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Hydrogen from Methane: Application of Microwave Discharge and Catalyst

This paper presents the results of experiments to produce hydrogen using methane pyrolysis with microwave (MW) discharge and steel catalyst in an applied research installation PM-6. The experiments were conducted to determine the fullest extent of methane decomposition in microwave discharge in the PM-6 installation using steel catalyst with varied nickel content. The results showed that the use of nickel catalysts provides the highest hydrogen yield under optimal conditions. Hydrogen production technology was implemented using catalyst in MW discharge with the maximum rate of methane conversion up to 32±2 % and hydrogen selectivity up to 85±1 % at MW discharge of 0.6 kW, methane flow rate of 0.25±0.05 l/min and argon 8.0±0.2 l/min. Overall, the results of the study highlight the importance of choosing catalysts to achieve optimal conditions for methane pyrolysis and create efficient technologies for hydrogen production. The data obtained from the experiments could be useful for the development of industrial pyrolysis plants, which will contribute to a more sustainable energy future and a reduced carbon footprint.

Keywords: hydrogen, methane pyrolysis, hydrogen energy, microwave discharge, methane conversion, carbon, hydrogen selectivity, catalyst.

Introduction

Hydrogen energy has been gaining increasing popularity in recent years. Hydrogen can be used for energy storage, accumulation, and delivery. In this context, hydrogen energy stands out as one of the most promising directions. Consequently, developed countries are developed their own hydrogen energy strategies to meet the annually increasing demand for clean energy [1, 2].

Hydrogen can be produced from various raw material sources using a wide range of technologies. Currently, natural gas is the primary feedstock for hydrogen production, accounting for 68 % of the total hydrogen production worldwide in 2023 [3]. Therefore, to ensure the sustainable use of hydrogen, it is necessary to develop efficient and environmentally friendly production methods. Methane pyrolysis is one such method, which, unlike traditional methods, significantly reduces CO_2 emissions [4–6]. Furthermore, research results have shown that methane pyrolysis is more economically advantageous compared to traditional hydrogen production methods due to lower feedstock costs, high product purity, and the possibility of utilizing by-products [7–10].

In this article, methane pyrolysis is implemented using a microwave discharge. The pyrolysis process in a microwave discharge allows for the achievement of the necessary temperatures and conditions for pyrolysis, significantly reducing reaction time and increasing the yield of target products [11]. It is necessary to optimize the process, particularly through the use of catalysts to enhance the efficiency of pyrolysis and achieve high selectivity for hydrogen.

Catalysts can be used to increase the efficiency of moment pyrolysis and research is currently underway into various materials for use in this process. One of the key factors in enhancing the conversion rate of methane is increasing the temperature of the catalyst since it heats the passing gas. As a result, excited atoms and molecules of methane passing through the catalyst, heated by the energy of the microwave discharge, undergo the decomposition of methane molecules under the influence of chemically active plasma particles (ions, electrons, free radicals).

In addition, the catalyst must have both good catalytic activity and a long service life, as well as low cost. In this regard, many studies of catalysts such as Ni and Fe have been conducted to improve the catalytic activity of methane pyrolysis [8, 12–19]. The main factors affecting the effectiveness of the catalyst application should be taken into account, such as temperature and time of gas contact with the catalyst, textural

properties, material composition and method of catalyst synthesis when choosing the material used in the catalyst.

The integration of methane pyrolysis with high-frequency discharge technologies and metallic catalysts can lead to the creation of closed cycles that promote cleaner and more sustainable hydrogen production. Thus, this article presents the current results of experimental studies on methane pyrolysis in a microwave discharge using a catalyst.

Experimental

Experiments were conducted using the setup for studying the plasma-chemical decomposition of methane in the microwave discharge PM-6 [20]. Data on the pyrolysis of methane have previously been obtained at the IAE RSENNC RK, and the results are presented in works [21, 22]. The schematic layout of the catalyst in the PM-6 installation is shown in Figure 1.

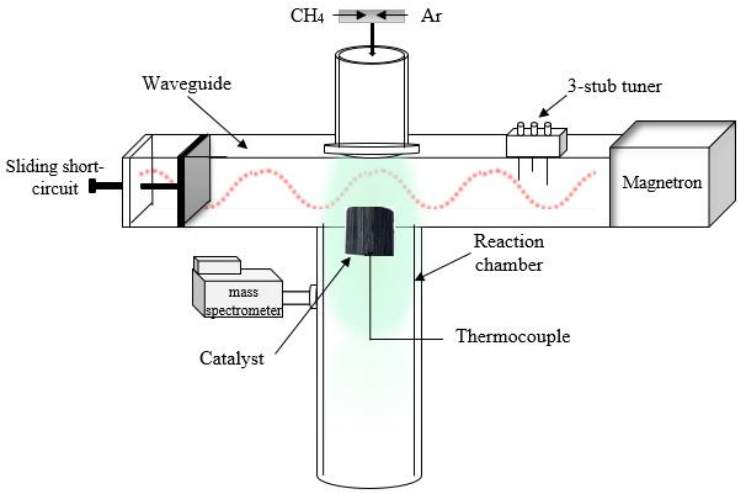


Figure 1. The schematic layout of the catalyst placement in the PM-6 installation

Astainless-steel metal sample was chosen as a catalyst according to literature data [5, 23, 24]. The research [7] has been shown that the use of a catalyst made of nickel and carbon-containing materials in the pyrolysis of natural gas can increase the formation of hydrogen in the mixture of gaseous products. In this regard, it was decided to apply a Ni coating to the steel catalyst.

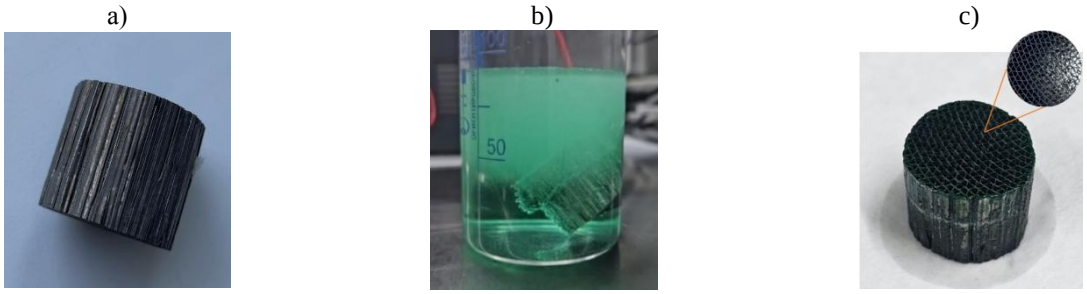
During this research, 3 series of experiments were fulfilled using the following materials:

– Stainless Steel catalyst (Initial);

– Stainless Steel catalyst, plated nickel catalyst;

– Stainless Steel catalyst electrochemically plated by nickel.

For nickel coating, the steel catalyst was placed in a nitric acid (HNO_3) solution to remove contaminants. Nickel nitrate ($Ni(NO_3)_2$) was used as the solution for electroless nickel coating. The composition of the liquid electrolyte in which the product was immersed for electroless coating also included nickel nitrate ($Ni(NO_3)_2$). Figure 2 shows a 20 mm long and 25.5 mm diameter steel catalyst.



a) Initial; b) During electrochemical deposition; c) Nickel plated;

Figure 2. Process of coating the initial catalyst

Upon coating, elemental analysis of the catalysts was performed using a Hitachi TM4000Plus scanning electron microscope (SEM) with an EDS attachment. Figure 3 shows the results.

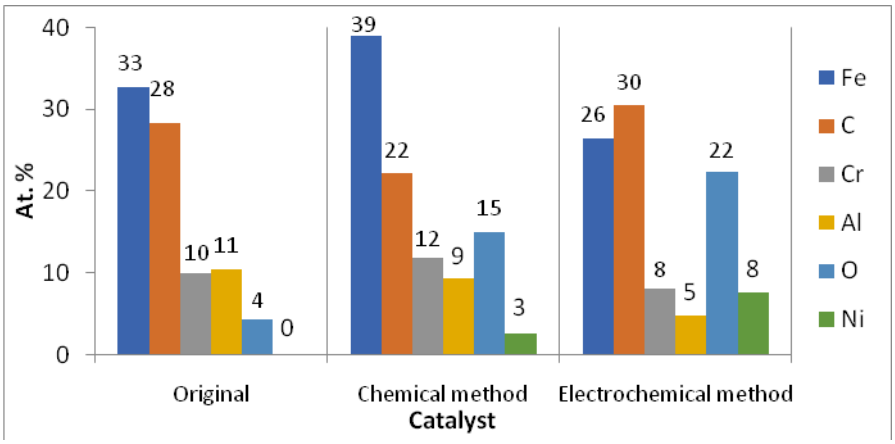


Figure 3. Elemental analysis of the catalyst surface after nickel coating

According to the results of elemental analysis, it is obvious that the electrochemical method of coating is more effective, since the composition of Ni in the catalyst increased by 5 at.%.

Experiments on methane decomposition in the PM-6 installation were conducted in a microwave discharge. A mixture of working gases of methane and argon was fed into a reaction chamber made of quartz glass with a length of 460 mm and an internal diameter of 26 mm. The reflected power was monitored on the magnetron control unit in all experiments and was reduced to "0". Argon was used as a plasma-forming gas for igniting the plasma, into which methane was smoothly fed. The carbon powder deposited on the catalyst (Fig. 4) on the wall of the quartz tube was mechanically removed and collected for analysis after each experiment.

a) catalyst

b) reaction chamber

Figure 4. View after the experiment

Results and Discussion

The reaction products were analyzed using a mass spectrometer "CIS100Gas Analyzers" based on partial gas pressures.

Based on the mass spectrometric analysis data, a qualitative and quantitative evaluation of the methane decomposition rate and hydrogen selectivity was performed.

The methane conversion rate K_{CH_4} is a qualitative value that determines the proportion of methane that has entered into a chemical reaction from the overall original methane. The methane conversion rate was calculated using the following formula:

$$K_{CH_4} = \frac{P_{CH_4}(\text{in}) - P_{CH_4}(\text{out})}{P_{CH_4}(\text{in})} \cdot 100\,\% \qquad (1)$$

.

Product selectivity S_{H_2} is a qualitative evaluation determining the proportion of methane from a chemical reaction was used to form the target product that is hydrogen. In this work, hydrogen selectivity was calculated using the formula [25]:

$$S_{H_2} = \frac{1}{2} \cdot \frac{P_{H_2}(\text{out}) - P_{H_2}(\text{in})}{P_{CH_4}(\text{in}) - P_{CH_4}(\text{out})} \cdot 100\ \% \qquad (2)$$

Figure 5 shows the partial pressure diagram of gases. In formulas (1) and (2) $P_{H_2}(\text{in})$ and $P_{CH_4}(\text{in})$ are the partial pressures of hydrogen and methane at the beginning of the experiment, $P_{H_2}(\text{out})$ and $P_{CH_4}(\text{out})$ are the partial pressures of hydrogen and methane at the end of the experiment.

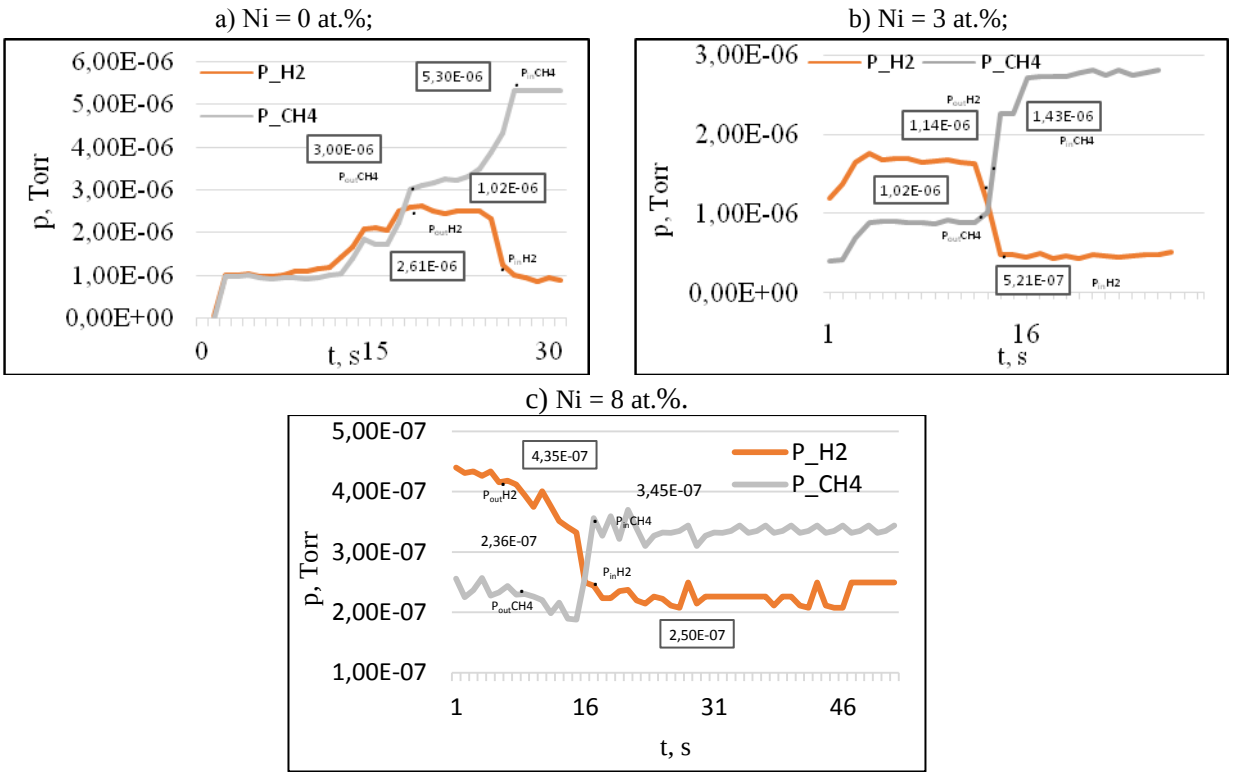


Figure 5. Diagram of partial pressures of gases in the result of the decomposition of methane with different nickel content in the catalyst

Table 1 shows the calculation data for the rate of methane conversion and hydrogen selectivity.

Table 1

Results of calculations of qualitative characteristics of conversion

#	W_{MW}, kW	Q_{Ar}, l/min	Q_{CH4}, l/min	T, °C	, %	, %
1	0.6	8.0±0.2	0.25±0.05	650±70	36±1	34±2
2					28±1	74±1
3					32±2	85±1
Note – W_{MW} — magnetron power, Q_{Ar} — argon consumption, Q_{CH4} — methane consumption, T — catalyst temperature, K_{CH_4} — methane conversion rate, S_{H_2} — hydrogen selectivity.						

Figure 6 shows a diagram of the dependence of the rate of methane conversion and hydrogen selectivity on the amount of nickel in the elemental composition of the catalyst.

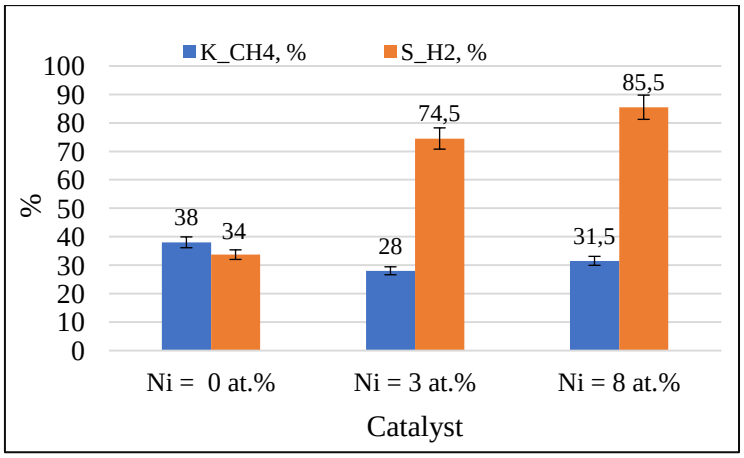


Figure 6. Diagram of the dependence of the rate of methane decomposition and hydrogen selectivity on at.% Ni in the catalyst

Figure 6 evidences that increasing the nickel content in the catalyst contributes to an increase in hydrogen selectivity.

Conclusions

Experiments were conducted to determine the maximum of methane decomposition rate in the microwave discharge using the PM-6 installation with a steel catalyst with varied nickel content. Three series of experiments were conducted with the same microwave discharge power, catalyst temperature, argon and methane flow rates. Based on the results of the experiments, it was found that increasing the Ni content in the steel catalyst to increase the rate of methane conversion and hydrogen selectivity is effective. The catalyst with 8 at.% nickel content, deposited electrochemically, showed the maximum hydrogen selectivity value.

Analysis of the reaction kinetics showed that the use of catalysts not only increases the rate of pyrolysis, but also helps improve the energy efficiency of the process.

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Метаннан сутегі өндірісі: аса жоғары жиілікті разряд пен катализаторды қолдану

Мақалада ПМ-6 қолданбалы зерттеу қондырғысында аса жоғары жиілікті (АЖЖ) разрядты және металл катализаторын қолдана отырып, метан пиролизі әдісімен сутегін алу бойынша тәжірибенің нәтижесі келтірілген. ПМ-6 қондырғысында аса жоғары жиілікті разрядта метан конверсиясының жоғары дәрежесін анықтау үшін, құрамында никель бар катализатор арқылы эксперименттер жүргізілді. Нәтижелер катализаторда никель құрамы жоғарылауы сутегі селективтілігінің жоғарылауына ықпал ететінін көрсетті. Метанның ең жоғары конверсия дәрежесі 32±2 % -ға дейін және сутегінің селективтілігі 85±1 % -ға дейін көрсеткіші, аса жоғары жиілікті разрядтың қуаты 0,6 кВт, метан шығыны 0,25±0,05 л/мин және аргон 8±0,2 л/мин қондырғы параметрлерінде қол жеткізілді. Жалпы алғанда, зерттеу нәтижелері метан пиролизінің оңтайлы шарттарына қол жеткізу және сутегі өндірісінің тиімді технологияларын құру үшін катализаторларды таңдаудың маңыздылығын көрсетеді. Тәжірибелерден алынған деректер өнеркәсіптік деңгейде метан пиролизін дамыту үшін пайдалы болуы мүмкін, бұл өз кезегінде энергияның тұрақты болашағына және көміртегі ізінің азаюына ықпал етеді.

Кілт сөздер: сутегі, метан пиролизі, сутегі энергетикасы, аса жоғары жиілікті разряд, метан конверсиясы, көміртек, сутегінің селективтілігі, катализатор.

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Водород из метана: применение СВЧ разряда и катализатора

В статье представлены результаты экспериментов по получению водорода методом пиролиза метана с применением сверхвысокочастотного (СВЧ) разряда и металлического катализатора с различным содержанием никеля на установке для прикладных исследований ПМ–6. Эксперименты проводились с целью определения наиболее полной степени разложения метана в микроволновом разряде на установке ПМ–6 с использованием стального катализатора с различным содержанием никеля. Результаты показали, что применение никелевых катализаторов обеспечивает наиболее высокий выход водорода при оптимальных условиях. Реализована технология получения водорода с использованием катализатора в СВЧ разряде с максимальной степенью конверсии метана до 32±2 % и селективностью водорода до 85±1 % при мощности СВЧ разряда 0,6 кВт, расходе метана 0,25±0,05 л/мин и аргона 8±0,2 л/мин. В целом, результаты исследования подчеркивают важность выбора катализаторов для достижения оптимальных условий пиролиза метана и создания эффективных технологий для производства водорода. Данные, полученные в ходе экспериментов, могут быть полезны для разработки промышленных установок пиролиза, что, в свою очередь, будет способствовать более устойчивому энергетическому будущему и снижению углеродного следа.

Ключевые слова: водород, пиролиз метана, водородная энергетика, сверхвысокочастотный разряд, конверсия метана, углерод, селективность водорода, катализатор.

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