FUNCTIONALITY AND SELECTIVITY OF A THERMOCATALYTIC CATALYST FOR HYDROGEN SULFIDE SENSING

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Hydrocarbon, gasoline, diesel fuel, propane / butane mixture, catalyst, platinum, palladium, a metal oxide, cobalt oxide, Nickel oxide, aluminium oxide, manganese oxide, carbon monoxide, oxide of cadmium.

ANNOTATSIYA:

The results of experiments studied the activity, selectivity and stability of catalysts based on metal oxides with the additive and without the addition of Pt and Pd in the oxidation process of hydrocarbons.

Experiments on the oxidation of combustible gases on different catalysts show the possibility of manufacturing the selective catalytic thermal sensors for detection of hydrocarbon vapors from industrial gaseous emissions (from the emissions of boilers, furnaces, exhaust gases, etc.)

Exhaust flue gases from combustion plants, exhaust gases from internal combustion engines and gaseous emissions from chemical technological processes, where constant control of the amount of carbon monoxide and hydrocarbons is required, is a complex multicomponent mixture containing water vapor, nitrogen and sulfur oxides that can poison oxide catalysts [1]. In this regard, special requirements are imposed on the catalyst of thermocatalytic sensors CO and CnHm.

When studying the catalytic activity in relation to the reaction of complete oxidation of hydrocarbons, we found that the highest degree of oxidation is achieved in the presence of a CoO-based catalyst where at 400 ^{0}C the degree of gasoline conversion is 89.5%. Complete 100% oxidation of hydrocarbons (gasoline vapors) in the presence of the most active metal oxides is achieved only at temperatures above 400^{0}C.

The use of high-temperature catalysts in the thermocatalytic method leads to a reduction in the operating life of the sensor [2, 3]. As was noted, maintaining high activity and service life of catalysts can be achieved by introducing Pt, Pd, etc. into their composition [4]. An analysis of literature data and preliminary experimental studies shows that the most promising thermocatalytic sensor catalyst for determining hydrocarbons in exhaust and flue gases are: oxides of cobalt, manganese, nickel, etc. with the addition of platinum group metals [2-4].

During experiments on selecting a catalyst for a low-temperature thermocatalytic hydrocarbon sensor, the catalytic characteristics of the following systems were studied: Pt/Al_2O_3, Pd/Al_2O_3, $Pt\text{-}NiO/Al_2O_3$, $Pt\text{-}MnO_2/Al_2O_3$, $Pt\text{-}CoO/Al_2O_3$, $Pt\text{-}CoO\text{-}MnO_2/Al_2O_3$, $Pt\text{-}CoO\text{-}NiO/Al_2O_3$, $Pt\text{-}NiO\text{-}MnO_2/Al_2O_3$.

The catalysts used in the work were pre-activated by heat treatment at 700 ^{0}C for 4 hours. Considering that the pre-treated catalyst may initially have a reduced or, conversely, increased activity, which can cause underheating or overheating, leading to undesirable physical and chemical changes, therefore, catalyst training was required. In this regard, before starting work, uninvestigated catalysts were trained under conditions limiting the reaction rate: at low temperature (150 ^{0}C and 300 ^{0}C) and low feed rate of the vapor-gas mixture. In the experiments, the mass of the catalyst loaded into the reactor was 10 g, onto the surface of which a vapor-gas mixture was supplied at a speed of 10 l/h. Experiments to study the activity and stability of catalysts for the oxidation of hydrocarbons were carried out at a temperature of 150-450 ^{0}C, a steam-gas mixture supply rate of 10 l/h and a vapor content of PBS - 0.25 vol%, gasoline - 0.23 vol%, diesel fuel - 0. 18 rev. %.

The results obtained show that on the studied catalysts, temperatures ensuring complete (100%) oxidation of AI-92 gasoline vapors have the following values: Pt/Al_2O_3-250^{0}C; $Pt\text{-}CoO\text{-}MnO_2/Al_2O_3$-300^{0}C; $Pt\text{-}CoO/Al_2O_3$-350^{0}C; Pd/Al_2O_3-350^{0}C; $Pt\text{-}NiO\text{-}MnO_2/Al_2O_3$ - 400^{0}C; $Pt\text{-}MnO_2/Al_2O_3$ - 400^{0}C; Pt- NiO-CoO-/Al_2O_3 -450 ^{0}C; $Pt\text{-}NiO/Al_2O_3$ more than 450^{0}C. Low yields of carbon dioxide were obtained on the $Pt\text{-}NiO/Al_2O_3$ catalyst, which indicates incomplete oxidation of hydrocarbons under the selected conditions. The increase in the temperature of complete oxidation of diesel fuel on selected catalysts compared to gasoline and PBS is explained by the fact that diesel fuel contains a large amount of hydrocarbons of various classes, which are characterized by a low oxidation rate.

The activity of the $Pt\text{-}NiO/Al_2O_3$, catalyst is lower than that of Pt/Al_2O_3, $Pt\text{-}CoO/Al_2O_3$, but this catalyst is more stable. The catalysts Pt/ Al_2O_3, Pt-CoO/ Al_2O_3, $Pt\text{-}CoO\text{-}MnO_2/Al_2O_3$ are characterized by high activity and stability under the conditions studied. In the presence of these catalysts, the degree of deep oxidation of gasoline at a temperature of 325 ^{0}C and a mixture feed rate of 10 l/h is 99.5-100.0%. These catalysts retained their original activity almost constantly during 1500 hours of operation. The studied catalysts for the oxidation of hydrocarbons can be arranged in the following sequence in terms of operational stability: $Pt/Al_2O_3 > Pt\text{-}CoO\text{-}MnO_2/Al_2O_3 > Pt\text{-}CoO/Al_2O_3 > Pd/Al_2O_3 > Pt\text{-}NiO\text{-}MnO_2/Al_2O_3 > Pt\text{-}MnO_2/Al_2O_3 > Pt\text{-}CoO\text{-}NiO/Al_2O_3 > Pt\text{–}NiO/Al_2O_3$. The results of studying the activity of catalytic systems during the oxidation of hydrocarbons show that for the thermocatalytic determination of hydrocarbons (PBS vapors, gasoline and diesel fuel in exhaust and flue gases, it is more advisable to use the following catalysts: Pt/Al_2O_3, $Pt-CoO-MnO_2/Al_2O_3$. Therefore, all further experiments were carried out on them.

The influence of the initial hydrocarbon products (diesel fuel and gasoline vapors) and oxygen on the oxidation of hydrocarbons was studied in the presence of Pt/Al_2O_3 and $Pt-CoO-MnO_2/Al_2O_3$ catalysts in the temperature range 250-350 ^{0}C. The experiments were carried out in the range of hydrocarbon concentrations of 500 - 2500 ppm and oxygen concentrations of 10, 21 and 99 vol.%. When studying the effect of oxygen concentration, a mixture of oxygen (O_2 -10 vol.%) and nitrogen (up to 90 vol.%), air (O_2 -21 vol.%) and technical oxygen (O_2 -99 vol.%) were used as an oxidizing gas. %). Experiments have shown that in the studied concentration range, oxygen in the mixture does not have a significant effect on the degree of fuel oxidation.

In the presence of $Pt-CoO-MnO_2/Al_2O_3$ at a temperature of 350 ^{0}C in the range of gasoline vapor concentrations of 500 - 1500 ppm, ~100% conversion of gasoline, diesel fuel and PBS vapors is observed. Consequently, the oxidation rate under these conditions is of zero order in relation to the studied concentrations of fuel vapor and oxygen. The results obtained are of particular importance if we keep in mind that the technique will be recommended for determining the concentration of petroleum product vapors in the exhaust gases of internal combustion engines and in the air near gas stations, KZP, oil depots, etc.

As is known, each combustible substance has its own specific oxidation temperature with atmospheric oxygen, which can be used to ensure selectivity of the catalytic oxidation of combustible substances on the surface of thermosensitive elements, using catalysts of the same composition. Our research has shown that the use of this method of ensuring selectivity involuntarily clutters and complicates the design of the device due to the use of two autonomous power supplies [4,5].

In this regard, in our work we used a method for ensuring the selectivity of thermocatalytic methods, based on the use of thermosensitive elements - sensors containing catalysts with different activities towards the components of the gas mixture. In this case, the output signal of the first element is proportional to the total concentration of hydrogen, carbon monoxide and hydrocarbons (methane, gasoline vapor, diesel fuel, etc.), the output signal of the second element is proportional only to the concentration of hydrogen and carbon monoxide, and the difference between the signals of the first and second elements is proportional to the concentration of the determined hydrocarbon component.

When developing selective thermocatalytic hydrocarbon sensors based on the use of measuring and compensation elements containing catalysts with inadequate activity to different components of the gas mixture, it was found that $Pd-NiO/Al_2O_3$ can be used as a comparative element catalyst. As we have found, in the presence of this catalyst, 100% oxidation of hydrogen and carbon monoxide in the presence of hydrocarbons is ensured. As a result of the experiments, highly efficient (active and stable) catalysts were selected for comparative ($Pd\text{-}NiO/Al_2O_3$) and measuring ($Pt\text{-}CoO\text{-}MnO_2/Al_2O_3$) sensitive elements of the TCS of hydrocarbons and optimal conditions for the oxidation of gasoline, diesel fuel and PBS vapors, optimal oxidation conditions which in the range of their concentrations 500 - 1500 ppm are the temperature of 275-350 ^{0}C and the rate of supply of the vapor-gas mixture to the surface of the catalyst, no more than 15 l/h.

Conclusion. As a result of the experiments, the activity, selectivity and stability of metal oxides with and without the addition of Pt and Pd in the process of hydrocarbon oxidation were studied. A selective method for thermocatalytic monitoring of carbon monoxide and hydrocarbons in a wide range of their concentrations in exhaust, flue and process gases has been developed.

Experiments carried out on the oxidation of flammable gases on various catalysts show the possibility of manufacturing selective thermocatalytic sensors for determining carbon monoxide and hydrocarbon vapors from industrial gaseous emissions (from boiler installations, roasting furnaces, vehicle exhaust gases, etc.).

When developing selective thermocatalytic sensors for hydrocarbons based on the use of measuring and compensation elements containing catalysts with inadequate activity to different components of the gas mixture, it was found that for the thermocatalytic determination of hydrocarbons in exhaust and flue gases, it is more expedient to use the following catalysts as a catalyst for the measuring sensitive element of the sensor: Pt/Al_2O_3, $Pt\text{-}CoO\text{-}MnO_2/Al_2O_3$.

$Pd\text{-}NiO/Al_2O_3$ can be used as a comparative element catalyst. As we have found, in the presence of this catalyst, 100% oxidation of hydrogen and carbon monoxide in the presence of hydrocarbons is ensured.

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