CHEMICAL SENSOR FOR MONITORING FIRE- AND EXPLOSION-HAZARDOUS CONCENTRATIONS OF NATURAL GAS

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Abstract: Methane explosions remain one of the most hazardous accidents in domestic and industrial facilities. The lower and upper explosive limits of methane are 5% and 16% respectively, which necessitates the development of chemical sensors to ensure explosion and fire safety. In this work, sensitive and selective thermocatalytic and semiconductor methane sensors were developed for natural gas monitoring. The catalytic composition based on $0.75In_2O_3–0.25Ag_2O$ and $0.25Fe_3O_4–0.75Ni_2O_3$ was synthesized using sol-gel technology, providing high sensitivity and selectivity towards methane in the presence of CO and H_2. Comparative studies conducted in the Gas Analysis Laboratory of Samarkand State University (1990–2019) demonstrated that the newly developed TCS-1 significantly outperforms previously designed analogs (TCS-2 and TCS-3) and commercial sensors in terms of selectivity and response time. The proposed sensors exhibit rapid response characteristics ($t_{0.1}$ = 3–4 s; t_p = 17–18 s) and a linear dependence of output signal on methane concentration in the range of 0.1–5.0% vol. The results confirm the applicability of the developed methane sensors for express control of natural gas concentrations in the atmospheric air of industrial and domestic facilities, as well as in transport cabins, ensuring reliable monitoring of fire- and explosion-hazardous gas levels.

Keywords: methane sensor, thermocatalytic sensor (TCS), semiconductor sensor, natural gas monitoring, explosion safety, sol-gel synthesis, gas analysis

One of the most dangerous types of accidents in domestic and industrial facilities are methane explosions. The lower explosive limit (LEL) of methane is generally considered to be 5%, while the upper explosive limit (UEL) is 16%. Therefore, the creation of chemical sensors and signaling devices ensuring explosion and fire safety of domestic and industrial facilities is of great importance [1,2]. In all developed countries, systematic research is being conducted to develop methods and sensors for monitoring toxic, fire-, and explosion-hazardous gases [3,4]. Particular attention is paid to the creation of rapid and sensitive sensors that provide reliable control of the explosiveness of gas mixtures in closed ecological systems.

The analysis of the development of gas sensors in industrially developed countries has shown that the most reliable means of preventing explosive hazards is the use of thermocatalytic and semiconductor sensors. With the development of transportation, energy, and the oil and gas industry on a global scale, the requirements for ensuring explosion safety in industrial and domestic facilities are becoming increasingly stringent.

This circumstance determines the relevance of theoretical and practical research aimed at developing rapid, sensitive, and selective sensors that provide reliable control of the explosiveness of gas mixtures in closed ecological systems.

The aim of this work is the development of sensitive, selective chemical sensors for methane and the creation, on their basis, of highly efficient signaling devices and gas analyzers for natural gas.

In the course of the experiments, the regularities of the oxidation of combustible substances were studied, and a catalyst composition for a selective thermocatalytic methane sensor was developed. Optimal conditions were selected that ensured the stability, selectivity, and high sensitivity of thermocatalytic determination of explosive concentrations of natural gas in the atmospheric air of closed ecological systems. Sensitive and rapid methods and reliable signaling devices were created for detecting methane leaks and accumulations in the atmospheric air of industrial and domestic facilities.

The regularities of the sol-gel synthesis of gas-sensitive nanocomposite materials were studied, and sensitive semiconductor methane sensors were created on their basis. Using both semiconductor and thermocatalytic sensors, a dual-channel methane gas analyzer was developed for continuous determination of natural gas in air and technological gases. The metrological and analytical characteristics of the methane analyzer were established. Using the developed catalysts (0.75In2O3–0.25Ag2O and 0.25Fe3O4–0.75Ni2O3), we fabricated a thermocatalytic sensor (TCS) that provides selective determination of CH4 in the presence of CO and H2.

The output signal of the measuring sensitive element (MSE) of the sensor (catalyst: 0.75In2O3–0.25Ag2O) is proportional to the total concentration of combustible gases (H2, CO, CH4). The output signal of the comparative sensitive element (CSE) is proportional to the concentration of the CO and H2 mixture without the selectively determined component (CH4). The difference between the signals of the first and second elements is proportional to the concentration of CH4.

The tests of the developed sensors included special experiments related to selecting the optimal supply voltage, establishing dynamic, calibration, and other characteristics of the sensor, as well as identifying the degree of its selectivity and stability.

The highest sensor signal for CH4 was observed at a supply voltage of 2.6 V. A comparison of sensor signals for CH4 and for natural gas showed that the supply voltage providing the highest signal of the TCS under identical conditions for natural gas (2.8–3.0 V) is higher than that for methane (2.6 V).

For the developed sensor, the response initiation time (t0.1) is 3–4 s, the time constant (t0.63) is no more than 9 s, the stabilization time (t0.9) is 13 s, and the total analytical signal output time (tp) of the sensor is within 17–18 s, which once again confirms the possibility of using the developed sensors for rapid monitoring of natural gas content. The dependence of the sensor signal on methane concentration in the studied range (0.1–5.0% vol.) has a linear character (Table 1).

The experimental results demonstrate the identical nature of the dependence of the developed TCS signal on the concentration of CH4 and natural gas. This confirms the possibility of using the developed TCS to monitor explosion-hazardous concentrations (in the range of 0–5.0% vol.) of natural gas in the atmospheric air of domestic and industrial premises, as well as in the cabins of vehicles.

As follows from the presented data, the proposed TCS significantly surpasses the commercially available sensor in terms of selectivity. During the experiments, the comparative characteristics of the developed selective thermocatalytic sensors over the period 1990–2019 were also studied in the Gas Analysis Laboratory of Samarkand State University [1]. The experimental results are given in Table 6.

As the data show, the developed TCS-1, fabricated using sol-gel technology based on the catalyst composition of 75% In_2O_3 + 25% AgO and 25% Fe_3O_4 + 75% Ni_2O_3, is more sensitive than the previously developed analogs (TCS-2 and TCS-3). Thus, the catalyst composition (0.75 In_2O_3 – 0.25 Ag_2O and 0.25 Fe_3O_4 – 0.75 Ni_2O_3) was selected for a selective and highly sensitive methane sensor.

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