

It was shown that the activity of catalysts will remain at a sufficiently high level (methane conversion degree (α_{CH_4}) = 80 – 90 %) if the oxidation of methane and other substances proceeds at temperatures no higher than 973 – 1,000 K, when significant amounts of Ni and Co aluminates are not formed in the composition of catalyst. Under influence of oxygen at higher temperatures (which can occur due to overheating), the amount of mixed oxides in the catalyst decreases in the oxidizing atmosphere according to the TPD data. Less active Ni and Co aluminates are formed during oxidation processes.

Studies have shown that the formed Ni and Cu aluminates can be reduced to the initial oxides or their mixtures under the influence of hydrogen at 973 – 1,223 K. The interaction of the reducing agent (hydrogen) in the TPR experiments with the above oxygen structures occurs easily, starting from 473 and up to 673 K. Adsorption of oxygen again on Ni - Cu - Cr catalysts after the decomposition of oxides occurs at low temperatures (325 K). This indicates a high reactivity of adsorbed and lattice oxygen of dispersed oxides, as well as their mixtures and its ability to easily reactivate.

The ability of the resulting $Cu(Ni)Al_2O_4$ aluminates to convert again to oxides under the influence of hydrogen (Table 2) was used to regenerate the catalyst.

Table 2: Influence of treatment conditions of the Ni - Cu - Cr catalyst on its activity in oxidative conversion of CH_4

| Catalyst treatment conditions | Temperature, K | | |
|---------------------------------|----------------|------------------------|------------------------|
| | Ni Start | Cu $\alpha = 50 \%$ | Cr $\alpha = 90 \%$ |
| Air, 873 K | 643 | 800 | 973 |
| Air, 1,373 K, 0.5 h | 773 | 923 | - |
| H ₂ , 1,223 K, 0.5 h | 663 | 823 | 973 |

The activity of catalyst was determined in the oxidative conversion of toluene, which was a frequently present substance in industrial gas emissions. The catalysts were tested at a space velocity of $5 \times 10^3 \text{ h}^{-1}$, in the temperature range of 523 – 723 K and a toluene concentration of 320 mg m^{-3} . The degree of toluene conversion increases on all catalysts as the temperature rises from 523 to 723 K. The degree of toluene conversion increases from 74 to 98.8 % at a temperature of 673 – 723 K on an optimal Ni - Cu - Cr/2 % Ce/ θ - Al_2O_3 catalyst. A further increase in temperature did not affect the degree of toluene conversion. It was found that the polyoxide Ni - Cu - Cr catalyst supported on 2 % Ce/ θ - Al_2O_3 provides 98.8 % conversion of toluene to CO_2 at optimum temperature of 723 K in the reaction of toluene oxidation at a space velocity of $5 \times 10^3 \text{ h}^{-1}$ and toluene content equal to 320 mg m^{-3} in the initial mixture with air.

The effect of space velocities of the deep conversion of toluene on effective Ni - Cu - Cr/2 % Ce/ θ - Al_2O_3 catalyst was studied. The toluene conversion on the catalyst was studied in the temperature range of 573 – 773 K with a toluene concentration in the initial mixture equal to 320 mg m^{-3} .

Experimental data showed that the toluene conversion degree decreases from 89.5 to 83.8 % on the Cu - Cr/2 % Ce/ θ - Al_2O_3 catalyst at 573 K and increase of the space velocity from 5 to $5 \times 10^3 \text{ h}^{-1}$. The 85.7 % toluene conversion degree was observed on the catalyst at space velocity of $10 \times 10^3 \text{ h}^{-1}$. A similar dependence of toluene conversion is observed at a temperature of 673 K. The toluene conversion degree in the indicated ranges of space velocities is higher than conversion at a reaction temperature of 573 K. The highest conversion of toluene to 98.5 % is achieved at a temperature of 723 - 773 K and a space velocity of $5 \times 10^3 \text{ h}^{-1}$. Toluene conversion decreases both at 573 and at 673 K at higher space velocities ($10 - 15 \times 10^3 \text{ h}^{-1}$).

The effect of toluene concentration in the initial mixture on its conversion on Ni - Cu - Cr catalysts was studied. Increase the toluene concentration in the initial mixture from 100 to 570 mg m^{-3} leads to a slight decrease of the toluene conversion degree on two-component Ni - Cu/2 % Ce/ θ - Al_2O_3 , Cu - Cr/2 % Ce/ θ - Al_2O_3 , and Ni - Cr/2 % Ce/ θ - Al_2O_3 catalysts. The highest toluene conversion degree (up to 98.8 %) is observed on the ternary Ni - Cu - Cr/2 % Ce/ θ - Al_2O_3 catalyst.

4. Conclusions

As a result of the studies, low-percentage oxide catalysts without noble metals for the purification of waste gases from toluene were developed and investigated by TPD, TPR, TPO, XRD, and TEM methods. It was shown that the resulting Ni and Cu aluminates can be reduced to the initial oxides or their mixtures under the influence of H_2 at 973 – 1,223 K. 98.8 % purification is provided on an effective thermal stable polyoxide catalyst while optimizing process parameters (temperature, space velocity, concentration of toluene in the gas mixture) of deep toluene oxidation at a space velocity of $5,000 \text{ h}^{-1}$ in the temperature range of 723 - 773 K at 100 - 570 mg m^{-3} toluene content in the initial reaction mixture.