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The effect of support type (Al_2O_3 , SiO_2) and features of the supported metal (Pd, Re, Ir) on the joint conversion of methane and n-pentane

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Abstract

Chemisorption of methane and catalytic performance in the joint conversion of CH_4 and $n\text{-C}_5\text{H}_{12}$ over supported monometallic catalysts were studied. The catalysts with Ir, Re or Pd supported on Al_2O_3 were shown to be more active in methane conversion as compared to the SiO_2 -supported catalysts. In the case of joint conversion of CH_4 and $n\text{-C}_5\text{H}_{12}$, the catalytic activity expressed in terms of the yield of aromatic hydrocarbons increases in the series of metals $\text{Re} \rightarrow \text{Ir} \rightarrow \text{Pd}$ irrespective of the type of support.

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1. Introduction

In recent years, the activation of C–H bond followed by conversion of low-molecular alkanes is the subject of many experimental and theoretical studies owing to its great scientific and industrial importance. The cleavage of C–H bond in methane is of particular interest, because this is the first step in the conversion of natural gas into more valuable chemical products. The available studies and publications on non-oxidative conversion of methane can be divided into two large groups.

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The first group includes the works dealing with the direct aromatization over zeolite catalysts at temperatures above 873 K. One of the first publications in the field was a paper by Wang L. and co-authors, who managed to subject methane to dehydroaromatization in a flow reactor over the Mo/H-ZSM-5 catalyst [1]. Although certain advances in investigation of zeolite catalysts were made in recent 20 years [2,3,4], the main difficulty related to the low stability of such catalysts (rapid deactivation) still remains unresolved.

The second group covers the studies on the two-step non-oxidative conversion of methane over metal oxide catalysts at lower temperatures. At the first step, methane is chemisorbed at 573-723 K with the cleavage of C–H bond and formation of molecular hydrogen. If the produced H₂ is removed from the system, the H-deficient CH_x (x < 3) fragments soon appear on the catalyst, and C–C bonds can emerge between the adjacent fragments. However, the precursors of C₂ hydrocarbons are bound quite strongly to the catalyst surface and cannot be desorbed by themselves. To remove them from the surface into the gas phase, holding in a hydrogen flow is necessary, often at a lower temperature – this is the second step [5, 6].

Recent studies revealed some catalytic systems able to activate the C–H bond in methane molecule for the both directions in non-oxidative conversion of methane. These are usually the transition metals.

By now, many studies were performed to elucidate the main regularities of methane conversion on individual atoms of metals as well as on their mono- and bimetallic clusters [7, 8]. The analysis of literature data showed that the main emphasis was made on such metals as Pt, Ru, Rh, Co, Mo, and Ni; however, some other transition metals also deserve attention.

This work is devoted to chemisorption of methane and its joint conversion with n-pentane in the presence of supported metal oxide catalysts (Pd/Al₂O₃, Re/Al₂O₃, Ir/Al₂O₃, Pd/SiO₂, Re/SiO₂, and Ir/SiO₂). The correlations between conversion of the indicated molecules and features of the supported metal, its particle size and type of support were revealed.

2. Experimental

2.1. Catalyst preparation

In the catalyst synthesis, the initial supports were represented by γ -Al₂O₃ (S_{sp} = 196.6 m²/g, V_{pore} = 0.42 cm³/g, d_{av} = 86 Å) and SiO₂ (S_{sp} = 320 m²/g, V_{pore} = 0.98 cm³/g, d_{av} = 122 Å). Re, Ir and Pd metals were deposited on aluminum and silicon oxides, which were precalcined in a flow of dry air at 773 K, by impregnation from solutions of HReO₄, H₂IrCl₆ and H₂PdCl₄. The samples dried at 393 K were calcined in flowing dry air at 773 K and then reduced in a hydrogen medium (Table 1).

Table 1. Content of metals in the tested samples.

Metal content on a catalyst, wt.%					
Pd		Re		Ir	
Al ₂ O ₃	SiO ₂	Al ₂ O ₃	SiO ₂	Al ₂ O ₃	SiO ₂
0.76	1.33	0.71	1.45	1.50	≈1.0

2.2. Characterization

The size and localization of supported metal were determined on a JEM-2100 JEOL electron microscope (accelerating voltage of 200 kV, crystal lattice resolution of 0.145 nm) with an INCA-250 Oxford Instruments energy dispersive X-ray spectrometer. To prepare samples for transmission electron microscopy (TEM) examination, they were ground in an agate mortar and then dispersed in ethanol at a frequency of 44 kHz. The

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