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Proceeding Paper

An Intelligent System for Predicting the Methanol Conversion Rate from the Direct Hydrogenation of CO_2 under Uncertainty †

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Abstract: In this study, an intelligent system was developed for the real-time monitoring of methanol conversion from the direct hydrogenation of CO_2 under the effect of uncertainty in process conditions. The modeling and simulation of methanol synthesis were conducted using Aspen Hysys, the first-principal modeling software. The Aspen model was then shifted into dynamic mode by introducing a $\pm 5\%$ uncertainty in key process conditions, i.e., temperature, pressure, and mass flow rate, to produce a dataset comprising 370 samples. The data samples were then employed to build a Gaussian Process Regression (GPR) model to predict the methanol conversion rate. The GPR model has a root-mean-square error (RMSE) of 0.83127 and a coefficient of determination (R^2) of 0.98078.

Keywords: machine learning; artificial intelligence; soft sensors; renewable methanol

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1. Introduction

The increasing carbon dioxide (CO_2) levels in the atmosphere have led to global climate change, emphasizing the need for effective carbon management strategies. Industrialized nations must collaborate on a global scale to implement measures aimed at reducing CO_2 emissions and limiting the temperature increase to 2 degrees Celsius [1]. Carbon capture and utilization (CCU) and renewable energy resources have emerged as potential solutions that can significantly contribute to mitigating the effects of climate change. Renewable energy technologies are crucial for reducing ongoing emissions, but it is difficult to achieve a shift to zero-net-emission energy by using exclusively renewable sources. In the end, CCU is often seen as a transitional technology for a decarbonized future energy economy [2]. CCU strategies not only directly address the existing CO_2 in the atmosphere but also convert captured CO_2 into methanol, aiding in combating climate change and laying the groundwork for a sustainable and cleaner-energy future [3]. Methanol, known for its versatility, holds the potential to serve as a renewable fuel, chemical feedstock, and energy carrier. Currently, the global market for methanol exceeds 94 million tons, with over 80% of it primarily produced from natural gas reforming [4]. Nevertheless, the process of converting CO_2 into methanol through direct hydrogenation is subject to economic, thermodynamic, and kinetic constraints due to the complexity of chemical reactions and the necessity for the precise control of process parameters to achieve optimal yields [5]. Soft sensors are emerging as a promising solution for overcoming the challenges associated with the conversion of CO_2 into methanol through direct hydrogenation. These sensors can collect, analyze, and interpret data from multiple sources, enabling the prediction ofoptimal conditions for the conversion process. Various case studies have reported on the implications of soft sensors in process industries regarding the real-time monitoring of quality [6], predictive maintenance [7], and energy efficiency [8,9]. However, no studies have been reported on the real-time monitoring of methanol conversion using machine-learning (soft sensor) algorithms. In this study, a soft sensor was developed for the databased prediction of methanol conversion from the direct hydrogenation of CO_2 from a cryogenic biogas upgrading process under the effects of uncertainty.

2. Process Description

The methanol production process begins with the introduction of feedstocks, including 76.46 kmol/h of CO_2 by-product and 535.22 kmol/h of hydrogen. The CO_2 by-product is a result of the upgrading of cryogenic biogas at 12.3 °C and 47.63 bar within the same plant, while hydrogen is supplied at 25 °C and 30 bar. Initial processing involves compressing these feeds to 78 bar, followed by amalgamation and heating to 210 °C. The combined stream is directed into a plug flow reactor for methanol synthesis. The reactor, comprising multiple tubes loaded with 44,500 kg of a $Cu/ZnO/Al_2O_3$ catalyst, facilitates an exothermic reaction. The reaction kinetics are modeled using Vanden Bussche and Froment's [10] approach, incorporating parameter modifications provided by Mignard and Pritchard [11], resulting in a reactor conversion rate of 61.94%. In the post-reaction section, the reactor effluent divides into two streams. The first, constituting 60% of the initial flow, is used for heating the incoming fresh feed. Simultaneously, the second stream preheats the feed intended for the distillation column. Subsequent stages involve the condensation and separation of water and methanol from unreacted gases in a knockout drum. The crude methanol undergoes a two-stage pressure reduction process facilitated by valves, reaching 1.2 bar. The remaining residual gases are efficiently removed using a flash tank. The resulting stream undergoes further heating up to 80 °C in a heat exchanger before entering a distillation column. At the column's base, water exits at 102 °C, while gaseous methanol, the desired product, exits at the top of the column at a pressure of 1 bar and a temperature of 46.84 °C.

3. Methodology

1. First-Principles Model: A first-principles model for methanol synthesis was developed using Aspen Hysys (https://www.aspentech.com/en/products/engineering/aspen-hysys, accessed on 1 April 2024). Model accuracy and real-world relevance were ensured through the incorporation of literature data [3,12].
2. Data Generation: Transitioning the Aspen Hysys model into dynamic mode, an interface between MATLAB and Aspen Hysys was established using actxserver. This dynamic mode introduced a ±5% uncertainty in critical process conditions (temperature, pressure, and mass flow rate), simulating real-world variability and resulting in a dataset of 370 samples. Table 1 presents some samples of generated data. Sample-1 represents the steady-state conditions of the Aspen Hysys model, while the rest were generated by inserting artificial uncertainty.
3. Soft Sensor Development: In the subsequent phase, a Matern GPR model served as a soft sensor and was developed using MATLAB 2023b. GPR's ability to handle small datasets, flexibility in capturing non-linear relationships, and suitability for sequential learning make it advantageous in this context. The dataset was divided into a ratio of 80:20 for training and testing, respectively. This partition facilitated the assessment of the model's generalization capability, leading to the creation of a robust soft sensor capable of predicting methanol syn thesis performance in diverse conditions.

Table 1. Generated data under conditions of uncertainty.

Data Samples	**Molar Flowrate (kmol/h)**		**Temperature (°C)**						**Pressure (kPa)**									
	CO_2	H_2	5.00	6.00	14.00	20.00	CO_2	H_2	1.00	2.00	5.00	9.00	10.00	12.00	19.00	CO_2	H_2	Conversion
1.00	76.46	535.22	210.00	284.00	35.00	42.37	80.00	12.30	7800.00	7800.00	7570.00	7480.00	7360.00	7360.00	7800.00	4763.00	3000.00	61.94
2.00	78.87	556.94	202.17	295.74	35.46	38.40	12.36	26.14	8162.61	7532.94	7926.24	7821.96	7349.24	7581.01	7520.67	4961.01	3087.66	58.83
3.00	82.49	565.61	192.78	306.07	37.00	35.15	12.66	25.86	8289.52	7285.25	8089.56	7455.76	7185.29	7236.96	7217.69	5057.67	3031.19	54.50
4.00	84.83	543.88	186.31	310.04	37.05	34.84	13.14	26.90	8450.52	7168.99	8004.65	7112.03	7563.63	7168.62	7193.36	5273.91	3122.98	51.24
5.00	74.14	521.90	212.44	283.24	34.48	36.30	12.36	26.04	7632.95	8000.62	7762.07	7390.57	7409.92	7047.83	7452.08	4895.97	3130.20	63.88

4. Results and Discussion

The GPR model was designed, trained, and tested in MATLAB 2023b. A total of 370 data points were produced by introducing artificial uncertainty in 17 operating conditions. From the generated data samples, 296 were randomly selected for model training, while the remaining 74 data samples were employed for model testing. A 5-k cross validation technique was employed to validate model performance. In this method, the dataset is divided into k subsets, or "folds", of approximately equal size; the model is trained on some of these subsets; and then its performance is evaluated with respect to the remaining data. The employed hyperparameters for the GPR model include a constant mean function, an Isotropic Matern 5/2 kernel function with a length scale of 147.098, and a noise level (sigma) of 4.4003. The hyperparameter of the model was identified through a trial-and-error method. To evaluate the performance of the developed soft sensor, the RMSE and R2 were used, calculated through Equations (1) and (2). Apart from the inherent distribution of samples for GPR training and validation, 74 data samples were intentionally kept from the model to evaluate its generalization for unknown data, as depicted in Figure 1. The GPR model has an R2 of 0.98078 and an RMSE of 0.83127 for the methanol conversion rate.

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=0}^{n}\left(Y_i^{exp} - Y_i^2\right)^2} \quad (1)$$

$$R^2 = 1 - \left[\frac{\sum_{i=0}^{n}\left(Y_i^{exp} - Y_i\right)^2}{\sum_i^n\left(Y_i^{exp} - Y_{avg}^{exp}\right)^2}\right] \quad (2)$$

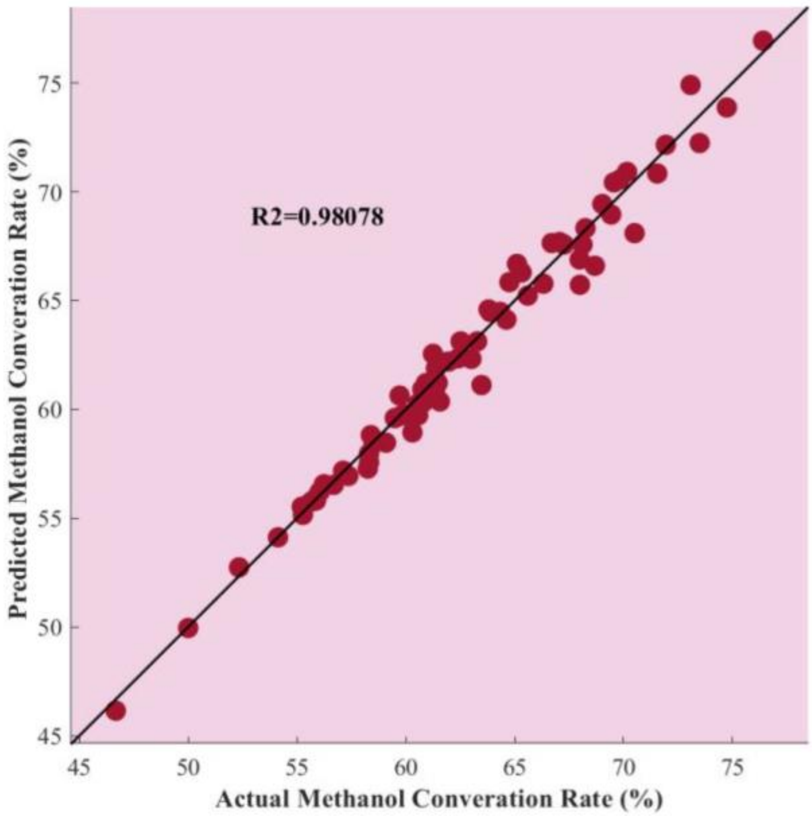

Figure 1. Actual vs. predicted methanol conversion rate.

The developed GPR model can be integrated into the plant's control system, which predicts methanol conversion rates with associated uncertainty based on continuous feed data from the plant. The predicted values are utilized as feedback for process control, allowing dynamic adjustments to be made to optimize the process. The GPR model periodically retrains with new data to adapt to evolving plant conditions, ensuring accurate predictions under varying uncertainties. This integrated soft sensor enhances process monitoring and control, facilitating improved decision-making and overall plant performance.

5. Conclusions

This work comprises the development of a soft sensor for predicting methanol conversion from the direct hydrogenation of CO_2 in the face of challenges of thermodynamic and kinetic constraints due to the complexity of chemical reactions. An Aspen model was simulated, presenting a methanol conversion rate of 61.94%. Then, 370 data samples were produced by shifting the model into dynamic mode through an interface with MATLAB and by introducing artificial uncertainty in the process conditions. From the generated data, a GPR model was developed, validated, and tested; the resulting R2 of 0.98078 and RMSE of 0.83127 show good model performance. The proposed soft sensor demonstrated good accuracy and can serve as a promising tool for real-time application in a renewable methanol synthesis plant.

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