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The ability of the Ni/Al_2O_3 catalyst in the CO_2 methanation process in terms of variations in CO_2 flow rate and catalyst

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ARTICLE INFO

Article history:
Received: 11 August 2024
Received in revised form: 21 November 2024
Accepted: 23 November 2024

Keywords:
CO_2, CO_2 flow rate, CO_2 methanation, methane (CH_4)

ABSTRACT

Carbon dioxide (CO_2) is one of the biggest contributors to the greenhouse effect. Based on data from the International Energy Agency (IEA), global carbon dioxide (CO_2) emissions from burning energy and industrial activities will reach 36.8 billion tons in 2022. The increase in emissions in 2022 will mainly come from burning coal and fuel oil (Fuel Oil). Carbon dioxide emissions from coal increased 1.6% while fuel oil emissions increased 2.5%. One effort to reduce carbon dioxide emissions is to convert carbon dioxide into methane gas (CH_4) which can be used as fuel. This research aims to produce methane gas from carbon dioxide using Ni/Al_2O_3 catalyst treatment and varying CO_2 flow rates. Based on the research results, the highest methane gas conversion yield was 49.23% with a variation of 0.05 L/minute with Ni_R4 catalyst treatment.

1. Introduction

Carbon Dioxide (CO_2) is one of the biggest contributors to the greenhouse effect. Based on the total CO_2 emissions released, there are three sectors that have the greatest influence on high CO_2 emissions, namely the electricity sector (42%), transportation (23%) and housing (6%) [1].

According to data released by the International Energy Agency (IEA) in 2022, burning oil and coal is the main cause of increasing carbon dioxide (CO_2) emissions, which are estimated to reach 36.8 billion tons in 2022, an increase of around 0.5 billion tons from 2021 [2]

The trend of increasing CO_2 abundance will continue if preventive efforts are not taken globally. One of these preventive efforts is to convert carbon dioxide (CO_2) into methane (CH4), or known as the carbon dioxide methanation process.

Utilizing CO_2 in situ during the hydrogenation process with nickel tallate for improved steam-based oil recovery [3]. Combustion synthesis uses a nickel catalyst for the methanation of carbon dioxide resulting in an operational catalyst that is stable under reaction conditions for at least 50 hours [4].

Hydrogenation of carbon dioxide to methane or methanation of carbon dioxide, known as the Sabatier reaction, is an exothermic reaction in which hydrogen and carbon dioxide react to form methane and water as byproducts [5]. Although it can be used at relatively low temperatures between 250°C and 400°C, this reaction is favorable. However, carbon dioxide methanation can only be achieved through an effective catalyst. [6].

The formation of CH_4 from CO_2 at low temperatures represents an important breakthrough in knowledge about the role and use of CO_2, although the conversion is still very low [7]. A variety of different catalysts have been used to study catalysts used in CO_2 methanation. In general, most heterogeneous catalysts in CO_2 methanation require certain temperature conditions to be active for methanation reaction selectivity [8].

Heterogeneous catalysts and homogeneous catalysts were used in this study because they are widely used in carbon dioxide methanation. Although homogeneous catalysts show adequate activity and selectivity, recovery and regeneration remain problems in CO_2 methanation [9].

The conversion of CO_2 into methane in water with a Ni nanoparticle catalyst synthesized in situ has succeeded in developing hydrogenation production from water and hydrogenation of CO_2 into methane [10]

Synthesis hydrothermal with in situ method from nanoparticles nickel/oxide as a catalyst in methanation where it is stirred for 20 hours, the highest %yield research results were 18.35% and 17.63% at a process temperature of 673K which produces little methane [11].

Tada et al., [17] have investigated CO_2 methanation with Ni sponges. When CO_2 methanation was carried out with the Ni sponge without pretreatment, the Ni sponge showed a CO_2 conversion of 83% at 250 °C under high space velocity. That is, by pretreatment at high temperature, the Ni sponge loses its activity in CO_2 methanation as well as surface defect sites. Thus, the loss of activity can be explained by the disappearance of surface defect sites by high temperature treatment.

Aditya et al., [15] Have conducted research on carbon dioxide methanation using a Ni/Al_2O_3 catalyst and determined the effect of variations in the Zn promoter and temperature. The research results showed that the highest value of methane gas was produced in situ in sample 15 with a Ni/Al_2O_3 catalyst mass of 8 grams and a Zn promoter of 5.5 grams.

Yuliani et al., [18] conducted a research experiment by producing methane gas from carbon dioxide in a fixed bed reactor. The best concentration of NaOH and carbon dioxide gas flow rate used in various variations of NaOH concentration is 3M, with the addition of NaOH concentration with the

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http://dx.doi.org/10.20527/k.v13i2.20235

highest analysis results, namely 9.25%Vol and a flow rate of 0.1 L/m. CO_2 methanation has become an indispensable reaction for converting toxic CO_2 into methane that can be used as an energy carrier or valuable chemical.

The study conducted by Zhong et al. [10] used a new and simple treatment to produce carbon dioxide methanation with a catalyst of nickel nanoparticles prepared in situ. This catalyst uses water as a hydrogen source and metal (Zn or Fe) as a reducing agent. The methane yield reached 98% of CO_2 or HCO_3 at 300 °C, and the in situ prepared nickel nanoparticle catalyst showed excellent catalytic activity and stability. In simple terms, CO_2 can be converted into methane very efficiently using materials found on Earth.

From the research above, it can be concluded that the time used to convert CO_2 takes a long time, the results of converting CO_2 to methane gas are still small, the temperature used is still too high. To overcome existing problems in order to increase the yield of methane gas products, this research will be carried out using Ni/Al_2O_3 catalysts with variations of nanoparticles which have been proposed by Zhong et al [10] in situ and varying the CO_2 flow rate to the ratio of the raw materials used and then circulated. in a catalytic fixed bed reactor.

2. Materials and Methods

2.1. Tools

A set of Fixed Bed Flug Flow Reactor Tools, measuring flask, watch glass, beaker, measuring pipette, spatula, rubber ball, stir bar and watch glass.

2.2. Materials

Carbon dioxide CO_2, Sodium hydroxide (NaOH), Ni/Al_2O_3 Catalyst, Aquadest, Deionized Water, and Zn Metal.

2.3. Research procedures

2.3.1. Preparation of NaOH solution and catalyst preparation

Make a solution of Sodium Hydroxide (NaOH) with a concentration of 4M in 40 grams using 250mL of distilled water. Then, 50 grams of Ni/Al_2O_3 catalyst activation treatment was carried out and 4 M NaOH solution was added, then heated for 2 hours in a water bath. After heating, the catalyst is rinsed with deionized water and dried using vacuum as a Ni_R0 catalyst treatment, likewise when treating Ni_R1 to Ni_R4 catalysts, the reaction product is recovered from the catalyst, then rinsed with deionized water and dried using vacuum.

2.3.2. Methanation equipment unit operation

After that, connect the methanation tool to the power supply with all the valves leading to the desired flow, then the Ni/Al_2O_3 catalyst and 10 grams of Zn metal are inserted into the reactor. The hose from the CO_2 tube and the NaOH hose are connected to the reactor. Next, raise the fuse until the light comes on and set the temperature at 100°C then press the on button at the same time turning on the NaOH flow pump. The CO¬2¬ flow rate to the reactor is varied at 0.1 L/minute and 0.05 L/minute. The gas that has been obtained is then analyzed to determine the gas contained and the percentage of CH_4 gas obtained.

3. Results and Discussion

3.1. Research result

Table 1. Gas analysis data is generated

Catalyst Treatment	CO_2 Flow Rate (L/minute)	Check-up result (%)		
		CH_4	CO_2	H_2 (ppm)
Ni_R0	0.1	30.34	66.73	1.622
Ni_R1		33.22	63.88	1.596
Ni_R2		38.71	58.52	1.467
Ni_R3		41.26	56.05	1.390
Ni_R4		43.67	53.74	1.286
Ni_R0	0.05	33.49	63.63	1.575
Ni_R1		37.47	59.72	1.501
Ni_R2		35.52	61.64	1.533
Ni_R3		41.68	55.64	1.372
Ni_R4		49.23	48.41	1.060

3.2. Discussion

3.2.1. Effect of catalyst treatment and CO_2 flow rate on methane gas (CH_4)

The activation behavior of the Ni/Al_2O_3 catalyst will increase the conversion optimally. Then, when CO_2 is flowed in the reactor, the CO_2 will continuously react with the NaOH solution and produce Methane Gas [12]. Figure 1 shows the results of the Methane Gas analysis (CH_4).

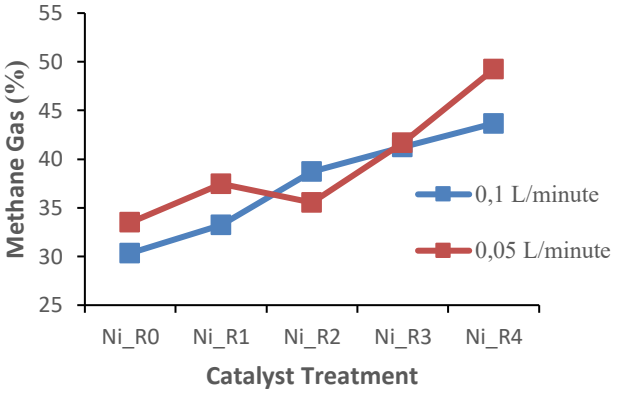

Fig. 1. Effect of catalyst treatment and CO_2 flow rate on CH_4 gas

From Figure 1, it can be seen that there is an increase in CH4 gas produced with a decrease in the flow rate of CO_2 used causing CH4 conversion to increase. The figure shows that the highest methane gas produced during the Ni_R4 catalyst treatment was 49.23% with a flow rate of 0.05 L/m at a temperature of 100°C and 50g of catalyst. However, during the eighth sample experiment with Ni_R2 catalyst treatment and a flow rate of 0.05L/m, a gas leak occurred at the reactor tube lid so that the conversion did not match the expected results. Thus, from the Figure above, it can be concluded that the more catalyst activation treatments and the lower the CO_2 flow rate, the higher the methane gas produced. This is because when the Ni_R0 catalyst treatment forms a slightly porous catalyst structure, the Ni_R4 treatment shows excellent activity and stability, so that catalyst activation can form and catalyze CO_2 reduction efficiently in hydrothermal conditions [10]. In addition, a large gas flow velocity results in a short contact time between the gas and the liquid so that the transfer of gas to the liquid is smaller [13].

3.2.2. Effect of catalyst treatment and CO_2 flow rate on CO_2 gas

The remaining carbon dioxide gas (CO_2) is CO_2 gas that does not react in the process of forming methane gas by circulation so it comes out of the reactor. Figure 2 shows the % CO_2 that does not act in the formation of methane gas.

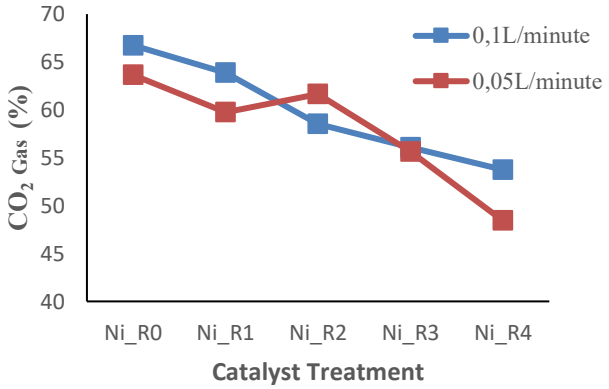

Fig. 2. Effect of catalyst treatment and CO_2 flow rate on CO_2 gas

Based on Figure 2, it can be seen that there is a decrease in the remaining % CO_2 gas which is inversely proportional to the increase in concentration in the production of methane gas. The figure shows that the highest conversion of CO_2 gas was in the Ni_R0 catalyst treatment with a flow rate of 0.1 L/m, namely 66.73% at a temperature of 100 °C and 50g of catalyst. Figure 2 shows the amount of CO_2 gas that does not react with H2. This is based on research by Zhong, et al [10] that the decrease in CO_2 yield at a longer reaction time may be because CO_2 is further reduced to CH_2 caused by the Ni nanoparticle catalyst which is active in reducing CO_2 to methane with the reaction system. So, in the research that has been carried out, the residual CO_2 gas decreases and the flow rate increases, meaning the CO_2 that reacts increases [10].

3.2.3. Effect of catalyst treatment and CO_2 flow rate on H_2 gas

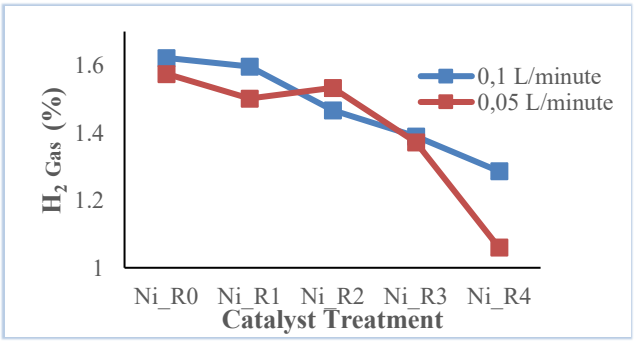

Fig. 3 Effect of catalyst treatment and CO_2 flow rate on H_2 gas

Based on Figure 3, it shows that the highest H_2 gas was at a flow rate of 0.1 L/m with Ni_R0 catalyst treatment, namely 1.622% ppm at a temperature of 100°C and 50 gr of catalyst. In research conducted by Yuliani et al. [18] shows that the best NaOH concentration used in various variations of NaOH concentration is 3M, with the addition of the NaOH concentration the highest analysis result is 9.25% Vol. Meanwhile, in this study, a 4M NaOH concentration was used, meaning that the higher the NaOH concentration, the more H_2 will react by adding 20% NaOH solution. [10]. Aditya K et al [16] also stated that the H_2 content absorbed was greater with the greater the flow rate.

In research that has been carried out at high flow rates, H2 has decreased, this is due to some of the H_2 not reacting with water. The higher the NaOH concentration, the CO_2 produced increases. According to Vas Bhat et al [14], in alkaline conditions the formation of bicarbonate reacts with OH^- to form CO^{32-}. The higher the CO_2 flow rate, the greater the amount of CO_2 absorbed. This is because more and more NaOH molecules are in contact with CO_2 gas. The amount of CO_2 absorbed at a certain time will reach a constant value [14].

4. Conclusion

Based on the results of the research that has been carried out, it was concluded that the highest methane gas produced during Ni_R4 catalyst treatment was 49.23% with a flow rate of 0.05 L/m at a temperature of 100°C and 50g of catalyst, so the more catalyst activation treatment and the lower the CO2 flow rate, the higher the methane gas produced.

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