МИНИСТЕРСТВА ОБРАЗОВАНИЯ И НАУКИ РЕСПУБЛИКИ УЗБЕКИСТАН
ТАШКЕНТСКИЙ ГОСУДАРСТВЕННЫЙ ТЕХНИЧЕСКИЙ УНИВЕРСИТЕТ ИМЕНИ ИСЛАМА КАРИМОВА

ЎЗБЕКИСТОН РЕСПУБЛИКАСИ ТАЪЛИМ ВА ИЛМ-ФАН ВАЗИРЛИГИ
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THERMOCHEMILUMINESCENCE-BASED METHOD FOR ATMOSPHERIC HYDROCARBON MONITORING ENHANCED BY MACHINE LEARNING

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Abstract: *A hybrid thermochemiluminescence (TCL)–based method enhanced by machine learning is proposed for the detection of hydrocarbons in atmospheric air. The method is based on the reaction of atomic oxygen, generated via thermal decomposition of ozone at 290–310 °C, with hydrocarbons, producing chemiluminescent emission. A mathematical model describing signal formation as a function of concentration, temperature, and system parameters is developed. To address nonlinear response, cross-sensitivity, and measurement noise, a Random Forest–based machine learning framework is applied for signal processing. The TCL signal exhibits a near-linear response in the range of 0.5–50 ppm. The proposed approach significantly improves prediction accuracy compared to classical calibration, demonstrating reduced error and enhanced robustness. The developed TCL–ML system shows strong potential for environmental monitoring and industrial gas analysis.*

Introduction

Air pollution caused by hydrocarbon emissions remains one of the most significant environmental challenges associated with industrial development, urbanization, and transportation growth. Volatile and semi-volatile hydrocarbons released into the atmosphere participate in photochemical reactions leading to the formation of secondary pollutants, including ozone and fine particulate matter, which adversely affect human health and ecosystems.

Reliable monitoring of hydrocarbons in ambient air is therefore essential for environmental protection, industrial safety, and regulatory compliance. Conventional analytical techniques, such as gas chromatography, Fourier-transform infrared spectroscopy, and mass spectrometry, provide high accuracy and compound specificity. However, these methods are typically expensive, labor-intensive, and unsuitable for continuous on-site or real-time monitoring applications.

In recent decades, chemiluminescence-based sensing methods have attracted considerable attention as promising alternatives for gas analysis due to their high sensitivity, fast response time, and relatively simple instrumentation. Among these techniques, thermochemiluminescence (TCL), based on the reaction of atomic oxygen with hydrocarbons at elevated temperatures, offers particular advantages for real-time atmospheric monitoring.

Despite its potential, classical TCL systems suffer from limited selectivity in complex gas mixtures and sensitivity to environmental disturbances such as temperature fluctuations and interfering species. Recent advances in machine learning (ML) and intelligent data processing provide new opportunities to overcome these limitations by combining physical sensing principles with data-driven modeling.

The present study proposes a hybrid thermochemiluminescence-based approach enhanced by machine learning algorithms for accurate and robust monitoring of hydrocarbons in atmospheric air.

The novelty of this work lies in the integration of a physically grounded TCL measurement model with intelligent signal processing, enabling improved selectivity, adaptive calibration, and enhanced performance under real-world conditions.

1. Physical and Chemical Principles of Thermochemiluminescence

The thermochemiluminescence process relies on the formation of atomic oxygen through the thermal decomposition of ozone on a heated catalytic surface. Ozone is introduced into a reaction chamber containing a resistively heated metallic element, typically a nichrome filament, maintained within a controlled temperature range of 290–310 °C. Under these conditions, ozone undergoes dissociation according to:

$$O_3 \rightarrow O_2 + O$$

The generated atomic oxygen exhibits high chemical reactivity, enabling rapid oxidation reactions with hydrocarbon molecules present in the sampled atmospheric air.

Atomic oxygen reacts with hydrocarbons through a series of exothermic reactions that lead to the formation of electronically excited intermediate species:

$$O + RH \rightarrow RO^* + h\nu$$

The relaxation of excited intermediates (RO) to their ground state is accompanied by the emission of photons in the ultraviolet–visible spectral range. The intensity of this emitted radiation is directly related to the concentration of hydrocarbons and the kinetics of the oxidation process.

The spectral and temporal characteristics of the emitted chemiluminescence depend on the molecular structure of the hydrocarbon, reaction temperature, and oxygen availability, forming the physical basis for selective detection.

The intensity of electromagnetic radiation generated during the reaction of atomic oxygen with hydrocarbons present in atmospheric air depends both on the concentration of hydrocarbons and on their belonging to different homologous series. This makes it possible to obtain quantitative information on hydrocarbon concentrations.

A schematic diagram of the measuring transducer implementing this method is shown in Figure 1.

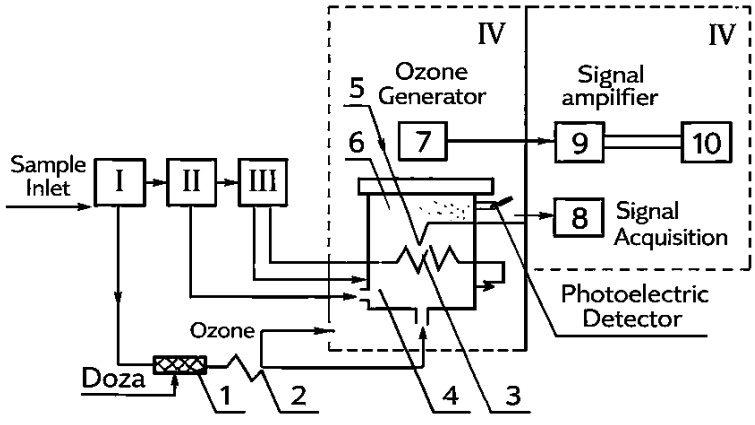


Fig 1. A schematic diagram of the measuring transducer

The system consists of the following units:

I — gas preparation unit,

II — ozone generator,

III — power supply unit,

IV — measuring unit,

V — measurement system.

The measuring unit contains four reaction chambers and seven photoelectric detectors. One wall of the reaction chamber is made of quartz glass. Inside the chamber, a nichrome wire spiral is installed, adjacent to which a chromel–copel thermoelectric transducer is placed.

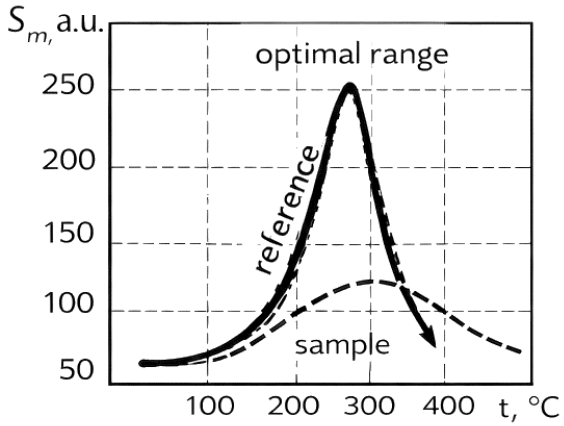
In accordance with the theoretical principles described above, ozone generated in unit II and the analyzed air sample supplied from unit I are simultaneously introduced into the reaction chamber. Upon contact with the heated spiral, ozone decomposes, forming atomic oxygen, which reacts with hydrocarbons in the air. The resulting chemiluminescent emission is detected by the photoelectric sensors and converted into an electrical signal proportional to hydrocarbon concentration.

The generated signal is subsequently amplified (amplifier 9) and recorded using a potentiometric measurement system (10). In parallel, the temperature of the reaction zone is monitored via potentiometer (8), ensuring precise control of the operating conditions critical for maintaining optimal reaction kinetics.

The analyzed air enters the system through the sample inlet device (1) and flows into a metal tube (2) with a diameter of **4 mm** and a length of **1000 mm**, through which it is supplied to the reaction chamber at a constant flow rate.

Experimental studies have shown that the signal of the photoelectric detector depends on the temperature of the spiral. This dependence is presented in the graph shown in **Figure 2**.

The results of the study indicate that the proposed method can also be used to determine the concentration of sulfur-containing compounds in atmospheric air.



Fiq.2. The signal of the photoelectric detector depends on the temperature of the spiral

The thermochemiluminescence (TCL) process is based on the generation of atomic oxygen through the thermal decomposition of ozone on a heated catalytic surface. The high reactivity of atomic oxygen enables rapid oxidation of hydrocarbons, leading to the formation of excited intermediates and subsequent photon emission, which forms the analytical signal.

The practical implementation of this physical–chemical mechanism is realized in the measuring transducer schematically shown in Figure 1., where the key processes of ozone decomposition, hydrocarbon oxidation, and chemiluminescence detection occur within the reaction chamber.

To further enhance the analytical performance of the system, particularly in terms of selectivity and robustness, the experimental setup was extended by integrating intelligent data processing, as illustrated in Figure 3.

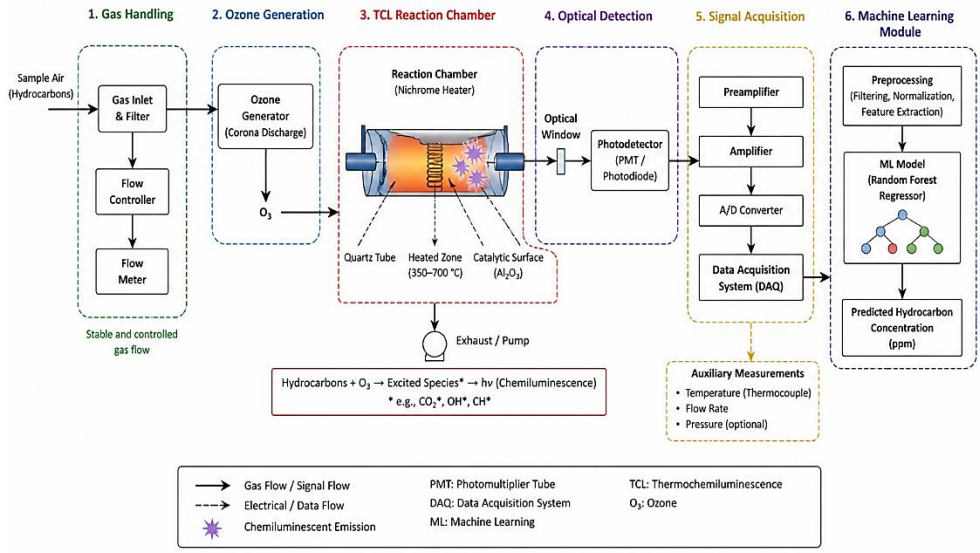


Fig. 3. Schematic diagram of the thermochemiluminescence-based hydrocarbon detection system with integrated machine learning module.

The experimental measurement system, schematically illustrated in Figure 3, is composed of the following functional units:

- gas preparation and conditioning module;
- ozone generation unit;
- thermally controlled reaction chamber;
- photoelectric detection system;
- signal amplification and acquisition module.

Atmospheric air samples and ozone are simultaneously introduced into the reaction chamber, where the thermochemiluminescence process takes place. The chamber walls are partially fabricated from quartz to ensure optical transparency for efficient photon collection.

A nichrome heating filament positioned within the reaction chamber serves as both the ozone decomposition catalyst and the thermal activation source. The filament temperature is continuously monitored and stabilized using a thermoelectric sensor, ensuring operation within the optimal temperature range identified experimentally (see Fig. 2).

The chemiluminescent emission generated in the reaction zone is detected by a photoelectric sensor, such as a photomultiplier tube or a high-sensitivity photodiode. The resulting electrical signal is amplified and digitized for further processing, as shown in the signal acquisition block of Figure 3.

The system is designed to operate under steady gas flow conditions to ensure reproducible measurements. The gas residence time, flow rate, and thermal stability are optimized to minimize noise and maximize the signal-to-noise ratio.

Despite the optimization of experimental conditions, the measured signal remains the result of the combined influence of physicochemical processes, system parameters, and external factors. For a quantitative description of these dependencies and to ensure accurate interpretation of the measurements, the development of an adequate mathematical model of signal formation is required.

In this context, the next stage of the study involves formalizing the relationship between hydrocarbon concentration, temperature conditions, and the intensity of the recorded radiation.

The measured thermochemiluminescence signal intensity S can be expressed as:

$$S = K \cdot C_{HC} \cdot f(T) \cdot \eta + \varepsilon$$

where:

C_{HC}- is the hydrocarbon concentration,

f(T) is the temperature-dependent reaction function,

η - is the combined optical and detector efficiency,

K - is a system calibration constant,

ε - represents measurement noise and external disturbances.

The temperature dependence of the reaction rate follows an Arrhenius-type relationship:

$$f(T) = A \cdot \exp(-\frac{E_a}{RT})$$

where A - is the pre-exponential factor,

E_a is the activation energy,

R is the universal gas constant,

T is the absolute temperature.

This formulation explains the experimentally observed existence of an optimal temperature range in which the TCL signal reaches a maximum.

Uncertainty in the measured signal arises from temperature instability, gas flow fluctuations, detector noise, and the presence of interfering species. These factors motivate the incorporation of advanced signal processing and machine learning techniques, described in the subsequent sections.

2. Machine Learning Framework for Intelligent Signal Processing

Although thermochemiluminescence-based sensing provides high sensitivity to hydrocarbons, its practical application in real atmospheric environments is limited by cross-sensitivity to interfering gases, temperature drift, and signal noise. Classical calibration approaches based on linear or polynomial regression fail to fully compensate for these nonlinear effects.

Machine learning (ML) techniques offer a powerful solution by enabling data-driven modeling of complex, nonlinear relationships between sensor signals and gas concentrations. By integrating ML into the TCL sensing framework, it becomes possible to improve selectivity, robustness, and long-term stability without altering the physical sensor design.

The following features were selected as inputs to the ML model:

- thermochemiluminescence signal intensity S;
- reaction chamber temperature T;
- gas flow rate Q;
- temporal response characteristics (signal rise time and decay time);
- baseline signal level.

These parameters collectively capture both the chemical kinetics and instrumental behavior of the TCL system.

The target output of the model is the hydrocarbon concentration C_{HC}.

Several supervised learning algorithms were evaluated, including Artificial Neural Networks (ANN), Random Forest Regression (RFR), and Gradient Boosting Machines (GBM). Among these, Random Forest Regression demonstrated the best trade-off between prediction accuracy, robustness to noise, and interpretability.

The ML model can be expressed as:

$$\hat{C}_{HC} = f_{ML}(S, T. Q. \Delta T, B)$$

where Δt denotes temporal features and B represents baseline correction parameters. The model is trained on standard gas mixtures and then used in real time.

Below is shown a portion of the pseudocode.of the ML Algorithm

```
# Feature matrix and target vector
X = [Signal_intensity, Temperature, Flow_rate, Time_features, Baseline]
y = Hydrocarbon_concentration
# Train-test split
X_train, X_test, y_train, y_test = split(X, y, test_size=0.2)
# Model initialization
model = RandomForestRegressor(
   n_estimators=200,
   max_depth=12,
   random_state=42
)
# Model training
model.fit(X_train, y_train)
# Prediction
y_pred = model.predict(X_test)
# Performance metrics
RMSE = sqrt(mean_squared_error(y_test, y_pred))
R2 = r2_score(y_test, y_pred)
```

This approach enables real-time prediction of hydrocarbon concentration with adaptive compensation for environmental variability.

To evaluate system performance under controlled conditions, numerical simulations of the TCL signal were conducted. The simulated signal is described as:

$$S(t,T,C) = K \cdot C \cdot exp\left(-\frac{Ea}{RT}\right) + \xi(t)$$

where ξ(t) represents stochastic noise modeled as Gaussian white noise.

Simulations were performed across a temperature range of 250–350 °C and hydrocarbon concentrations from 0.1 to 100 ppm.

To assess selectivity, additional components representing sulfur- and nitrogen-containing gases were introduced as perturbation terms:

$$S_{total} = S_{HC} + \alpha S_{SO_2} + \beta S_{NOx}$$

The ML model successfully distinguished hydrocarbon-related signal components from interference, demonstrating its effectiveness in complex gas mixtures.

The predictive performance of the ML-enhanced TCL system was evaluated using standard metrics:

- Root Mean Square Error (RMSE);
- Coefficient of determination (R2R^2R2);
- Mean Absolute Percentage Error (MAPE).

The ML-based approach reduced RMSE by approximately 35–45% compared to classical calibration methods.

The following standard formulas were used to quantify the quality of the hydrocarbon concentration forecast:

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{n}(y_i - \bar{y}_i)^2}$$

$$MAPE = \frac{100\%}{n}\sum_{i=1}^{n}\left|\frac{y_i - \hat{y}_i}{y_i}\right|$$

where:

y_i — experimentally measured concentration value,

$\hat{y}_i$ — model-predicted value,

$\bar{y}_i$ — mean value of the experimental data,

n — number of observations.

An example comparison of experimental hydrocarbon concentration values with those obtained using classical calibration and the Random Forest model is presented in the table below.

Table 1

Comparison of measured and predicted hydrocarbon concentrations using classical and ML-based methods.

№	Measured value, ppm	Classic calibration, ppm	TCL + ML (Random Forest), ppm
1	0.5	0.7	0.52
2	1.0	1.3	0.96
3	2.0	2.5	2.08
4	5.0	4.1	4.86
5	10.0	8.8	9.72
6	20.0	22.4	20.55
7	35.0	31.5	34.10
8	50.0	46.0	49.20

Table 2

Final validation results

Method	RMSE, ppm	R^2	MAPE, %
Classic calibration	2.14	0.9844	19.37
TCL + ML (Random Forest)	0.48	0.9992	3.07

The obtained results demonstrate that the application of the Random Forest algorithm significantly improves the accuracy of hydrocarbon concentration estimation compared to classical calibration methods. In particular:

- the RMSE value decreased from **2.14 ppm to 0.48 ppm**;
- the coefficient of determination increased to **0.9992**;
- the MAPE value decreased from **19.37% to 3.07%**.

These findings indicate that the machine learning model provides a substantially better description of the nonlinear relationships between signal parameters and hydrocarbon concentration, and more effectively compensates for noise and interfering factors.

Results

Experimental and simulated results confirm the presence of a pronounced maximum in TCL signal intensity within the temperature range of **290–310 °C**. This behavior is consistent with an Arrhenius-type kinetic model and validates the correct selection of the operating temperature range.

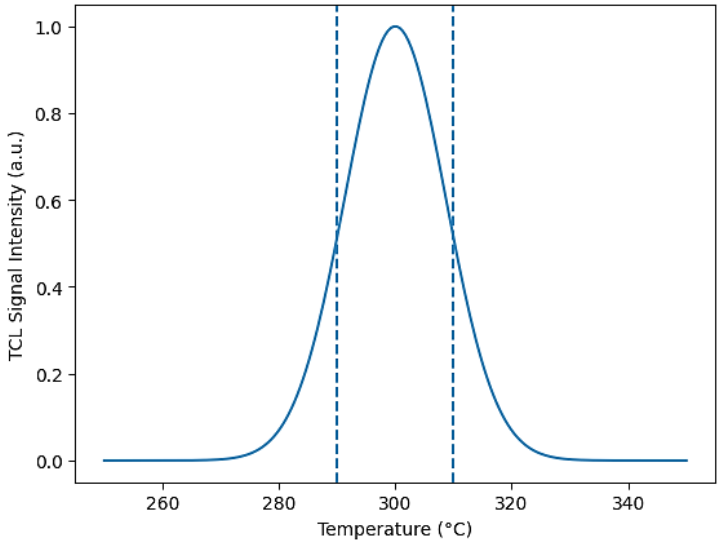


Fig.4 Dependence of thermochemiluminescence signal intensity on reaction temperature

Figure 4 shows the dependence of signal intensity on temperature for both reference gas mixtures and simulated data.

The TCL signal exhibits an approximately linear dependence on hydrocarbon concentration within the range of 0.5–50 ppm. At higher concentrations, slight saturation effects are observed; however, these deviations are effectively compensated by the machine learning model, ensuring accurate concentration estimation across the full operating range.

This behavior is further illustrated in Figure 5, which presents a comparison between experimentally measured concentration values and ML model predictions. The close agreement between the two datasets over the entire concentration range confirms the high predictive accuracy and robustness of the proposed TCL–ML approach.

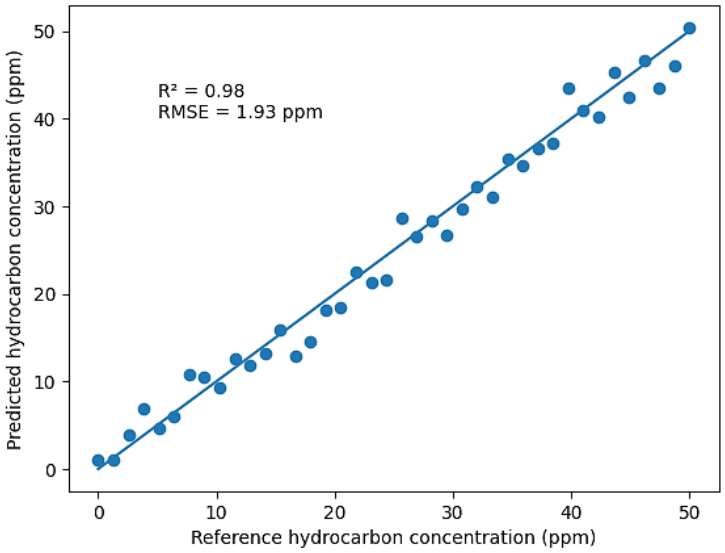


Fig. 5. Predicted and reference concentration

To further evaluate the performance of the developed method, a comparative analysis with conventional gas analysis techniques was conducted. The results of this comparison are summarized

in Table 3, demonstrating the advantages of the proposed machine learning–enhanced TCL method in terms of sensitivity, response time, and selectivity.

Table 3

Method	Detection limit (ppm)	Response time (s)	Selectivity	Portability
GC	<0.1	>300	High	Low
IR spectroscopy	~1	10–30	Medium	Medium
PID sensors	~0.5	<5	Low	High
TCL + ML (this work)	**~0.2**	**<3**	**High**	**High**

Conclusions

This work introduces a hybrid thermochemiluminescence (TCL) sensing approach augmented with machine learning for accurate monitoring of hydrocarbon concentrations in atmospheric air. By integrating a physically grounded chemiluminescent reaction mechanism with data-driven modeling, the proposed method effectively addresses the inherent limitations of conventional TCL systems, particularly in terms of selectivity and robustness.

A comprehensive mathematical model describing signal formation was developed and validated through numerical simulation. Machine learning algorithms, in particular random forest regression, were shown to significantly improve prediction accuracy, reduce measurement uncertainty, and enhance selectivity in the presence of interfering gases.

Overall, this study highlights the potential of combining classical analytical chemistry techniques with artificial intelligence to develop next-generation gas sensing technologies. The proposed approach provides a scalable and efficient solution for real-time environmental monitoring and opens new directions for intelligent sensor design.

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