ANN-Based Surrogate Modeling for Prediction and Optimization of Carbon Conversion to Methanol Plant Under Uncertainty

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ANN-Based Surrogate Modeling for Prediction and Optimization of Carbon Conversion to Methanol Plant Under Uncertainty

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THESIS ACCEPTANCE CERTIFICATE

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MASTER'S THESIS WORK

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3.	TEE-820	Process Intensification	Elective	3.0	A
4.	RM-898	Research Methodology	Additional	2.0	Q
5.	PSE-802	Optimization and Decision Analysis	Compulsory	3.0	B+
6.	PSE-823	Advanced Process Dynamics and Control	Compulsory	3.0	A
7.	CSE-801	Computational Fluid Dynamics	Elective	3.0	B+

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MASTER'S THESIS WORK

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Dedication

I have devoted my thesis to my family and teacher's constant support, encouragement, love, and honor.

Acknowledgment

All praise and honor are rightfully directed towards "ALLAH," the unquestionable creator of this world, who has bestowed upon us the precious gift of comprehension and ignited our unwavering curiosity about the world as a whole. We extend our warmest and most respectful greetings to the revered leader of this world and the hereafter, "Prophet Mohammed (PBUH)," a source of boundless knowledge and blessings for all of humanity, as well as for the Uma.

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Abstract

Artificial Neural Networks (ANN) utilization as surrogates within particle swarm optimization (PSO) and genetic algorithm (GA) frameworks for methanol flow rate optimization under uncertainty is explored in this work. First, Aspen Plus model with steady-state conditions of the CO_2 hydrogenation to methanol process was developed. The process model was then transformed to a dynamic mode by inserting ±5% uncertainty in the process and 3880 data samples were generated. An ANN model, developed using MATLAB 2022a, was trained using these data samples. ANN achieved an impressive accuracy having a root mean square error (RMSE) of 26.83 and correlation coefficient (R) of 0.995 while testing for unseen data during cross-validation of the model. Then the ML model ANN was used as a surrogate in the PSO and GA for optimization methods to identify optimal conditions that maximize the methanol flow rate amidst uncertainty. Results show consistent improvements over the standalone Aspen model, with PSO slightly outperforming GA. Validation in Aspen Plus confirms the efficacy of the proposed methodology. This study highlights the potential of ANN-based surrogate modeling and its application in intelligent data-driven optimization for complex chemical processes under uncertainty, ultimately contributing to more efficient production systems.

Keywords: Carbon Conversion, Artificial Neural Network, Artificial Intelligence, Particle Swarm Optimization, Genetic Algorithm, Surrogate modeling, Machine learning.

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Nomenclature

Yi: Predicted value

Yi ^{exp}: Actual value

AI: Artificial intelligence

ANN: Artificial neural networks

BASF: Badische Anilin–und Soda Fabrik (a chemical company)

BR: Bayesian Regularization

BR: Boilup ratio

CCS: Carbon capture and sequestration

f: Activation function

FP: First principle

GA: Genetic algorithm

LM: Levenberg-Marquardt (an optimization algorithm)

ML: Machine learning

PSO: Particle Swarm Optimization (an optimization technique)

R: Correlation coefficient

RMSE: Root means squared error.

RR: Reflux ratio

SA: Stand alone

SCG: Scaled Conjugate Gradient (an optimization algorithm)

Chapter 1

Introduction

1.1 Background

Lately, the global focus has shifted towards exploring environmentally-friendly solutions to address pressing environmental challenges. Environmental issues arise because of the increased demandfor energy requirements. Nowadays most energy is generated using non-fossil energy sources, but fossil fuels are still used as the main energy supply source [1]. So, we need to develop reasonable and sustainable fossil fuel with less environmental concerns [2]. Carbon dioxide (CO_2) is a greenhouse gas generated from fossil fuels whose increased emission has causeda greenhouse effect that results in global warming, acidification, and a certain degree of sea level rise with 1^oC rise of average temperature since before the industrial revolution of the planet with the ten warmest years recorded occurring since 1998 [3-5]. As per a recent report, by 2100, our atmosphere could hold up to 570 ppm of CO_2, resulting in a 1.9 ^oC temperature rise and a 3.8-meter sea level increase[6]. When it comes to various emission sources of CO_2, an anthropogenic emission source coal-fired power plants stand out as significant contributors to global CO_2 emissions, releasing around 2 billion tons of CO_2 annually as shown in Fig 1. Thus, it is of utmost importance to deal with rising levels ofCarbon dioxide (CO_2).

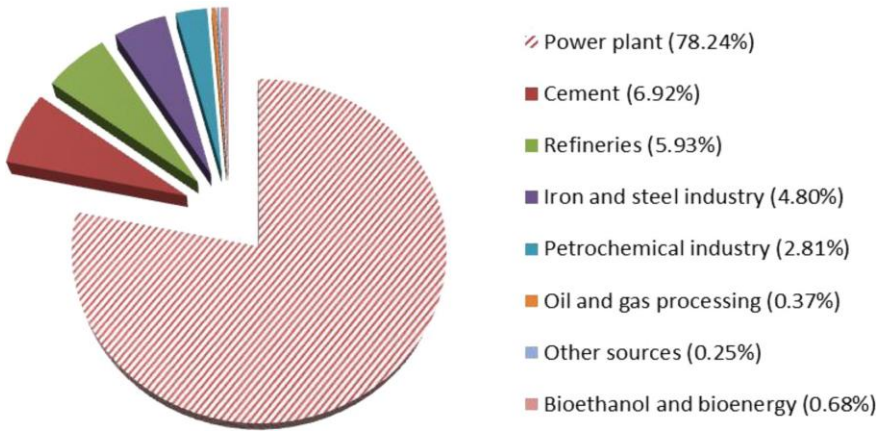


Figure 1: Breakdown of the dominant CO_2 emission sources [7]

Carbon dioxide (CO_2) can effectively be mitigated by four possible pathways: (1) enhancing current process efficiency (2) using eco-friendly or sustainable energy resources (3) Trapping carbon dioxide emissions at their origin and depositing them beneath Geological formations, known as carbon capture and sequestration (CCS), and (4) Utilizing the collected CO_2 for make useful and marketable products [8-10]. Among all the alternatives, the CO_2 utilization methods stands out as the most intriguing option for addressing climate change in the long term because the price of CO_2 capture can be recovered by making the market-desired end product from capture Carbon dioxide (CO_2) utilization can either be physical where its chemistry is same or dissolved in the mixture (i.e., process fluid, solvent, dry ice, refrigerant, welding medium, or fire extinguisher), or chemical where the CO_2 molecule loses its identity and is transformed into another end product (i.e. dimethyl carbonate (DMC), methanol, ethanol, di-methyl ether (DME), methane, syngas, and carbonates) [11] as shown in figure 2.

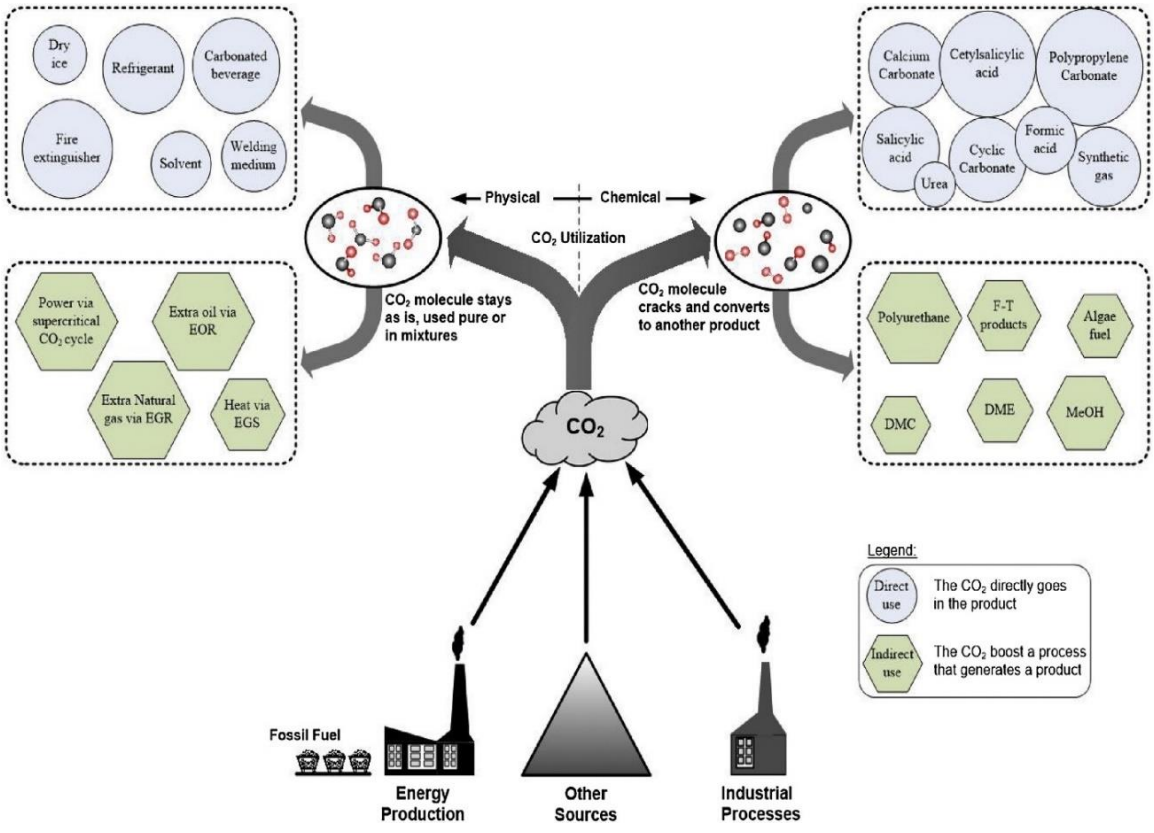


Figure 2: CO_2 different chemical utilization pathways [11]

Out of all the chemical conversion product methanol is one of the most important products produced from chemical utilization of CO_2 which is both a significant fuel and chemical industry raw material [12]. With an energy density of 22.7 MJ/kg, it is approximately half that of gasoline. It appears as a colorless, water-soluble liquid with a boiling point of 64.6 ^oC and a freezing point of 97.6 ^oC [13]. Unlike gasoline, methanol (MeOH) is highly toxic but is not considered carcinogenic. As a result, MeOH is a preferred fuel choice for internal combustion engines [14]. The process of methanol synthesis requires cost-effective raw materials and highly efficient chemical processes. The transformation of carbon dioxide into methanol through hydrogenation is considered a great industrial opportunity [15]. Although there are numerous sources for affordable CO_2, the availability of low-priced hydrogen is somewhat constrained.

Methanol is a widely used commodity in various industrial processes. It finds application in producing formaldehyde [16], methyl formate [17], methyl tertiary butyl ether [18], DME [19], DMC [20], acetic acid [21], biodiesel [22], ethanol [23], propanol [24], methyl amines [25], ethylene glycol [26]. Additionally, it serves as a common industrial solvent in numerous applications. Additionally, methanol possesses excellent combustion qualities, making it suitable as a vehicle fuel, despite having roughly half the energy density of gasoline. Moreover, it is a cleaner option compared to traditional fossil fuels. It can be blended with gasoline in various proportions, ranging from minor additives to higher concentrations like M85 (85% methanol and 15% gasoline). Vehicles running on pure methanol (M100) are also feasible and even more streamlined [27]. However, it's important to note that methanol's toxicity is a significant concern when considering its use as fuel. Ingesting large quantities of methanol can be extremely harmful, leading to blindness and potentially even death.

Now ML is used in the study because the industry undergoing a transformation towards AI-driven smart manufacturing, commonly referred to as Industry 4.0, machines are now capable of autonomous communication and collaboration.

Machine learning is at the forefront of AI, allowing systems to create mathematical models from training data. This helps them learn and make predictions, even in unfamiliar situations [28]. The precision comes from carefully selecting features and effective training, making ML a valuable

tool for understanding complex environmental phenomena with variations over time and space [29-31].

ML has a wide impact, touching all aspects of life and being a key part of Industry 4.0. Inspired by how the human brain learns, ML gives computers the ability to handle complex problems by learning and adapting to different inputs. It has many advantages, like dealing with complexity, improving computational efficiency, handling uncertainty, and aiding decision-making [31]. ML's progress has benefited various scientific and engineering fields, including materials science, bioengineering, construction management, and transportation engineering [32].

Numerous studies have explored the diverse applications of AI and machine learning in various industries. These encompass real-time monitoring [33, 34], predictive maintenance [35, 36], quality control [37, 38], energy efficiency enhancements [39, 40], data mining and analytics [41], drug discovery and development [42], streamlining industrial processes [43], safeguarding against process-aware attacks on industrial control systems [44], optimization [45], monitoring and diagnosing faults in supervised processes [46], IoT-based Smart Agriculture systems [47], industrial tomography [48], predicting Heart Failure Disease [49], earthquake engineering [32], and more.

1.2 Conventional methanol production process

On commercial scale methanol is typically produced by catalytically converting synthesis gas(gaseous mixture of CO, CO_2 and H_2). In 1905, Sabatier introduced the initial synthetic method for generating methanol from CO and H_2. [50]. Synthesis gas, which serves as the primary feedstock for methanol production is most commonly derived from the reforming of fossil fuels, with natural gas being the predominant source in today's use [9].In 1923, Badis-Che Anilin–und Soda Fabrik (BASF) patented a methanol-producing method referred to as "high-pressure methanol synthesis" [51]. This process utilized syngas generated through coal gasification. Operating within a temperature range of 300^{o}C to 400^{o}C and under pressures ranging from 250 to 350 atm, it employed a zinc/chromium oxide catalyst. It's worth noting, however, that this catalyst also yielded lightweight hydrocarbons and methane with a selectivity of around 2–5 wt% [52]. In 1966, Imperial Chemical Industries (ICI) introduced a process, operating at lower temperatures and pressures (300^{o}C and 100 atm), using cleaner

sulfur-free syngas from methane steam reforming. This allowed for a more efficient Cu/ZnO catalyst and eliminated the co-production of lighter hydrocarbons due to the lower reaction temperature [50]. Lurgi later developed a method operating under milder conditions (250–350^{o}C, 40–50 atm). Initially, methanol was made via CO catalytic hydrogenation using CO/H_2 or $CO/CO_2/H_2$ as a feed gas [53]. During the 1990s, research revealed that the hydrogenation of CO_2 occurred at a faster rate when compared to CO, making it the preferred reaction even when utilizing a feed gas composed of CO, CO_2, and H_2. [54].

1.3 CO_2 to methanol process

1.3.1. Overview

Methanol can be made from CO_2 using two different methods: the one-step and two-step processes. In the one-step process, methanol is directly produced through the hydrogenation of CO_2 (Eq.(1)). In the two-step process, a series of reactions is employed. Initially, CO_2 undergoes conversion into CO by means of the Reverse Water Gas Shift (RWGS) reaction, as illustrated in Equation (2). Then,the generated CO is transformed into methanol by adding hydrogen (Eq.(3)) [55].

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \quad \Delta H_{298} = -49.5 \text{ kJ/mol}_{CO_2} \qquad (1)$$

$$CO_2 + H_2 \rightarrow CO + H_2O \quad \Delta H_{298} = 41.2 \text{ kJ/mol}_{CO_2} \qquad (2)$$

$$CO + 2H_2 \rightarrow CH_3OH \quad \Delta H_{298} = -90.7 \text{ kJ/mol}_{CO} \qquad (3)$$

The methanol-producing reactions (Eq.(1)) and (Eq.(3)) are exothermic, releasing heat, while the reverse water-gas-shift (RWGS) reaction (Eq.(2)) occurs together and is endothermic.

Many recent investigations have been carried out regarding the production of methanol through the hydrogenation of CO_2. Joo et al.[56] explored a two-step process for methanol production and determined that it results in higher yields compared to the one-step method. Mignard et al.[57] introduced synthesis process of methanol that relies on utilizing CO_2 extracted from coal power plant flue gas and electrolytic hydrogen. The effectiveness of this method in achieving substantial CO_2 reduction hinges upon the presence of waste heat from the power

plant, which provides the necessary thermal energy. When these heat sources are not available, the capacity for CO_2 abatement is essentially negligible. Mignard et al.[58] conducted a study in which the energy effectiveness of manufacturing methods involving the hydrogenation of CO_2 to produce ethanol, gasoline, and methanol is compared. The methanol production process exhibited the maximum level of efficiency. Pontzen et al.[59] carried out tests to contrast the production of methanol from CO_2 using a $Cu/ZnO/Al_2O_3$ catalyst with the traditional syngas production process. The CO_2-dependent method displayed reduced efficiency in comparison to the conventional method. Soltanieh et al.[60] performed an economic evaluation of the simultaneous production of methanol and electricity using captured CO_2 and hydrogen devoid of carbon content. Van Der Ham et al.[61] developed a fluidized-bed membrane reactor system for converting CO_2 into methanol. Although the process successfully reduced CO_2 emissions, it faced challenges in terms of economic feasibility.

1.3.2 Catalysts

Cu and Zn have been identified as the primary components of catalysts essential for methanol production from CO_2, often supplemented with additives like Al, Si, Cr, Zr, B, Ga, and others [62]. For commercial methanol production from CO_2, the prevalent catalyst of choice is typically $Cu/ZnO/Al_2O_3$, as examined by various researchers [57-59, 63]. However, it's worth noting that this catalyst exhibits reduced efficiency when supplied with CO_2 compared to a mixture of CO/CO_2 [9]. In pursuit of catalysts better suited for CO_2 feedstock, substantial research efforts have been dedicated. In many instances, the proposed catalysts are built upon copper and zinc oxides, incorporating additional elements like ZrO_2, GaO_3, and SiO_2 over alumina. Guo et al. [64] conducted a study to examine how the preparation process in- fluences the performance of a $Cu/ZnO/ZrO_2$ catalyst. Meanwhile, Zhang et al. [65] explored how the addition of zirconia affects the γ-Al_2O_3-supported Cu-based catalyst. Additionally, Raudaskoski et al. [51] conducted a comprehensive review of research papers focusing on copper-based catalysts containing zirconia.

1.4 Objectives

Objective of this work includes:

- Integration of First principal model and Matlab for parameters uncertainty.

- ANN model Development for data-based prediction of methanol production plant under uncertainty.
- Employing an ANN model as a surrogate within PSO and GA frameworks to achieve enhance methanol production in the presence of uncertainty.

1.5 Thesis Outline

Thesis follows the following pattern: In Chapter 1, we provide an Introduction and background to the research topic. This is followed by Chapter 2, where we present an extensive literature review on the subject of CO_2 conversion to different products, especially methanol. Chapter 3 delves into the research methodology employed in the development of a predictive and optimization framework for Methanol flow rate. Moving on to Chapter 4, we present the outcomes of our research, including quantification of Methanol flow rate and discussions pertaining to the optimization framework.

Chapter 2

Literature Review

2.1 Literature Review

Numerous studies have explored the utilization of machine learning in the conversion stage of carbon dioxide (CO_2) management. These studies aim to enhance the efficiency of CO_2 conversion through a variety of computational techniques and make the process efficient. The literature in this field is often categorized based on the diverse range of products that can be synthesized.

2.1.1 Methanol Production:

Khademi et al.[66] used an artificial neural network approach to Investigate a Conventional model with two jacketed reactors connected in series for methanol production. The ANN gives good predictions for Conventional Methanol Production with Reactor Outlet: MSE=0.0042, AARD= 0.0035 and Unit outlet: MSE=0.0042, AARD= 0.0040 and Modified Methanol Production with Reactor outlet: MSE=0.009, AARD= 0.0024 and Unit outlet: MSE=0.008, AARD= 0.0024. Chuquin-Vasco et al.[67] utilized a neural network (ANN) to forecast the methanol flux at the exit of a carbon dioxide dehydrogenation facility, employing the $Cu/ZnO/Al_2O_3$ catalyst. They compared the ANN's performance when trained with three distinct algorithms: Levenberg–Marquardt (LM), Bayesian regularization (BR), and scaled conjugate gradient backpropagation (SCG). Notably, the ANN trained with the Levenberg–Marquardt (LM) algorithm displayed exceptional predictive accuracy, achieving an RMSE (Root Mean Square Error) of 0.0085 and an overall regression coefficient of 0.9442.

Vanjari et al.[68] conducted a comparative analysis of different ML models to investigate the direct catalytic CO_2 hydrogenation process for methanol production. The GBRT and ANN models demonstrated superior performance compared to other ML models, achieving R^2=0.95, RMSE=2.06 and R^2=0.94, RMSE=2.29 for CO_2 conversion, and R^2=0.95, RMSE=6.06 and

R^2=0.95, RMSE=6.05 for methanol selectivity, respectively. The study explores various catalysts and operating conditions to directly convert CO_2 and hydrogen into methanol.

2.1.2 Synthesis Gas (Syngas) Production:

Istadi et al.[69] introduced an innovative method that integrates a genetic algorithm and an artificial neural network (ANN-GA) for the modeling of the "Catalytic-dielectric barrier discharge" process, which aims to produce synthesis gas (syngas) by converting methane and CO2. Their study employed a dielectric barrier discharge system to investigate the impacts of different operational parameters. The ANN-GA gives good prediction for all process parameter with CH_4 conversion:R^2=0.997 and % E=8.29%, C_2^+ selectivity:R^2=0.998 and % E=20.39%, H_2 selectivity:R^2=0.991 and % E=14.86%, C_2^+ yield: R=0.997 and % E=21.59% and H_2/CO ratio:R^2=1 and % E=12.53%. Hossain et al.[70] explored the utilization of artificial neural networks (ANN) for the modeling of hydrogen-rich syngas production from methane dry reforming on Ni/$CaFe_2O_4$ catalysts within a continuous flow fixed bed stainless steel reactor. The investigation encompassed two different neural network architectures, namely the multi-layer perceptron (MLP) and the radial basis function (RBF). The ANN-MLP-based model achieved superior coefficient of determination (R^2) values when compared to the ANN-RBF-based model. Specifically, the ANN-MLP model achieved R^2 values of 0.9726 for H_2 yield, 0.8597 for CO yield, 0.9638 for CH_4 conversion, and 0.9394 for CO_2 conversion. Ayodele et al.[71] employed an artificial neural network (ANN) to assess the influence of various process parameters on the production of synthesis gas through dry reforming of methane over a cobalt catalyst. This investigation was conducted within a fixed-bed stainless steel reactor. The ANN, trained using the forward propagation algorithm, exhibited excellent predictive abilities for CH4 conversion, CO_2 conversion, and the rates of H_2 and CO production, achieving R^2 values of 1 for all these variables. Ayodele et al.[72] Ayodele and colleagues (Reference 72) carried out a research study aimed at assessing how various process parameters affect the conversion of carbon dioxide (CO2) and methane (CH4) in reforming reactions using Nickel (Ni) catalysts. They employed various configurations of multilayer perceptron (MLP) and nonlinear auto-regressive exogenous (NARX) neural network models to investigate this impact. The models' performance was assessed based on their capacity to predict the conversion of CO_2 and CH_4. The NARX model, specifically the one trained with Bayesian Regularization (BR-NARX), exhibited

the most exceptional performance, achieving an impressive coefficient of determination (R^2) of 0.998 and a very low mean squared error (MSE) of 3.24×10–9. The BR-NARX accurately predicted the thermocatalytic conversion of CH_4 and CO_2.

2.1.3 C_2 and C_2^+ Hydrocarbon Production:

Ayodele et al.[73] utilized the Radial Basis Function artificial neural network to model and predict the thermo-catalytic CO_2 oxidative coupling of methane to C_2 hydrocarbons. The Radial Basis Function ANN demonstrated excellent performance with a sum of square error (SSE) of 0.224 and a coefficient of determination (R^2) of 0.990. Moreover, the model's high R^2 values of 0.989 and 0.998 for predicting the selectivity and yield of C_2 hydrocarbons respectively, highlight the robustness and accuracy of the Radial Basis Function ANN. Istadi et al.[69] developed a hybrid artificial neural network-genetic algorithm (ANN-GA) to model Investigated Catalytic-dielectric barrier discharge for the production of C_2^+ hydrocarbons through the conversion of CO_2 and methane. The research utilized a dielectric barrier discharge system to explore the effects of various process parameters. The ANN-GA give good prediction for all process parameter with CH_4 conversion:R^2=0.997 and E=8.29%, C_2^+ selectivity:R^2=0.998 and % E=20.39%, H_2 selectivity:R^2=0.991 and % E=14.86%, C_2^+ yield: R=0.997 and % E=21.59% and H_2/CO ratio:R^2=1 and % E=12.53%. Istadi et al.[74] develop a hybrid artificial neural network-genetic algorithm (ANN-GA) model, to study the effect of process parameter on Noncatalytic dielectric barrier discharge (DBD) method for the production of C_2^+ hydrocarbons from carbon dioxide (CO_2) and methane (CH_4). The model gives good predictions with MSE=0.0034 and R =0.9938, 0.9955, 0.9877, 0.9978 for CH_4 conversion, C_2^+ selectivity, H_2 selectivity and C_2^+ yield respectively.

2.1.4 Biomass Productivity:

Hossain et al.[75] apply the comparison of three artificial intelligence (AI) modeling approaches including boosted regression tree (BRT), artificial neural networks (ANN), and support vector regression (SVR) to Study Bio fixation to enhance biomass productivity using air and CO_2 to support microalgae growth. Comparing results SVR outperforms all ML approaches in prediction with R^2=0.779, MAE=0.013, MARE=1.2587, RMSE 0.0296, FB=0.0061 for biomass productivity and R^2=0.911, MAE=0.0128, MARE=0.4131, RMSE=0.0189, FB=0.0088 for CO_2

fixation (RCO_2). Kushwaha et al.[76] introduced a novel prediction model to forecast the carbon dioxide (CO_2) fixation of microalgae, employing a hybrid approach that combines the adaptive neuro-fuzzy inference system (ANFIS) and genetic algorithm (GA). The analysis revealed that the GA-ANFIS model outperformed the standalone ANFIS model. The GA-ANFIS model demonstrated superior prediction capability, with RMSE, R^2, and AARD values of 0.00056, 0.98457, and 0.032156 respectively.

2.1.5 Lower Olefins(LO) Production

Chandana et al.[77] designed a machine learning (ML) framework to model and design catalysts for the direct hydrogenation of CO_2 to lighter olefins, focusing on the relationship between structural composition and operating parameters. The study employed artificial neural network (ANN) models using Bayesian-Regularization (BR) and Levenberg-Marquardt (LM) backpropagation learning algorithms to predict catalyst activity. These ANN models were then compared with other ML models, including linear, tree-based, and kernel-based approaches. Among the various ML models investigated, the ANN-BR model demonstrated the best performance in predicting CO_2 conversion and LO (lighter olefins) selectivity, showing minimal deviation from experimental data with R values of 0.90 and 0.8, RMSE values of 8.43 and 16.73, and AAD values of 5.8 and 9.5 for test data, respectively.

2.1.6 Methane Production

Yılmaz et al.[77] presented a novel random forest (RF) model to analyze an extensive dataset comprising 4051 data points from 527 distinct experiments collected from 100 published articles. The study focused on catalytic CO_2 methanation from CO_2 and H_2. The RF model demonstrated promising results in predicting CO_2 conversion, yielding a root mean square error (RMSE) of 12.7 and an R^2 value of 0.85.

Wiheeb et al.[78] develop an Artificial Neural Network (ANN) to study CO_2 Conversion in a falling film reactor. This study shows that the ANN model gives a very close estimation of CO_2

conversion. The average error as predicted by ANN was 0.6% which is within the acceptable level.

Alsaffar et al.[78] conducted a comprehensive investigation into the impact of process parameters on hydrogen production through catalytic methane dry reforming. Ten distinct ANN models were constructed, with variations in the number of hidden neurons ranging from 1 to 10. The model that exhibited the most favorable performance was the one with a topology of 4-9-2.

In short, the convergence of AI, especially ML, with industrial processes represents a significant technological advancement. It redefines how industries operate, enabling greater efficiency, sustainability, and innovation. Various researchers have integrated AI with methanol production. But to the best knowledge of the authors, no work has been reported in CO_2 hydrogenation to methanol product under uncertainty.

Chapter 3

Process Description and Methodology

In this section, a brief general overview of process description, ANN, GA, PSO, and methodology will be discussed.

3.1 Process description

In this process (CO_2 hydrogenation to methanol), two primary feed gases, carbon dioxide (CO_2) and hydrogen (H_2), are subjected to compression with intercooling prior to entering the adiabatic reactor. Within this reactor, a $Cu/ZnO/Al_2O_3$ is used catalyst to facilitate a chemical conversion, potentially increasing the selectivity of methanol product. Unreacted gases from the reactor are efficiently recycled, with a fraction released into the atmosphere to prevent impurity buildup. Following reactor operation, phase separation is employed to distinguish untreated gases from liquid condensate. This phase separation step is integral to the process, ensuring the efficient management of gas and liquid components.

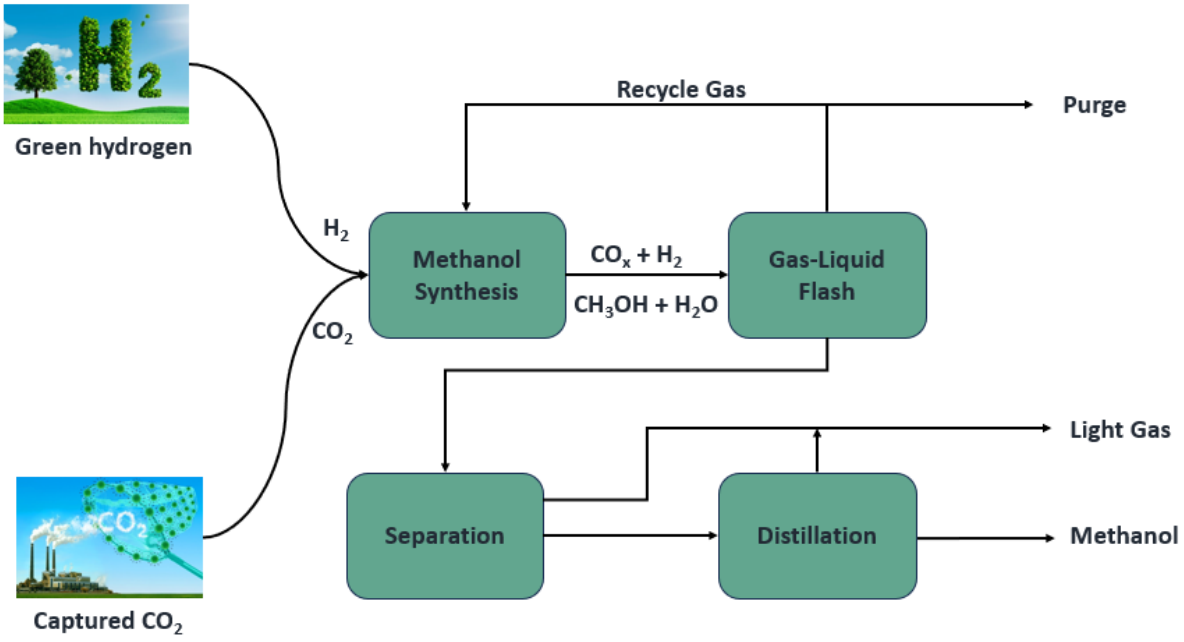


Figure 3: Bloc diagram of the process

The final crucial stage involves a distillation column. The crude methanol stream, obtained from prior phases, undergoes depressurization and further phase separation to eliminate any residual gases and water content. This comprehensive process results in the production of the desired methanol product while optimizing resource utilization and minimizing waste. Figure 3 represents an overview of the process.

3.2. Artificial neural networks

An artificial neural network (ANN) is a subset of machine learning algorithms that draws inspiration from the human brain for its computational model [79]. It consists of a network of interconnected neurons with predefined functions, arranged into layers known as the input, hidden, and output layers [80, 81]. They use artificial neurons to receive inputs, apply activation functions, and predict output. In situations where deriving precise analytical correlations for highly nonlinear phenomena is challenging, artificial intelligence techniques, especially ANNs, come into play [80]. They make predictions using real-world data without needing any relationship between inputs and outputs [82]. Figure 4 represents ANN's general structure.

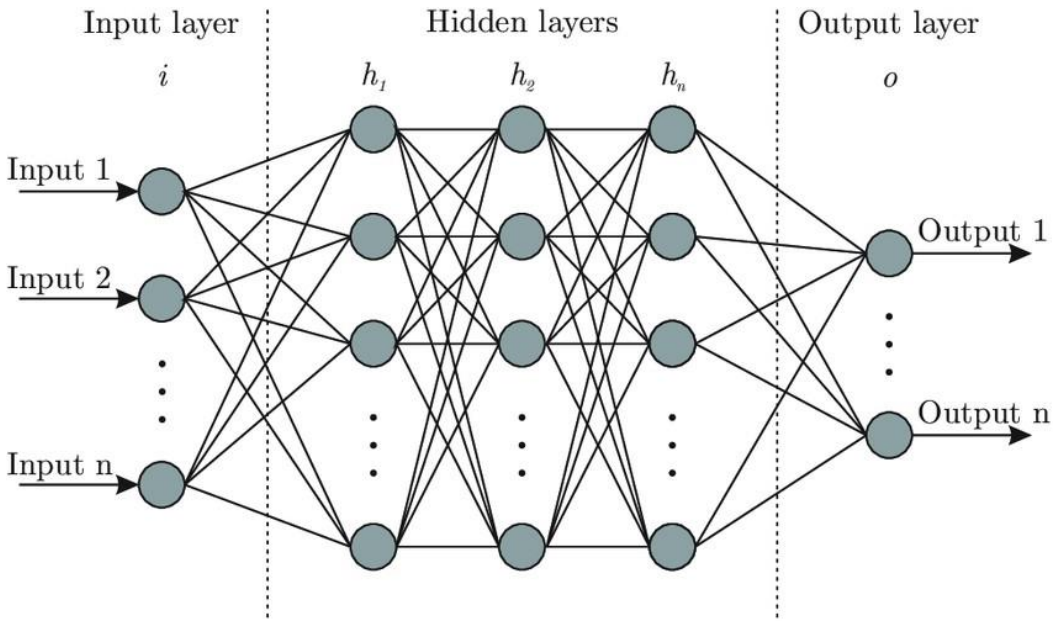


Figure 4: General ANN structure

3.2.1 The Levenberg-Marquardt Method

This method addresses nonlinear programming problems by reducing the sum of squared errors between data points and a model function. This minimization process is achieved by iteratively adjusting a set of parameters through a combination of Gauss-Newton and gradient descent updates, as expressed in equation (4).

$$[\boldsymbol{J}^{\top}\boldsymbol{W}\boldsymbol{J} + \boldsymbol{\lambda}(\boldsymbol{J}^{\top}\boldsymbol{W}\boldsymbol{J})\,]\boldsymbol{h}_{|\boldsymbol{m}} = \boldsymbol{J}^{\top}\boldsymbol{W}(\boldsymbol{y} - \hat{\boldsymbol{y}}) \qquad (\mathbf{4})$$

The process begins by varying these parameters in the steepest-descent manner, gradually reducing the total squared error using gradient descent. This method assumes that the least square function is approximately quadratic in the vicinity of the current parameter values. By minimizing this quadratic approximation, the Gauss-Newton approach effectively reduces the sum of squared errors.

The damping parameter $\boldsymbol{\lambda}$ plays a crucial role in the update process. A small $\boldsymbol{\lambda}$ leads to a Gauss-Newton update, while a large $\boldsymbol{\lambda}$ results in a gradient descent update. To ensure stability and convergence, $\boldsymbol{\lambda}$ starts with a large value, causing initial updates to be relatively small steps along the steepest-descent path. As the algorithm progresses and the solution improves, $\boldsymbol{\lambda}$ is gradually reduced, guiding the approach towards the Gauss-Newton method and moving closer to a local minimum.[83]

3.3 Genetic algorithm

The Genetic Algorithm (GA) is a bio-inspired optimization method that draws its inspiration from Charles Darwin's theory of evolution, initially proposed by Holland and later expanded upon by Goldberg and Holland [84, 85]**.** Genetic algorithms mimic nature's selection and inheritance to solve complex optimization problems. In GA, every parameter corresponds to a genetic element, and solutions are encoded as chromosomes. The process kicks off by establishing an initial population, wherein the fitness of each individual is assessed using an objective function. The algorithm proceeds to generate fresh populations iteratively through processes like selection, crossover, and mutation until it converges towards an optimal solution, guided by predefined termination criteria, such as a specified threshold for solution differences or a set number of iterations [82]. In short, GA is a meta-heuristic method that continuously refines a population of solutions, driven by principles of natural selection. It selects parents from

the current population based on fitness, produces offspring through crossover and mutation, and evolves toward the best solution until termination conditions are met [86**]**. These genetic operators, including selection, crossover, mutation, and elitism, guide GA iteratively to explore unvisited regions of the search space in pursuit of better solutions [87, 88]**.** Figure 5 depicts a general flowchart of a genetic algorithm, outlining the steps and processes within the algorithm.

3.3.1 Genetic Algorithm Operators

The functions of all the genetic operators are as follow:

3.3.1.1 Population

The initial population was randomly generated, and each conceivable solution is referred to as a chromosome, as demonstrated in Table 1.

$$P = \{p_1, p_2, \dots, p_{pop\_size}\} \quad (5)$$

$$p_i = \left[p_{i_1}\ p_{i_2}\ \cdots\ p_{i_j}\ \cdots\ p_{i_{no-vars}}\right] \quad (6)$$

$$para^{\,j}_{min} \leq p_{i_j} \leq para^{\,j}_{max} \quad (7)$$

Table 1: Chromosomes

Chromosome No. 1	1011000101110010
Chromosome No. 2	1001010110111001

In equation (5), pop_size represents the total population size, while in equation (6), no_vars denotes the number of variables to be optimized. The symbols $para^{j}_{min}$ and $para^{j}_{max}$ correspond to the minimum and maximum values of the parameter p_{i_j}.

3.3.1.2 Determination of Parents and Offspring

During the selection process, the algorithm identifies which chromosomes will serve as parents for reproduction and mating. Additionally, it determines the number of offspring that each selected chromosome will generate.

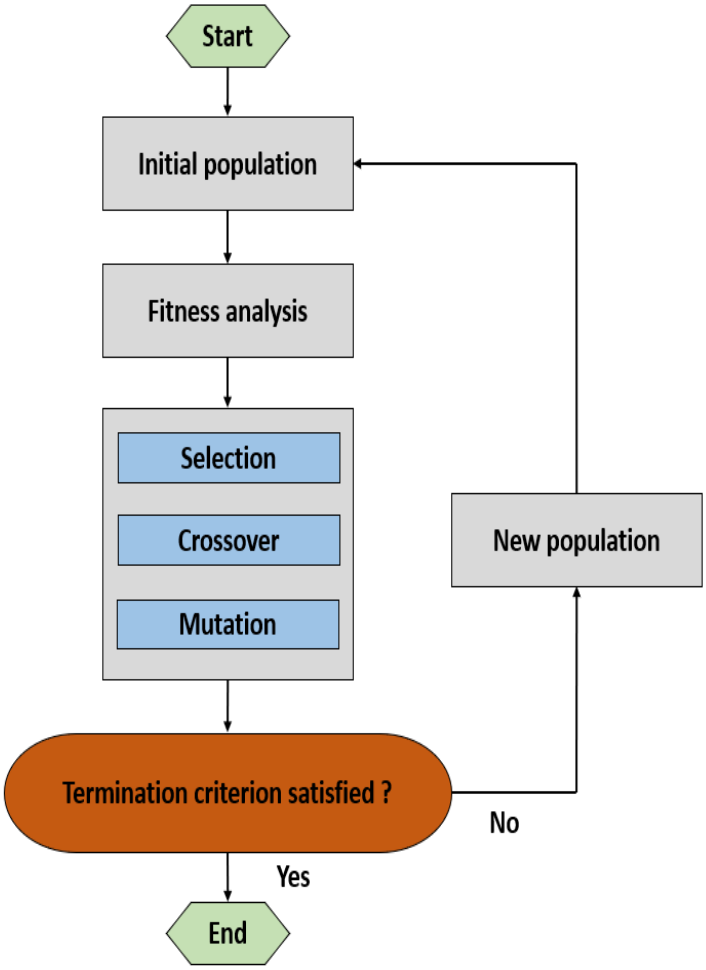


Figure 5: Schematic representation of Genetic Algorithm

Objective of Selection: The primary objective of the selection process is to give preference to individuals with higher fitness levels. This is often summarized by the principle that "the better an individual's fitness, the greater its likelihood of becoming a parent [89]. Several well-known selection methods are as follows:

Tournament Selection: This method is widely regarded as one of the most common and efficient techniques in the field of genetic algorithms due to its simplicity and effectiveness [90]. The tournament selection process involves randomly selecting individuals from the broader population. These selected individuals then engage in a competition to determine which one possesses the highest fitness value. The victor of this competition is chosen as a parent for the

next generation. Typically, individuals compete in pairs, forming binary tournaments or "tournament size." Tournament selection ensures diversity by offering an equal opportunity to all individuals, although it may slightly slow down convergence. Notably, tournament selection is adept at utilizing computational resources, particularly when implemented in parallel. It also demonstrates resilience against domination by a few individuals, thus enhancing its robustness. Moreover, it eliminates the need for fitness scaling or sorting procedures [91].

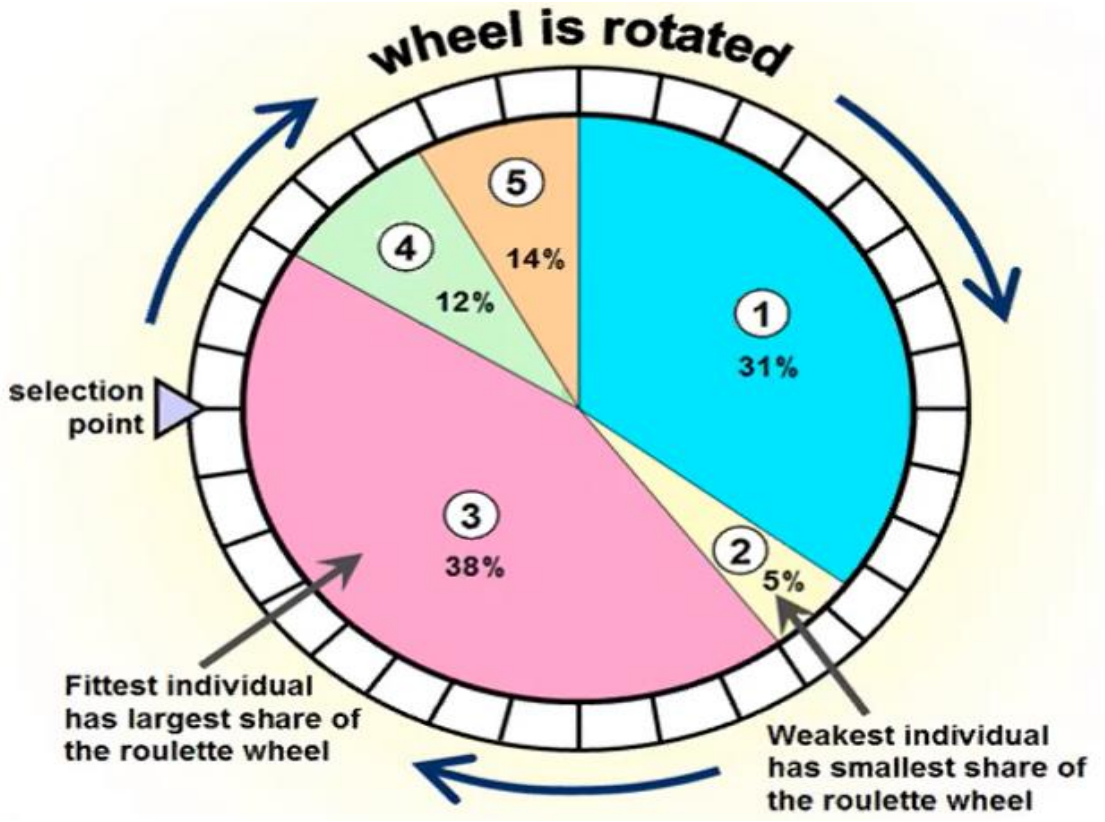


Figure 6: Roulette wheel selection

Proportional Roulette Wheel Selection: In this method, possible solutions are represented as segments on a roulette wheel, with the size of each segment determined by their respective fitness values. The wheel is then spun randomly to select the solutions that will participate in the creation of the next generation, as depicted in Figure 6. Rank-based selection is a variation of this

approach where individuals are assessed based on their ranks rather than their absolute fitness values, ensuring that every individual has a chance of being chosen [92].

Rank Selection: It is employed for parent selection, involving the utilization of a ranking mechanism. Within this context, the fitness value is utilized to assign rankings to individuals within the population. The individual with the highest fitness receives the highest rank (n), while the lowest-ranked individual is assigned a rank of 1. Each chromosome's ranking is determined based on its expected value [93].

3.3.1.3 Crossover

This process involves merging the genetic data from two or more parents to generate offspring. Genetic algorithms commonly employ crossover operators such as single-point, double-point, and uniform. In Single-Point Crossover, a random crossover point is designated, and the genetic information of two parents is exchanged beyond that specific point, as visually represented in Figure 7 [86].

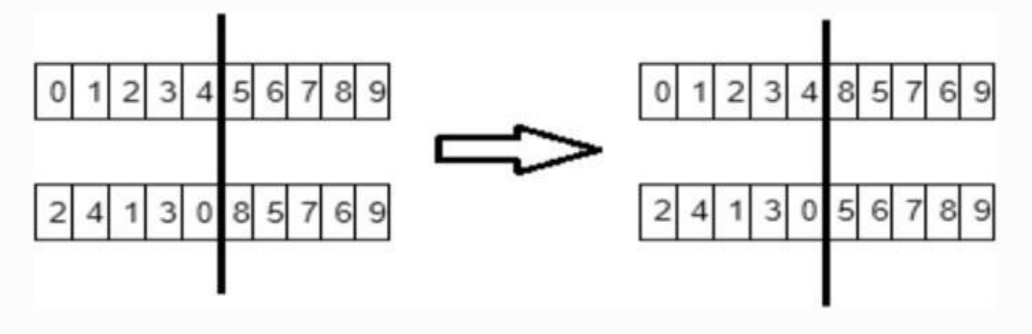


Figure 7: Single point crossover

Double Points Crossover: In this method, entails the random selection of two or more crossover points. The exchange of genetic information between parents occurs based on the segments created, as exemplified in Figure 8 [86].

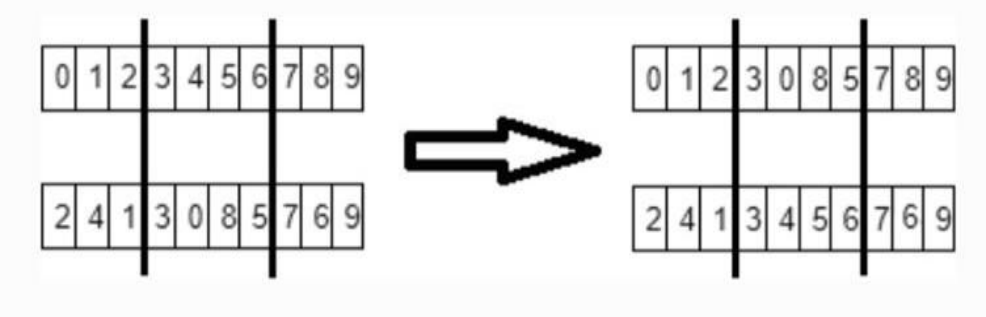


Figure 8: Double points crossover

Uniform crossover: The parental individual cannot be divided into separate segments. Instead, each parent is considered to represent each gene independently. The choice of whether to swap a gene with its corresponding gene at the same position in another chromosome is determined through a random process, as depicted in Figure 9 [86].

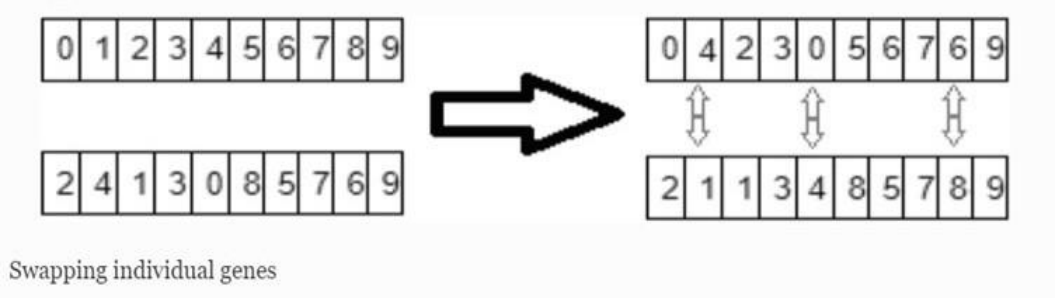


Figure 9: Uniform crossover

3.3.1.4 Mutation

Mutation plays a vital role in preserving genetic diversity from one generation to the next. During the mutation process, genes within the chromosomes undergo changes. This alteration can lead to variations in the characteristics of chromosomes inherited from their parents. Notably, the mutation process generates three additional offspring [94]**.** In practice, within the Genetic Algorithm (GA), this operator prevents solutions from becoming identical and enhances the likelihood of avoiding local optima. Refer to Figure 10 for a conceptual depiction of this

operator. After the crossover (replication) stage, minor modifications in some randomly selected genes can be observed in the diagram [95].

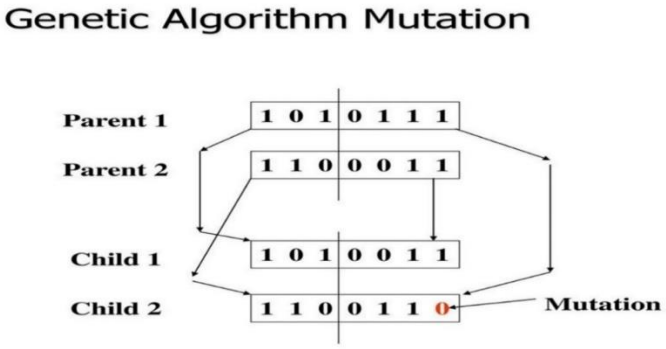


Figure 10: After the crossover phase, the mutation operator changes one or more genes in the children's solutions

3.4.Particle Swarm Optimization

Particle swarm optimization (PSO) is a robust exploration technique that draws inspiration from the collective movement patterns observed in birds and fish. [96]. It was initially developed by Kennedy and later refined by Clerc [97]. In PSO, a group of particles explores a search space, and in each step, the algorithm assesses how well each particle is positioned. Particles are drawn toward the best positions discovered within the group. This cooperative exploration continues, with particles adjusting their positions based on their neighbors and using a defined fitness function to guide their movement [98, 99].

One of the notable strengths of PSO is its rapid convergence, making it stand out among other evolutionary algorithms. This adaptability has led to its application in various engineering challenges [100]. PSO primarily employs a population-based approach, where random particles explore the search space, continually updating their positions. Each particle independently seeks

optimal solutions while considering the positions of nearby particles and using a fitness function as a guide [96, 101].

3.5 Surrogate Model

A surrogate model, also referred to as a meta-model, serves as a valuable analytical tool for establishing a statistical link between the input and output behaviors of complex systems. These surrogate models are generally categorized into two main types based on their approach to approximation: (i) model-driven and (ii) data-driven, often called black box models. Within the model-driven framework, known as the Reduce Order Model (ROM), the primary aim is to reduce computational costs by employing lower-order equations for approximating the original equations. However, it's important to note that implementing this approach often necessitates access to the source code of the simulation, a requirement that is typically unattainable when using commercial software. In contrast, in the data-driven surrogate model approach, the primary emphasis is on constructing the model using input data and their corresponding output responses. The development process of a surrogate model involves several well-defined steps:

- Sampling the Design Space: The initial phase involves systematically sampling the design space to identify the input parameters from various datasets.
- Executing Simulations or Conducting Experiments: Subsequently, the simulator is run, or experiments are carried out to generate the outcomes corresponding to the input parameters.
- Selecting and Training the Surrogate Model: A surrogate model is chosen and then trained using the available training data, which includes both input and output.
- Evaluating Model Performance: The model's performance is rigorously assessed based on test data. If the model's accuracy falls short of expectations, the entire process is reiterated, commencing from the initial design space sampling.

In essence, a surrogate model fulfills the essential role of establishing a statistical link between the input parameters and the corresponding system outputs. These surrogate models are broadly divided into two types: data-driven and model-driven. The model-driven approach, or ROM, seeks to reduce computational costs through the application of

lower-order equations, while the data-driven approach relies on input-output data to construct the model. The process of creating a surrogate model involves systematic design space sampling, simulator execution or experimentation, the selection and training of the surrogate model, and ongoing evaluation to ensure accuracy[102].

3.6 Methodology

The methodology used in this study is summarized in Figure 11 and is briefly detailed below.

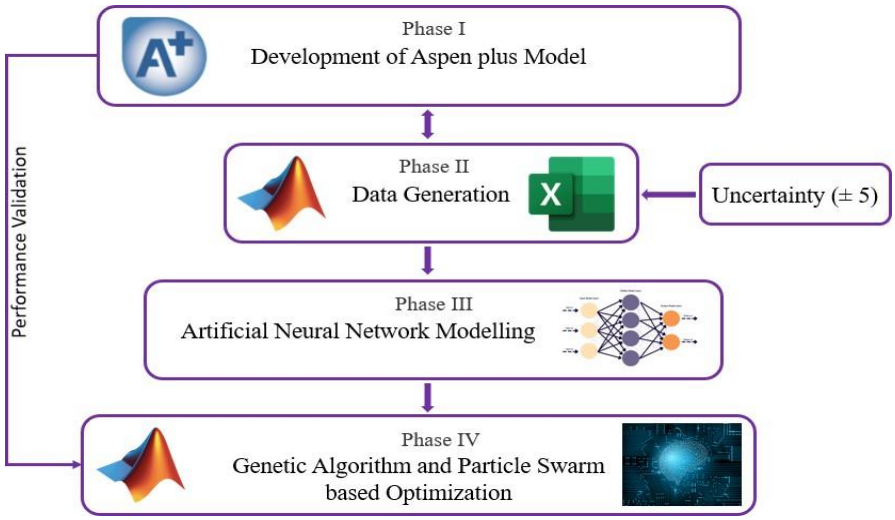


Figure 11: Methodology

Phase I: First principle (FP) for CO_2 hydrogenation into methanol was developed in Aspen plus with a little modification by changing the heat exchanger with heater [8].

Phase II: Aspen plus model was integrated with Matlab to shift the steady state model into dynamic mode. A total of 3880 datasets were generated by introducing ±5 % uncertainty in the steady state process input variables. i.e., mass flow rates, pressures, temperatures, split fraction, RR, and BR.

Phase III: Matlab 2022a was used to build and validate the ANN model. Model selection, training, and validation was part of the modeling.

- **Model selection:** A feed-forward neural network was chosen and trained using the Levenberg-Marquardt back propagation (trainlm) algorithm. The dataset was divided,

with 80% of the samples designated for training, and the rest 20% was evenly split between testing and validation of the model.

- **Training and validation:** The model's performance was calculated by two measures: root-mean-squared error (RMSE) and correlation coefficient (R). These values were determined using the following equation (8) and (9).

$$RMSE = \sqrt{\frac{1}{n}\sum_{i}^{n}\left(Y_i^{exp} - Y_i\right)^2} \qquad (8)$$

$$R = 1 - \frac{\sum_{i=0}^{n}(Y_i^{exp} - Y_i)}{\sum_{i}^{n}(Y_i^{exp} - Y_{avg}^{exp})} \qquad (9)$$

Y_i^{exp} and Y_i stand for the actual and the expected outcomes results, and n indicates the total test samples.

The RMSE (Root Mean Square Error) is always a non-negative value, and a smaller RMSE indicates more accurate model predictions. The coefficient of determination, typically denoted as R-squared, falls within the range of 0 to 1. A value of 0 suggests that the output variable cannot be effectively predicted from the regressor variables, while a value of 1 signifies that the response variable is entirely predictable from the regressor variables.

Phase IV: The ANN was used as a surrogate in a GA and PSO framework to optimize a system under uncertainty, with the goal of optimizing the methanol flow rate at the product stream. The ideal settings that resulted in the methanol flow rate at the product stream were determined using the GA and PSO methods. The proposed optimization's effectiveness was confirmed through the utilization of the optimized outcomes in the Aspen Plus model.

Chapter 4

Results and Discussion

In this segment, Section 4.1 focuses on Aspen model development, Section 4.2 on the ANN Model, and Section 4.3 on GA and PSO based optimization and the ANN utilization as surrogate components within GA and PSO frameworks.

4.1 Aspen Model

For this study, a model proposed by Van-Dal et al. [9] was regenerated by using the commercial software Aspen Plus V11 and was used to design and simulate the process. RedlicheKwonge Soave equation of state with modified HuroneVidal mixing rules (RKSMHV2) was used to calculate the thermodynamic properties of streams at high pressure (>10 bar). For streams at low pressure (<10 bar), NRTL-Rk model was employed.

Carbon dioxide (CO_2) with a flow rate of 88.0 t/hr was introduced at a pressure of 1 bar and a temperature of 250C, and then it underwent a four-stage compression network (CP-1, CP-2, CP-3, and CP-4) to elevate its pressure to 78 bar. Throughout the compression process, there is intercooling(HX-1, HX-2, and HX-3), and the heat extracted can be redirected to the CO_2 capture plant. Simultaneously, hydrogen was supplied to the methanol production plant with a flow rate of 12.1 t/hr at a pressure of 30 bar, and the same temperature of 250C. This hydrogen stream was compressed to 78 bars using a single-stage compressor (CP-5). The hydrogen and CO_2 gases were then carefully mixed in a dedicated mixer (MIX-1) and subsequently combined with a recycled stream (stream 19) in another mixer (MIX-2). The resulting mixed feed (stream 12) was heated to a temperature of 2100C and then introduced into the fixed bed adiabatic reactor. This reactor was packed with 44,500 kg of a commercial $Cu/ZnO/Al_2O_3$ catalyst. The output stream (stream 14) exiting the reactor was to preheat the feed of the distillation column (stream 25) to 80^0C in a heat exchanger (HX-5), and then its temperature was further reduced to 35^0C in the heat exchanger (HX-6).

Furthermore, in the flash tank (KO-1), untreated gases (stream 17) are effectively separated from the water-methanol condensate (stream 20). A minor portion (1%) of the un- treated gases (PURGE) is released into the atmosphere to prevent the buildup of impurities in the system. The

remaining 99% of the untreated gases were compressed back to 78 bars in CP-6 and returned to the process as a recycle stream. The liquid stream exiting the flash tank (KO-1), referred to as crude methanol, was de- pressurized to 1.2 bar using two valves (VLV-1 and VLV-2) and subsequently introduced into another flash tank (TKFL1) to eliminate any residual gases completely. The resulting stream was then heated to 800C in the heat exchanger (HX-5). The heated stream (stream 25) was further directed to a distillation column (DT-1), where water (stream 27) was extracted from the bottom at a temperature of 1020C, with minimal methanol content.

The desired methanol product (stream 26) was obtained from the top of the column at a pressure of 1 bar and a temperature of 64.060C, in gaseous form with small amounts of water and some unreacted gases. Methanol was compressed to 1.2 bar using CP-7 (to accommodate the pressure losses in the following equipment) and then cooled to 400C in heat exchanger HX-8. At this point, the methanol is directed to a second flash tank (KO-2), where non-reacted gases exit from the top (stream 31), while the final methanol product emerges from the bottom (stream 30) in liquid form. A detailed schematic for the process is given in Figure 12.

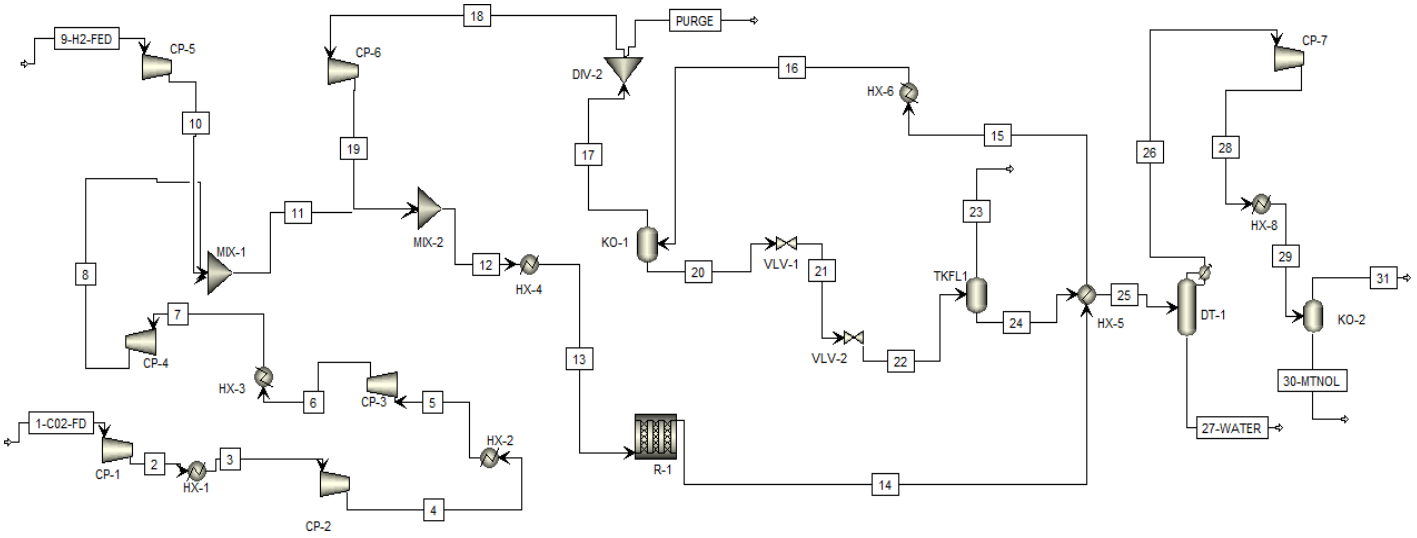


Figure 12: Process flowsheet of CO_2 hydrogenation into Methanol [9]

To assess the model's validity, a parallel investigation was conducted concerning similar methanol production processes. The findings revealed that the overall CO2 levels in this study

closely aligned with the literature data reported in Table 2, demonstrating a high degree of concordance with previous research.

Likewise, an inquiry into the MeoH (% w) purity within analogous processes was under- taken by consulting relevant literature, and the results are elegantly presented in Table 3. This rigorous comparison serves to underscore the robustness and credibility of our study.

Table 2: Overall CO_2 conversion of similar MeoH processes

	[9]	[103]	[104]	[105]	[106]	[107]	Current Work
CO_2 conversion [%]	93.4	95.2	98.4	93.9	99.7	97.3	95.4

Table 3: Produced MeoH (%w) purity

	[103]	[104]	[106]	[107]	[108]	Current Work
MeoH (%w) purity	100	99.8	99.04	99.9	99.9	99.04

The kinetic used in this paper is that of the Rearranged model of Vanden Bussche and Froment [109] as presented in Van-Dal and Bouallou [9] where pressure is expressed in Pa and temperature is expressed in K. For these kinetic models, the Reaction rates are given by equations 10 and 11, and the Parameters of the rearranged kinetic model are given in Table 4.

Methanol generation:

$$r_{CH3OH} = \frac{K_1 P_{CO2} P_{H2} - K_6 P_{H2O} P_{CH3OH} P_{H2}^{-2}}{(1 + K_2 P_{H2O} P_{H2}^{-1} + K_3 P_{H2}^{0.65} + K_4 P_{H2O})^3} \quad \left[\frac{Kmol}{Kg_{cat} s}\right] \qquad (10)$$

Reverse water gas shift reaction:

$$r_{RWGS} = \frac{K_5 P_{CO2} - K_7 P_{H2O} P_{CO} P_{H2}^{-1}}{(1 + K_2 P_{H2O} P_{H2}^{-1} + K_3 P_{H2}^{0.65} + K_4 P_{H2O})} \quad \left[\frac{Kmol}{Kg_{cat} s}\right] \qquad (11)$$

Table 4: Parameters of the rearranged kinetic model

K_1	A_1	-29.87
	B_1	4811.2
K_2	A_2	8.147
	B_2	0
K_3	A_3	-6.452
	B_3	2068.4
K_4	A_4	-34.95
	B_4	14928.9
K_5	A_5	4.804
	B_5	-11797.5
K_6	A_6	17.55
	B_6	-2249.8
K_7	A_7	0.131
	B_7	-7023.5

4.2 ANN Model

The ANN model was constructed using MATLAB 2022a. An uncertainty of ±5% was introduced into the 12 process variables documented in Table 5. A comprehensive dataset comprising 3880 data samples was meticulously generated. Among these, 3104 data points were allocated for training the model, another 388 for validation purposes, and an additional 388 for testing the model's performance. The training of the ANN was carried out employing the Levenberg-Marquardt backpropagation (trainlm) training algorithm, while the behavior of the neural network was governed by the utilization of the Tansig activation function. The ANN gives good results with an R-value of 0.994 and RMSE of 28.59 as shown in Figure 13.

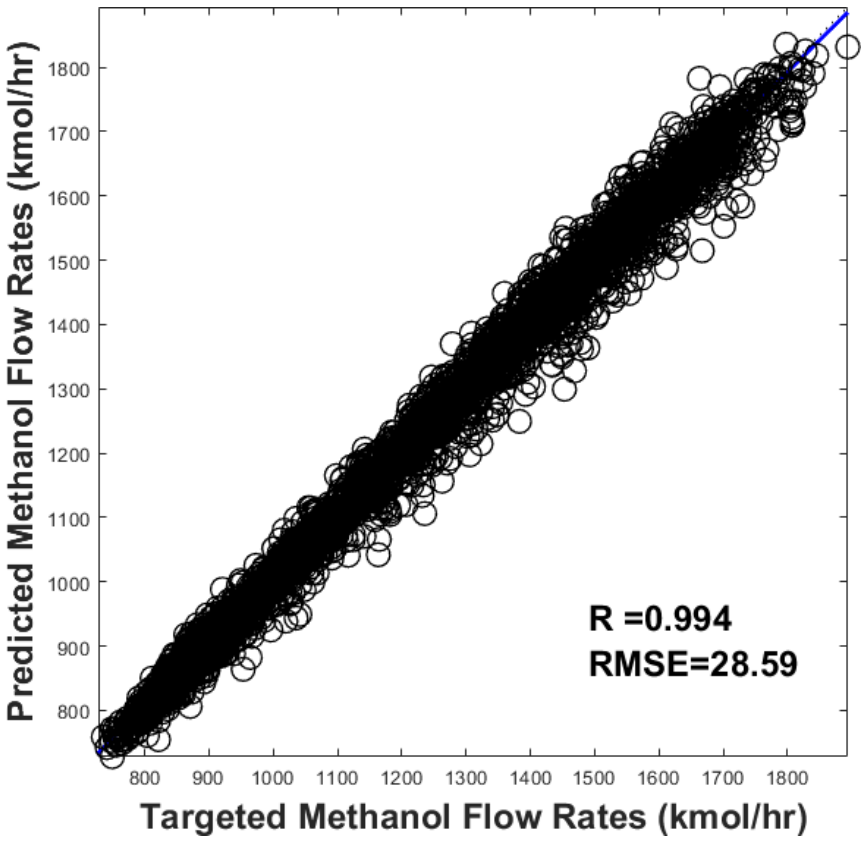


Figure 13: Targeted vs predicted flowrate of ANN during training

Table 5: Steady State conditions

Process Input variables											
Mass Flow Rate(tons/hr)		Temperature (^{o}C)				Pressure (bar)			Split fraction	RR	BR
C02-FEED	H2-FEED	HX-4	KO-1	TKFL1	HX-5	HX-4	KO-1	TKFL1	DIV-2	DT-1	
88	12.1	210	35	22	80	75	73.4	1.2	0.001	1.2	0.6

In addition to the automatic allocation of data samples by the ANN for training and testing purposes, an extra set of 125 data samples (referred to as test samples) was intentionally reserved and kept unknown from the model. This was done to evaluate the model's ability to generalize, as illustrated in Figure 7. For these predictions of methanol flow rates,the ANN model

achieved a remarkable correlation coefficient (R) of 0.995, coupled with a root mean square error (RMSE) of 26.83.

Thus, it is evident from the visual representations in Figure 6 and Figure 14 that both illustrations exhibit a remarkable degree of consistency, not only for the known databut also for the unknown data presented to the ANN. This compelling agreement underscores the robust predictive capabilities of our ANN model.

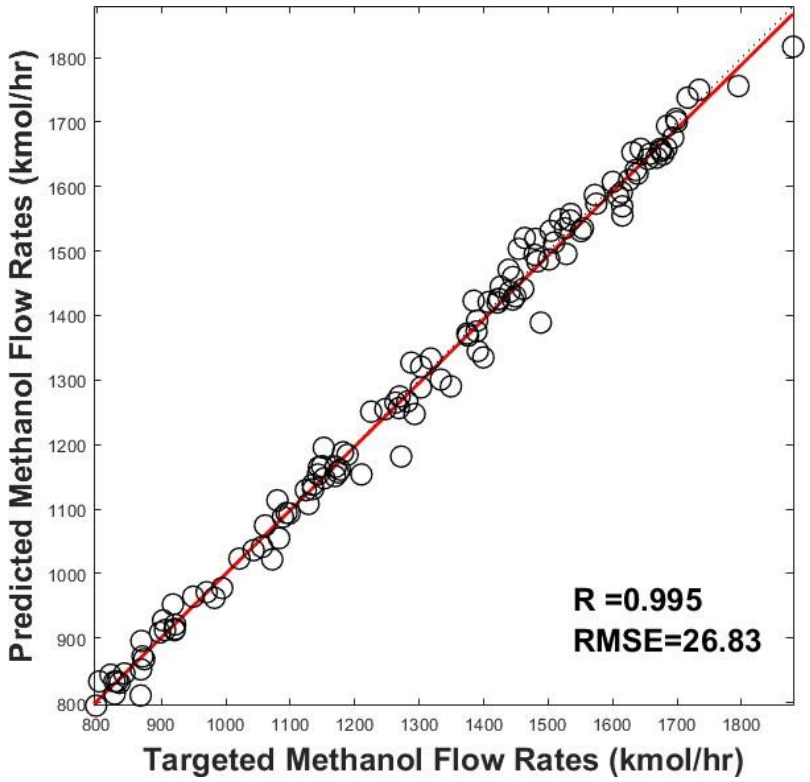
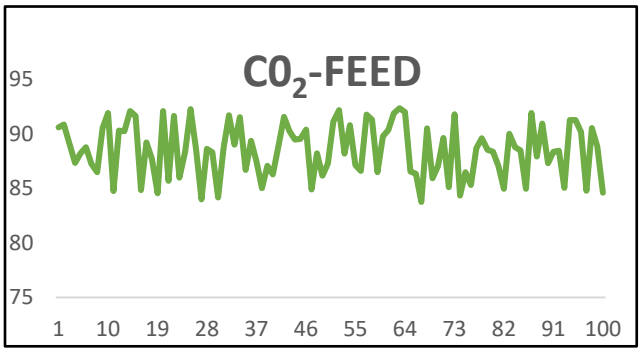
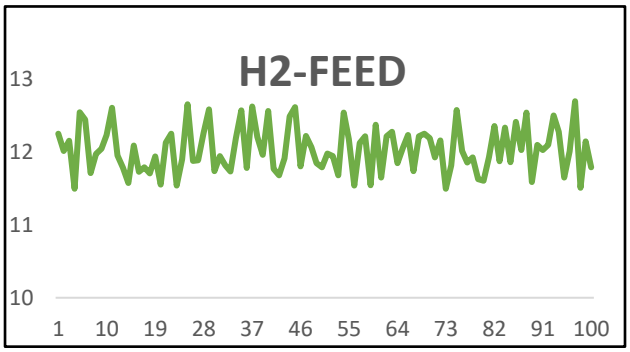


Figure 14: Targeted vs predicted flowrate of ANN during testing.

All the 12 graphs in figure 15 give a graphical representation of uncertainty that we bring in the 12 input variables of the process mentioned in table 5 in order to transfer the Aspen plus model from steady state to dynamic state.


C0$_2$-FEED
95
90
85
80
75
1 10 19 28 37 46 55 64 73 82 91 100


H2-FEED
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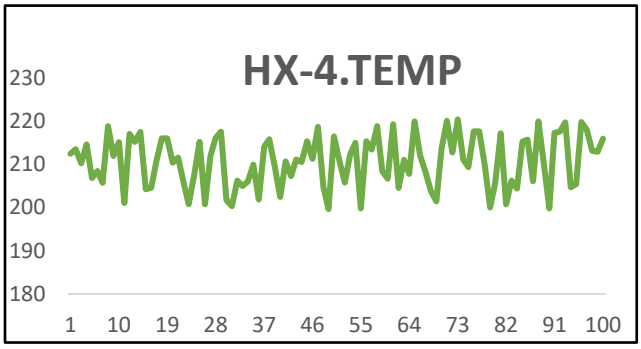
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210
200
190
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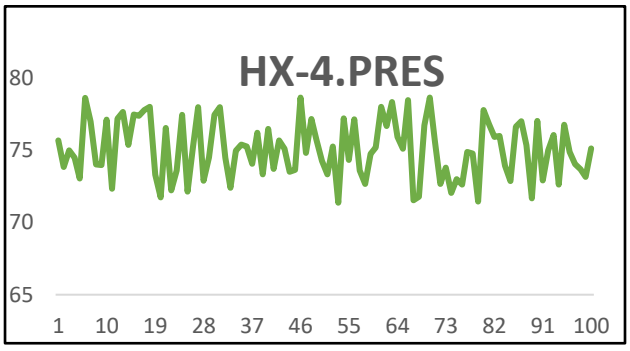
1 10 19 28 37 46 55 64 73 82 91 100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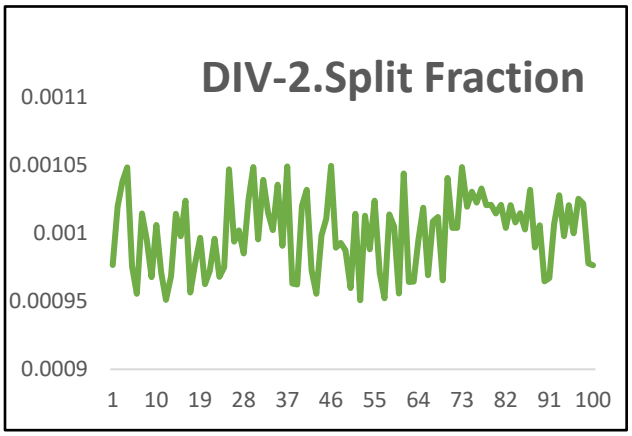
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0.001
0.00095
0.0009
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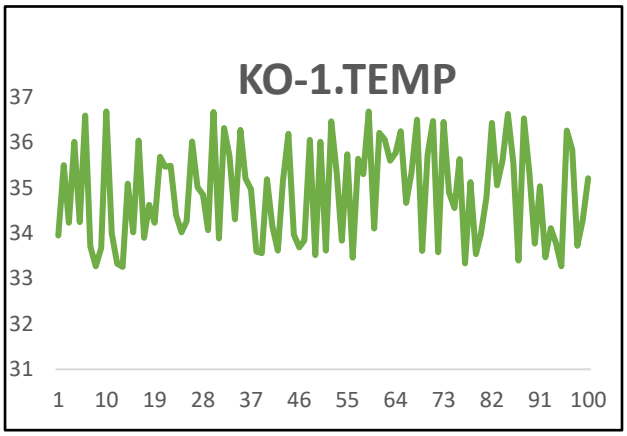
KO-1.TEMP
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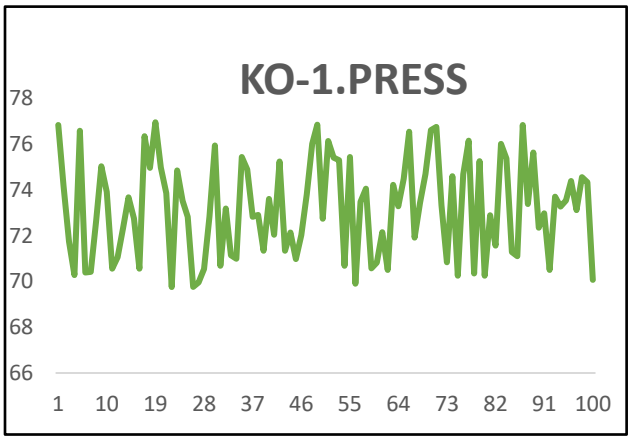
1 10 19 28 37 46 55 64 73 82 91 100


KO-1.PRESS
78
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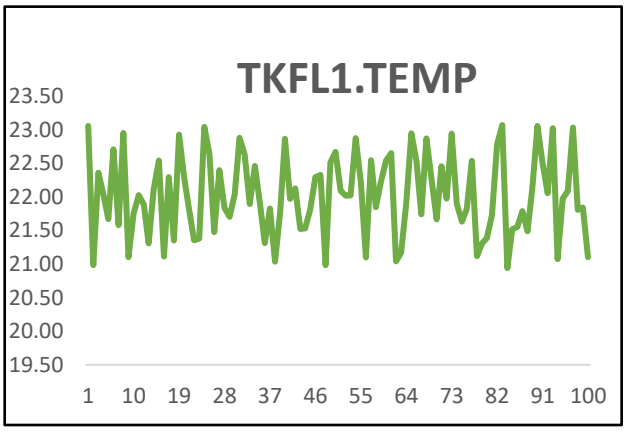
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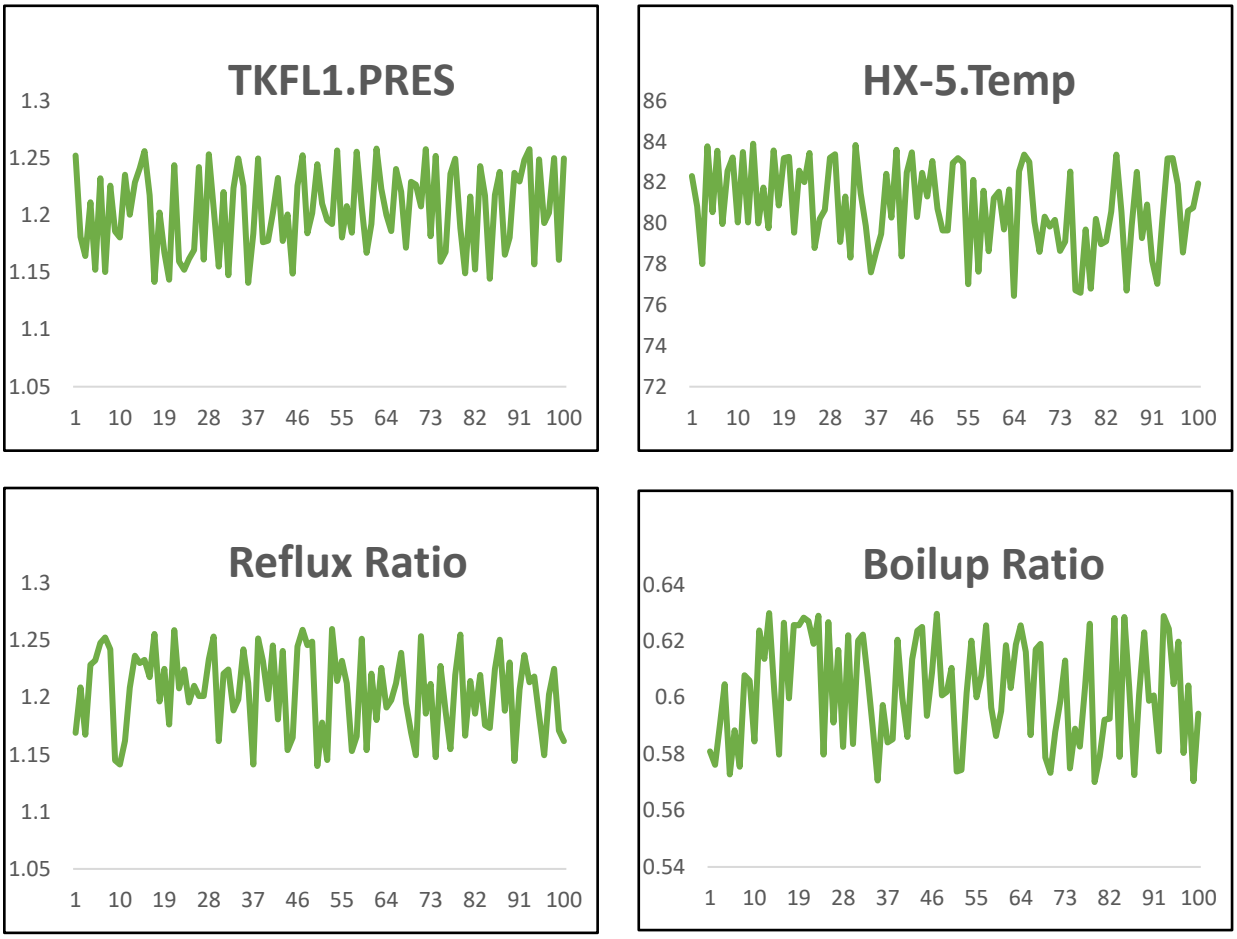

Figure 15: Graphical representation of Uncertainty in 12 Process input variables for 100 datasets lying on X-axis with an input variable fluctuating on Y-axis

4.3 Genetic algorithm and particle swarm based optimization

GA and PSO employ an ANN-trained model as a surrogate to enhance the optimization of Methanol flow rates under conditions of uncertainty. Table 6 provides a comprehensive comparison of the Methanol flow rates produced by the process for three different frameworks: Standalone (SA), GA, and PSO. The SA model refers to the Aspen first principle (FP) model without any optimization under uncertainty. Remarkably**,** both the GA and PSO-based frameworks consistently outperform the SA model across all test data samples in terms of Methanol flow rates.

For instance, in data sample 1, the SA model predicts a Methanol flow rate of 1479.02 kmol/hr, while the GA and PSO optimizations significantly improved methanol flow rates to 1794.94

kmol/hr and 1867.45 kmol/hr, respectively. A similar trend is observed in data sample 2, where the SA model estimates a Methanol flow rate of 1317.86 kmol/hr, but the GA and PSO optimizations elevate it to 1754.62 kmol/hr and 1813.76 kmol/hr, respectively. This consistent enhancement in performance underscores the effectiveness of both GA and PSO in optimizing Methanol flow rates under uncertainty.

Table 6: Comparison of SA, GA, and PSO for Methanol stream(product)

	SA Methanol Stream flow(kmol/hr)	GA optimized Methanol Stream flow(kmol/hr)	PSO optimized Methanol Stream flow(kmol/hr)
Data Sample 1	1,479.02	1,794.94	1,867.45
Data Sample 2	1,317.86	1,754.62	1,813.76
Data Sample 3	1,438.58	1,851.68	1,885.78
Data Sample 4	1,668.18	1,784.50	1,786.39
Data Sample 5	1,389.80	1,805.58	1,792.01
Data Sample 6	1,614.92	1,825.18	1,837.57
Data Sample 7	1,317.86	1,754.62	1,823.76
Data Sample 8	1,528.23	1,775.15	1,822.39
Data Sample 9	1,461.28	1,829.78	1,878.34
Data Sample 10	1,448.87	1,815.86	1,777.78

The effectiveness of our framework was assessed through a cross-validation process. Specifically, by feeding the Aspen model with optimized process conditions obtained via both the GA and PSO techniques. The goal was to determine the absolute error and % increase in methanol flow rate associated with each approach. In Table 7, you can observe a performance evaluation comparing the GA and PSO models.

Table 7: GA and PSO performance validation

Data Sample	GA optimized	Aspen model	Absolute Error (%)	% increase	PSO optimized	Aspen model	Absolute Error (%)	% increase
1	1,794.94	1,834.45	2.15	21.36	1,867.45	1,864.16	-0.18	26.26
2	1,754.62	1,758.26	0.21	33.14	1,813.76	1,812.08	-0.09	37.63
3	1,851.68	1,839.69	-0.65	28.72	1,885.78	1,863.52	-1.19	31.09
4	1,784.50	1,755.62	-1.64	6.97	1,786.39	1,780.63	-0.32	7.09
5	1,805.58	1,804.32	-0.07	29.92	1,792.01	1,815.21	1.28	28.94
6	1,825.18	1,814.48	-0.59	13.02	1,837.57	1,814.41	-1.28	13.79
7	1,754.62	1,758.26	0.21	33.14	1,823.76	1,812.08	-0.64	38.39
8	1,775.15	1,773.61	-0.09	16.16	1,822.39	1,807.20	-0.84	19.25
9	1,829.78	1,841.64	0.64	25.22	1,878.34	1,863.80	-0.78	28.54
10	1,815.86	1,795.53	-1.13	25.33	1,777.78	1,758.19	-1.11	22.70

Upon reviewing Table 6, it becomes apparent that PSO holds a slight advantage over GA in terms of performance. For instance, in the first data sample, PSO demonstrates an absolute error of -0.18% and an increase in Methanol flow rate of 26.26%, while GA exhibits a larger absolute error of 2.15% and an increase in Methanol flow rate of 21.36%. Similarly, in the second data sample, PSO's absolute error stands at -0.09% and an increase in the methanol flow rate of 37.63%, whereas GA's absolute error is notably higher at 0.21% and an increase in the methanol flow rate of 33.14%. Thus, these comparisons show that using PSO not only gives accurate prediction but also results in an increased methanol flow rate that is higher than GA during optimization. Figures 16 and 17 give a comparison of SA with GA and PSO, having the highest absolute error(%) of 2.15 and 1.28 for GA and PSO respectively. Figure 18 gives a comparison of the Percentage(%) increase in original value by GA and PSO with PSO clearly outperforming GA. Figure 19 gives an Absolute error Comparison of SA and PSO clearly showing that GA has low overall absolute error as compared to PSO.

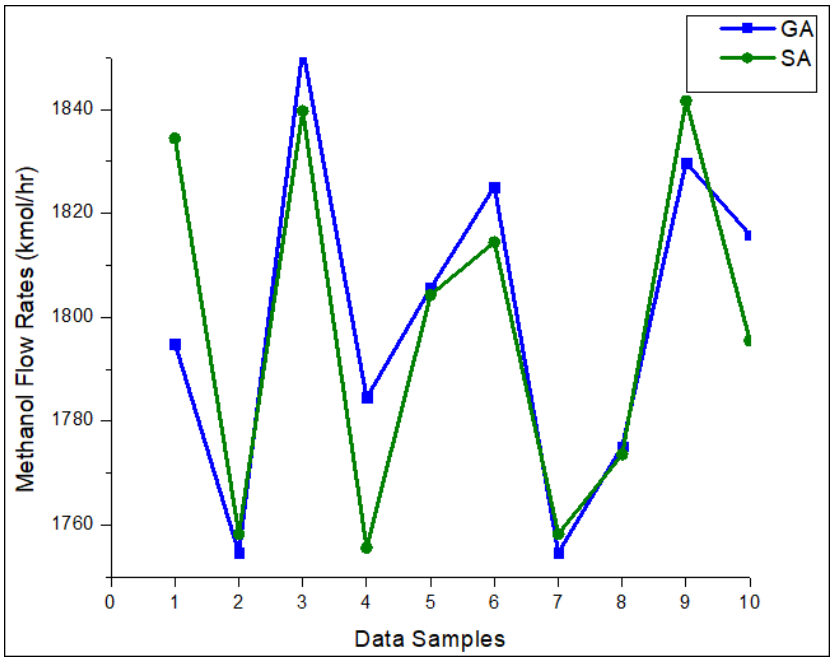

Figure 16: Comparison of SA and GA

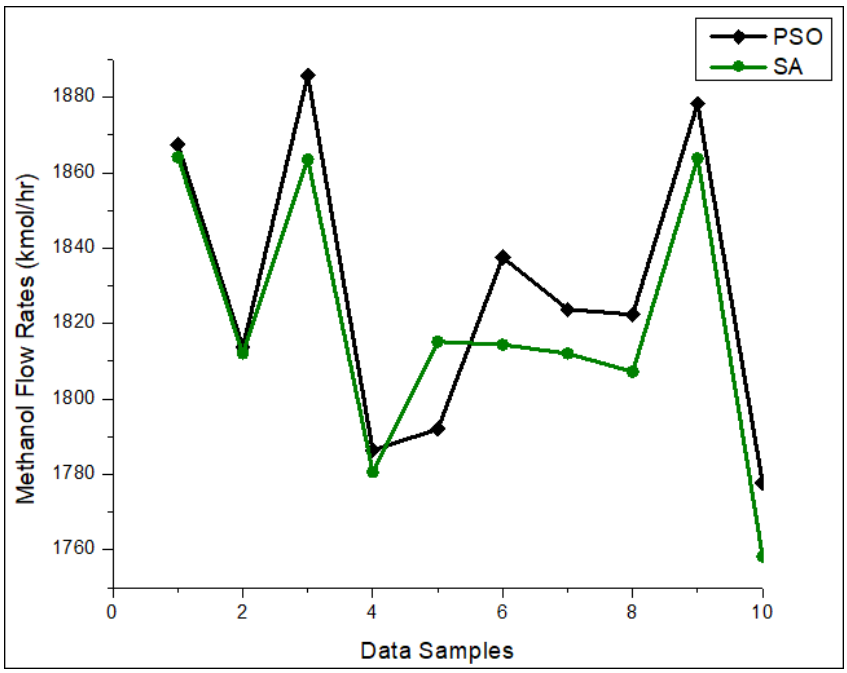

Figure 17: Comparison of SA and PSO

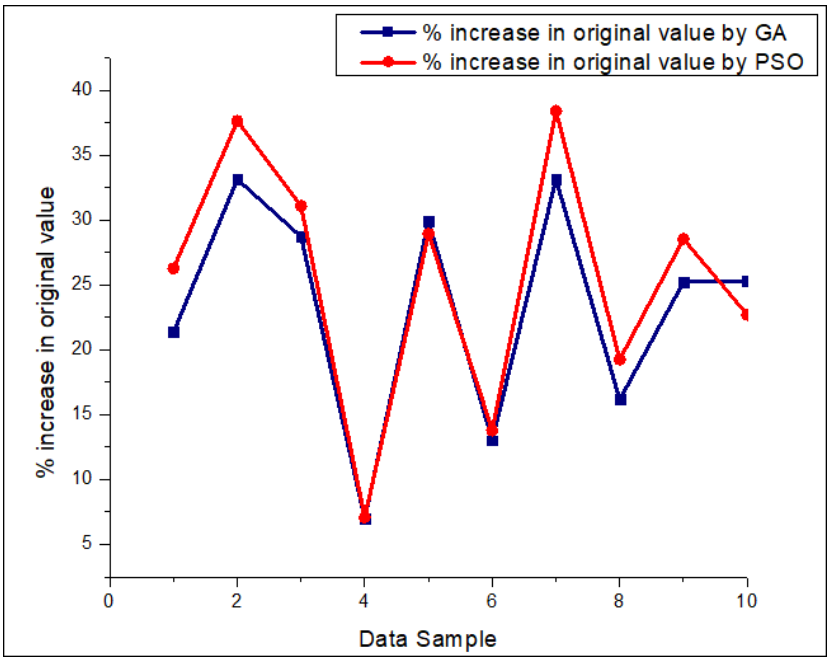

Figure 18: % increase comparison of GA and PSO

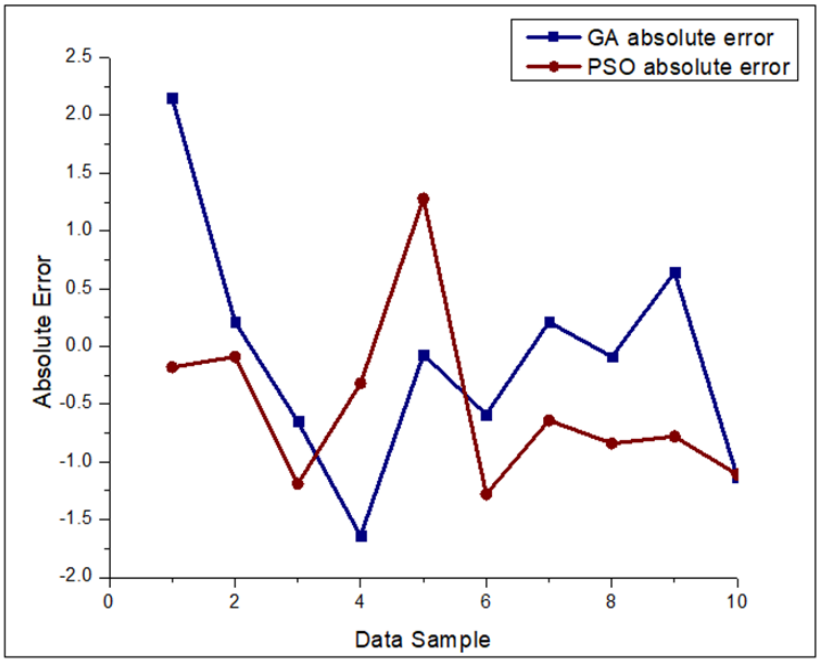

Figure 19: Absolute error Comparison of SA and PSO

Conclusions

This research paper delves into the realm of artificial neural networks (ANN) as a means to optimize methanol flow rate under uncertainty. The study's methodology involves a systematic approach, starting with the creation of a first-principal model for CO_2 hydrogenation into methanol. To accommodate the uncertainties inherent in real-world processes, the study integrates Aspen Plus and MATLAB, transitioning from steady state to dynamic modeling. The resulting ANN model proves its ability by achieving remarkable predictive accuracy with a correlation coefficient(R) of 0.996 and a root mean square error(RMSE) of 26.81 during cross-validation for unseen data. This ANN model, now a reliable surrogate, takes center stage in the genetic algorithm (GA) and particle swarm optimization (PSO) frameworks, consistently enhancing methanol flow production despite the presence of uncertainty. PSO,in particular, emerges as a slightly superior optimization method. In practical terms, checking if the optimized conditions work well in the Aspen Plus model confirms that this methodcan be used in the real world. This research shows how powerful artificial intelligence can bein making complicated chemical processes better. It combines basic modeling(First principle), ANN surrogate models, and smart optimization algorithms, providing a strong base for using data-driven methods to improve the monitoring, prediction, and control of complex chemical processes under uncertain conditions. This way of doing things could change how efficient and eco-friendly chemical production is.

Looking forward to this, there are several promising directions for future research of this work. One path is to use real data from actual plants instead of the Aspen model. This will help ussee how well this approach works in real industrial settings and deal with uncertainty in practice. Another area to investigate is to explore the performance of alternative machine learning algorithms beyond ANN, such as support vector machines and Gaussian processes, in surrogate modeling for optimization under uncertainty. Moreover, expanding the dataset to include additional process variables and levels of uncertainty could lead to more robust ANN models. Advanced control strategies could be designed based on the integrated Aspen-ANN model to enhance the regulation of methanol production amidst dynamic uncertainty. This research can also be extended to optimize other chemical production processes involving CO_2 utilization under uncertain conditions, making it a versatile contribution to the field.

Finally, the implementation of real-time optimization on pilot or commercial-scale CO2 to methanol plants could provide practical insights into the proposed strategy's effectiveness in real-world scenarios. In short, this study establishes a foundation for intelligent optimization in the realm of chemical production under conditions of uncertainty, offering numerous avenues for further advancements and applications.

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