

Observation the initial radiation to the surface of mixed nano catalyst on oxidation processes

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Abstract. In this paper the oxidation/conversion process of carbon-monoxide has been investigated on the surface of catalysts ($T_{Al_2O_3}$, $T_{Al_2O_3+CuO}$ and $RT_{Al_2O_3+CuO}$) which has been modified by radiation. The catalyst samples have been radiated in various absorption doses by the source of ^{60}Co isotope device in order to analyze distribution of anion centers on the surface. It was found out by the examples of investigation, the size and element concentration of anion centers (atoms O^-) depending on absorption dose. The results show that the amount of paramagnetic centers on surface have increased. The average activation energy of process decrease and conversion happens faster. The triangular oxide can also form on the square lattice of nano-catalyst of Al_2O_3 under CO oxidation conditions, similar features are observed. Furthermore, oxides are unstable outside the O_2 atmosphere, indicating the presence of active O atoms, crucial for oxidation catalysts.

Key words: conversion rate, paramagnetic centers, absorption, radiation dose, initial radiation, nano-catalyst.

Introduction.

Investigation of oxidation processes on the surface of nano- heterogenic catalyst modified by the gamma radiation have demand for implementation. The emerged anion centers, influence of hole centers in radiolysis-catalytic transitions and the working mechanism of these process is today's actual challenge. Adsorption and desorption processes on the surface of the dielectric oxides, emerging of O^- centers, transmission of CO to CO_2 is depending on amount of atomic oxygen [1,2]

The major objective of the research is define the more cheaper and the accessible material for catalyst which is modified on radiation and capable of neutralizing the exhaust emissions from different sectors of industry and transport while executing conversion processes according to the exhaust temperature of internal combustion [3].

Methodology:

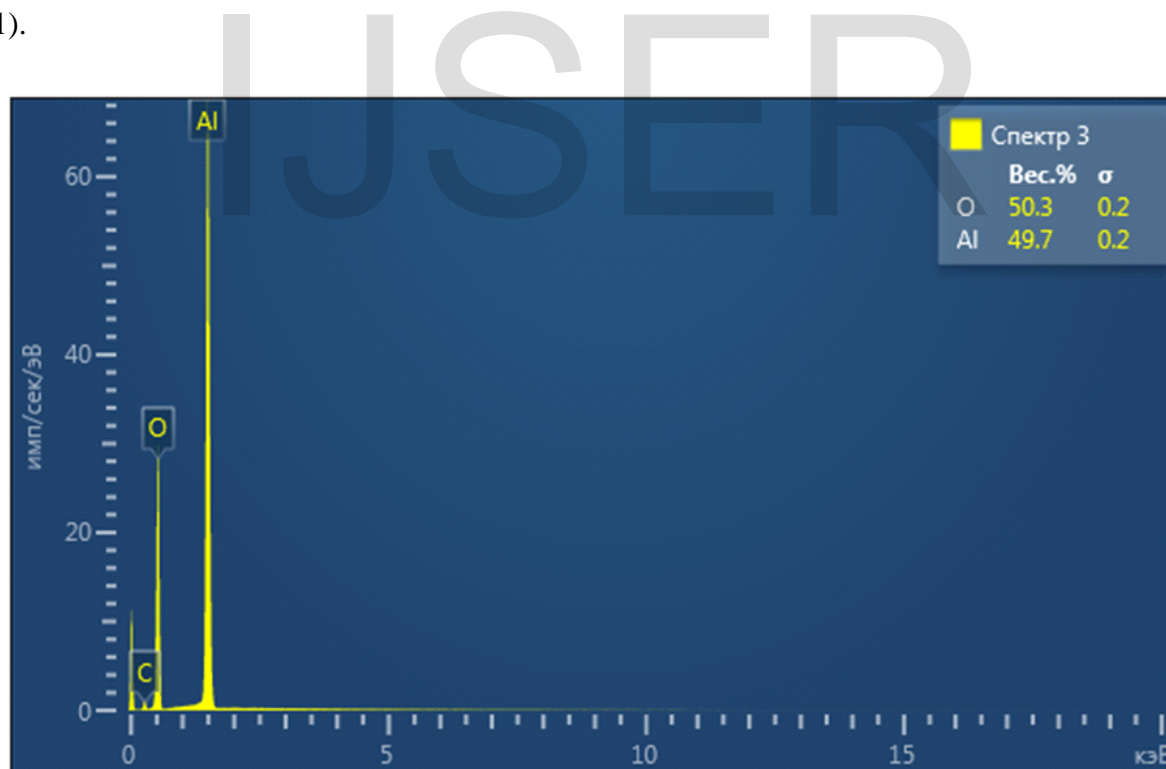
The initial surface modification process: Mixed nano Al_2O_3+CuO catalyst with $d = 20-30$ nm (nano scale) and $m = 7.32$ g total weight has been taken as a research material which is modified by γ -radiation in the ^{60}Co isotope device under normal room temperature. Active

centers emerged on the surface of catalyst and their sum element amount has been analyzed by SEM (Scanning Electron Microscope, Zeiss, SIGMA/VP). The main goal of initial modification to research the change of catalyst activeness in the conversion processes, radiation endurance of emerged centers while could use of long time in repetitive processes. Research works, oxidation processes have been carried out insulated reactor of flow mixed gas, while the gas concentration has been analyzed in gas chromatograph [3].

Discussing results:

The catalyst have been radiated in an air condition, room temperature with the dose of $P=0.158$ Gy/s during $\tau=17$ hr. [3]. As a result of modification by the gamma radiation, activity on the surface of catalysts, alteration of density and morphologic nature of an active anion centers(O⁻) has been increased. The study of new conversion processes on the surface have been acquired. Samples have been zoomed 1.0 million times and measurements in the scale of 1.3 nm have been taken. The element content of Al₂O₃/CuO catalyst, as white color in the range of 6 nm diameter.

The catalyst samples have been radiated in various absorption doses in order to analyze distribution of anion centers on the surface. As it seen by the pictures, their size and element content of anion centers depending on absorption dose. It found out that the amount of paramagnetic centers on surface have increased dependent on radiation absorption dose (pic. 1).

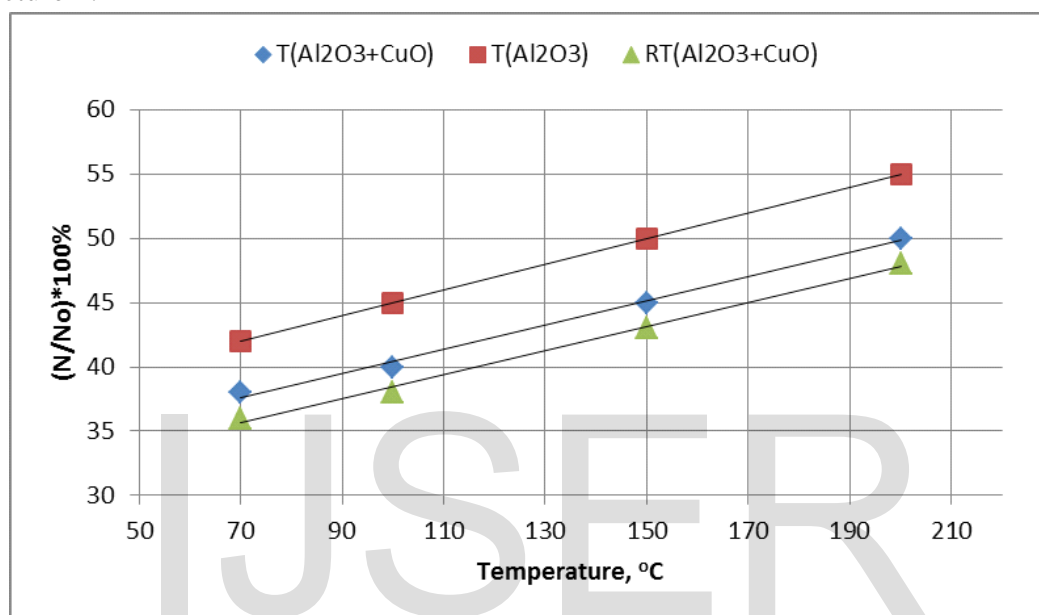


Picture 1. Dependence of mass fraction and atomic quantity of [O] - % on the surface of nano- catalyst of Al₂O₃/CuO.

Dependence curve of mass fraction and atomic quantity of atomic oxygen on the surface of catalyst in various absorption doses has been defined.

As it seen by the picture 1, atomic mass fraction of oxygen(O⁻) has shown steady increase from 46% to 54% total in the interval of D=0÷20 kGy dose. Increase line stabilizes in higher dose intervals while the process of mono layer generation starts.

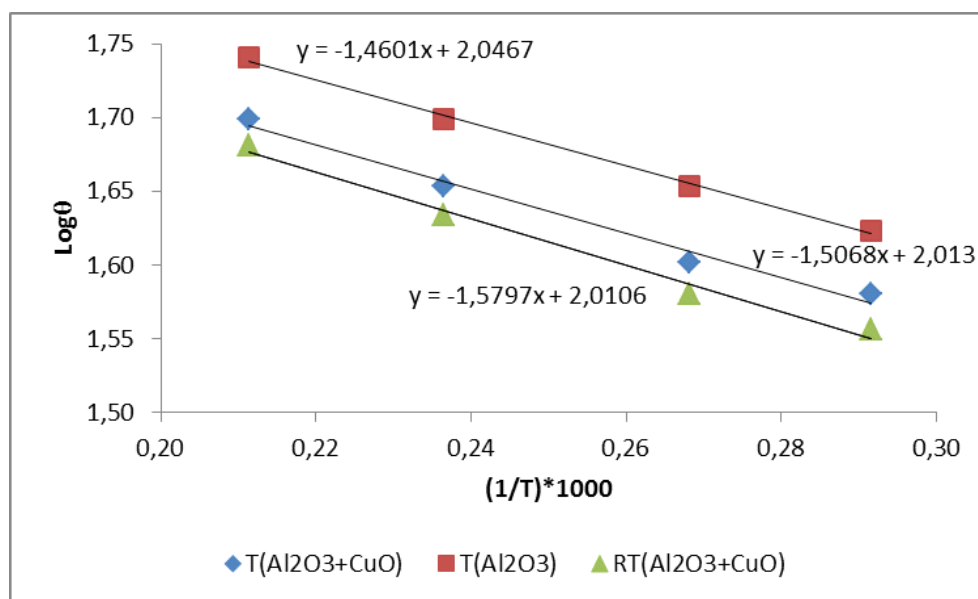
Experimental dependences of the conducted research (pic. 1) shows that oxidation/ conversion rate of carbon monoxide on the surface of nano-catalysts (T(Al₂O₃), T(Al₂O₃+CuO) and RT(Al₂O₃+CuO)) has been investigated on three types of catalyst surface in different temperatures. Firstly, thermally modified pure nano-Al₂O₃ catalyst, secondly, thermally modified mixed nano-Al₂O₃+CuO and finally mixed Al₂O₃+CuO catalyst which has modified by γ -radiation. For all three cases the dependence curves has been displayed in the picture 2.



Picture 2. Comparative curves of oxidation/conversion ((N/No)*100%) of carbon monoxide on the surface of nano catalyst by the dependence on temperature.

As it is figured in the picture, in all three cases the conversion rate increases so as the temperature increase, tangent alphas of curves are relatively different. Thus, in the conversion on pure Al₂O₃ catalyst tg=0,10 in Al₂O₃+CuO tg=0.092 and in irradiated Al₂O₃+CuO catalyst tg=0.085, which has caused small reduction in activation energy in mixed and modified systems while the reaction speed is increase. As a result of mixed catalysts modified by surface gamma radiation have higher activeness while the conversion rate has increased 7 -10 % [4-5].

Arrhenius coordinates of conversion/oxidation processes on the surface of all three types of catalyst and the results of experimental assessment has been illustrated in picture 3. CO conversion for all three cases - pure nano Al₂O₃, mixed nano Al₂O₃+CuO and mixed Al₂O₃+CuO catalyst that has been initially modified by radiation - is shown in Arrhenius coordinates.



Picture 3. The temperature dependence on CO conversion (log) on the surface of nano-catalyst in Arrhenius coordinates (1-Al₂O₃, 2-T Al₂O₃+CuO, 3-RT Al₂O₃+CuO)

It is found out that the energy activation on the surface of catalyst is $E_{T-Al_2O_3} = -6.68$ kC/mol, $K_0 = 5.23 \cdot 10^7 \text{ sec}^{-1}$, mixed with CuO energy activation $E_{T-Al_2O_3+CuO} = -6.89$ kC/mol, $K_0 = 5.13 \cdot 10^7 \text{ sec}^{-1}$, on the catalyst of initial modification on radiation has lower energy activation of $E_{RT-Al_2O_3+CuO} = -7.23$ kC/mol, $K_0 = 5.03 \cdot 10^7 \text{ sec}^{-1}$.

Conclusions

It was found out that the mass fraction of O atoms is emerged on the surface of mixed nano Al₂O₃ and Al₂O₃-CuO via increase of conversion rate by depending on absorption dose. In this case, CO can react with the O atoms of the surface oxide at Mars-van Krevelen reaction mechanism. The initial radiation are create additional anion centers [O] on the surface of catalyst which is influence to increase the speeds of conversion reaction as well as increase conversion rate. The initial radiation catalyst have higher activeness while the conversion rate at more than 7 -10% higher. The average activation energy is decrease while conversion happens more faster.

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