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Proceeding Paper

Performance Evaluation of Various Ni-Based Catalysts for the Production of Hydrogen via Steam Methane Reforming Process †

Sudeep Noorambala Subramanya [1,2], Vaka Sai Charan Reddy [3] and Vasudeva Madav [1,2,*]

[1] Department of Mechanical Engineering, National Institute of Technology Karnataka, Surathkal, Mangalore 575025, India; sudeepnsharsha@gmail.com
[2] Maire Tecnimont Centre For Research in Waste Recycling and Circular Economy, National Institute of Technology Karnataka, Surathkal, Mangalore 575025, India
[3] Associate, PwC, Hyderabad 500081, India; vakasaicharan123@gmail.com
* Correspondence: vasu@nitk.edu.in; Tel.: +91-9008889796

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Abstract: Steam methane reforming (SMR) approaches are highly recognised and pivotal in industrial H_2 production, contributing over 40% to global hydrogen production. The prime objective of this study is to optimise the significant parameters involved in the SMR process to achieve the utmost conversion of CH_4 to H_2. To attain this, a sophisticated one-dimensional unsteady-state heterogeneous plug flow reactor (PFR) model was methodically constructed and simulated using the Aspen HYSYS V11 software. The study comprises an exhaustive comparison of seven diverse sets of catalysts, primarily categorised based on the different weight percentages of Ni in Ni/Al_2O_3 catalysts, along with various promoters incorporated to enhance the conversion rate in the SMR process. This comprehensive evaluation identifies the most operative catalyst configuration for optimising CH_4 conversion. The results obtained through the simulations revealed that CH_4 conversion intensifies with an increase in temperature, while it weakens with higher pressures within the catalyst set considered for the study. The analysis yielded promising conclusions by comparing the simulated CH_4 conversion percentages at various temperatures with data from the existing literature. The maximum absolute error encountered was only 3.72%, signifying the accuracy and reliability of the developed model. Moreover, the Mean Absolute Error (MAE) calculated was a low 1.42%, suggesting the robustness of the proposed approach. The findings lay the foundation for future innovations and improvements in the field, ultimately fostering more efficient and sustainable hydrogen generation. As the demand for clean energy grows, the optimisation of the SMR process becomes increasingly vital, making this study a crucial step towards meeting global energy needs while minimising environmental impact.

Keywords: steam methane reforming; plug flow reactor; Aspen HYSYS; catalysts; mean absolute error

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1. Introduction

In recent decades, there has been a significant increase in demand for energy. The increased usage of fossil fuels has been observed across various industries, resulting in higher levels of greenhouse gases and environmental pollution in the Earth's atmosphere [1]. To meet global energy demands and reduce adverse effects, research is being done on alternative energy sources [2]. The dwindling supply of coal and oil has sparked a surge in the pursuit of renewable and alternative energy sources. In today's modern landscape, renewable energy sources have garnered a significant following for their versatility in various applications. One such source, biogas, has emerged as a promising contender [3].

The CH_4 contribution per mole in generating greenhouse gas is 25 times more than that of a mole of CO_2, and the amount of energy produced by utilising H_2 is about 2.4 times that of CH_4 for combustion purposes. Therefore, the conversion and utilisation of CH_4 to H_2 have received a tremendous amount of research attention recently [4–6]. As per the Energy Policy Act of 1992, the interest in H_2 as an alternate fuel has recently increased dramatically. The preference for H_2 from food waste is considered a promising renewable energy source due to its ability to be used for household purposes, clean-burning potential, fuel cell vehicles, and 2–3 times higher efficiency prospects than petroleum products [7].

There are different leading methods for reforming methane: steam methane reforming (SMR), partial CH_4 oxidation (POx), dry CH_4 reforming (DR), and auto thermal CH_4 reforming (ATR) [8]. Among the CH_4 reforming technologies, SMR has gained research attention [4] because it has been reported that steam reforming of CH_4 present in the natural gas process produces approximately 40% of the total global demand for H_2, and it is one of the least expensive ways of producing H_2, thus contributing its share to cleaner energy production [6]. SMR yields four mols of H_2 and one mol of CO_2 for each CH_4 group and has a high H_2 yield efficiency of up to 74% [8]. The SMR equation is represented below:

$$CH_4 + H_2O \rightarrow CO + 3H_2,\ \Delta H\ 298\ K = 206\frac{kJ}{mol} \tag{1}$$

$$CO + H_2O \rightarrow CO_2 + H_2,\ \Delta H\ 298\ K = -41\frac{kJ}{mol} \tag{2}$$

The numerous transitions and catalysts made with noble metals and used for Equation (1) (syngas gas generation) and Equation (2) (water–gas shift reaction) combination processes have been reported. Metals like platinum (Pt), rhodium (Rh), and ruthenium (Ru) are considered noble due to properties such as their exceptional catalytic activities and maturity against coke resistance, However, they are excessively costly to deploy in industrial SMR applications. Interestingly, Ni as a base metal catalyst shows good performance at an affordable price. Therefore, Ni-based catalysts are widely utilized on an industrial scale for reforming process applications. However, Ni-based catalysts are easily suspectable to deactivation by metal sintering, oxidation, and coking [8–10].

Various catalyst characteristics include CH_4 selectivity, activity and conversion, H_2 yield, thermal stability, and coke resistance. Each catalyst tailored to specific applications, based on its desired characteristics, is presented in Table 1 [6,11].

Table 1. Catalyst enhancements and their effects.

S. No.	Catalyst and Modifications	Key Findings
1	$Ni/SiO_2Al_2O_3$	1. Good thermal stability, H_2 selectivity, coke resistance
2	$Ni_{0.03}Mg_{0.97}O$ solid solution	1. Good stability for longer periods of time
3	Zinc addition to Ni/Al_2O_3	1. Enhances the activity with better stability 2. Enhanced hydrogen selectivity
4	Cu addition to Ni/Al_2O_3	1. Enhanced catalyst stabilization 2. Low coke formation
5	15Ni-1Cu-5Zn/γ- Al_2O_3	1. Optimum catalyst towards hydrogen yield. 2. Lower selectivity to CO
6	K as a promoter to Ni/Al_2O_3	1. Increased coke resistance. 2. Promote reactivity
7	Ni-0309S/γ- Al_2O_3	1. Highly active with good repeatability 2. No Carbon deposition.

2. Materials and Methods

Mathematical modelling is critical in developing chemical reactions at elevated temperatures and pressure. It aids in comprehending the experimental processes and procedures observed by testing SMR reactor models with software that is firmly established. With axial dispersion, the SMR process is represented by a mathematical model with heterogeneous and one-dimensional properties, accounting for mass transfer in solid as well as gas phases, balanced energy distribution covering the reactor arrangements, and reaction kinetics in Aspen HYSYS [12,13].

The model being developed followed the assumptions given below:

- 1D heterogeneous plug flow model with adiabatic operation.
- The reactant flow is realistic to the ideal gas law.
- The temperature of the reaction and reactant concentration gradients are negligible in the direction of the radius. As a result, accountable changes in temperature and concentration are only in one direction, specifically in the axial direction.
- The bed porosity and size of catalyst particles are uniform.
- The catalyst particle temperature gradient is neglected.

Uniform inlet conditions were ensured. To model the CH_4 reforming reactions, five sets of rate equations need to be interpreted: continuity, momentum, energy, and species transport equations describing mass, energy, and momentum transfer are expressed as depicted below [14,15]:

Continuity Equation:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho U)}{\partial X} + \frac{\partial(\rho V)}{\partial Y} = S_m \tag{3}$$

X and Y direction momentum equation:

$$\frac{\partial(\rho U)}{\partial t} + U\frac{\partial(\rho U)}{\partial X} + V\frac{\partial(\rho U)}{\partial Y} = -\frac{\partial P}{\partial X} + \frac{1}{Re}\left[\frac{\partial^2 U}{\partial X^2} + \frac{\partial^2 U}{\partial Y^2}\right] \tag{4}$$

$$\frac{\partial(\rho V)}{\partial t} + U\frac{\partial(\rho V)}{\partial X} + V\frac{\partial(\rho V)}{\partial Y} = -\frac{\partial P}{\partial X} + \frac{1}{Re}\left[\frac{\partial^2 V}{\partial X^2} + \frac{\partial^2 V}{\partial Y^2}\right] \tag{5}$$

where P = stream pressure, and V = gas stream viscosity.

Each equation's right-hand side has three terms: the first for pressure forces and the second and third for viscous forces.

Energy Equation:

$$\rho\frac{\partial e}{\partial t} = -P \text{ div } u + \text{div}(k \text{ grad } T) + \phi + S \tag{6}$$

$$\text{div } u = \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \tag{7}$$

$$\text{grad } T = \frac{\partial T}{\partial x} + \frac{\partial T}{\partial y} \tag{8}$$

$$\phi = \left\{\left[2\left(\frac{\partial y}{\partial x}\right)^2 + \left(\frac{\partial v}{\partial y}\right)^2\right] + \left(\frac{\partial y}{\partial x} + \frac{\partial y}{\partial x}\right)^2\right\} \tag{9}$$

where e = internal specific energy, k = thermal conductivity of the species, ϕ = energy of dissipation rate for a given volume, and S = work done/unit volume by body forces.

The first term on the right-hand side of the equation represents the rate of work done for a considered volume, while the second term represents the rate of heat transfer for a considered volume through reaction conduction.

Species transport equation:

$$\frac{\partial(\rho Y_i)}{\partial t} + \frac{\partial(\rho Y_i)}{\partial x} + \frac{\partial(\rho Y_i)}{\partial y} = -\left[\frac{\partial(J_{i,x})}{\partial x} + \frac{\partial\left(J_{i,y}\right)}{\partial x}\right] + (S_i) \tag{10}$$

$$\left(J_{i,x}\right) = -\rho D_i \frac{\partial Y_i}{\partial x} \tag{11}$$

$$\left(J_{i,y}\right) = -\rho D_i \frac{\partial Y_i}{\partial y} \tag{12}$$

where Y_i = mass fraction, D_i = co-efficient of diffusion, J_i = mass flux of the reactant component, and i, and S_i = net chemical reactions production rate of the reactant species [16].

Modelling and Simulating Steps for Aspen HYSYS

The first step involves selecting the species—CH_4, H_2O, CO, CO_2, and H_2. Peng–Robinson's equation of state is selected to solve the chemical kinetics that are present as a function of pressure and temperature. The set of syngas generation and water–gas shift reaction equations are given as the set of reactions that occur during the SMR process. The respective chemical kinetic parameters presented in Tables 2 and 3 are given as input to the rate equations and are solved. According to the format required for Aspen HYSYS, the constants obtained after solving are given to the respective reactions.

Table 2. Properties of Ni-based SMR catalyst bed.

Catalyst	Name	d_p (m)	ρ_{cat} (kg/m^3)	ρ_{bed} (kg/m^3)	εb
Catalyst—1	$Ni/MgAl_2O_4$	2.00×10^{-3}	1870	1122	0.4
Catalyst—2	Ni-0309S/γ-Al_2O_3	1.75×10^{-3}	3737	2429	0.35
Catalyst—3	$Ni/K_{1.4}Al_{10.9}O_{17.2}$	5.60×10^{-3}	1687	877	0.48
Catalyst—4	18 wt% NiO/α-Al_2O_3	1.20×10^{-3}	1870	1122	0.4
Catalyst—5	15.4 wt% Ni/α-Al_2O_3	1.60×10^{-3}	1274	726	0.43
Catalyst—6	10.34 wt% Ni/α-Al_2O_3	2.15×10^{-4}	2200	1320	0.4
Catalyst—7	8.6 wt% Ni/γ-Al_2O_3	5.40×10^{-3}	2355	1154	0.51

Table 3. Catalyst Arrhenius kinetic parameters.

Catalyst	E_1 (J/mol)	E_2 (J/mol)	E_3 (J/mol)	A_1 (mol bar$^{0.5}$/kg$_{cat}$ s)	A_2 (mol/kg$_{cat}$ bar s)	A_3 (mol bar$^{0.5}$/kg$_{cat}$ s)
Catalyst—1	240,100	67,130	243,900	1.17×10^{15}	5.43×10^{5}	2.83×10^{14}
Catalyst—2	209,500	70,200	211,500	9.048×10^{11}	5.43×10^{5}	2.14×10^{9}
Catalyst—3	218,550	73,523	236,850	5.83×10^{11}	2.51×10^{4}	4.67×10^{13}
Catalyst—4	257,010	89,230	236,700	5.19×10^{12}	9.90×10^{6}	1.32×10^{13}
Catalyst—5	217,010	68,200	215,840	5.79×10^{12}	9.33×10^{6}	1.29×10^{13}
Catalyst—6	216,722	67,966	227,941	9.78×10^{14}	5.29×10^{5}	2.57×10^{14}
Catalyst—7	240,100	67,130	243,900	9.49×10^{15}	4.39×10^{6}	2.29×10^{15}

In the Aspen Hysys solver as shown in Figure 1, the CH_4 and H_2O feed is set at the required temperature and pressure, and the S/C ratio is maintained at 3. The CH_4 volume flow rate is set at 25 lit/hr. These two streams are mixed, and the output of this pre-feed is sent as the inlet to a heater in which the output is set to the required temperatures, while the pressure drop is kept at 0. This is then sent to a plug flow reactor whose length is 49.5 cm, and the inner diameter is set to 1.2 cm. The other bed catalyst properties mentioned in Table 4 are also given in the plug flow reactor and simulated.

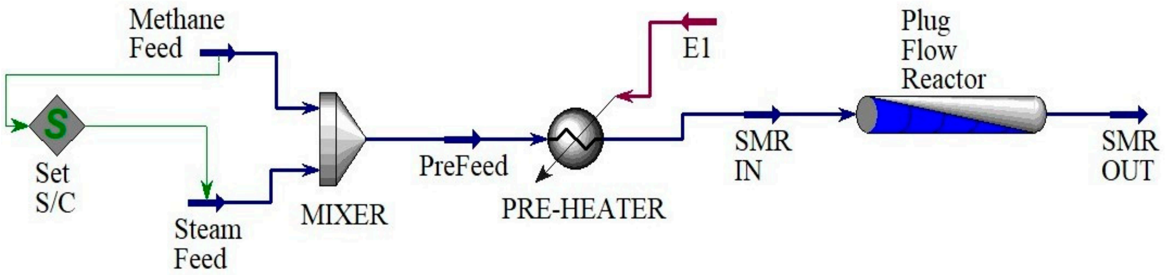


Figure 1. SMR process modelled in Aspen HYSYS.

Table 4. Comparison of CH_4 conversion (%) at different temperatures.

Temperature (K)	Author [17]	Present Work	Absolute Error (%)
	CH_4 Conversion (%)		
873	87.09	90.328	3.72
923	95.92	92.7310404	3.32
973	97.88	95.0713494	2.87
1023	98.88	97.144423	1.75
1073	99.73	98.9914669	0.74
1123	99.89	99.9374718	0.05
1173	99.99	99.999617	0.01
1223	99.97	99.9998265	0.03
1273	99.74	99.9998411	0.26

3. Results

The reactor model-simulated data generated from the Aspen HYSYS was validated in contradiction to steady-state industrial-generated data [17]. The packed catalyst bed adiabatic reactor model was presumed to abide by the non-ideal SMR plug flow characteristics, utilizing the catalyst present in the conditions used for the validation. The side-by-side comparison of results obtained from the literature and the simulated values were grouped and are shown in Table 4. It was observed that the Maximum Absolute Error (MAE) of 3.72% and the Relative Mean Error (RME) of 1.42% were found. The reliability and accuracy of a developed Aspen HYSYS SMR model with a low absolute error of 3.72% indicates that the model's predictions are very close to the actual values reported in the literature, indicating calculated CH_4 conversion percentages are within an average deviation of just 3.72% from the reference data points. Such a small absolute error suggests that the SMR model is proficient in capturing the primary behaviours and trends of the Ni-based catalytic system. Furthermore, the RME of 1.42% underpins the SMR model's reliability. RME signifies the average extent of the errors between the model's calculations and the actual values. In this work, 1.42% shows the model's consistently accurate performance across various temperature and pressure data points. The forecasted results of the outlet of the SMR reactor with the molar composition of the reformed product gases are equated with the industrial statistics and are shown in Table 5. This is predominantly significant in catalytic processes like the SMR process, where even minute errors in estimation can have significant implications for process optimization and productivity.

Table 5. Mole fractions of products at outlet over various temperatures.

Mole Fractions of Products at the Outlet of the Reactor									
		773 K		873 K		973 K		1073 K	
	Exit	This work	[4]	This work	[4]	This work	[4]	This work	[4]
P = 10 bar S/C = 3	CH_4	0.265	0.26	0.206	0.203	0.132	0.126	0.065	0.05
	CO	0.002	0.004	0.014	0.015	0.053	0.061	0.108	0.115
	H_2	0.166	0.174	0.291	0.3	0.414	0.434	0.528	0.563
	H_2O	0.526	0.524	0.429	0.421	0.338	0.314	0.246	0.222
	CO_2	0.041	0.038	0.06	0.061	0.063	0.065	0.053	0.05

4. Discussion

H_2, CO_2, CO, H_2O, and CH_4 are the species that exist as reactants and products in the SMR reaction. For the syngas generation and water–gas shift reactions, the rate equations are determined in relation to the concentrations of the species adsorbed. By using equilibrium relations, a few of these concentrations were eliminated and balanced by considering the sites which are active, and even including those unreacted vacant constituents and those enclosed by species adsorbed. This results in the obtaining of rate equations in terms of the constituent's partial pressures in the gas phase, and these rate equations result from the reacting species adsorbed [18].

The SMR reaction rate is greatly influenced by the temperature of the operation and gas concentrations. The molar steam to CH_4 ratio is kept constant to inspect the impact of pressure and temperature. Similarly, the pressure effect is investigated by considering the temperature and constant molar steam to the CH_4 ratio. The below-mentioned equation is used to estimate the CH_4 conversion.

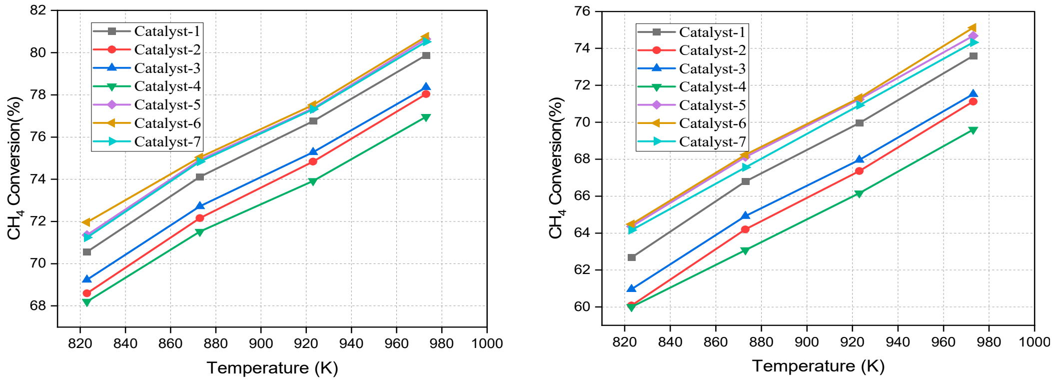
$$\mathrm{CH_4\ Conversion}(\%) = \frac{n_{CH_4,in} - n_{CH_4,out}}{n_{CH_4,in}} \times 100 \tag{13}$$

where n represents the molar rate flow of the CH_4 through the reactor.

When the temperature rises, the CH_4 conversion in the plug flow SMR reaction becomes stronger due to the higher kinetic energy of molecules. This leads to more collisions and successful reactions. At elevated temperatures, the activation energy barrier for the reaction is more easily overcome, promoting the formation of products. However, if the pressure is too high, the conversion weakens because it affects the equilibrium, which is pushed towards the reactants. This reduces the yield of desired products and reaction kinetics. The partial pressures of the reactants increase, which can potentially lead to increased rates of reverse reactions. Therefore, finding the right balance between temperature and pressure is crucial to achieving the most efficient SMR process without any secondary reactions or catalyst deactivation.

4.1. Effect of Temperature

The temperature has a significant impact on the SMR process sensitivity. In accordance with Le Chatelier's principle, it has been observed that, at equilibrium, in the SMR reaction during Syngas generation, the rate showed an increase with the increase in the reactor operating temperature [19,20]. The conversion rates of CH_4 with the effect of temperature are demonstrated in Figure 2a,b. The observations from figures states that the temperature has a constructive effect, i.e., the conversion of CH_4 intensifies with an intensification in temperature, and the CH_4 conversion remains high at higher temperatures.

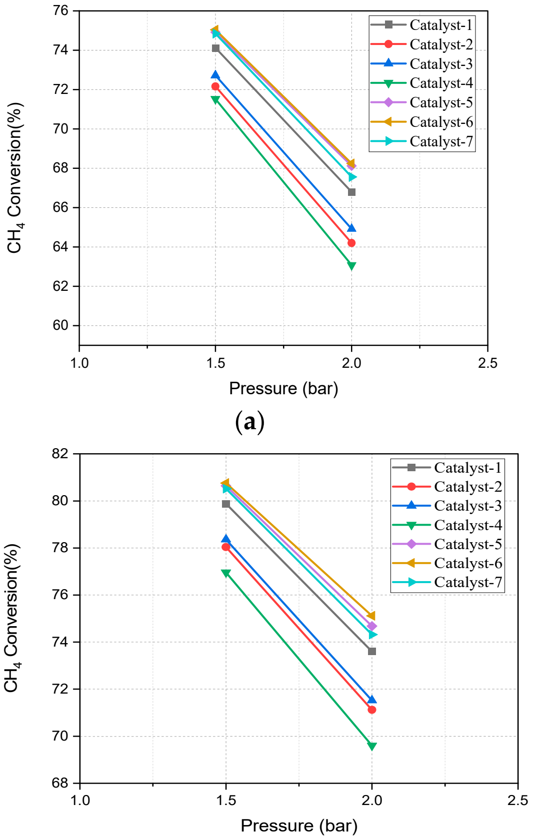


(**a**) (**b**)

Figure 2. (**a**,**b**): Effect of the temperature on CH_4 conversion (%) at (**a**) 1.5 and (**b**) 2 bar.

4.2. Pressure Effect on SMR Reaction

To understand the effect of the reactant's pressure parameter on the CH_4 conversion rates, the S/C ratio is constant at 3.0 during the reaction, and the pressure variations are plotted for the parameters against a constant temperature, as shown in the following, Figure 3a,b. From the plots, it can be inferred that for all the seven catalysts used for analysis, the conversion rates of CH_4 go down as the pressure of reactants increases. This suggests that CH_4 conversion is favoured at lower pressures for the set of catalysts considered in this work. In SMR reactions at higher pressures, the number of moles on the product side is more; this causes the equilibrium shift towards the reactants, resulting in reduced CH_4 conversion.



(**a**)

(**b**)

Figure 3. (**a**,**b**): Effect of the pressure on CH_4 conversion (%) at (**a**) 873 K and (**b**) 973 K.

In Figure 4 the conversions of CH_4 to H_2 at the reformer exit of different sets of catalysts (catalysts 1–7) are forecasted under the operating conditions of 823 K, 1.5 bar, and S/C of 3.0 for an adiabatic reactor. A simulation with catalyst 6, which is 10.34 wt% Ni/Al_2O_3, generates the highest CH_4 conversions of 71.96% and 43.95 at 823 K and 1.5 bar pressure due to its fast kinetics. The energy for activation to generate syngas using catalyst 6 is low, and thus the rate constant value is high. Therefore, the rate of s of CH_4 selectivity and conversion to H_2 is considerably higher for catalyst 6 compared to the other catalysts used in the current work. Catalyst 4 shows the lowest CH_4 conversion (68.2 %), with operating conditions of 823 K, 1.5 bar, and S/C of 3.0 compared to the other catalysts. Considering the following order of catalyst activity in terms of CH_4 conversion, Catalyst 6 > Catalyst 5 > Catalyst 7 > Catalyst 1 > Catalyst 3 > Catalyst 2 > Catalyst 4 for optimum conversion. The SMR process is an extremely endothermic reaction that requires the reactor to be operated under adiabatic conditions.

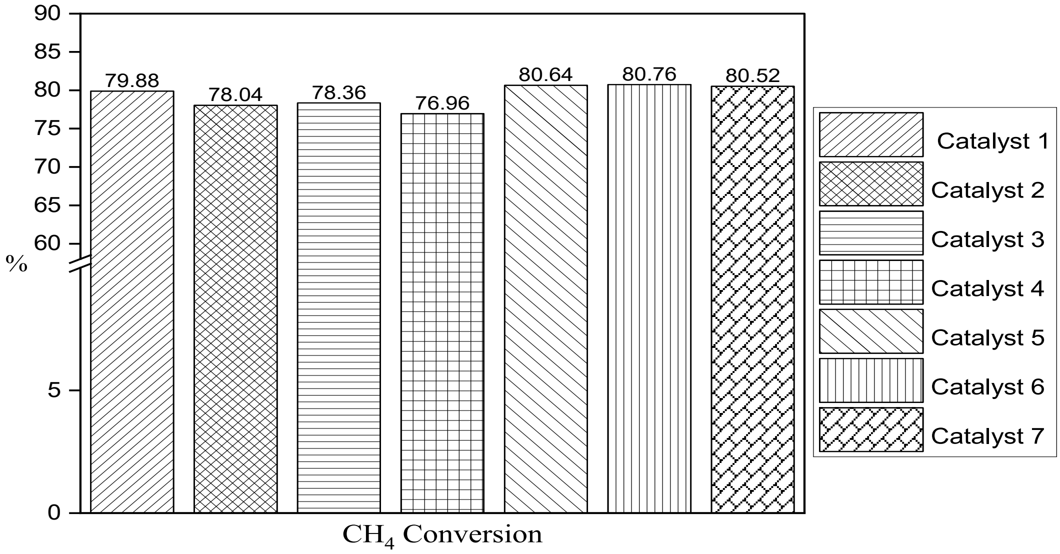


Figure 4. Comparison in terms of CH_4 conversion (%) for various catalysts at 823 K, 1.5 bar, S/C of 3.0.

5. Conclusions

The technicalities involved in an SMR process were studied from the extensive literature on catalysts that can be used in the SMR process, with seven different Ni-based catalysts. The catalysts were simulated in the operating conditions of pressures 1.5 and 2 bar, a molar S/C ratio of 3, and temperature varying from 873 to 923 K to obtain higher CH_4 conversion rates to H_2. A heterogeneous 1D adiabatic plug flow reactor model is deployed to compare the performance of seven Ni-based metal commercial catalysts in the SMR process. The reactor with the chemical kinetics of the respective catalysts is modelled and simulated in Aspen HYSYS. The modelling results were in decent agreement with chemical equilibrium calculations based on the literature. The reactor model was then used for various catalysts to evaluate their performance under operating conditions that are suitable in the scenario of establishing a small-scale H_2 production plant. Regarding the catalyst's performance, fast reaction kinetics gave better results under the operating conditions. Catalyst 6, a Ni-based catalyst, showed superiority over all the other catalysts in CH_4 conversion, of 80.76% at 923 K, 1.5 bar, S/C of 3, and CH_4 flow rate of 25 lit/hr. A 10.34 wt% $Ni/\alpha\text{-}Al_2O_3$ is preferred for CH_4 conversions to H_2 for the discussed operating conditions.

The Aspen HYSYS SMR simulation with an optimum 10.34 wt% $Ni/\alpha\text{-}Al_2O_3$ catalyst at 923 K and 1.5 bar is conducive to addressing global energy problems by leading to clean H_2 production methods. This method helps in reducing emissions, enhances energy efficiency by conserving resources, and influences policies and industrial practices. These exercises collectively contribute to a more sustainable energy landscape and support the evolution towards a low-carbon and ecologically responsible future.

Author Contributions: S.N.S. and V.S.C.R. dedicated myself every facet of the study, investing my time and expertise to ensure the successful completion of this article. The said authors meticulously devised experimental designs, gathered and analyzed data, and derived valuable insights from findings. The collective contributions of both the first and second authors have profoundly impacted this research article, shaping its core essence. Additionally, the authors express their deep gratitude for the invaluable guidance and mentorship provided by V.M., our esteemed research supervisor, whose expertise and support proved instrumental in steering our efforts in the right direction. Their contribution significantly enriched the quality and coherence of our work. The authors take immense pride in this comprehensive study and its potential to advance the understanding of our field. All authors have read and agreed to the published version of the manuscript.

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