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Development of a Deep Learning-Enhanced Chemiluminescence Method for Trace Formaldehyde Detection in Water Samples

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Abstract: Formaldehyde, a widely encountered organic pollutant and recognized Group I carcinogen, poses significant threats to human health even at trace concentrations. This study presents a chemiluminescence-based method for the sensitive detection of trace formaldehyde in water, enhanced by deep learning algorithms for signal processing and concentration prediction. The detection system utilizes an optimized gallic acid reagent system, combined with a photomultiplier tube (PMT) for signal acquisition. Chemiluminescence signals are processed using a convolutional neural network (CNN) to denoise, extract features, and predict formaldehyde concentrations in real time. The method achieves a detection limit of 5.96×10^{-5} μg/mL, outperforming conventional phenol reagent methods in terms of simplicity, speed, sensitivity, and accuracy. Its capability for online and portable detection makes it a promising approach for real-time water quality monitoring and environmental analysis.

Keywords: Chemiluminescence; Formaldehyde Detection; Gallic Acid; Deep Learning; Environmental Monitoring.

1. Introduction

Formaldehyde is a common organic pollutant widely present in daily life, often used for disinfection, sterilization, and preservation. It can be found in various sources such as building materials, leather products, automotive interiors, cosmetics, and even alcoholic beverages. Formaldehyde has been classified as a Group I carcinogen by the International Agency for Research on Cancer (IARC) [1]. Its adverse effects on human health include respiratory irritation, tissue degeneration, and carcinogenicity [2-3]. Long-term exposure to low concentrations of formaldehyde can cause chronic respiratory diseases, impair DNA repair mechanisms, lead to menstrual disorders in women, and negatively impact cognitive development and memory in adolescents [4]. Acute exposure to high concentrations can result in suffocation, respiratory paralysis, and even death, with vulnerable populations such as children, the elderly, and pregnant women facing particularly elevated risks.

Formaldehyde pollution in the environment has become a significant public health concern, prompting extensive research into sensitive, accurate, and rapid detection technologies. Traditional formaldehyde detection methods include the phenol reagent method [5], acetylacetone method [6], gas chromatography (GC) [7], liquid chromatography (LC) [8], ion chromatography (IC) [9], and fluorescence methods [10]. In recent years, newer approaches such as sensor-based detection [11], chemiluminescence analysis [12], potentiometric techniques [13], and microfluidic chip technology [14] have emerged. Furthermore, to enhance sensitivity and specificity, hybrid technologies have been developed, including flow injection-chemiluminescence methods [15], high-performance liquid chromatography-chemiluminescence coupling [16], capillary electrophoresis-chemiluminescence methods [17], and molecular imprinting-chemiluminescence techniques [18].

With the rapid development of artificial intelligence and data science, deep learning has shown great potential in enhancing chemical sensing and analytical techniques. In particular, convolutional neural networks (CNNs) and recurrent neural networks (RNNs) have been successfully applied to process complex analytical signals, extract hidden features, and improve detection accuracy and robustness in the presence of environmental noise. By integrating deep learning models with chemiluminescence detection, it is possible to develop an intelligent formaldehyde detection platform that achieves both high sensitivity and real-time analysis.

In this study, trace formaldehyde in water samples was selected as the target analyte. An in-house developed chemiluminescence online detection system was used, incorporating an optimized reagent system consisting of ethylene glycol (cosolvent), gallic acid, hydrogen peroxide, sodium hydroxide, and a sensitizer. The influence of reagent mixing order and the length of the liquid collection tube on chemiluminescence intensity was systematically evaluated. In addition, a deep learning-based signal processing framework was introduced to denoise, extract temporal features, and predict formaldehyde concentrations in real time. Finally, the proposed method was compared with the traditional phenol reagent method and standard instrument methods to validate its performance.

2. Experimental

2.1 Reagents and Apparatus

Gallic acid, sodium hydroxide, hydrogen peroxide, ammonium ferric sulfate, ethylene glycol, hexamethylenetetramine, sodium hydroxymethyl sulfonate, ethylenediaminetetraacetic acid (EDTA), cetyltrimethylammonium chloride, zinc nitrate hexahydrate, nickel nitrate hexahydrate, silver nitrate, chromium trichloride, potassium chloride, and manganese chloride tetrahydrate were purchased from Chengdu Kelon Chemical Reagent Factory. Among these, the purity of potassium chloride and sodium hydroxide was chemically pure, the purity of gallic acid was 99%, and the purity of hydrogen peroxide was 30%, with all other reagents being analytical grade. Tetrabutylammonium iodide, polyvinylpyrrolidone, sodium dodecyl sulfate, lauryl polyoxyethylene ether, cetyltrimethylammonium bromide, sodium methyl sulfonate, potassium hydrogen phthalate, octadecyl trimethylammonium chloride, and trisodium citrate dihydrate were purchased from SIGMA Reagent Company (USA), with tetrabutylammonium iodide at 99% purity.

The analytical balance (XY-A/B/C) was supplied by Shanghai Shunyu Hengping Scientific Instrument Co., Ltd., while the chemiluminescence detection system (ATMOS-DSI) was developed by the Analysis and Test Center Laboratory of Sichuan University of Light Chemical Technology. A 1200UV-visible spectrophotometer from Aoyi Instruments (Shanghai) Co., Ltd. was also used in the study.

2.1.1 Experimental Setup

The schematic diagram of the experimental device is shown in Figure 1. The system is based on a custom-built chemiluminescence detection platform [19], where the reaction reagent and formaldehyde sample solution are combined via a pipeline and transported into the reactor using a peristaltic pump. The mixed solution enters a reaction interface composed of hydrophilic material, ensuring that the trace solution spreads rapidly across the surface to initiate a stable chemiluminescence reaction. The waste solution is subsequently discharged into a collection bottle.

A photomultiplier tube (PMT) converts the emitted chemiluminescence into an electrical signal, which is acquired by the main control circuit and transmitted to a computer software system for real-time processing, recording, and display. Under optimized conditions, the luminescence intensity correlates with the formaldehyde concentration, and the system software generates corresponding signal data. Formaldehyde concentration can thus be quantified based on the chemiluminescence intensity.

2.1.2 Intelligent Signal Processing Enhancement

To further enhance detection accuracy and reduce noise interference, a deep learning-based signal processing module was integrated into the data processing pipeline. The time-series chemiluminescence signal collected by the PMT was pre-processed using baseline correction and smoothing algorithms. The pre-processed signal was then fed into a convolutional neural network (CNN) model, trained to extract critical signal features and predict formaldehyde concentration in real time. This intelligent enhancement not only improved the sensitivity and robustness of the detection system, but also enabled adaptive calibration under varying environmental conditions.

2.1.3 Reactor Design

The reactor was fabricated using Teflon material, with a long reaction bed made from microfine polyester fiber. This porous fiber surface contains micro-grooves, significantly increasing the contact area between the reaction reagent and the target analyte. The detection reagent is evenly spread over the fiber surface by gravity and liquid diffusion, forming a thin liquid film. The formaldehyde sample is then pumped into the reactor, where it reacts with the reagent on the liquid film surface, emitting light that is captured by the PMT. The resulting chemiluminescence signal, after pre-processing and deep learning analysis, is used to quantify the formaldehyde concentration with improved accuracy and noise suppression.

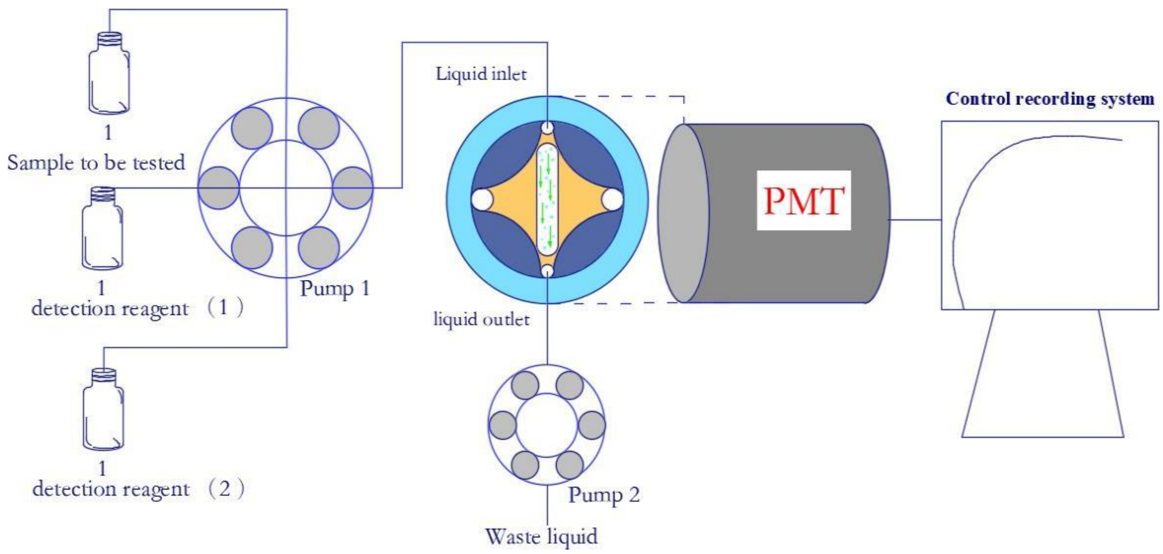

Figure 1. Schematic diagram of flow path detection; PMT is a photomultiplier tube.

2.2 Parametric Optimization

Gallic acid was dissolved in a glycol-water system, where the presence of glycol significantly improved the dispersion of gallic acid, ultimately influencing the chemiluminescence intensity of the system. As shown in Figure 2, with increasing glycol concentration, the chemiluminescence intensity of a 1.0×10^{-3} mg/mL formaldehyde solution gradually increased, reaching a maximum signal intensity of 78 mV when the glycol concentration was 30% (v/v). Therefore, a glycol concentration of 30% (v/v) was selected as the optimal condition.

The effect of gallic acid concentration on the chemiluminescence intensity was also investigated across a concentration range of 1.0×10^{-3} to 4.0×10^{-2} mol/L. As presented in Figure 3, the maximum luminescence intensity of 90 mV was observed when the gallic acid concentration reached 0.030 mol/L. Consequently, 0.030 mol/L was determined to be the optimal concentration of gallic acid.

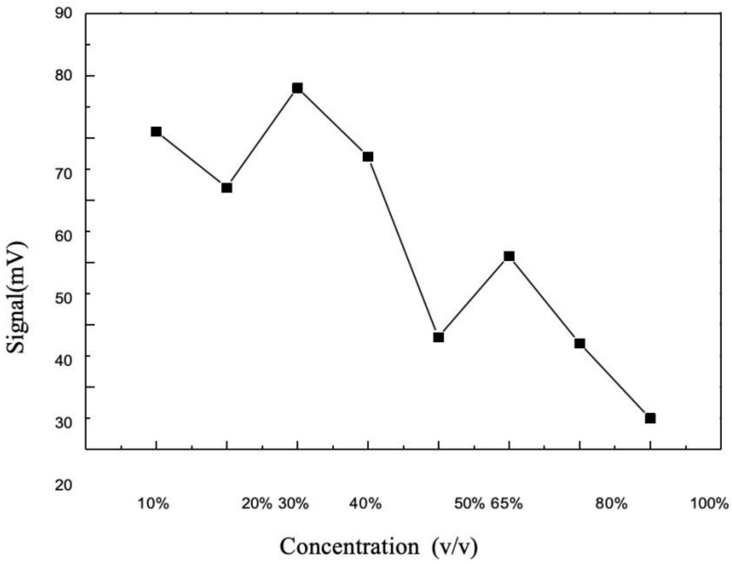

Figure 2. The effect of the concentration of ethylene glycol on CL intensity

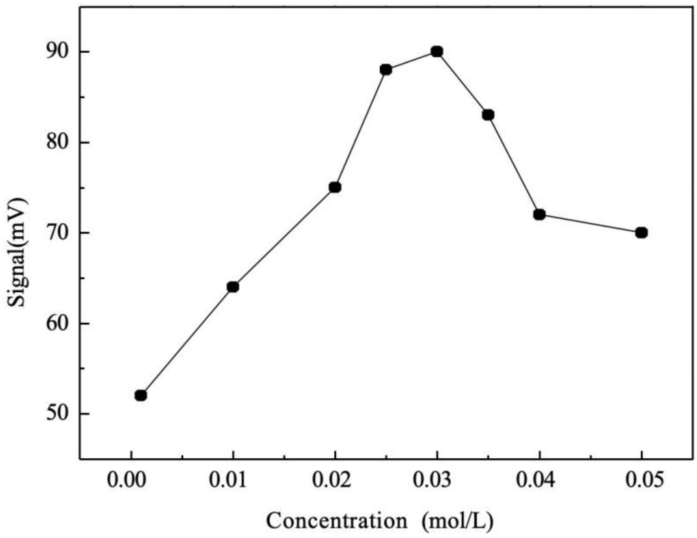

Figure 3. The effect of the concentration of gallic acid on CL intensity

Hydrogen peroxide served as the oxidant in the reaction system. To optimize its concentration, the effect of hydrogen peroxide concentration on luminescence intensity was investigated within the range of 0 to 0.7 mol/L. As shown in Figure 4, the chemiluminescence intensity increased with increasing hydrogen peroxide concentration, reaching a maximum value of 95 mV at 0.20 mol/L. Therefore, 0.20 mol/L was selected as the optimal hydrogen peroxide concentration.

Since the chemiluminescence reaction involving gallic acid is pH-dependent, the effect of sodium hydroxide concentration on luminescence intensity was evaluated over the range of 0 to 0.5 mol/L. As shown in Figure 5, the luminescence intensity peaked at 98 mV when the sodium hydroxide concentration was 0.12 mol/L. Therefore, 0.12 mol/L was chosen as the optimal sodium hydroxide concentration.

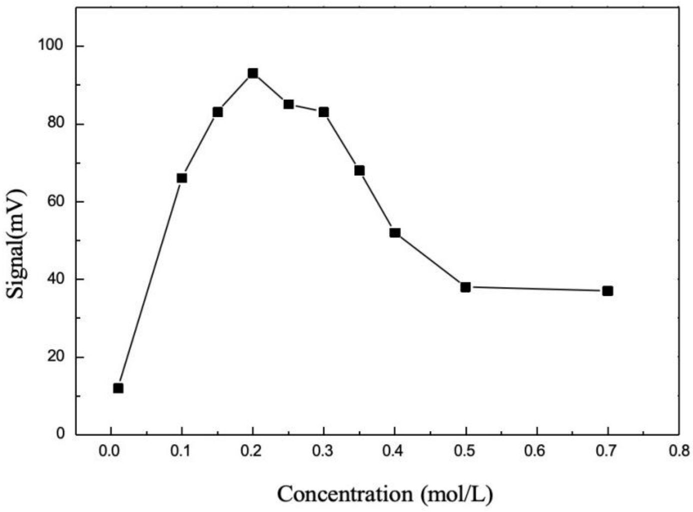

Figure 4. The effect of concentration of hydrogen peroxideon CL intensity

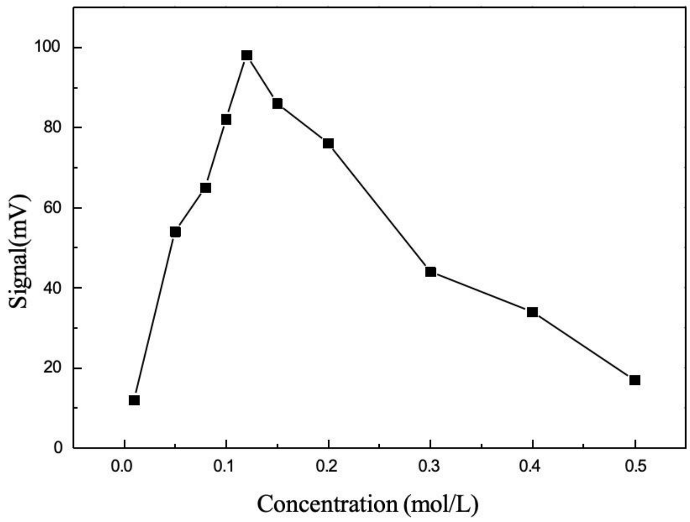

Figure 5 . The effect of concentration of sodium hydroxideon CL intensity

In addition, the effects of different sensitizers—including sodium alkyl sulfate (SAS), tetrabutylammonium iodide (TBAI), potassium hydrogen phthalate (KAP), disodium ethylenediaminetetraacetate (EDTA), cetyltrimethylammonium chloride (CTAC), and trisodium citrate (Citrate)—on the chemiluminescence intensity of the gallic acid system were systematically studied. As shown in Figure 6, TBAI significantly enhanced the luminescence intensity compared to the blank control, indicating that TBAI was the most effective sensitizer for this system.

Further investigation into the effect of TBAI concentration revealed that the luminescence intensity reached a maximum value of 181 mV when the TBAI concentration was 0.40% (v/v), as shown in Figure 7. Thus, 0.40% (v/v) was selected as the optimal concentration of TBAI.

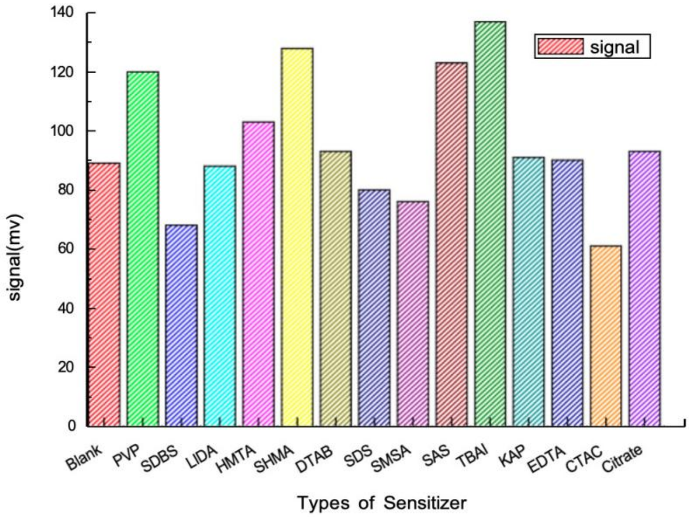

Figure 6. Curves of luminescence intensity and signal-to-noise ratio under different sensitizers.

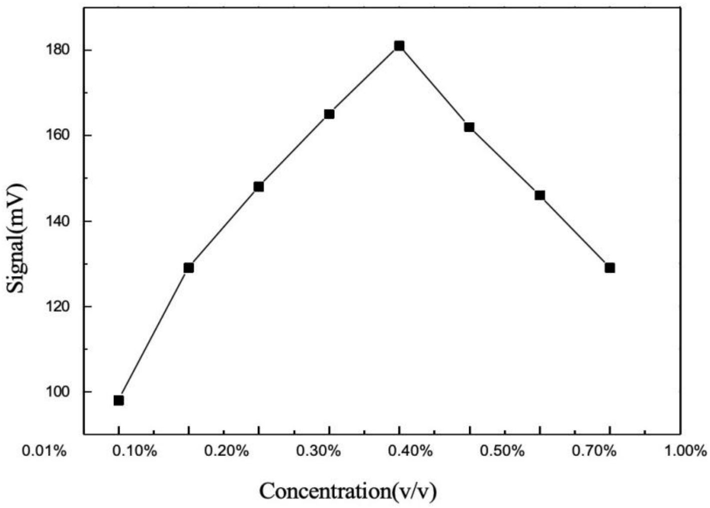

Figure 7. The effect of concentration of tetrabutylammonium iodide on CL intensity.

Finally, the influence of the liquid collector length on the chemiluminescence reaction was assessed. As shown in Figure 8, the highest luminescence intensity was observed when the liquid collector length was 7 cm. Therefore, the optimal liquid collector length was determined to be 7 cm.

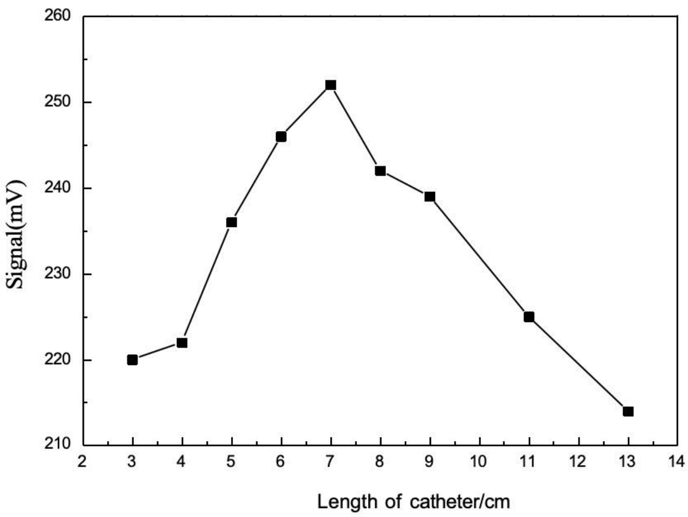

Figure 8. The luminescence intensity varies with the length of the collector.

3. Analytical Performance

To evaluate the reliability and accuracy of the proposed experimental method, a comparative analysis was conducted using the conventional phenol reagent method [20] and the chemiluminescence-based instrument method developed in this study. As shown in Figure 9, the linear regression equation for the phenol reagent method was:

$$y = 0.344x + 0.0464$$

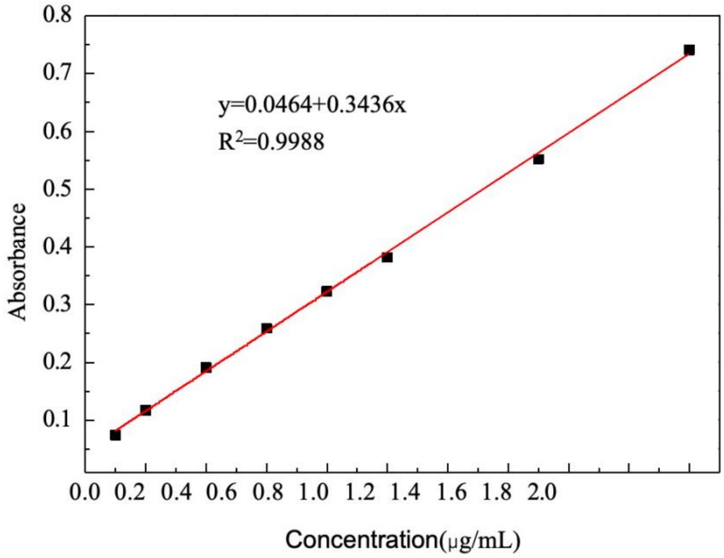

Figure 9. Standard graph of phenol reagent method.

Under optimized experimental conditions, the chemiluminescence intensity obtained from the instrument method was plotted against the formaldehyde concentration. As shown in Figure 10, the linear regression equation for the instrument method was:

$$y = 243.624x + 5.865$$

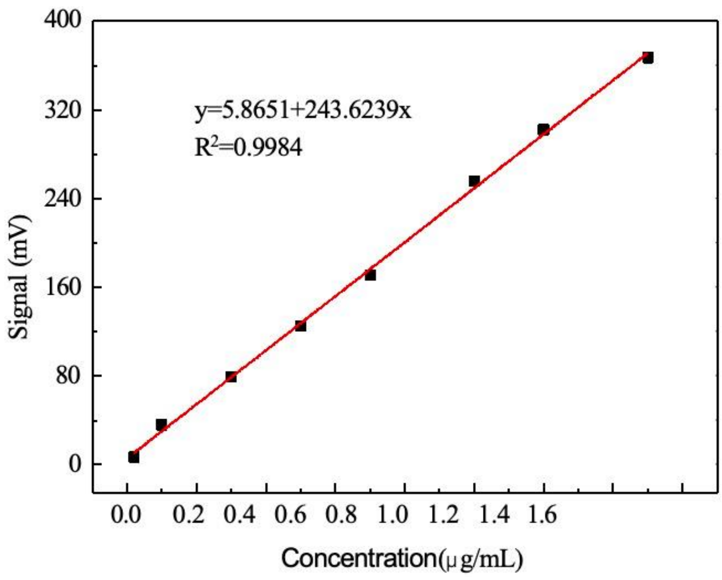

Figure 10. Standard graph of instrumental method

The correlation coefficient (R2R^2R2) for this method was 0.998, with a detection limit of 5.96×10^{-5} μg/mL and a relative standard deviation (RSD) of 0.92%.

To further validate the accuracy of the proposed method, four formaldehyde standard solutions with different concentrations were prepared and measured in parallel using both the phenol reagent method and the instrument method, with each sample analyzed in triplicate. The results are summarized in Table 1, demonstrating good agreement between the two methods.

Table 1. Table of Test results of four kinds of samples by phenol reagent Method and Instrument method (n=3)

Test sample(μg/mL)	Phenol reagent method(μg/mL)	Instrumental method(μg/mL)
sample1(0.500)	0.489	0.493
sample2(0.800)	0.758	0.781
sample3(1.200)	1.161	1.217
sample4(1.500)	1. 451	1.546

4. Conclusion

In this study, a sensitive and efficient method for detecting trace formaldehyde in water samples was developed based on the principle of interfacial chemiluminescence, with an optimized gallic acid reagent system. The proposed method achieved a detection limit of 5.96×10^{-5} μg/mL, demonstrating excellent sensitivity. Comparative analysis with the traditional phenol reagent method confirmed that the proposed method offers significant advantages in terms of simplicity, speed, sensitivity, and accuracy.

Furthermore, the method is well-suited for online monitoring and portable field detection, providing strong potential for practical applications in environmental monitoring and water quality assessment, particularly for the detection of trace formaldehyde contamination in diverse aquatic environments.

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