Вісник ЛДУБЖД
Bulletin of Lviv State University of Life Safety
https://journal.ldubgd.edu.ua/index.php/Visnuk
ISSN 2078-4643 (print), ISSN 2708-1389 (online)

UDC 665.521.004.17
DOI: 10.32447/20784643.32.2025.22

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ASSESSMENT OF THE COMPOSITION OF A MULTI-COMPONENT EXPLOSIVE MIXTURE USING THERMOCATALYTIC SENSORS

Introduction. When using control devices based on the thermocatalytic control method in explosion hazard monitoring systems for man-made objects, which make it possible to obtain an integrated assessment of explosiveness for all possible combustible components, the cost of explosion protection systems is significantly reduced and the reliability of detecting explosive concentrations of combustible components is increased. However, to simplify the process of finding and eliminating the source of danger, it is advisable to know which combustible component triggered the emergency alarm.

Purpose. The aim of the study is to find solutions that allow to clarify the composition and causes of the formation of an explosive mixture when thermocatalytic sensors with a non-selective platinum-palladium catalyst are used in monitoring systems for technogenic objects.

Methods. Theoretical analysis of the oxidation processes of combustible components of fuel-air mixtures on sensitive elements of thermocatalytic sensors with a non-selective platinum-palladium catalyst, based on classical principles of electrical engineering, thermodynamics and catalysis. experimental studies of serial thermocatalytic sensors when used for integral control of the explosion hazard of a multicomponent fuel-air mixture.

Results. The studies made it possible to determine the nature of the dependence of the relative value of the output voltage of thermocatalytic sensors with a non-selective platinum-palladium catalyst with an increase in the preheating temperature of the sensitive elements, provided that these sensors are used for integrated control of the explosive hazard of the environment. Taking into account the revealed nature of the dependence of the relative value of the output voltage of the sensors with increasing current through the sensitive elements for different types of explosive mixtures, a method for assessing the composition of the mixture in the integrated control of explosiveness is proposed, which allows determining which combustible component leakage led to a dangerous situation. To implement this method, an algorithm for the operation of the gas analyser in the mode of assessing the composition of an explosive mixture was developed.

Conclusion. The proposed method for assessing the composition of the mixture in the integrated control of explosiveness by thermocatalytic sensors with a non-selective platinum-palladium catalyst allows timely detection of sources of leakage of explosive gases or vapours of volatile combustibles, which allows timely warning of personnel about the threat of explosion, taking measures to prevent leaks and eliminating possible sources of ignition of explosive mixtures. In view of this, further work on its practical implementation in explosion hazard monitoring systems is advisable.

Keywords: hazard, combustible gases, fuel, monitoring systems, sensors, integrated explosion hazard assessment.

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ОЦІНКА СКЛАДУ БАГАТОКОМПОНЕНТНОЇ ВИБУХОНЕБЕЗПЕЧНОЇ СУМІШІ ПРИ ВИКОРИСТАННІ ТЕРМОКАТАЛІТИЧНИХ ДАТЧИКІВ

Проблема. При використанні в системах моніторингу вибухонебезпечності техногенних об’єктів засобів контролю заснованих на термокаталітичному методі контролю, які дають можливість отримання інтегральної оцінки вибуховості за усіма можливими горючими компонентами, суттєво зменшується вартість систем вибухозахисту та збільшується надійність виявлення вибухонебезпечних концентрацій горючих компонентів.

Проте для спрощення процесу пошуку і усунення джерела небезпеки доцільно знати, який горючий компонент обумовив спрацювання протиаварійної сигналізації.

Метою роботи є пошук рішень, що дозволяють уточнити склад та причини утворення вибухонебезпечної суміші при використанні в системах моніторингу техногенних об'єктів термокаталітичних датчиків з неселективним платино-паладієвим каталізатором.

Методи дослідження. Теоретичний аналіз процесів окиснення горючих компонентів паливо-повітряної суміші на чутливих елементах термокаталітичних датчиків з неселективним платино-паладієвим каталізатором, що базуються на класичних положеннях електротехніки, термодинаміки та каталізу, експериментальні дослідження серійних термокаталітичних датчиків при їх використанні для інтегрального контролю вибухонебезпечності багатокомпонентної паливоповітряної суміші.

Основні результати дослідження. Виконані дослідження дозволили встановити характер залежності відносного значення вихідної напруги термокаталітичних датчиків з неселективним платино-паладієвим каталізатором при збільшенні температури попереднього розігріву чутливих елементів за умови використання цих датчиків для інтегрального контролю вибухонебезпечності середовища. З врахуванням виявленого характеру залежності відносного значення вихідної напруги датчиків при збільшенні струму через чутливі елементи для різних видів вибухонебезпечних сумішей, запропоновано метод оцінки складу суміші при інтегральному контролі вибухонебезпечності, що дозволяє визначити витоки якого горючого компоненту призвели до небезпечної ситуації. Для реалізації цього методу розроблено алгоритм роботи газоаналізатора в режимі здійснення процедури оцінки складу вибухонебезпечної суміші.

Висновки та конкретні пропозиції автора. Запропонований метод оцінки складу суміші при інтегральному контролі вибухонебезпечності термокаталітичними датчиками з неселективним платино-паладієвим каталізатором дозволяє своєчасно виявити джерела витоків вибухонебезпечних газів чи парів летких горючих речовин, що дозволяє своєчасно попередити персонал про загрозу вибуху, прийняти заходи з попередження витоків та усунути можливі джерела запалення вибухонебезпечних сумішей. Зважаючи на це доцільне подальше проведення робіт з практичного його впровадження в системах моніторингу вибухонебезпечності.

Ключові слова: небезпека, горючі гази, паливо, системи моніторингу, датчики, інтегральна оцінка вибухонебезпечності.

Introduction. Explosions of gas-air and fuel-air mixtures pose a danger to humans and the environment, as well as cause the destruction of buildings, damage to process equipment and machinery, which leads to significant material losses [1]. To reduce the risk caused by explosions, man-made facilities where explosive substances are stored or used are equipped with technical means for early detection of hazards. Such equipment allows timely detection of leaks of explosive gases or significant concentrations of volatile combustible vapors in the air and warns personnel of the threat of an explosion. This allows timely measures to prevent leaks and eliminate possible sources of ignition of such mixtures [2]. Among these technical means, gas analyzers are usually used, which automatically continuously monitor the content of explosive gases or vapors in places of their possible release into the air [3-4].

Nowadays, with the development of information technology, safety monitoring systems for man-made facilities are becoming increasingly important, in which, in order to assess the hazard, predict risks and take measures to minimize or eliminate them, technical means for early detection of hazards continuously monitor the content of explosive gases or vapors in places of their possible release into the air [5-6].

The issue of explosive hazards at man-made facilities is especially relevant nowadays, when, as a result of military operations, shelling, and the fall of wreckage from downed air targets, the threat of damage to the integrity of tanks and transportation routes with explosive gases and volatile flammable liquids is significantly increasing. Given the increased risk of explosions and fires at man-made facilities, research aimed at preventing them is highly relevant.

Literature review. Much attention is paid to the development of gas analyzers designed to monitor the explosiveness of fuel-air mixtures. Among the available control devices that are used in safety monitoring systems for man-made facilities, the most common gas analyzers are based on thermocatalytic and semiconductor methods [7-8].

In coal mine explosion protection systems, sensors based on the thermocatalytic method of control are mainly used [4]. This is due to their simple design, affordability, and ability to operate stably for a long time in difficult underground conditions. Modern serial thermocatalytic sensors have two identical-sized thermocouples made in the form of miniature balls (pelletizers) of γ-aluminum oxide, inside which there are spirals of platinum micro-wires. One of the thermocouples is catalytically active (working), and the other is passive (comparative). A non-selective platinum-palladium catalyst is used to activate the working element.

Depending on the preheating temperature of the thermocouples, the oxidation reaction of combustible gases and vapors on a platinum-palladium catalyst can occur in the kinetic and diffusion regions [4]. In gas analyzers used to monitor the explosive hazard of the mine atmosphere, in order to increase sensitivity, increase the speed of the explosion protection system and minimize errors due to changes in operating

conditions, thermocatalytic sensors usually operate at a temperature corresponding to the diffusion region of the reaction. For example, in coal mine explosion protection systems, the preheating temperature of the sensor thermocouples is close to 400 ^{0}C. This ensures that the methane oxidation reaction takes place in the diffusion region. At this temperature of the thermocouples, combustible components characterized by a lower self-ignition temperature are able to oxidize both on the working and on the reference thermocouple. Since the main explosive component of the mine atmosphere is methane gas, and the presence of other hydrocarbons in small quantities has little effect on the explosiveness of the mixture, the choice of this temperature regime for the sensors is appropriate in this case.

Early warning systems for other man-made facilities mainly use thermocatalytic and semiconductor (metal oxide) sensors. The latter are selective, relatively cheap and simple in design. Their advantage is high sensitivity. However, these sensors are quite sensitive to overloads [7] and are not stable enough in time. They are widely used in ventilation systems, air purification systems, and portable gas analyzers [8]. Thermocatalytic combustible gas sensors, which are used in explosion monitoring systems [9], are usually customized for a specific combustible component, such as methane, butane, propane, hexane, etc. When using such sensors, provided that various combustible gases and vapors can enter the air, it is necessary to install measuring devices configured for specific types of explosive substances. This complicates the explosion protection system, increases its cost and maintenance costs. In addition, the error in measuring the concentration of a particular component, or the triggering of an alarm for that component, is significantly affected by the presence of other combustible vapors or gases.

At the same time, the lower limit of ignition of an explosive mixture, with the simultaneous presence of various combustible components in the air, is determined not by the content of a single component, but by their total content and the explosive properties of each of these components. Paper [10] shows that when choosing a certain temperature regime for the working thermocouple of a thermocatalytic sensor with a non-selective platinum-palladium catalyst, it is possible to carry out an integrated unambiguous assessment of explosiveness. The conditions under which thermocatalytic sensors allow for an integrated assessment of the environment in the presence of a multicomponent fuel-air mixture have been established, and the power supply mode of thermocatalytic sensors with stabilization of the temperature of the working thermocouple has been substantiated, which ensures minimal alarm error in the systems for monitoring the explosiveness of man-made objects [11].

Explosion-hazardous concentrations of petroleum product vapors are formed during various processes related to the processing, transportation, storage, and use of petroleum products. At storage facilities and fuel-filling stations, the greatest danger of explosion-hazardous mixtures is associated with the evaporation of gasoline from tanks [12], [13]. The introduction of promising technical measures aimed at reducing gasoline vapor emissions into the environment during transportation [14], [15] and minimizing the intensity of gasoline evaporation during storage [16]--[18] has helped mitigate the risks. However, these measures do not eliminate the threat of explosions. This risk remains particularly significant in extreme situations, such as those related to military operations and psychophysiological factors [19]-[21].

The use of tools that enable an integrated assessment of explosiveness for all possible combustible components significantly reduces the cost of explosion protection systems and increases the reliability of detecting explosive concentrations. However, to simplify the process of finding and eliminating the source of danger, it is desirable to know which combustible component caused the alarm to be triggered. In view of this, it is relevant to conduct research aimed at finding solutions that would allow to assess the composition of an explosive mixture in the integrated control of explosiveness.

The purpose of the article. The aim of the work is to find solutions that allow to clarify the composition and causes of the formation of an explosive mixture when using thermocatalytic sensors with a non-selective platinum-palladium catalyst in monitoring systems for technogenic objects.

Research methods. Theoretical analysis of the oxidation processes of combustible components of the fuel-air mixture on the sensing elements of thermocatalytic sensors with a non-selective platinum-palladium catalyst, experimental studies of serial thermocatalytic sensors when used for integrated control of the explosiveness of a multicomponent fuel-air mixture.

Presentation of the main material. According to the integrated assessment of the explosiveness of a multicomponent fuel-air mixture, the preheating temperature of the working thermocouple of a thermocatalytic sensor with a non-selective platinum-palladium catalyst for the studied sensors was 350 ^{0}C [10]. Under such conditions, the catalytic oxidation reaction of such a mixture component as methane proceeds in the kinetic region, and such components as ethane, propane, and butane - in the transition region. The oxidation reaction of fuel components characterized by a lower self-ignition temperature

(hexane, heptane, etc.) in this mode of operation of the sensor occurs in the diffusion region [4].

According to the pre-explosive concentration of the combustible component c, it can be assumed that each combustible component diffuses to the surface of the working element independently, at a rate determined by its diffusion coefficient. The thermal effect of oxidation on the working element of the fuel component P sensor can be represented as [4]

$$P = QC_{rk}\gamma_e, \quad (1)$$

where Q is the lower heat of oxidation of the fuel, J/kg; C_{rk} is the fuel concentration in the reaction chamber, kg/m^3;γ_e is the effective diffusion conductivity of the element, m^3/s.

The effective diffusion conductivity of the element depends on its size, the effective oxidation of fuel on its surface, and the rate of fuel flow to the element surface [4].

$$\gamma_e = 10^{-2} k_e \beta_e F_e, \quad (2)$$

where k_e is the fuel oxidation efficiency coefficient; β_e – is the mass transfer coefficient, m/s; F_e – Fe is the elemental surface area, m^2.

In turn, the concentration of the combustible component in the reaction chamber is

$$C_{rk} = \frac{\gamma_f}{\gamma_f + \gamma_e} C_a, \quad (4)$$

where C_a is the concentration of the combustible component in the air, kg/m^3; γ_f – is the diffusion conductivity of the filter, m^3/s.

When the oxidation reaction proceeds in the kinetic region, an increase in the heating temperature of the working thermocouple leads to a significant increase in the fuel oxidation efficiency coefficient. In turn, the mass transfer coefficient, which depends on the diffusion properties of the fuel component, is generally proportional to the molecular diffusion coefficient of the fuel in air [4], which also increases slightly with increasing temperature (proportional to $\sqrt{T}$)). This causes the fact that during the oxidation reaction in the kinetic region, an increase in the temperature of preheating of the thermocouples leads to a significant increase in the output voltage of the thermocatalytic sensor. Thus, according to the results of experimental studies of the operation of sensors in a methane-air mixture, given in this work, when the temperature of the sensor preheating changes from 341 ^{0}C to 427 ^{0}C, the output signal of the sensor increases almost 2.5 times.

If the oxidation reaction of the combustible component occurs in the diffusion region, the oxidation efficiency coefficient is ke=1, and a further increase in the preheating temperature of the working thermocouple, on the contrary, leads to a decrease in the sensor output signal [4].

This nature of the change in the sensor output signal allows not only to carry out an integrated assessment of the explosiveness of a multicomponent fuel-air mixture [9], but also to assess the composition and causes of the formation of an explosive mixture when using thermocatalytic sensors with a non-selective platinum-palladium catalyst in monitoring systems for man-made objects by controlling the preheating temperature of the working element of the thermocatalytic sensor.

To evaluate the possibility of assessing the composition of an explosive mixture when using thermocatalytic sensors with a non-selective platinum-palladium catalyst in monitoring systems for technogenic objects by changing the temperature regime of the thermocatalytic sensor, observations were made of the change in the output signal of serial thermocatalytic sensors when the current through the thermocouples changes from 175 to 200 mA, For the study, 10 sensors with a non-selective platinum-palladium catalyst used in mines were used. The sensors were included in the measuring bridge and installed in a small-sized chamber, which was first filled with clean air (to set the bridge zero), and then with certified gas mixtures with concentrations of methane, butane and hexane in the air close to 0.1 LEL (lower ignition limit of the mixture). In the steady-state mode, at a current of 175 mA through the thermocouples, the value of the output signal of the measuring bridge U0 was determined, and then, at time t = 0, the current instantly increased to 200 mA. Fig. 1 shows the characteristic dependences of the change in the relative value of the output voltage of the measuring bridge, obtained as a result of the experiment, for different mixtures.

At the initial time t = 0, regardless of the type of mixture, an instantaneous increase in the bridge output signal by 14-15% is observed, which is naturally associated with an increase in the bridge supply voltage with an increase in the current through the thermocouples by 15%.

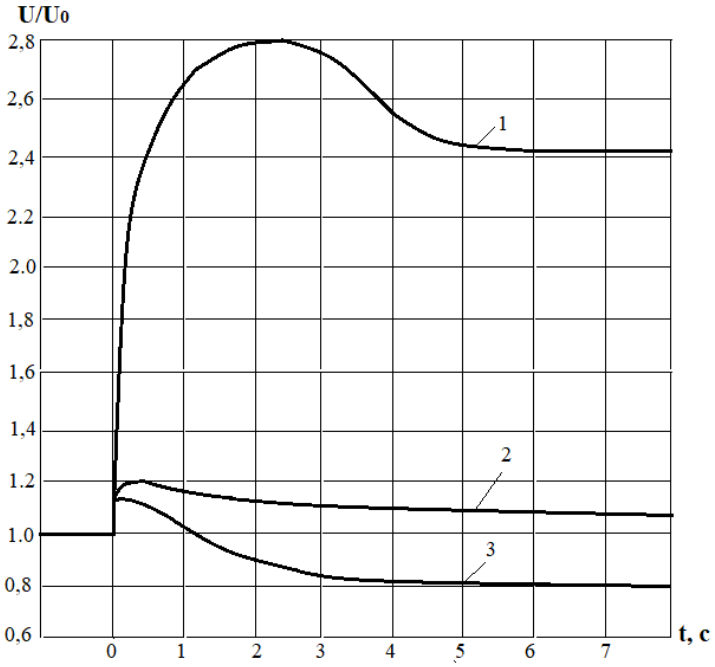

Fig. 1. Dependence of the relative value of the output voltage of the sensors when the current through the thermocouples is increased from 175 mA to 200 mA when measuring pre-explosive concentrations of combustibles: 1 - methane; 2 - butane; 3 - hexane.

Subsequently, the nature of the dependence of the relative value of the output voltage of the sensors with an increase in the current through the thermocouples for different mixtures is significantly different. When the sensors operate in a methane-air mixture, with an increase in the current through the thermocouples in the first 2-3 seconds, an almost threefold increase in the output signal of the bridge is observed, followed by a slight decrease and its stabilization at a level 2.4 times higher than the initial one. This is due to a significant increase in the effective diffusion conductivity of the working thermocouple during the transition of the methane oxidation reaction from the kinetic to the diffusion region. The initial surge in the output signal is explained by the presence of a higher concentration of methane in the reaction chamber, which was accumulated in the reaction chamber before the acceleration of the oxidation reaction, compared to the new steady-state concentration value, due to the presence of the filter element and its effect on the methane content in the reaction chamber [4].

When the sensors operate in a mixture of butane and air, with an increase in the current through the thermocouples in the first seconds, an increase in the output signal of the bridge by 20% is observed, followed by a decrease and stabilization at a level 1.05-1.1 times higher than the initial one. This indicates a slight acceleration of the butane oxidation reaction from the transition region to the diffusion region.

When the sensors operate in a mixture of hexane and air, after a slight initial surge in the bridge output signal caused by an increase in the voltage of the measuring bridge, the output signal quickly decreases and stabilizes at 0.8 of its initial value after almost 3 s. This is due to the fact that the rate of hexane oxidation reaction in this case does not increase, and an increase in the preheating temperature leads to an increase in heat loss from the working element, as well as partial oxidation of hexane on the comparative thermocouple of the sensor at a high temperature of its heating.

Under the condition of an integrated explosion hazard assessment, this behavior of the sensor output signal with an increase in current through the thermocouples allows us to propose a method for assessing the composition of the mixture, which allows us to determine which combustible component leaks led to a dangerous situation.

The essence of the proposed method for assessing the composition of the mixture is as follows:

- the algorithm of operation of the gas analyzer designed for an integrated assessment of explosiveness in addition to the existing setup and operating modes [10] is supplemented by the mode of assessing the composition of an explosive mixture;

- in the setup mode, by alternately supplying the sensor with clean air and a calibration mixture of hexane in the air, the preheating temperature of the working thermocouple of the sensor is automatically set to 350 ^{0}C [11], the sensitivity of the analyzer to hexane is determined, and the alarm thresholds of the gas analyzer are determined;

- in the operating mode, a stable preheating temperature of the sensor's working thermal element of 350 ^{0}C is maintained and continuous monitoring of the explosive hazard of the environment is carried out;

- when explosive gases or vapors appear in the controlled environment and the sensor output signal reaches a value corresponding to 0.1 LEL (lower ignition limit), a command is given to turn on the alarm;

- after the alarm is activated, the gas analyzer is switched to the mode of evaluating the composition of the explosive mixture.

In this mode:

- a ban is imposed on switching off the alarm for the period of evaluation of the mixture composition;

- the current through the thermocouples is increased by 15%, which ensures an increase in the temperature of the working thermocouple of the sensor to almost 400 ^{0}C;

- a stable preheating temperature of the sensor's working thermal element of 400 ^{0}C is maintained;

- provided that a stable temperature regime is maintained with a time delay of about 4 s, the output signal of the sensor is determined and this value is compared with its previous value (before the temperature regime change);

- in case of its decrease, the gas analyzer displays information that the explosive mixture is formed due to the evaporation of gasoline (kerosene or other combustible components with a low auto-ignition temperature);

- if it increases by more than 1.5 times, information is displayed that the formation of an explosive mixture is caused by the leakage of light hydrocarbons (methane, ethane);

- Otherwise, information is displayed that the cause of the explosive mixture is the accumulation of butane or propane in the air;

- reduces the current through the thermocouples by 15%, which ensures that the temperature of the working thermocouple of the detector drops to the previous value;

- provided that a stable temperature regime is maintained with a time delay of about 5 s, which ensures the completion of transient processes in the sensor caused by a change in temperature, the gas analyzer is switched to the operating mode and continuous monitoring of the explosive hazard of the environment is carried out.

The algorithm of the gas analyzer operation in the mode of evaluating the composition of an explosive mixture is shown in Fig. 2

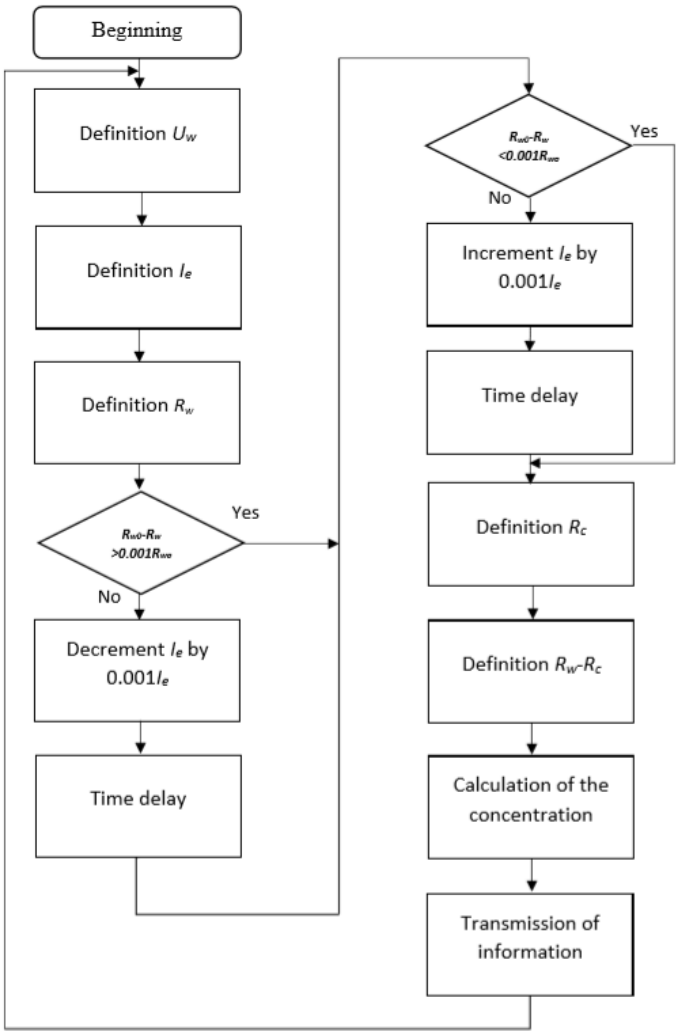

Fig. 2. The algorithm of the gas analyzer operation in the mode of implementing the procedure for assessing the composition of an explosive mixture.

Conclusions. The performed studies made it possible to establish the nature of the dependence of the relative value of the output voltage of thermocatalytic sensors with a non-selective platinum-palladium catalyst with an increase in the preheating temperature of the thermocouples, provided that they are used for integrated control of the explosive hazard of the environment. In the presence of combustible components with a relatively low auto-ignition temperature (hexane, heptane, gasoline vapors, etc.) in the mixture, when the preheating temperature of the working thermocouple is increased from 350 ^{0}C to 400 ^{0}C, a decrease in the sensor output signal of up to 20% is observed, in the presence of components with a relatively high auto-ignition temperature (methane, ethane), the sensor output signal increases significantly, and in the presence of propane and butane it practically does not change.

Taking into account the revealed dependence of the relative value of the output voltage of the sensors with an increase in the current through the thermocouples for different mixtures, a method for assessing the composition of the mixture is proposed, which allows determining the leakage of which combustible component led to a dangerous situation, on the basis of which an algorithm for the operation of the gas analyzer in the mode of implementing the procedure for assessing the composition of an explosive mixture has been developed.

Further research in this direction consists in conducting experimental studies of the operation of gas analyzers designed for integrated explosion hazard control in the mode of mixture composition assessment.

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Науково-методична стаття.
Надійшла до редакції 29.08. 2025.
Прийнято до публікації 26.11.2025.