

Introduction

Nowadays, there are three widely used gas sensing technologies for detecting combustible gases in the environment below the explosive threshold: optical, catalytic and semiconductor. Typically, each sensor is calibrated for detecting a separate gas. However in industry the gas mixtures of combustible hydrocarbons and vapors as well as hydrogen are common. In this case multisensor gas detectors, optical spectrometers and other equipment for multigas analysis in the environment should be used. This ‘multisensor’ approach complicates the task on gas sensing by exploiting complex gas equipment and measurement.

The state-of-the-art approaches for estimating the level of explosiveness of combustible gas mixtures start with the identification of all single gases and their concentrations composing the mixture. This process is followed by the calculation of LEL of gas mixture that is then compared with the theoretical threshold value of particular mixture. Based on the comparison, the level of explosion hazard is already estimated. At the same time the identification of separate gases in the mixture for the inference on potential explosion is an intermediate task which is essential in traditional approaches for calculating LEL of gas mixture. That is why the assessment of potential explosiveness of gas mixture without the identification of separate gases in the mixture is a promising solution in this context.

In this paper, we have offered a methodology for assessing the degree of explosiveness of hydrocarbon gas mixtures with hydrogen without recognizing the type of hydrocarbons, their quantity and concentration by controlling the burning process in the typical catalytic sensor. According to its fundamental feature, a catalytic sensor practically reacts on the total gaseous and vapor composition of the environment. This feature opens the way for an assessment of the explosiveness of gas mixtures, regardless of their composition.

Experimental

The proposed method is based on the well-known fact that the combustion heat of various hydrocarbons at a concentration of 100% LEL in air differs less than 10% if the same volume of hydrocarbons is burned. Therefore, it is possible to determine the explosiveness of hydrocarbon gas mixture regardless of its exact composition by measuring the amount of heat released during the mixture burning in a constant volume. In the same time, the combustible heat of hydrogen is strongly different from hydrocarbons. However, the hydrogen starts to burn at a much lower temperature than hydrocarbons. Hence it can be concluded that using a two-stage impulse heating voltage of a catalytic sensor, where the voltage at a low stage is sufficient for burning hydrogen, but not sufficient for burning other gases, it is possible to separate hydrogen from the hydrocarbon gases.

In this paper we investigated the combustion of hydrocarbon mixtures which consist of propane, butane and hydrogen (in the range of 20%–60% lower explosive limit) in the regime of diffusion restriction of gas leakage. This is important in order to ensure the main condition of correct measurements- the burning rate of the mixture should be much greater than the rate of their leakage inside the sensor chamber.

We use the catalytic sensor manufactured by NTC IGD, Russia. The sensor height is 9.5 mm and diameter 9 mm, power consumption is 75 mW at 1.4 V in the continuous measurement mode. Its low power consumption is achieved by applying a heater implemented as 10 μm platinum micro wire in glass insulation. The sensor is a pellistor with spiral as heating element that is coated by the platinum – palladium catalyst.

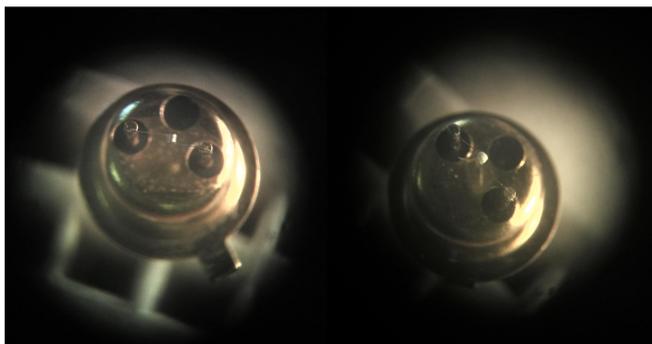


Fig. 1. The catalytic sensor manufactured by NTC IGD, Russia

The measurements of sensor response were carried out according to the voltage divider circuit (Fig.2). For taking a measurement the sensor R_s is embedded in a voltage divider circuit with resistor R (10 Ω in our case). The sensing circuit is controlled by the ADuC831 MCU which generates a specific heating profile for the sensor. The two-stages heating profile with different duration and voltage amplitude was developed to heat the sensor and provide the measurements (Fig.3). The supply voltage is the voltage applied to the divider. Since there is no one arm in the bridge circuit, the measurement of the voltage drop across the sensor does not calculated from the zero value. This reduces the accuracy of the measurements. Therefore, to emulate the bridge circuit we introduced a compensating voltage (equal to the voltage on the sensor in the air) (Fig. 3) which plays the role of the equivalent of the second arm of the bridge circuit. The measured value is the difference between the compensation voltage and the voltage on the sensor.

This allowed to divide the catalytic sensor response onto hydrogen (first stage) and hydrocarbons mixture (second stage). The heating were carried out in the temperature range 0–450 °C. As well as it allows to exclude the influence of uncontrolled environmental factors (temperature, humidity and pressure) on the result of sensor response measurements and optimization of power consumption.

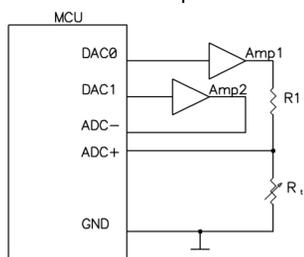


Fig.2. Sensing circuit

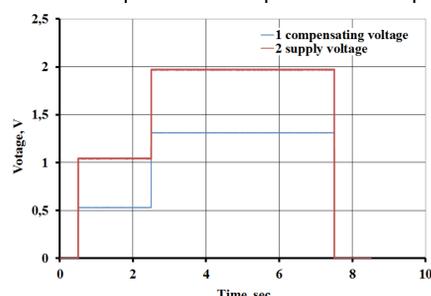


Fig. 3. Dependence of the applied voltage on time of two –stage sensor heating profile.

Results and Discussion

Heat of combustion for an identical volume of limiting hydrocarbons and hydrogen is shown in Fig. 4.

The measurement is fulfilled by the next way. We place the sensor in the atmosphere with clear air and measure the amount of heat emitted during the measurement procedure. Next, we carry out the same experiment, but in the presence of a combustible gases and measure the amount of heat emitted in the same time interval.

The duration of stages of 2 and 5 seconds were chosen on the basis of the fact that it is necessary to completely combust gases in the working chamber of the sensor.

Fig. 5 shows the dependence of sensor sensitivity on the applied voltage for different combustible gases (S-shaped characteristics). It is seen that due to its physical properties the hydrogen burns in the entire range of the applied voltage started from the small values. Combustion of other hydrocarbons begins with a voltage of 600 mV. So, we have chosen the voltage of first stage and second stage 0.6 V и 1.4 V, respectively.

Results of experiment where we estimate the explosiveness of gas mixture consisting of three gases in the air are shown in Table I. As the initial gases, we used two calibration hydrocarbon gases (propane, butane) and hydrogen with known concentrations. The concentration of individual components in mixture was controlled by the gas flow rate. These results are compared with the theoretical values that are calculated using a well-known equation (1) for a combustible gas mixture from an industrial standard.

$$\varphi_d = \sum_{k=1}^n \varphi_k / \sum_{k=1}^n (\varphi_k / \varphi_{p_k}) \quad (1)$$

where φ_d is LEL for a mixture of combustible gases, φ_k is the concentration of k-th component of the mixture in % vol., φ_{p_k} is LEL of the k-th component in the mixture with air in% vol., n is the number of combustible components of the mixture.

In total we evaluated seven mixtures and the experimental results are in line with the theoretical values. The measurement error is approximately 6-8 %.

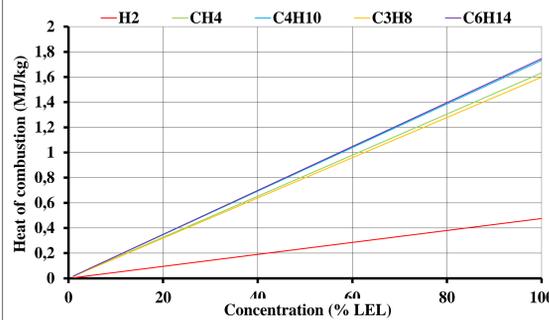


Fig.4. Heat of combustion for an identical volume of limiting hydrocarbons and hydrogen.

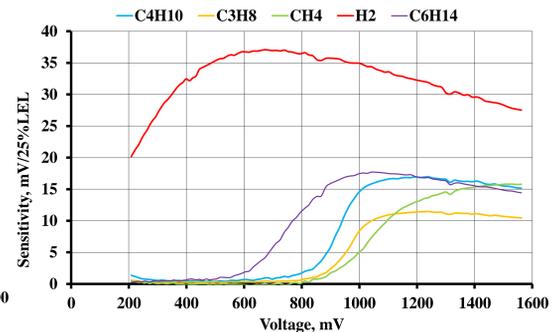


Fig.5. Dependence of the sensor sensitivity on the applied voltage

Table I. Estimation of Mixture Explosiveness Consisting of Three Gases

| | Gas type and concentration, %vol. | | | Concentration in mixture, %vol. | | | Theoretical LEL, %vol. | Mixture concentration, %vol. | Theoretical LEL, %LEL | Experimental LEL, %LEL |
|----|-----------------------------------|------------------------------------|--------------------------------------|---------------------------------|-------|-------|------------------------|------------------------------|-----------------------|------------------------|
| | Gas 1 | Gas 2 | Gas 3 | Gas 1 | Gas 2 | Gas 3 | | | | |
| 1. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,34 | 0,56 | 0,07 | 2,090 | 0,958 | 46 | 47 |
| 2. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,59 | 0,14 | 0,16 | 2,588 | 0,897 | 35 | 37 |
| 3. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,37 | 0,45 | 0,12 | 2,129 | 0,931 | 44 | 47 |
| 4. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,69 | 0,05 | 0,15 | 2,863 | 0,894 | 31 | 33 |
| 5. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,07 | 0,07 | 0,57 | 1,526 | 0,712 | 47 | 50 |
| 6. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,05 | 0,26 | 0,45 | 1,561 | 0,771 | 49 | 50 |
| 7. | 0.96 H ₂ | 1.01 C ₃ H ₈ | 0.665 C ₄ H ₁₀ | 0,66 | 0,16 | 0,11 | 2,767 | 0,921 | 33 | 35 |

Conclusions

We proposed a method for estimating the explosiveness of a mixture consisting of hydrocarbons of unknown composition using the typical catalytic sensor. The measurements were carried out for three-component mixtures of explosive gases (propane, butane and hydrogen).

The obtained results demonstrate a good agreement between the experimental values of LEL for hydrocarbon mixtures and the theoretical values. We also performed the estimation of error and its minimization. It is important to emphasize that the offered methodology can be used for sensing as single gases as well as combustible gas mixtures of unknown composition and allow to isolate the hydrogen from hydrocarbons mixture.

Alternative ways for flammable mixtures analysis are typically associated with the application of multi wavelength IR sensor. However IR detectors do not allow to detect the hydrogen.

The obtained results have got great potential for the industries dealing with the combustible gases detection in outdoor or harsh environments.

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