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Thermocatalytic and Semiconductor Sensors for Monitoring Gas Mixtures

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Abstract: Today in the world, special attention is paid to the creation of express and inexpensive chemical sensors for reliable and unambiguous control of leaks and accumulations of natural gas. Analysis of the development of gas sensors in industrially developed countries has shown that the use of thermocatalytic and semiconductor sensors is the most promising for preventing explosion. The composition of the catalyst of the measuring and compensating sensing element of the methane sensor is established in the work. Using a technique to ensure selectivity with the use of the above catalysts, a thermocatalytic sensor was manufactured for the selective determination of methane in the presence of carbon monoxide and hydrogen.

Keywords: thermocatalytic, circumstance, element, measuring, hydrogen.

With the introduction of new technologies and the development of analytical control, the requirements for the sensitivity and selectivity of methods for determining substances are increasing. A promising modern direction in the development of sensors for the analysis of the concentration of flammable explosive and toxic gases is the use of semiconductor and thermocatalytic sensors. Considering the above, the development of new highly efficient sensors for fire and explosive gases, in particular methane, remains an urgent problem.

With the development of various sectors of the economy, especially the oil and gas industry, transport and energy on a global scale, the requirements for ensuring explosion safety of industrial and household facilities are being tightened. This circumstance determines the relevance of research aimed at the development of express, sensitive and selective sensors that provide reliable control of the explosion hazard of gas mixtures of closed ecological systems.

In this work, using selected optimal conditions and selective catalysts of measuring and compensating elements of the TCS, high sensitivity and selectivity of the determination of CH4 from the composition of atmospheric air of closed ecological systems is ensured. The principle of operation of this control method is the flameless combustion of CH4 on the surface of the catalyst and the measurement of the amount of heat released in this case [1,2]. When exposed to methane molecules on the surface of the catalyst of the measuring element, a reaction occurs, accompanied by the release of heat and a change in its resistance. At the same time, the resistance of the comparative element does not change, since methane oxidation is not observed on it due to the absence of a deep oxidation catalyst. As a result, the bridge is unbalanced and the output voltage proportional to the concentration of methane in the analyzed mixture becomes the controlled analytical signal of the sensor Therefore, in the developed sensor, the concentration of the imbalance. Since the measuring element is coated with deep oxidation catalysts, the reaction proceeds, as well as with conventional gorenje, in accordance with the formula [1]:

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CH₄+ 2O₂= CO₂+ 2H₂O + 805,2 кДж/моль

(1.)

When heat is released, the resistance R of the spiral changes by the value ΔR . The resistance of the spiral is determined by the formula:

$$R = R_0 (1 + \alpha \Delta T)$$
 (2.)

where R0 is the resistance of the spiral at $T = 25 \degree C$; α is the temperature coefficient of resistance of the platinum wire, ΔT is the temperature change of the spiral. Since platinum has a sufficiently large temperature coefficient, the temperature increase caused by oxidation causes a significant change in the resistance of the measuring element. Usually, in existing sensors, the concentration of methane CH4 = 1% vol causes an increase in the temperature of the catalytically active (measuring) element by 20 - 300C [3]. This ensures a relatively high sensitivity of the developed methane sensor. The oxidation reaction of combustible gases on heterogeneous catalysts can occur in the kinetic and diffusion regions [4]. The rate of methane oxidation reaction in the kinetic region is usually represented as a function of volume-molar concentrations of methane and oxygen.

 $W = kF \mathfrak{I}(C_{CH4}C_{O2}) \qquad (3)$

where w is the reaction rate, mol/s; k is the reaction rate constant, c-1; Fe is the active surface of the catalyst, m2. The value of k does not depend on the concentration of methane in the air and increases with increasing temperature.

It was found that the catalyst 0.75in2o3-0,25Ag2O is the most optimal for the measuring sensor element of the thermocatalytic sensor of methane. As a catalyst for a comparative element, it is advisable to use 0.25 Fe3O4-0.75ni2o3. This composition ensures high selectivity of CO and H2 oxidation in the presence of methane. Using the method of ensuring selectivity [5] with the use of the above catalysts, we manufactured a thermocatalytic sensor for the selective determination of methane in the presence of carbon monoxide and hydrogen (Fig. 1.).

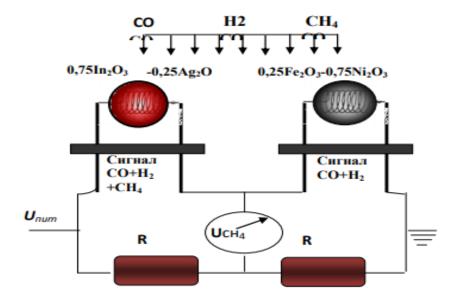


Рис.1. Селективный термокаталитический сенсор метана.

The output signal of the measuring sensor element of the sensor (catalyst: 0,75In2O3-0,25Ag2O) is proportional to the total concentration of combustible gases (hydrogen, carbon monoxide and hydrocarbons), the output signal of the comparative sensor element is proportional to the concentration of a mixture of gases (hydrogen and carbon monoxide) without a selective detectable



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component (methane), and the difference between the signals of the first and second the concentration of elements is proportional to the concentration of the component being determined-methane.

The results of the determination of methane and natural gas are shown in Figure 2 and Table 1. As follows from the data given, a thermocatalytic sensor with a compensation element catalyst is characterized by high selectivity in the determination of methane-containing gas in the presence of

CO and H2. (fig. 2 and Table 1)

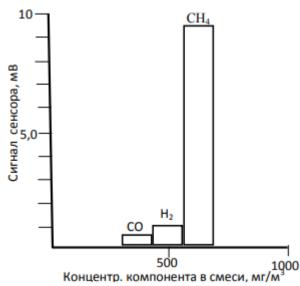


Fig.2. Selectivity of the thermocatalytic sensor in the determination of methane in the presence of hydrogen and carbon monoxide

Таблица 1.

№	Состав газовой смеси, %об	Найдено природ газ, об%		
п/п		x±Δx	S	Sr*10 ²
1	Природ. газ (0,50)+воздух(ост)	0,48±0,06	0,05	1,2
2	Природ. газ (0,50)+СО(1,00)+возд(ост)	0,52±0,03	0,03	1,6
3	Природ. газ (0,50)+H ₂ (1,00)+возд(ост)	0,51±0,03	0,04	1,7

Результаты определения селективности сенсора природного газа (n=5, P=0,95)

Thermocatalytic sensors of methane are based on the principle of flameless oxidation of combustible gases on the surface of a catalyst deposited on a thermistor, usually a platinum spiral. The heat released during the oxidation of methane heats up the platinum wire, which leads to a change in the resistance of the sensor. The advantages of catalytic sensors are ease of manufacture, small size and power consumption, almost linear output signal. To measure methane concentrations, two sensing elements are usually used: working and comparative (compensatory). Two sensing elements are included in one branch of the Wheatstone bridge and placed in the test medium, two permanent resistors are included in the second branch of the bridge. Catalytic sensors have good selectivity to combustible gases, but initially they are calibrated only for methane, for other combustible gases, a recalculation of the readings is required. Sensors of this type work well with pre-explosive concentrations of combustible gases and vapors, selectivity to combustible gas is their main advantage. Their affordability and ease of maintenance make them the most common sensors



for monitoring the explosion safety of the environment. The reaction chamber of the first thermocatalytic sensors was a cylinder made of a double metal mesh having an inner diameter of about 15 mm and a height of 15-20 mm. The design of the thermocatalytic sensors currently being manufactured has significantly smaller dimensions. The reaction chamber is made of ceramic or cermet with an internal diameter of 5-6 mm. [6].

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