Effect of Temperature and Pressure Variation on CO_2 Methanation Performance in a PFR Reactor Using Ni/Al_2O_3 Catalyst

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Abstract

This study investigates the effect of feed temperature and pressure on the performance of CO_2 methanation using simulation results from Aspen HYSYS. The analysis focuses on the variation of CO and CH_4 mole fractions in the product stream under temperature ranges of 350–500°C and pressure ranges of 100–300 kPa. The results show that temperature has a minimal effect on both CO and CH_4 compositions, with only slight fluctuations observed, indicating that the system operates equilibrium conditions. In contrast, pressure exhibits a more noticeable influence, where increasing pressure leads to a gradual decrease in CO mole fraction and a corresponding increase in CH_4 mole fraction. These findings are consistent with the exothermic nature of the methanation reaction and Le Chatelier's principle, confirming that higher pressure favors methane formation. Overall, the study highlights that pressure is a more significant operating parameter than temperature in enhancing CO_2 conversion to CH_4 under the investigated conditions.

Keywords:

CO_2 methanation; temperature effect; pressure effect; methane production; process optimization

1. Introduction

Global carbon dioxide (CO_2) emissions continue to rise due to heavy reliance on fossil fuels, exacerbating climate change and underscoring the urgent need for effective carbon mitigation efforts. CO_2 is recognized as the primary greenhouse gas emitted by the energy sector, making the adoption of sustainable and low-carbon technologies increasingly important [1]. One potential solution is the Power-to-Gas (PtG) approach, which converts excess renewable electricity into gaseous fuel, enabling improved energy storage and cross-sector utilization [1]. In this context, CO_2 methanation plays a crucial role by converting captured CO_2 along with green hydrogen into methane via the Sabatier reaction [2]. Furthermore, Synthetic Natural Gas (SNG) serves as a valuable energy carrier as it can be distributed through existing natural gas networks and stored in large quantities. Consequently, combining PtG systems with CO_2 methanation and SNG production offers a practical strategy to reduce emissions while facilitating the transition to a more sustainable energy future [3].

Catalyst development for the CO_2 methanation reaction (Sabatier reaction) generally focuses on the use of transition metals such as Ni, Fe, and Co, as well as noble metals such as Ru, with Ni/Al_2O_3 based catalysts remaining the primary choice due to their balance between catalytic activity and relatively low cost [4]. Nevertheless, the limitations of Ni catalysts namely, suboptimal activity at low temperatures and a tendency toward sintering and carbon formation have driven the development of bimetallic catalysts and the use of promoters [5]. The addition of metals such as Fe to Ni systems has been reported to produce a synergistic effect through the formation of a Ni-Fe alloy, which enhances CO_2 adsorption and activation, thereby improving both conversion and catalyst stability compared to monometallic Ni [6]. In addition, oxide-based promoters such as CeO_2 and La_2O_3 play a role in increasing the number of oxygen vacancies, which accelerates the CO_2 activation mechanism [7]. In terms of operating conditions, most studies report that the reaction proceeds optimally at temperatures of 250 - 400℃ and H_2/CO_2 ratio of 3 - 5, where increases in temperature and the H_2/CO_2 ratio boost conversion to near equilibrium, but CH_4 selectivity tends to decrease at high temperatures due to the dominance of the reverse water gas shift (RWGS) reaction [8]. Although various studies have demonstrated performance improvements through catalyst modification, research integrating the simultaneous effects of operating parameters such as temperature and pressure on Ni-Fe/Al_2O_3 catalyst systems in PFR type reactors remains limited; therefore, further research is needed to identify optimal operating conditions capable of simultaneously maximizing CO_2 conversion and CH_4 selectivity [9].

A comparison of catalyst performance from various studies is presented in Table 1.

Research on CO_2 methanation has demonstrated that nickel (Ni)-based catalysts remain the most widely used due to their high activity and excellent selectivity toward CH_4 formation [10]. Several studies report that CO_2 conversion can reach approximately 96–97% with CH_4 selectivity exceeding 99% under operating conditions of 300–400 °C in fixed-bed or plug flow reactors (PFR) [2]. Furthermore, the development of bimetallic catalysts such as Ni–Fe/Al_2O_3 has shown improved performance compared to monometallic Ni catalysts, where the synergistic interaction between Ni and Fe enhances the stability of active sites and suppresses CO formation, enabling CO_2 conversion above 95% with CH_4 selectivity greater than 98% [11]. In addition, modified catalyst systems such as Ni–Mg–Al have been reported to achieve very high CH_4 selectivity (>99%) even at relatively low temperatures (~275 °C), indicating their potential for improved energy efficiency [12].

Table 1. Comparison of catalyst performance from various studies

Catalyst	Temperature (℃)	H_2/CO_2	CO_2 Conversion	CH_4 Selectivity
Ni/Al_2O_3	300 - 400	4 – 5	High (80 - 90%)	High
Ru/Al_2O_3	250	4	Medium	100%
Ni–Fe	250 - 350	4	Higher than Ni	High

Ni–Ce/La	300 - 400	4 - 5	Increase	High
Ni–Co	350 - 450	3 - 5	Increase	High

Despite these promising results, several limitations remain critical challenges in CO_2 methanation processes. The highly exothermic nature of the reaction leads to hotspot formation at the reactor inlet, resulting in non-uniform temperature distribution and inefficient catalyst utilization. At elevated temperatures (>350 °C), the reverse water–gas shift (RWGS) reaction becomes more dominant, reducing CH_4 selectivity due to increased CO production. Moreover, although Ni–Fe/Al_2O_3 catalysts exhibit promising performance, most existing studies are limited to single-parameter variation, and there is still a lack of comprehensive research addressing the simultaneous optimization of key variables such as temperature and pressure in PFR systems. This highlights a significant research gap, particularly in understanding the non-linear interactions among these parameters and their combined effect on hotspot formation, reactor performance, and long-term stability [13].

Recent studies have extensively investigated Ni-Fe/Al_2O_3 catalysts due to their enhanced catalytic activity compared to monometallic Ni catalysts, particularly in reactions such as CO_2 methanation and reforming. Previous works mainly focused on catalyst characterization, kinetic studies, or the effect of individual operating parameters such as temperature, pressure, and feed composition. However, these studies are generally limited to either simplified reactor systems or partial optimization approaches involving only a few variables. Furthermore, limited attention has been given to the integration of bimetallic Ni-Fe catalysts with detailed Plug Flow Reactor (PFR) modeling under multi-variable optimization conditions [14].

This study presents an improved Aspen HYSYS simulation of CO_2 methanation in a Plug Flow Reactor (PFR) within a Power-to-Gas (PtG) framework to address rising CO_2 emissions. As an exothermic and equilibrium-limited process, its performance is strongly influenced by operating conditions, particularly temperature and pressure, which affect conversion and methane selectivity. The system integrates renewable hydrogen with captured CO_2 to produce Synthetic Natural Gas (SNG), supported by Ni-based and Ni–Fe/Al_2O_3 catalysts that enhance activity and stability.

Overall, the results highlight the importance of multi-variable optimization to improve conversion and support sustainable energy development.

2. Methods

To optimize the carbon dioxide methanation process, Aspen HYSYS is employed for process simulation and analysis. The system consists of five main components: carbon dioxide (CO_2), hydrogen (H_2), methane (CH_4), water (H_2O), and carbon monoxide (CO). The Peng-Robinson equation of state is applied as the thermodynamic fluid package to accurately calculate the non-ideal properties of the gas mixture. The process begins with a total feed flow rate of 55 cm^3/min, comprising a volumetric feed ratio of H_2 to CO_2 at 4:1. This reactant mixture is directed into a Plug Flow Reactor (PFR) operating isothermally at a temperature of 673 K and a total pressure of 115 kPa [15].

The internal configuration of the PFR is meticulously defined to reflect the physical constraints and fluid dynamics of the experimental fixed-bed setup. The tubular reactor features a bed length of 30 cm and an inner bed diameter of 3 mm [15]. It is packed with a Ni-Fe/Al_2O_3 catalyst. To accurately simulate the catalytic bed hydrodynamics and potential pressure drops, the catalyst particles are specified with a diameter range of 125-250 µm, an assumed bulk solid density of 400 kg-cat/m3, and a bed void fraction of 0.42 [15].

The core of the simulation relies on a highly detailed kinetic model derived from experimental parameters. The reaction network is simulated using the Langmuir-Hinshelwood/Hougen-Watson (LHHW) mechanism, which is broken down into three interdependent equilibrium-driven reactions:

The primary CO2 methanation

$$CO_2+4H_2\leftrightarrow CH_4+2H_2O\Delta_r H^0_{298k} \quad (1)$$

The intermediate Reverse Water Gas Shift (RWGS) reaction

$$CO_2+H_2\leftrightarrow CO+H_2O\Delta_r H^0_{298k}=41\ kJ.mol^{-1} \quad (2)$$

The subsequent CO methanation

$$CO+3H_2\leftrightarrow CH_4+H_2O\Delta_r H^0_{298k}=-206\ kJ.mol^{-1} \quad (3)$$

By incorporating the specific activation energies and Van't Hoff adsorption constants for each constituent (CO, H_2O, CO_2, and H_2), the simulation intricately captures the competitive adsorption on the active catalyst sites and the transient formation of carbon monoxide [15].

As a strategic approach to process optimization, a comprehensive parametric study is implemented within the simulation environment to evaluate the reactor's performance under varying operational boundaries. Key variables specifically the reactor temperature and operating pressure are systematically manipulated [15]. The responses to these variations are monitored against critical performance indicators, primarily the overall CO_2 conversion rate and the net CH_4 yield. This optimization framework aims to identify the most favourable operating window that maximizes synthetic natural gas production efficiency while simultaneously observing the kinetic limitations of the catalytic bed [15].

3. Results and Discussion

3.1 Comparison Between Basic and Modified Processes

Figure 1 shows the Aspen HYSYS flowsheet of the CO_2 methanation process, where hydrogen (H_2), carbon dioxide (CO_2), and nitrogen (N_2) are mixed in a mixer (MIX-100) before entering a plug flow reactor (PFR-100). The reactor operates at a pressure of 115 kPa with an inlet temperature of 400°C and an outlet temperature of approximately 277°C. The decrease in temperature along the reactor indicates the exothermic nature of the CO_2 methanation reaction, where heat is released as the reaction proceeds [15].

The main reaction involved is the Sabatier reaction ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$), which is highly exothermic [15]. According to thermodynamic principles, exothermic reactions are favored at lower temperatures, while higher temperatures enhance reaction rates. However, in this system, the temperature profile suggests that heat release plays a significant role in influencing reactor performance. These operating conditions are consistent with the kinetic study by literature journal, where CO_2 methanation over a Ni/Al_2O_3 catalyst was investigated at temperatures between 350–450°C under near-atmospheric pressure [15].

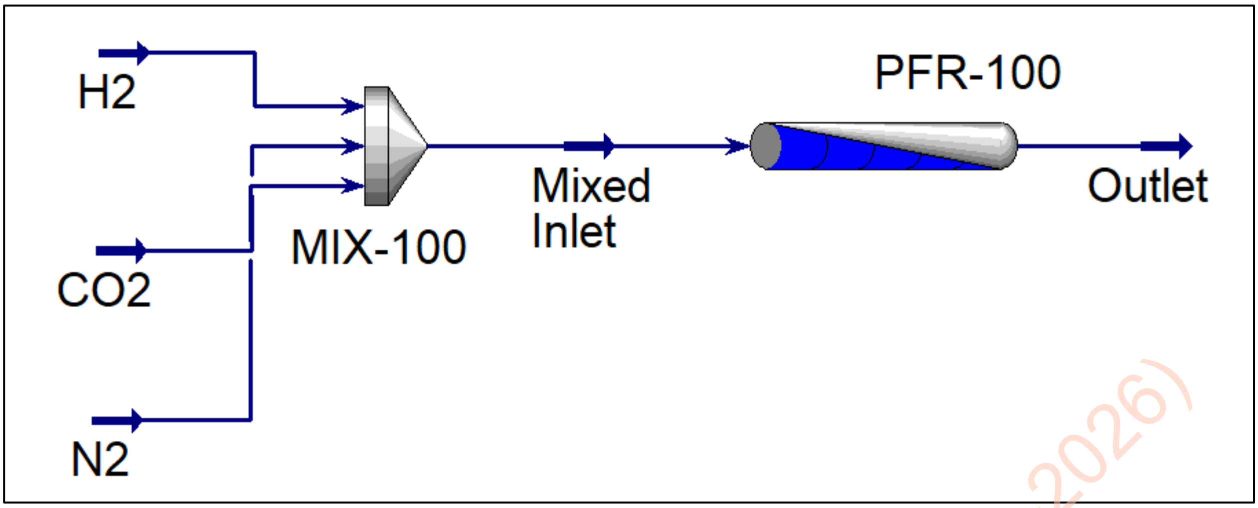

Figure 1. Aspen HYSYS flowsheet of the CO_2 methanation process.

The temperature and pressure balances of the modified process are presented in the Figure 2 and 3. The graph in Figure 2 shows that an increase in temperature leads to a decrease in CO_2 conversion. Theoretically, this behavior is explained by the exothermic nature of the methanation reaction ($CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$), where increasing temperature shifts the equilibrium toward the reactants according to Le Chatelier's principle, thereby reducing CO_2 conversion. Although higher temperatures increase the reaction rate constant (Arrhenius law), thermodynamic limitations dominate at elevated temperatures, leading to lower equilibrium conversion [2]. In addition, higher temperatures favor competing reactions such as the Reverse Water Gas Shift (RWGS), which further reduces methane formation and overall CO_2 conversion. This phenomenon has been widely reported, where methanation is favored at lower temperatures and conversion decreases at higher temperatures due to equilibrium constraints [3].

Meanwhile, the graph in Figure 3 shows that increasing pressure results in higher CO_2 conversion. This can be explained by the stoichiometry of the methanation reaction, where the number of gas moles decreases from 5 moles (reactants) to 3 moles (products). Based on Le Chatelier's principle, increasing pressure shifts the equilibrium toward the products, thus enhancing CO_2 conversion [16]. Furthermore, higher pressure increases the partial pressure of reactants, which improves adsorption on the catalyst surface and enhances reaction rates in heterogeneous catalytic systems. As a result, both thermodynamic and kinetic effects contribute to the increase in conversion at higher pressures. Recent thermodynamic and simulation studies confirm that CO_2 methanation is strongly favored at higher pressures and lower temperatures, leading to improved conversion and selectivity toward methane [1].

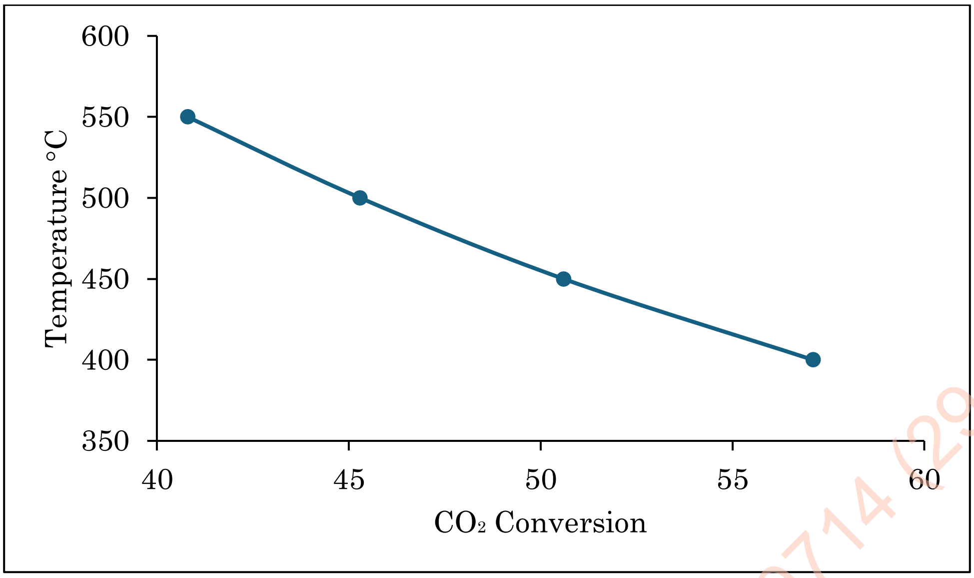

Figure 2. Effect of temperature on CO_2 conversion.

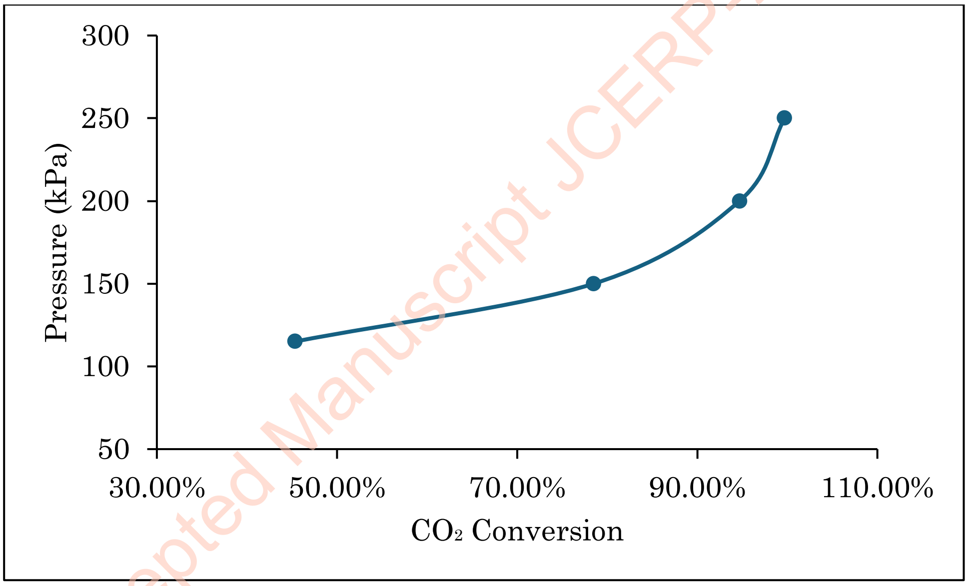

Figure 3. Effect of pressure on CO_2 conversion.

3.2 Thermodynamics Review

CO_2 methanation rate equations

$$r_{CO_2\ meth} = \frac{K_{CO_2\ meth}\, K_{H_2}\, K_{CO_2}\, P_{H_2}\, P_{CO_2}\left(1-\frac{P_{CH_4}P_{H_2O}^{2}}{PH_2^{4}P_{CO_2}\, K_{eq\,\cdot\, CO_2\ meth}}\right)}{\left(1+K_{CO_2}P_{CO_2}+K_{H_2}P_{H_2}+K_{H_2O}\, P_{H_2O}+K_{CO}P_{CO}\right)^{2}} \quad (4)$$

$$k_1 = k_{CO_2}math\cdot k_{H_2}\cdot k_{CO_2}\ (gmol^{-1}\cdot mm^{-1}\cdot g^{-1}) \quad (5)$$

where,

$$k_{CO_2}math=k_{0_{CO_2}}math\cdot\exp\left(\frac{-EQ\ CO_2math}{RT}\right) \tag{6}$$

$$k_{H_2}=k_{0_{H_2}}\cdot\exp\left(\frac{-EQ\ CO_2math}{RT}\right) \tag{7}$$

- Reversible reaction

$$r_{CO_2\ meth} = (k_{0_{CO_2}}math \cdot k_{0_{H_2}} \cdot k_{0_{CO_2}})\cdot\exp\left(\frac{-EQ\ CO_2meth\text{-}Q_{H_2}\text{-}Q_{CO_2}}{8{,}314\times673}\right) \tag{8}$$

$$r_{CO_2\ meth} = (1{,}14 \times 10^{8}) \times (5{,}20 \times 10^{-5}) \times (1{,}07)\cdot\exp\left(\frac{-110\text{-}52\text{-}9{,}72}{8{,}314\times673}\right) \tag{9}$$

$$r_{CO_2\ meth} = 6151{,}2518$$

- Irreversible reaction

$$r_{CO_2\ meth} = \frac{(k_{0_{CO_2}}math \cdot k_{0_{H_2}}\cdot k_{0_{CO_2}})}{-EQ\ CO_2meth}\exp\left(\frac{-EQ\ CO_2meth\text{-}Q_{H_2}\text{-}Q_{CO_2}}{8{,}314\times673}\right) \tag{10}$$

$$r_{CO_2\ meth} = \frac{(1{,}14\times10^{8})\times(5{,}20\times10^{-5})\times(1{,}07)}{5{,}8\times10^{-3}}\exp\left(\frac{-110\text{-}52\text{-}9{,}72}{8{,}314\times673}\right) \tag{11}$$

$$r_{CO_2\ meth} = 1{,}0606$$

RWGS rate equations

$$r_{RWGS} = \frac{K_{RWGS}\ K_{CO_2}P_{CO_2}\left(1\text{-}\frac{P_{CO}\ P_{H_2O}}{P_{H_2}P_{CO_2}K_{eq\ \cdot\ RWGS}}\right)}{\left(1+K_{CO_2}P_{CO_2}+K_{H_2}P_{H_2}+K_{H_2O}\ P_{H_2O}+K_{CO}P_{CO}\right)^2} \tag{12}$$

- Reversible reaction

$$r_{RWGS} = (k_{RWGS}\ k_{CO_2})\cdot\exp\left(\frac{-EQ\ CO_2RWGS\text{-}Q_{H_2}\text{-}Q_{CO_2}}{8{,}314\times673}\right) \tag{13}$$

$$r_{RWGS} = \left(1{,}78\times10^{6}\right)\times(1{,}07)\cdot\exp\left(\frac{-110\text{-}52\text{-}9{,}72}{8{,}314\times673}\right) \tag{14}$$

$$r_{RWGS} = 1{,}84704 \times 10^{6}$$

- Irreversible reaction

$$r_{RWGS}=\frac{(k_{RWGS}\ k_{CO_2})}{-EQ\ CO_2meth}\exp\left(\frac{-EQ\ CO_2RWGS\text{-}Q_{H_2}\text{-}Q_{CO_2}}{8{,}314\times673}\right) \tag{15}$$

$$r_{RWGS} = \frac{\left(1{,}78\times10^{6}\right)\times(1{,}07)}{5{,}8\times10^{3}}\cdot\exp\left(\frac{-110\text{-}52\text{-}9{,}72}{8{,}314\times673}\right) \tag{16}$$

$$r_{RWGS} = 318{,}45445$$

CO methanation rate equations

$$r_{CO\ meth} = \frac{k_{CO\ meth}\ k_{H_2}k_{CO}P_{H_2}P_{CO}\left(1\text{-}\frac{P_{CH_4}P_{H_2O}}{P_{H_2}P_{CO}K_{eq\ \cdot\ CO\ meth}}\right)}{\left(1+k_{CO_2}P_{CO_2}+k_{H_2}P_{H_2}+k_{H_2O}\ P_{H_2O}+k_{CO}P_{CO}\right)^2} \tag{17}$$

- Reversible reaction

$$r_{CO_2} = (k_{CO_2}meth \cdot k_{H_2} \cdot k_{CO}) \cdot \exp\left(\frac{-EQ\ CO_2 - Q_{H_2} - Q_{CO_2}}{8{,}314 \times 673}\right) \tag{18}$$

$$r_{CO_2} = \left(2{,}23 \times 10^{8}\right) \times (5{,}20 \times 10^{-5}) \times (2{,}39 \times 10^{-3}) \cdot \exp\left(\frac{-110-52-9{,}72}{8{,}314 \times 673}\right) \tag{19}$$

$$r_{CO_2} = 0{,}000999536$$

- Irreversible reaction

$$r_{CO_2} = \frac{(k_{CO_2}meth \cdot k_{H_2} \cdot k_{CO})}{-EQ\ CO_2 meth} \exp\left(\frac{-EQ\ CO_2 - Q_{H_2} - Q_{CO_2}}{8{,}314 \times 673}\right) \tag{20}$$

$$r_{CO_2} = \frac{\left(2{,}23 \times 10^{8}\right) \times (5{,}20 \times 10^{-5}) \times (2{,}39 \times 10^{-3})}{5{,}8 \times 10^{3}} \cdot \exp\left(\frac{-110-52-9{,}72}{8{,}314 \times 673}\right) \tag{21}$$

$$r_{CO_2} = 4{,}63393 \times 10^{-8}$$

[15]

3.3 Effect of Temperature and Pressure Variation on CO_2 Conversion in CO_2 Methanation

The effect of temperature on CO_2 convertion in methanation process, ad preented in Fig 2, indicates that increasing temperature within the range of 350 – 600 °C result only minor variations in CO and CH_4 mole fractions. The relatively flat trend suggests that system operates close to thermodynamic equilibrium, where further increases in temperature no longer significantly improve conversion. This behavior is consistent with thermodynamic analyses reported in recent studies, which state that CO_2 methanation is an exothermic reaction and that increasing temperature shifts equilibrium is an toward the reactants, thereby reducing conversion effiency [17]. Futhermore, pressure also influences reaction kinetics by increasing the partial pressure of reactans, particularly hydrogen, which enhances the adsorption and surface reactions on catalyst, thereby accelerating methane formation.

In addition, the limited impact of temperature can be attributed to the increasing contribution of the Reverse Water Gas Shift (RWGS) rreaction at elevated temperatures the RWGS reaction is endothermic and becomes more favorable as temperature increases, producing CO and reducing methane selectivity. This is consistent with the presence of CO in Fig 2 despite temperature variation. According to kinetic studies, CO_2 methanation proceeds through both direct and indirect pathways, where the indirect route involves RWGS followed by CO methanation, creating competition between reactions [15]. Moreover, recent reviews highlight that higher temperatures tend to promotes side reactions and reduce CH_4 selectivity, particularly when the system approaches equilibrium conditions [3]. Therfore, increasing temperature does not significantly enhance CO_2 conversion due to both equilibrium limitations and competing reaction pathways.

In contrast, the effect of pressure on CO_2 conversion, as shown in Fig 3, is considerably more significant. Increasing pressure from 50 – 300 kPa leads to a clear decrease in CO mole fraction and simulataneous increase in CH_4 mole fraction. This observation aligns with Le Chatelier's principle, as the methanation reaction involves a reduction in the number of gas moles, making higher pressure favorable for methane formation. Thermodynamic studies confirm that elevated pressure enhances both CO_2 conversion and CH_4 yield, especially at moderate temperatures, due to the shift in equilibrium toward products [17]. Furthermore, pressure also influences reaction kinetics by increasing the partial pressures of reactants, particularly hydrogen, which enhances adsorption and surface reactions on the catalyst, thereby accelerating methane formation.

Overall, the combined analysis of Fig 2 and Fig 3 demonstrates that pressure is a more domintant parameter than temperature in improving CO2 conversion during methanation. While temperature effects are constrained by equilibrium limitations and the promotion of RWGS at higher values, pressure consistently enhances both thermodynamic favorability and reaction kinetics. This conclusion is supported by optimization studies which rank pressure as the most influential parameter effecting CO_2 conversion and CH_4 yield [12]. Therefore, the system can be classified as equilibrium-limited, where increasing temperature alone is insufficient to significantly improve performance, and pressure optimization becomes the key strategy for maximizing methane production efficiency.

Conclusion

The CO_2 methanation process presents a promising approach to reduce greenhouse gas emissions through the integration of Power-to-Gas (PtG) systems and synthetic natural gas (SNG) production. Based on the analysis, temperature and pressure significantly influence CO_2 conversion, where temperature increases tend to reduce conversion due to the exothermic nature of the reaction and equilibrium limitations. In contrast, increasing pressure enhances CO_2 conversion by shifting the equilibrium toward the products and improving reactant adsorption on the catalyst surface. The results also indicate that the system operates close to thermodynamic equilibrium, where further temperature increases do not significantly improve performance and may instead promote competing reactions such as RWGS. Moreover, catalyst development, particularly Ni-based and Ni–Fe/Al_2O_3 systems, plays a crucial role in improving conversion and stability, although challenges such as sintering and carbon formation remain. Overall, this study highlights that pressure is a more dominant parameter than temperature in enhancing CO_2 conversion, emphasizing the importance of multi-variable optimization to achieve efficient and sustainable methanation performance.

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