INVESTIGATION OF THE EFFECT OF Fe ON Ni CATALYSTS FOR CARBONATE CONVERSION OF METHANE

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Abstract

In this work, an activated Ni catalyst for carbonate conversion of methane was prepared by placing γ-Al_2O_3 in solutions of nickel and iron nitrates. The time dependence of methane conversion was studied for different catalysts. In these processes, CO_2/CH_4=1:1,41 ratio was obtained. As a result of the research conducted, the methane effect of Fe on Ni/Al_2O_3 catalyst for conversion following possible mechanism was proposed.

The purpose of the work is to study the effect of Fe on the conversion of methane to carbonate, and the dependence of these processes on catalysts of different compositions and time.

Keywords: Methane conversion, methane, aluminium oxide, catalyst, conversion, nickel, iron.

Introduction

Synthesis gas is a mixture of carbon monoxide and hydrogen. The ratio of $CO:H_2$ varies from 1:1 to 1:3 depending on the synthesis gas production method. Synthesis gas production was considered one of the most important tasks of modern gas chemistry. Different H_2/CO ratios of synthesis gas can produce different valuable products.

Carbonate conversion

$$CH_4 + CO_2 \leftrightarrow 2CO + 2H_2 \Delta H = +247 \text{ kJ/mol}$$

To study the effect of ZrO_2 on the activity of the Ni-catalyst, $10Zr/Al_2O_3$ (without nickel) $5Ni5Zr/Al_2O_3$ $10Ni10Zr/Al_2O_3$ catalysts were prepared by adding γ-Al_2O_3 additives to the aqueous solution. Studying the conversion of $CH_4 + CO_2$ mixture on Pt/ZrO catalyst with the addition of molecular oxygen to the reactive system gave good results [1-6].

Co-catalysts are similar to nickel systems in terms of activity and stability. Like CoOMgO solid solutions and perovskites, cobalt oxide catalysts lose their activity when the composition of $Ca_{0,8}Sr_{0,2}Ti_{0,8}Co_{0,2}O_3$ changes. The most active among $Co/MgO/SiO_2$ catalysts is the system containing 50% MgO, the ability of Cobalt to reduce coke formation is known from the literature [7-10].

Experimental Part

In the work, $Fe(NO_3)_3 \cdot 6H_2O$ 94% nickel-free salt was used as a source of Fe. To evaluate the activity of the Fe catalyst first **$5Ni/Al_2O_3$** **$5Ni5Fe/Al_2O_3$** and **$10Ni5Fe/Al_2O_3$** samples were prepared, and the experimental results of the process of converting methane into carbon dioxide were studied. A 30% solution of catalysts was swallowed for 12 hours. Then, the catalyst was separated from the solution and dried at 350-450 °C for 6 hours in a nitrogen stream and reduced to a state of 7-9 mm granules.

Crystal 2000 (flame ionization detector) chromatograph was used.

Results and Discussion

According to the literature, the introduction of Fe into a Ni catalyst affects its activity and selectivity [11-16]. We studied the effect of Fe addition on the Ni/Al_2O_3 sample. A temperature of 800 °C was chosen to compare the catalytic properties of the samples.

Table 1.

Catalyst	Temperature °C	Time, h	Product yield %				conversion, %	
			H_2	CO	CO_2	CH_4	CH_4	CO_2
$5Ni/Al_2O_3$	800	2	40,9	48,5	8,3	2,3	90,4	76,8
$5Ni5Fe/Al_2O_3$	800	0,3	20,2	23,4	30,6	25,8	23,7	36,1
		4	21,8	22,3	33,4	22,5	32,3	29,0
		10,7	26,1	25,7	27,5	20,7	34,4	38,6
		13	29,0	30,6	19,9	20,5	33,8	54,7
$5Ni5Fe/Al_2O_3$	850	4,6	36,8	32,6	19,9	10,7	61,0	49,1
		8	37,7	30,8	22,0	9,5	65,6	43,9
$10Ni5Fe/Al_2O_3$	800	1	26,4	25,5	28,7	19,4	37,8	35,0
		3	25,4	24,5	29,1	21,0	33,3	34,7
		6	29,5	25,5	25,8	19,1	38,2	41,2
		8	28,7	25,1	27,1	19,1	38,4	38,4
$10Ni10Fe/Al_2O_3$	800	1	31,9	28,3	22,8	17,0	42,3	45,2
		4	30,5	26,3	25,0	18,2	38,8	40,7
$5Ni10Fe/Al_2O_3$	800	1	24,8	24,2	30,0	20,9	39,7	38,9
		14,5	26	26,3	27	20,7	34,6	38,8

As can be seen from the graphs presented in Figure 1, an increase in coke formation is observed in catalysts with a high content of nickel. When the nickel content of the Ni-Fe catalyst is 5%, the coke formation decreases, and then the coke formation increases with the increase of the iron catalyst content.

But iron reduces the activity of the catalyst, as we can see from Table 1, which leads to a decrease in conversion. Thus, to further reduce coke formation, it is preferable to use a catalyst with a low content of nickel and iron. It should also contain a small amount of iron to stop the decrease in catalyst activity.

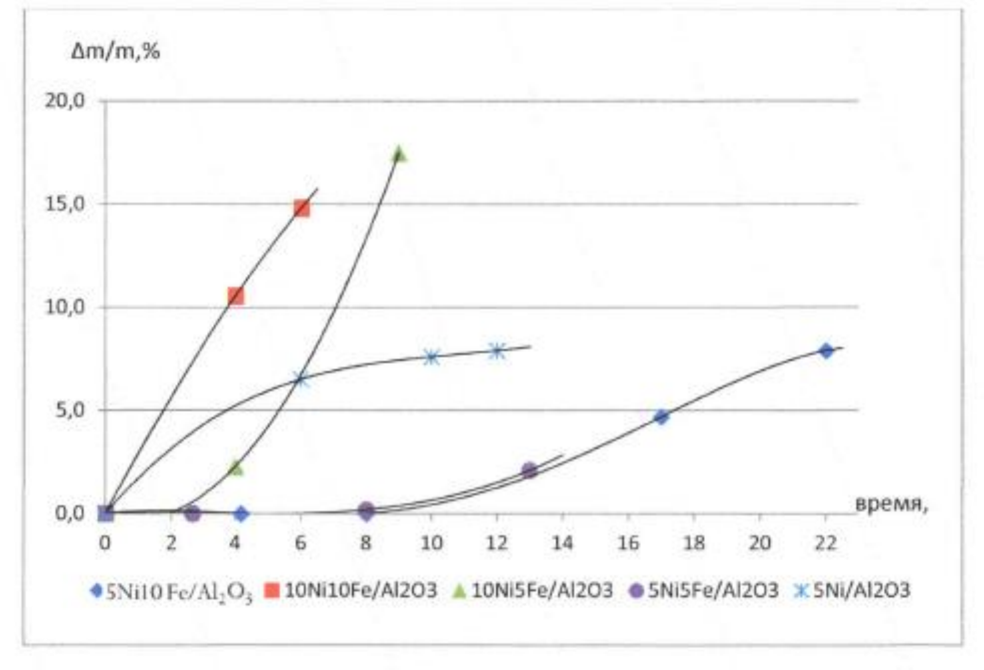

Figure 1. Time dependence of catalyst mass increase
(CO_2: CH_4 = 1.42, т = 800 °C).

Conclusion

1. The time dependence of methane conversion was studied for different catalysts.
2. $Fe(NO_3)_3 \cdot 6H_2O$ 94% salt was used as a source of Fe in the work. To evaluate the activity of the Fe catalyst first **5Ni/Al_2O_3 5Ni5Fe/Al_2O_3** and **10Ni5Fe/Al_2O_3** samples were prepared, and the experimental results of the process of converting methane into carbon dioxide were studied.
3. As a result of the research conducted, methaneconversionThe following the possible mechanism of the reaction was proposed.

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