasizes the maximization of information gain to improve parameter estimation and reduce uncertainty in physics-based model predictions. The MBDoE approach can also be applied to design experimental design campaigns online [8].

In contrast, GOOED focuses solely on achieving target KPIs, such as maximizing conversion, without prioritizing information gain or uncertainty reduction, making it

highly effective for applications where achieving maximum performance is the primary objective.

The goal of the study is to evaluate the relative merit of the different experimental design approaches and establish which among the three techniques (BO, MBDoE, and GOOED) represents the optimal experimental design scenario by examining the reaction for methane total oxidation in an automated flow reactor as a case study.

A comparative analysis of the three techniques as experimental design scenarios is carried out to quantify their relative performance in terms of (i) conversion of methane; (ii) uncertainty in the evaluation of the optimal conversion; (iii) confidence level of parameter estimates, and (iv) number of experiments required for the maximum KPI to be achieved. The analysis demonstrates how the three techniques can distinctly balance convergence speed and experimental robustness ultimately guiding the choice of more effective experimental design campaigns.

# 2 METHODOLOGY

## 2.1 Generic Model and System Description

The system under consideration is represented by a set of differential and algebraic equations (DAEs), where the measured variables $\mathbf{y}$ can be sampled at discrete time points:

$$\mathbf{f}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{u}, \boldsymbol{\theta}, t) = 0 \qquad \mathbf{y} = g(\mathbf{x}, \mathbf{u}, \mathbf{v}) \quad (1,2)$$

where **f** is a set of model equations, **x,** $\dot{\mathbf{x}}$ and are $N_x$ -dimensional vectors of state variables and their first derivatives respectively, **u** is a $N_u$ -dimensional vector of control variables, $t$ is time, **θ** is a $N_\theta$-dimensional vector of model parameters, **y** is a $N_y$-dimensional vector of response variables that are measurable.

Eq. (1), and (2) establish the fundamental structure upon which the specific methane oxidation reactor model is developed in Section 2.2

## 2.2 Methane Oxidation: System & Reactor Model

Building on the general modeling framework from Section 2.1, this section details a specific application to methane oxidation in flow reactor system as presented by Bawa et al. [12]. In their research, they presented a reactor model characterized by the following set of ordinary differential algebraic equations:

$$\begin{aligned}
\frac{dx_1}{dw} &= \frac{R.u_1}{u_2 P_{avg}} . (-r) \quad x_1(0) = u_4 \\
\frac{dx_2}{dw} &= \frac{R.u_1}{u_2 P_{avg}} . (-2r) \quad x_2(0) = u_3.u_4 \\
\frac{dx_3}{dw} &= \frac{R.u_1}{u_2 P_{avg}} . (r) \\
\frac{dx_4}{dw} &= \frac{R.u_1}{u_2 P_{avg}} . (2r) .
\end{aligned} \qquad (3)$$

For the methane oxidation system, the control variables (**u**) is defined in Eq. 4, while **y** represents the response variables for $CH_4$, $CO_2$, $O_2$ measured at the reactor outlet. The state variables **x,** control variables **u** and model parameters **θ** are defined for the reactor model as follows:

$$\begin{aligned}
\mathbf{x} &= [x_1, x_2, x_3, x_4] \\
\mathbf{u} &= [u_1, u_2, u_3, u_4] \\
\boldsymbol{\theta} &= [\theta_1, \theta_2, \theta_3, \theta_4, \theta_5, \theta_6]
\end{aligned} \qquad (4)$$

where $x_1$ $x_2$ $x_3$ $x_4$ are mole fraction of methane, oxygen, carbon dioxide and water, respectively, and $u_1, u_2, u_3, u_4$ represent temperature [℃], flow rate of the feed $[Nml\ min^{-1}]$, oxygen-to-methane mole fraction $R_{O_{2/CH_4}}$ $[mol\ mol^{-1}]$, and inlet methane mole fraction $[mol\ mol^{-1}]$ respectively. These controls form the design vector $\boldsymbol{\varphi}$ which is bounded within the experimental design domain as shown in Table 1. In the model $R$ $[J\ mol^{-1}\ K^{-1}]$ is the universal gas constant, $r$ $[mol\ g^{-1}\ min^{-1}]$ the reaction rate according to the kinetic model, and $w$ $[g]$ is the catalyst mass along the reactor. The experimental design examined the effects of key control variables on methane oxidation by varying them within defined ranges (Table 1).

**Table 1**. Experiment control variables

| Control Variables | Range | Units |
|---|---|---|
| Temperature | 250-350 | ℃ |
| Mass flow rate | 20-30 | $Nml\ min^{-1}$ |
| Oxygen to methane mole ratio | 2-4 | $mol\ mol^{-1}$ |
| Methane mole fraction | 0.005-0.025 | $mol\ mol^{-1}$ |

The Mars Van Krevelen (MVK) model reaction rate expression (Eq. 5) was adopted as the true model in this study as a representation of the system under consideration:

$$r_{ch4} = \frac{k_1 k_2 P_{CH_4} P_{O_2}}{k_1 P_{O_2} + 2\, k_2 P_{CH_4} + \frac{k_1 k_2}{k_3} (P_{O_2} P_{CH_4})} \qquad (5)$$

Eq. 5 was identified and validated in [11], where the authors calibrated the MVK model from flow reactor data. The authors also stressed that the oxidation of methane involved three steps: 1. Surface oxidation 2. Surface Reduction. 3. Desorption of product. Kinetic model parameters of the MVK model (i.e. rate constants $k$, and activation energies $E_a$) were identified and formulated with respect to these steps thus giving rise to six parameters, $k_1$ , $k_2$ , $k_3$, $E_{a_1}$, $E_{a_2}$, $E_{a_3}$. Furthermore, these parameters were reparameterised to the vector form **θ** and estimated as **θ** = [5.77, 6.72, 5.87, 9.51, 10.17, 7.98]. These parameter values are used as the true parameter vector in this work.

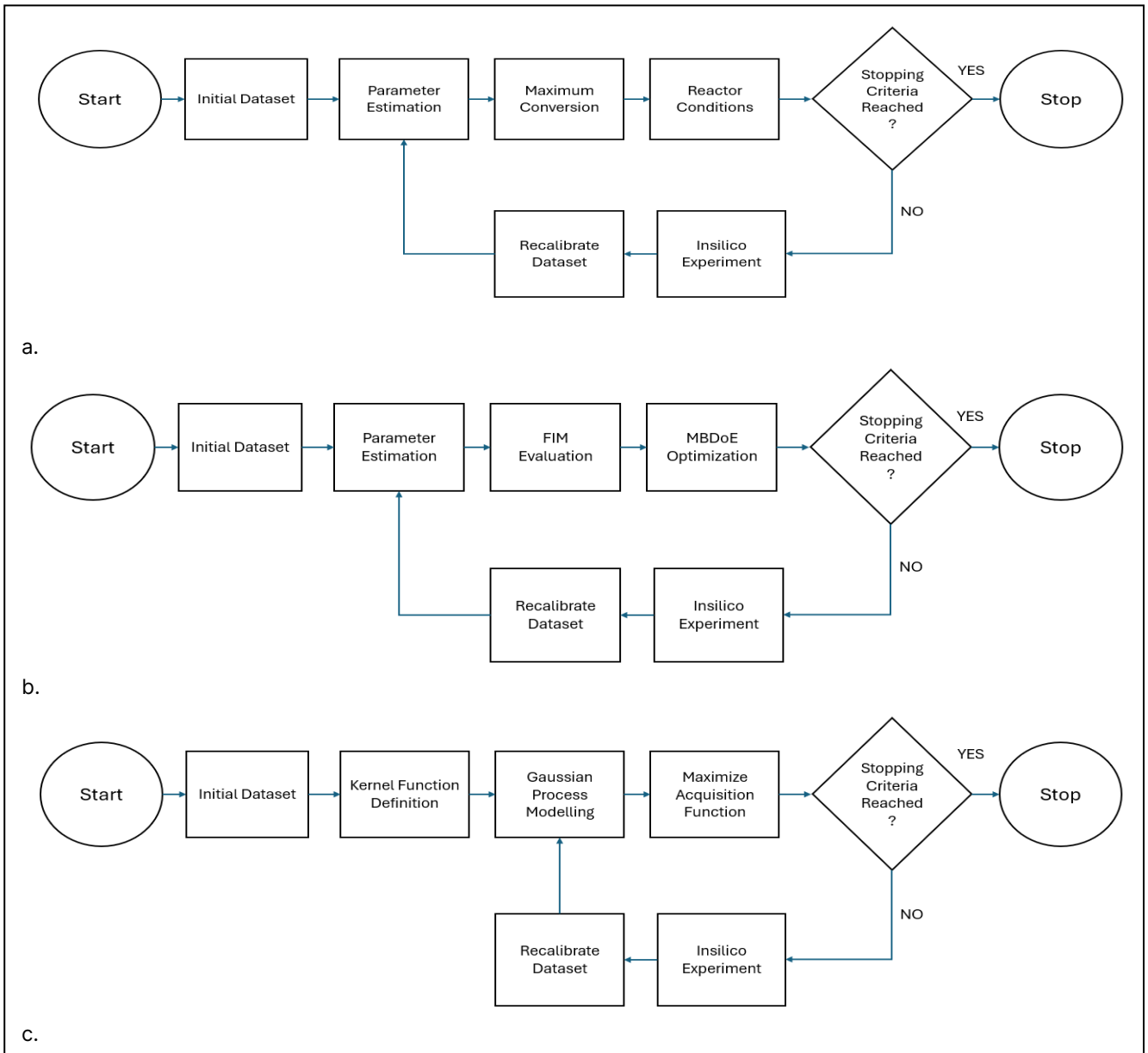


**Figure 1.** Procedural Framework showing workflow procedures for (a) GOOED (b) MBDoE and (c) BO scenarios.

## 2.3 Procedural Approach for GOOED, MBDOE, and BO

The procedural approach for the three experimental design scenarios is tailored to their distinct objectives as shown in Figure 1. GOOED (a) and MBDoE (b) are both exploitative approaches. While they have distinct objectives, they both focus on identifying the optimal point, albeit with different goals in mind. The objective of (a) is to maximise a specific KPI, whereas (b) focuses on the improvement of model predictions by minimising uncertainty in parameter estimation and (c) BO balances exploration and exploitation through adaptive learning to optimize system performance without using mechanistic modelling insights on the system. The stopping criteria used to terminate the procedures for any of the scenarios can be one of the following: a. experimental budget termination b. satisfied $t$ −test on model parameters.

The objective function that describes maximizing methane conversion and used by both GOOED and BO is shown in Eq. 6 below.

$$\mathbf{u}^* = argmin_{\mathbf{u}\in U} - h\,(\mathbf{u}) \tag{6}$$

where $h\,(\mathbf{u})$ is the predicted conversion at any experimental condition $\mathbf{u}$, The algorithms mostly perform minimization, so we minimize $-h\,(\mathbf{u})$, which is the same as maximizing $h\,(\mathbf{u})$, $\mathbf{u}^*$ is the optimal condition at maximum conversion. The design space, $U$, is the boundary

conditions of the control variables shown in Table 1.

### 2.3.1 GOOED

In the GOOED approach the objective is to maximize the conversion of methane. The objective function Eq. 7 which is a slight modification of Eq. 6 is shown below.

$$\mathbf{u}^{*}_{\boldsymbol{GOOED}} = argmin_{\mathbf{u}\in U} - h\,(\mathbf{u}) \tag{7}$$

The steps defining the GOOED procedure are illustrated in Figure 1a and summarised below.

1. *Initial Dataset*: An initial dataset from 12 preliminary experimental runs, created using statistical design of experiments (DoE) methodology as described in [11], serves as the starting point for the procedure.
2. *Parameter Estimation*: Using computational methods and Eq. 6, the observations are utilized to estimate model parameters and estimates are evaluated for statistical significance.
3. *Maximize Methane Conversion.* The optimization of methane conversion is guided by the experimental conditions defined in Table 1; the objective function is represented in Eq. 7. The system undergoes an optimization routine to identify the maximum achievable conversion of methane.
4. *Optimal Reactor Conditions.* During this process, the corresponding operating conditions that yield the maximum conversion are also determined.
5. *In Silico Experiment*: Simulations are run to forecast how the reaction will behave under the determined optimal conditions.
6. *Iterate Until Stopping Criterion*: The procedure continues, updating the dataset and improving predictions of the optimal conversion based on feedback from each experimental run.
7. *Procedure termination*: The process stops when the stopping criterion is met, i.e. the maximum experimental budget is exhausted, or the *t*-test results are statistically significant – both of which are the stopping criteria.

### 2.3.2 MBDoE

In the MBDoE approach, the objective function is designed to maximize the determinant of the Fisher Information Matrix (FIM), $H_{\hat{\theta}}$ this function is defined in Eq. 8 which represents dynamic systems.

$$\mathbf{H}_{\hat{\boldsymbol{\theta}}}(\hat{\boldsymbol{\theta}}, \boldsymbol{\varphi}) = [\mathbf{V}^{0}_{\hat{\theta}}]^{-1} + \sum_{i=1}^{n_{sp}} \left(\frac{d\hat{\mathbf{y}}}{d\hat{\boldsymbol{\theta}}}\right)_i^{T} \sum_{y}^{-1} \left(\frac{d\hat{\mathbf{y}}}{d\hat{\boldsymbol{\theta}}}\right)_i \tag{8}$$

where $\mathbf{V}^{0}_{\hat{\theta}}$is the $N_\theta\ x\ N_\theta$ prior variance-covariance matrix of model parameters, while $(\frac{d\hat{y}}{d\hat{\theta}})$is the $N_y\ x\ N_\theta$ matrix with first-order derivatives of model responses with respect to the parameters at time point *i*. The parameter estimates are obtained from Eq. 11, which provides the initial values for MBDoE based on the model and available data. To assess the quality of these estimates, Eq. 8 computes the Fisher Information Matrix (FIM) which allows to predict the expected precision of parameter estimates. The *t*-values are calculated as metric of parameter precision (see Section 2.4.1), and statistical conditions are checked to ensure the validity of the estimates.

The procedural approach for MBDoE (Fig 1b) is the following

1. *Initial Dataset*: Same as in GOOED
2. *Parameter Estimation:* Same as in GOOED
3. *FIM Evaluation:* The evaluation of the expected FIM was computed at the current value of the parameter estimates.
4. *Optimal Design of Experiment:* Based on the estimated parameters and uncertainties, new experimental conditions are determined by maximizing the determinant of the expected FIM [8].
5. *In Silico Experiment:* The new experimental conditions are applied, the experiment is simulated generating new data in silico, and the experimental dataset is updated.
6. *Iterative Process:* The process continues iteratively until the t-test results are statistically significant, or the experimental budget is terminated both of which are the stopping criteria.

### 2.3.3 Bayesian Optimization (BO)

In the third scenario, we adopted the BO technique due to its ability to effectively find the optimized value of the objective function. The objective function which is a modification of Eq. 6 is defined below in Eq. 9

$$\mathbf{u}^{*}_{\boldsymbol{BO}} = argmin_{\mathbf{u}\in U} - h\,(\mathbf{u}) \tag{9}$$

The choice of a composite kernel function Eq. 10 combining constant kernel *C*, and RBF kernel to effectively capture both global scaling and local variability in the relationship between experimental conditions $\mathbf{u}$ and the conversion of methane (Eq. 14). This combination helps the GP to represent system behaviour and optimize control variables for maximum conversion efficiency.

$$k(\mathbf{u}, \mathbf{u}') = C.RBF\ (\mathbf{u}, \mathbf{u}') \tag{10}$$

where $\mathbf{u}, \mathbf{u}'$ are two different sets of experimental conditions.

The procedural approach for the BO (Fig 1c) is outlined below:

1. *Initial Dataset*: Same as in GOOED
2. *Define the Kernel and Gaussian Process (GP) Model*: We defined the GP with a composite kernel function Eq. 10 where $\mathcal{C}$ is a constant kernel with an initial value of 1.0 (bounded between $10^{-3}$ and $10^{3}$), and $RBF$ (Radial Basis Function) has a length scale initialized at 1.0 with bounds between $10^{-2}$ and $10^{2}$). The hyperparameters of the kernel are optimized during model training by maximizing the log marginal likelihood using gradient-based optimization technique [13].
3. *Train the GP Model*: The GP model is trained using an initial dataset to learn the relationship between the input conditions and the corresponding system response variables [14]. To account for noise in the data, the GP model incorporates the noise and variance from the preliminary dataset.
4. *Cross-Validation*: To ensure the GP model generalizes well, 5-fold cross-validation is performed on the initial datasets. The data is split into 5 subsets, where the model is trained on 4 subsets and tested on the remaining one [15], repeating the process 5 times. The average performance metric (e.g., RMSE) is used to assess model robustness and prevent overfitting.
5. *Define the Acquisition Function (AF)*: An AF is chosen to guide the search for optimal experimental conditions in our case we chose the Expected Improvement (EI).
6. *Optimize the AF*: The chosen acquisition function is then optimized this leads to selection of the next experimental condition $\mathbf{u}_{\text{new}}$ to test.
7. *In Silico Experiment*: At this new experimental condition $\mathbf{u}_{\text{new}}$, the experiment is simulated generating new data in silico, and the experimental dataset is updated.
8. *Procedure termination*: The process stops when the stopping criterion – the experimental budget is exhausted.
9. The next experiment is conducted at the selected reactor conditions, $\mathbf{u}_{\text{new}}$ The process continues iteratively until the experimental budget of 20 runs is reached, at which point the process stops.

## 2.4 Parameter Estimation

Parameter estimates (indicated as $\theta$) from the experimental data were computed by minimizing the difference between measured responses ($y$) and predicted responses ($\hat{y}$) through the negative log-likelihood function [6].

$$L(\hat{\boldsymbol{\theta}}) = \frac{N}{2}\log(2\pi) + \frac{n_{sp}}{2}\log\left(det|\boldsymbol{\Sigma}_{\boldsymbol{y}}\right) + \frac{1}{2}\sum_{i=1}^{n_{sp}}[\boldsymbol{y_i} - \hat{\boldsymbol{y}}_{\boldsymbol{i}}\,(\hat{\boldsymbol{\theta}})]^T\ \boldsymbol{\Sigma_y}^{-1}[\boldsymbol{y_i} - \hat{\boldsymbol{y}}_{\boldsymbol{i}}(\hat{\boldsymbol{\theta}})] \quad (11)$$

$n_{sp}$ is the number of sampling points considering all the Ne performed experiments, namely $n_{sp} = \sum_{i=1}^{N_e} N_{sp_i}$ is the number of sampling points in the $i_{th}$ experiment), $N$ is the total number of experimental measurements calculated as $N$. In GOOED and MBDoE the $\hat{y}_i$ is computed from the mechanistic model. In BO $\hat{y}_i$ is computed from GP predictions.

### 2.4.1 Uncertainty in Parameter Estimates

Parameter precision is quantified using the $N_\theta\ x\ N_\theta$-dimensional covariance matrix ($V_\theta$), approximated by the inverse of the observed Fisher Information Matrix ($H_\theta$) (Eq. 8). From the covariance matrix, the 95% confidence interval for the $ith$ parameter is calculated using Eq. 12 as the square root of the variance element $v_{\theta,ii}$, multiplied by the *t*-value at the 95% confidence level:

$$95\%\ \text{CI} = \sqrt{\boldsymbol{v_{\theta,ii}}} \times \text{t (95\%, DoF) for i = 1, ..., } N_\theta \quad (12)$$

$$t_i = \frac{\hat{\boldsymbol{\theta}}_{\boldsymbol{i}}}{95\%\ Confidence\ Interval_i} \quad (13)$$

The *t*-value for each parameter was calculated by dividing the parameter estimate by the 95% confidence interval, as shown in Eq. 13. A high *t*-value indicates a reliable parameter estimate, while a low *t*-value suggests that the confidence interval may include zero, implying the parameter might not be statistically significant and could be excluded from the model, and the corresponding uncertainty is then computed for each parameter estimates.

## 2.5 Conversion of Methane

The conversion of methane is the KPI used in this study, and it is defined in Eq. 14 as the ratio

$$\alpha = \frac{Y_{CH_4} - y}{Y_{CH_4}}, \quad (14)$$

where $Y_{CH_4}$ and $y$ are the inlet and outlet concentration of methane respectively [12]. To account for uncertainties in methane conversion, error propagation of the uncertainties associated with the conversion was performed using statistical methods [16].

## 2.6 Preliminary Dataset

The preliminary dataset used in this study was derived from a series of experiments designed using DoE as reported in [11], aimed at investigating the kinetics of complete methane oxidation over the $Pd/Al_2O_3$ catalyst.

## 2.7 Performance Metrics for the Design Scenarios

Results are compared in terms of (a) conversion and

associated uncertainty; (b) operating conditions in the design space; (c) parameter estimation significance after the stopping criteria are reached; (d) number of experiments required to satisfy stopping criteria. Simulations for the three different scenarios were all implemented in Python including SciPy library functions, *scalar* and *minimize* functions while a custom class called Gaussian Regressor was specifically developed to solve the BO scenario.

## 3 RESULTS & DISCUSSION

This section compares the three (3) different scenarios using the performance metrics from 2.7.

### 3.1 Methane Conversion and Associated Uncertainty

Table 2 illustrates the conversion percentages and associated uncertainties for GOOED, MBDoE, and BO under the evaluated scenarios. Key observations are the following:

**GOOED:** Exhibited the highest conversion with values as high as 97 – 99%, but with high uncertainty, highlighting variability in the results.

**MBDoE**: Delivered a moderate conversion values hovering at 45% with low uncertainty values ensuring reliability and consistency.

**BO**: Achieved an optimal conversion (~87%) with moderate uncertainty, obtaining a balance between performance and reliability. Table 2 compares the scenarios, showing how GOOED maximizes conversion, MBDOE minimizes uncertainty, and BO essentially balances both objectives.

**Table 2:** Methane Conversion and Corresponding Uncertainty for GOOED, MBDOE, and BO Scenarios

| Exp | GOOED Scenario | MBDOE Scenario | BO Scenario |
|---|---|---|---|
| 13 | 99.00±94.00 | 42.90 ±0.57 | 43.9 ±9.81 |
| 14 | 99.04±64.00 | 88.90 ±0.37 | 87.45 ±5.78 |
| 15 | 97.70 ± 4.00 | 40.30 ±0.18 | 88.01 ±5.08 |
| 16 | 97.93 ± 4.26 | 37.00 ±0.16 | 87.71 ±4.98 |
| 17 | 97.95 ± 4.26 | 46.40 ±0.13 | 87.72 ±4.85 |
| 18 | 98.07 ± 4.28 | 79.20 ±0.08 | 87.72 ±4.78 |
| 19 | 98.07 ± 4.04 | 48.40 ±0.06 | 87.72 ±4.65 |
| 20 | 98.06 ± 4.13 | 44.80 ±0.05 | 88.01 ±4.08 |

### 3.2 Operating Conditions in the Design Space

Figures 2a, 2b, and 2c illustrates the distribution of the reactor conditions in the experimental design space for GOOED, MBDOE, and BO campaigns. Key observations include:

**GOOED:** The reactor conditions identified reflects a strong exploitation of known high-performing conditions, with no emphasis on uncertainty reduction**.**

**MBDoE:** The reactor conditions for this scenario show transition from exploration to exploitation ensures improved system understanding while maintaining reliability in predictions**.**

**BO:** The reactor conditions selected by BO demonstrate a balance between exploration and exploitation by achieving average conversion rates while exploring under-characterized areas for further improvement. The oxygen-to-methane ratio for the simulated reactor condition for the three scenarios is reported in Table 3.

**Table 3.** Simulated Oxygen to Methane Mole Ratio in (mol/mol)

| Exp | GOOED | MBDOE | BO |
|---|---|---|---|
| 13 | 3.8 | 3.9 | 3.5 |
| 14 | 4 | 4 | 3.9 |
| 15 | 4 | 4 | 3.9 |
| 16 | 4 | 4 | 4 |
| 17 | 3.7 | 4 | 3.9 |
| 18 | 4 | 2 | 3.9 |
| 19 | 3.9 | 2.1 | 4 |
| 20 | 3.8 | 2.1 | 3.9 |

### 3.3 Parameter Estimation Statistics

For the evaluation metrics based on significance of parameter estimates, we focused on only two of the scenarios: GOOED, and MBDOE. This is because BO is a data-driven process and has no kinetic model parameter representation.

**GOOED:** At the end of the twenty (20) experiments, the t-test value for the 4th parameter failed the test as its value were less than the reference *t*-value as shown in Table 4.

**MBDoE:** For this scenario, the parameter estimates all passed the t-test evaluation successfully as shown in Table 4, this occurred at the 17th experiment making it one of the most efficient scenarios based on minimum number of experiments, and uncertainty. Table 4 shows that *t*-values obtained from MBDoE were statistically significant, including the critical parameter $\boldsymbol{\theta_4}$, at the end of the 17$^{th}$ experiment as their values were all greater than the reference.

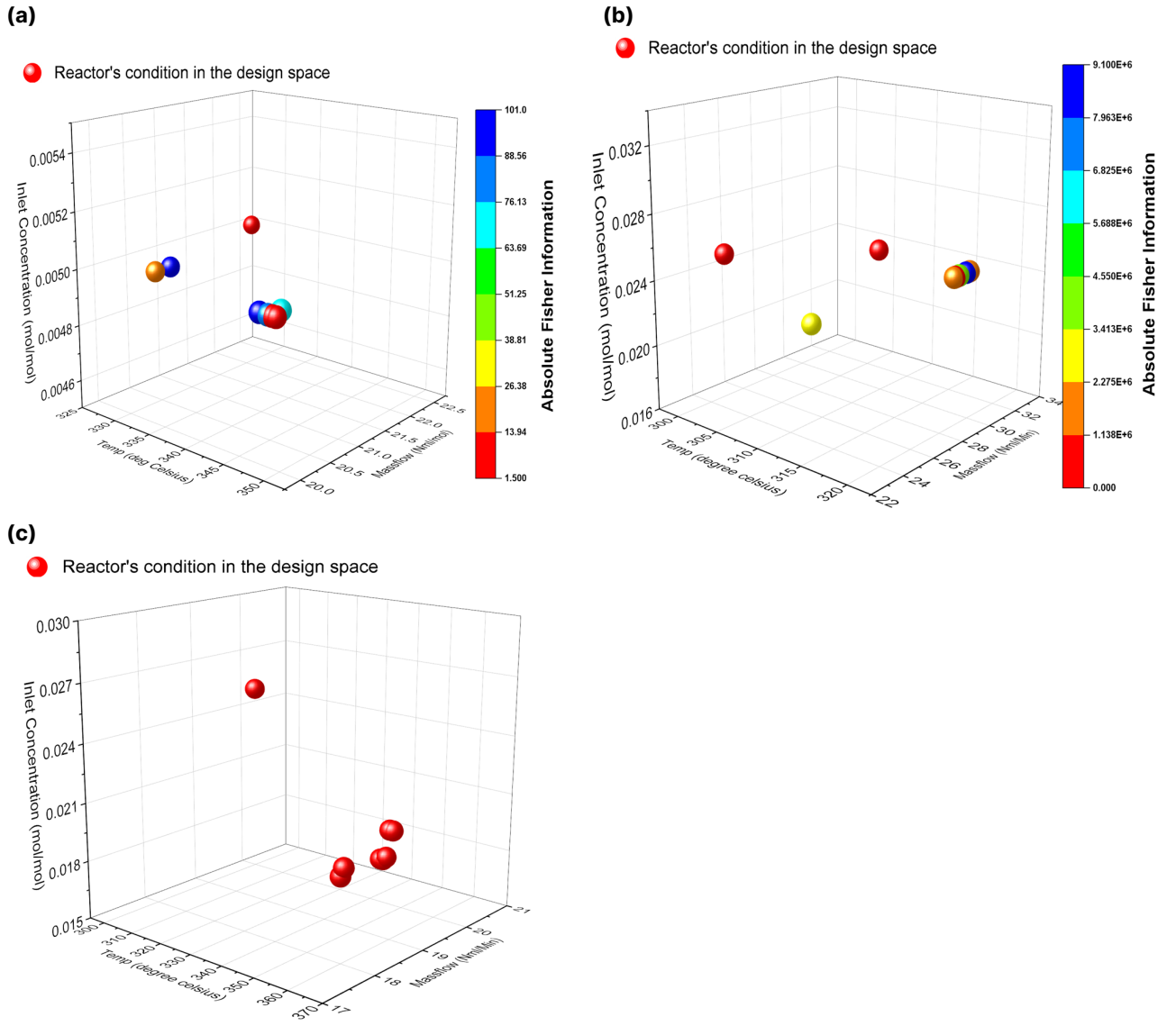


**Figure 2.** Plot of reactor conditions within the design space for the three scenarios: (a) GOOED, (b) MBDOE, (c) BO, respectively. The subplot (c) shows no Fisher information as they are not computed for the BO scenario

**Table 4.** Parameter estimation results for GOOED & MBDoE after stopping criteria successfully terminated (the reference *t*-value is $t^{ref}$ = 1.67, the parameter failing the t-test is indicated in boldface).

| S/N | Parameters | GOOED Estimates | *t*-value | MBDoE Estimates | *t*-value |
|---|---|---|---|---|---|
| 1 | $\boldsymbol{\theta_1}$ | 6.08 | 13.02 | 5.92 | 17.48 |
| 2 | $\boldsymbol{\theta_2}$ | 7.43 | 1.91 | 6.86 | 2.24 |
| 3 | $\boldsymbol{\theta_3}$ | 5.54 | 6.41 | 4.06 | 3.77 |
| 4 | $\boldsymbol{\theta_4}$ | 7.52 | 0.94 | 9.84 | 1.74 |
| 5 | $\boldsymbol{\theta_5}$ | 9.84 | 11.69 | 10.52 | 111.17 |
| 6 | $\boldsymbol{\theta_6}$ | 7.70 | 1.715 | 6.96 | 6.23 |

## 3.4 Number of Experiments

The minimum number of experiments (see Table 5) required to achieve optimal conditions were based on the pre-defined stopping criteria. The MBDOE achieved the lowest number of experimental runs as the parameters were successfully estimated at the 17th experiment.

**Table 5**. Minimum no. of experiments required

| **Scenarios** | GOOED | MBDOE | BO |
|---|---|---|---|
| **No of Experiment** | 20 | 17 | 20 |

## 3.5 Optimizing Conversion with Reduced uncertainty

In this study, the MBDOE demonstrated reduced

uncertainty, and required the fewest experiments. At the 17th experiment, it satisfied the stopping criteria. To validate the strength and performance of this scenario (MBDOE), we optimized the system to achieve maximum conversion at the 17th experiment for this scenario and the method achieved a predicted conversion of 95% which is a good conversion value for the system.

### 3.6 Best Scenario performance

The best scenario included the scenario providing the highest conversion value of methane, lowest value of conversion uncertainty and minimum number of experiments. Table 6 shows that MBDOE performed better than the other scenarios, as it had maximum predicted conversion with reduced uncertainty and satisfactory *t*-values with only 17 runs.

**Table 6**. Scenarios and Performance Metrics

| Scenarios | Conversion (%) | Uncertainty (%) | No. of Experiment | *t*-value Results |
|---|---|---|---|---|
| GOOED | 98 | ± 22.87 | 20 | Failed |
| MBDOE | 95 | ± 0.2 | 17 | Passed |
| BO | 87 | ± 5.5 | 20 | N/A |

## 4 CONCLUSION

In comparing GOOED, MBDoE, and BO, distinct differences emerge across conversion, parametric uncertainty, reactor conditions and experimental efficiency. GOOED achieves the highest conversions (98%) but the prediction of the KPI is affected by a significant uncertainty. MBDoE balances exploration and exploitation, delivering a high conversion (~95%) with the lowest uncertainty, requiring only 17 experiments to meet the parameter precision termination criterion. BO, while needing 20 experiments like GOOED, provides a moderate conversion (~87%) associated with a higher uncertainty, because of balancing exploration and exploitation in the design space. Overall, MBDoE proves to be the most efficient method as it shows minimum variance, reduced uncertainty across experiments, maximum conversion of methane and minimum number of experiments required to achieve maximum conversion.

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