

Article

Laser-Induced Graphene-Based Gas Sensor with PEDOT:PSS/Gold–Platinum Nanocomposites for Highly Sensitive Detection of Methane

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Abstract: Methane is a common intestinal gas that has been linked to a variety of gastrointestinal disorders. In this study, we prepared a LIG-based electrochemical sensor modified with PEDOT:PSS/Au-Pt nanocomposites for high-sensitivity methane detection. LIG's high porosity, flexibility, and excellent electrical conductivity are beneficial for electrochemical detection. The PEDOT:PSS film is electrodeposited on the electrode surface to amplify the signal further. The synergistic effect of bimetallic nanocomposites can improve the electrochemical catalytic activity and increase the specific surface area and the advantages of active sites. The experimental results show that the LIG sensor has a wide linear detection range (2–500 ppm), a low detection limit (about 0.36 ppm), high sensitivity, and good repeatability. The sensor is easy to manufacture, is low-cost, operates at room temperature, has high sensitivity, has a low detection limit, and is expected to be used for methane breath detection in the future.

Keywords: electrochemical gas sensor; laser-induced graphene; methane detection; nanocomposites; methanogens



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1. Introduction

Methane (CH₄) is prevalent in people's daily lives [1] and is the main component of natural gas and is a major greenhouse gas. In recent years, studies have found that methanogens in the human gut can produce methane gas through anaerobic digestion of ingested food [2] and are closely related to some gastrointestinal diseases, such as constipation, small intestinal bacterial overgrowth (SIBO), colorectal cancer, inflammatory enteritis, etc. [3–5]. With increasing awareness and recognition of methane and its relationship to gut health, to highlight the importance of methanogens, a new term has been proposed in clinical guidelines published by the American College of Gastroenterology (ACG) in 2020 [6], namely intestinal methanogen overgrowth (IMO). The methane produced by methanogens in the intestine is mainly discharged from the body through the intestinal exhaust, and the rest is discharged by pulmonary respiration through the circulatory system [7]. This makes it possible to diagnose IMO by detecting methane in the breath.

At present, molecular biological methods are some of the most common methods for detecting methanogens [8,9], such as polymerase chain reaction (PCR), quantitative real-time PCR (qPCR), 16S rRNA gene sequencing, fluorescence in situ hybridization (FISH),

etc. These methods have high sensitivity and specificity and do not require culture time, but they are expensive, require professional technical personnel, and are prone to false positives. To solve these problems, the detection of methane in the breath can be used to monitor the presence of intestinal methanogens indirectly.

Breath testing [10–12] is convenient, safe, noninvasive, and low-cost, making it easy for patients to accept. Human cells cannot produce methane gas, so the methane gas detected in a breath sample must come from methanogens. At present, the methods used in the laboratory for detecting methane in exhaled gas are usually gas chromatography or mass spectrometry [13,14]. The equipment is large, expensive, and complex, and it is difficult to realize portable breath detection. In addition, metal oxide semiconductor methane sensors [15–17] have high sensitivity and a low cost, but the selectivity is poor and the operating temperature is high; infrared methane sensors [18–20] have the advantage of high selectivity, but they cost more and have a larger volume. In 2024, Kong et al. [21] proposed a near-infrared sensor for real-time methane detection with a volume of 233.3 cm³, and the volume of the electrochemical sensor in this paper is about 12 mm × 15 mm × 0.6 mm. After comparison, it can be seen that the volume of our electrochemical sensor is very small. Electrochemical sensors [22] have the advantages of low cost, high sensitivity, good selectivity, and can work at room temperature, but there are few reports on the detection of methane by electrochemical methane sensors. Based on this, we propose an electrochemical methane sensor based on laser-induced graphene (LIG) electrodes.

Laser engraving can use the photochemical and photothermal effects generated by laser radiation on the surface of the material to convert precursor materials, such as polyimide (PI), directly into graphene [23–25]. The resulting graphene has a highly porous surface structure and high specific surface area [26], which greatly increases the gas adsorption capacity. At the same time, LIG has excellent electrical conductivity [27] and chemical stability [28], which can ensure the long-term stability and reliability of the sensor. Moreover, the patterned preparation of LIG is simple and low-cost, and large-scale production can be realized.

To further amplify the signal, we modified Poly (3,4-ethylenedioxythiophene): Polystyrene sulfonate (PEDOT:PSS) films and Au-Pt nanocomposites on the electrode surface. PEDOT:PSS is a widely used conductive polymer material with good electrical conductivity and excellent stability in a variety of chemical environments [29–32]. The high surface area and high conductivity of PEDOT:PSS can improve the sensitivity of the sensor and permit fast electron transmission capability, so that the sensor can quickly respond to gas concentration changes, suitable for real-time detection [33]. Zheng et al. [34] prepared a reduced graphene oxide (rGO)/PEDOT:PSS nanocomposite film for hydrogen sensing with a maximum sensitivity of 4.2% and a fast recovery and response time. Many studies have confirmed that Au and Pt have high catalytic activity [35,36], which can effectively promote the oxidation–reduction reaction of methane gas on the electrode surface, improve the sensitivity and response speed of the sensor, and, at the same time, these two metals have good electrical conductivity, which is conducive to improving the signal transmission efficiency. The Au-Pt nanocomposite can further improve the catalytic activity, create a large specific surface area, and result in more active sites through synergistic effects [37] and, therefore, achieve low-concentration methane sensing. We compared PEDOT:PSS/Au-Pt nanocomposites with other materials in the literature. Hingangavkar et al. [38] proposed a gas sensor based on WO₃-GO material for the detection of NO₂, which has excellent selectivity, repeatability, and stability. The material was prepared by the hydrothermal method, and the hydrothermal reaction was carried out at 100 °C for 4 h. In this work, the PEDOT:PSS/Au-Pt nanocomposites were prepared by the electrochemical method, which

can be achieved at room temperature in about 15 min, and can achieve low-concentration gas sensing.

In this work, we demonstrate an electrochemical sensor for the detection of methane gas based on a LIG electrode, which is modified with Au-Pt nanocomposites and further amplifies the signal using the PEDOT:PSS membrane. And we use room temperature ionic liquid (RTIL) as the electrolyte. The schematic diagram of the electrode is shown in Figure 1.

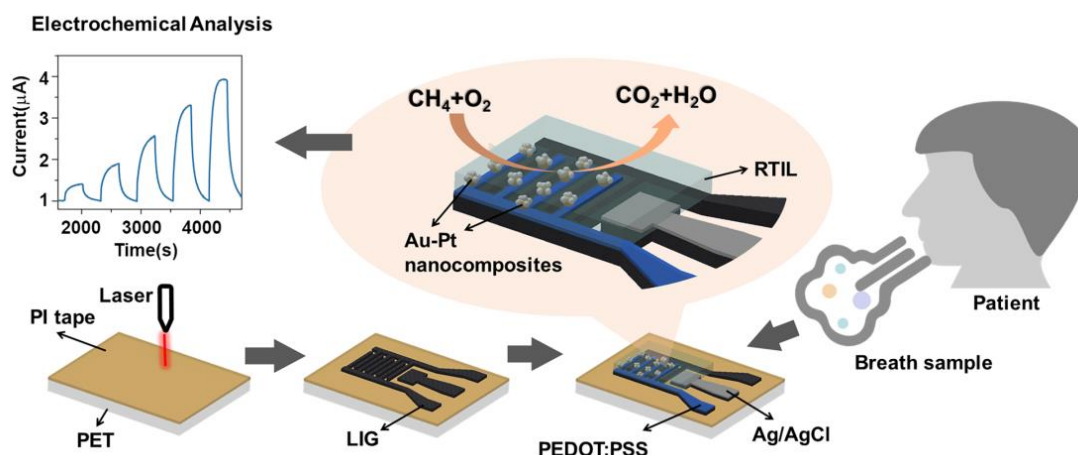


Figure 1. Schematic diagram of LIG electrode preparation.

2. Materials and Methods

2.1. Reagent and Apparatus

The chemicals used in the experiment were analytically pure and used without further treatment. Gold chloride trihydrate ($\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$), potassium hexachloroplatinate (IV) (K_2PtCl_6), 3,4-ethylenedioxythiophene (EDOT), and RTIL 1-Butyl-1-methylpyrrolidinium Bis (trifluoromethanesulfonyl)imide were all obtained from Shanghai Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). PSS was obtained from Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China). PI tape (0.055 mm) was obtained from Hangzhou Shengneng Packaging Materials Co. Ltd. (Hangzhou, China), the polyethylene terephthalate (PET) board (0.5 mm) was obtained from Hengda Automation Technology Co., Ltd. (Taizhou, China), and conductive silver paste was obtained from Shenzhen Xinwei Electronic Materials Co., Ltd. (Shenzhen, China). Ag/AgCl paste was purchased from Shanghai Julong Electronic Technology Co., Ltd. (Shanghai, China). All water used for the experiment was deionized water (DI water), which was made by a water purification system obtained from Shanghai CanrexAnalytic Instrument Co., Ltd. (Shanghai, China).

The CO_2 laser engraving machine (50 W) was obtained from Liaocheng Julong Laser Equipment Co., Ltd. (Liaocheng, China). All electrochemical measurements were performed at the electrochemical workstation CHI660E (Shanghai Chenhua Instrument Co., Ltd., Shanghai, China). The Pt counter electrodes and Ag/AgCl reference electrodes were obtained from Shanghai Chenhua Instrument Co., Ltd. (Shanghai, China). The standard air and methane were from Hangzhou Jingong Special Gas Co., Ltd. (Hangzhou, China). The required concentration of methane gas was configured using a gas mixer (MCQ GB100, MCQ Instruments, Rome, Italy). Various characterizations of LIG and modified materials were used. A field emission scanning electron microscope (SEM) SU-8010 and SU-3500 (Hitachi Limited, Tokyo, Japan), X-ray photoelectron spectrometer (XPS) Thermo Scientific Escalab250Xi (Thermo Fisher Scientific, Waltham, MA, USA), Raman spectrometer Renishaw inVia Qontor (Renishaw, Wotton-under-Edge, UK), infrared spectrometer Thermo Nicolet 6700 (Thermo Fisher Scientific, Waltham, MA, USA), and

High-Resolution Transmission Electron Microscopy (HRTEM) JEM-2100 (Japan Electronics Co., Ltd., Amagasaki, Japan).

2.2. Fabrication of LIG

Firstly, the PET base plate was ultrasonically cleaned with ethanol and water, and after drying, the PI tape was smoothly pasted on the clean PET base plate to avoid bubbles as much as possible. Then, the tape surface was washed with ethanol and deionized water, respectively, and dried. Before laser engraving, CorelDRAW X4 (Version 14.0.0.701) was used to draw the desired interfinger electrode pattern. The left interfinger was the working electrode (WE), the right interfinger was the counter electrode (CE), and the square area in the middle was the reference electrode (RE). The electrode structure was based on the literature [39], and the appropriate improvement was made on this basis. The width and interval of the interfinger were 400 μm , the length of the interfinger was 5.4 mm, and the size of the entire LIG electrode was 12 mm $\times$ 15 mm. The pattern was then carved onto the PI tape using a CO₂ laser engraving machine with a laser wavelength of 10.6 μm and CorelLASER (Version 2013.02) software to obtain the desired graphene electrodes. The parameters of laser engraving were optimized, and the optimization process was shown in the Supplementary Materials. The best parameters were finally determined as a power of 9.5% and a stepping speed of 55 mm/s. The physical diagram of the LIG electrode is shown in Figure S1A.

2.3. Electrochemical Synthesis of PEDOT:PSS Films and Au-Pt Nanocomposites

We use an external Ag/AgCl reference electrode and Pt counter electrode when electroplating the electrodes. The electroplating method was based on the literature [40] and improved appropriately. The electrode was immersed in EDOT:PSS aqueous solution (0.02 M EDOT, 1% PSS (*w/v*)) and electroplated for 150 s at a potential of 0.9 V to form PEDOT:PSS film. After cleaning, the electrode was immersed in 5 mM HAuCl₄/0.5 M H₂SO₄ solution and electroplated for 300 s at a potential of -0.1 V. After cleaning again, the electrode was immersed in 5 mM KPtCl₄/0.5 M H₂SO₄ solution and electroplated for 300 s at a potential of -0.3 V to form the Au-Pt nanocomposites.

2.4. Sensor Design and Implementation

The LIG electrode was carefully pasted to a custom PCB board and dried under an infrared lamp. The reference electrode was then coated with the Ag/AgCl paste to stabilize the potential and dried. After that, WE, RE, and CE are connected to the corresponding pads with conductive silver paste and dried. After all connections are completed, the physical picture is shown in Figure S1B. Finally, it was packaged with insulating adhesive to avoid the subsequent diffusion of ionic liquid to the non-working area. The gas chamber was designed according to the electrode size, and the resin was used as the material to manufacture the gas chamber by 3D printing. The gas chamber is shown in Figure S1C. Before detecting the gas, 5 μL of RTIL 1-Butyl-1-methylpyrrolidinium Bis(trifluoromethanesulfonyl)imide was dropped in the working area, and then, the gas chamber was covered in the corresponding region of the PCB board. The gas chamber and PCB board were sealed with glue to prevent air leakage. The gas chamber was then connected to the gas mixer. Then, the electrochemical workstation was connected to the electrode. For subsequent gas testing, a standard air cylinder and a 1000 ppm methane cylinder were used to configure the corresponding methane gas concentration as shown in Figure S2. We use chronoamperometry (CA) as the electrochemical method for detecting the gas concentration. CA is a constant voltage method; the principle of CA for gas detection is as follows: the applied constant potential causes the target gas to undergo oxidation or reduction reactions on the electrode surface, which generates a Faradaic current and leads to current changes. The gas

concentration is usually proportional to the current generated, so it is possible to convert the current change into the gas concentration.

3. Results and Discussion

3.1. LIG Characterization

We performed a series of characterizations of LIG. Figure 2A shows that the electrode surface is porous, making it ideal for gas detection. This is due to the photothermal effect of the laser focusing on the PI surface, resulting in an instantaneous increase in local temperature, forming LIG accompanied by the release of gas products, thus forming a porous fluffy structure. We explored the influence of different laser power levels on the surface of the LIG electrode, and the results are shown in Figure S3. We selected the optimal power for subsequent experiments. Figure 2B shows a cross-section of the LIG electrode; it can be seen that the thickness of the LIG is about 50 μm . A Fourier transform infrared spectrometer (FT-IR) and XPS were used to characterize the composition changes during PI conversion to LIG. By comparing the infrared spectra before and after laser engraving (Figure 2C), it can be found that the typical characteristic peaks of PI almost disappear after the formation of LIG, including C=O (1711 cm^{-1}), C=C (1496 cm^{-1}), C-N (1368 cm^{-1}), and C-O (1231 cm^{-1}). It has been confirmed that during the conversion of PI to LIG, the high temperature generated by the laser destroys the C-N, C-O, and C=O bonds in the carbon precursor PI, and the remaining C atoms will rearrange into the graphite structure [41]. The results of the XPS total spectrum (Figure 2E) show that LIG has a higher content of C elements and a lower content of N and O elements. The XPS fitting peak results are shown in Figure 2D: C-C (284.8 eV), C-N (286 eV), C-O (286.6 eV), and C=O (289 eV). Using Raman spectroscopy to characterize LIG (Figure 2F), three typical characteristic peaks of graphene can be observed, marked on the figure, namely the D peak, G peak, and 2D peak. These parameters are closely related to the structure of graphene.

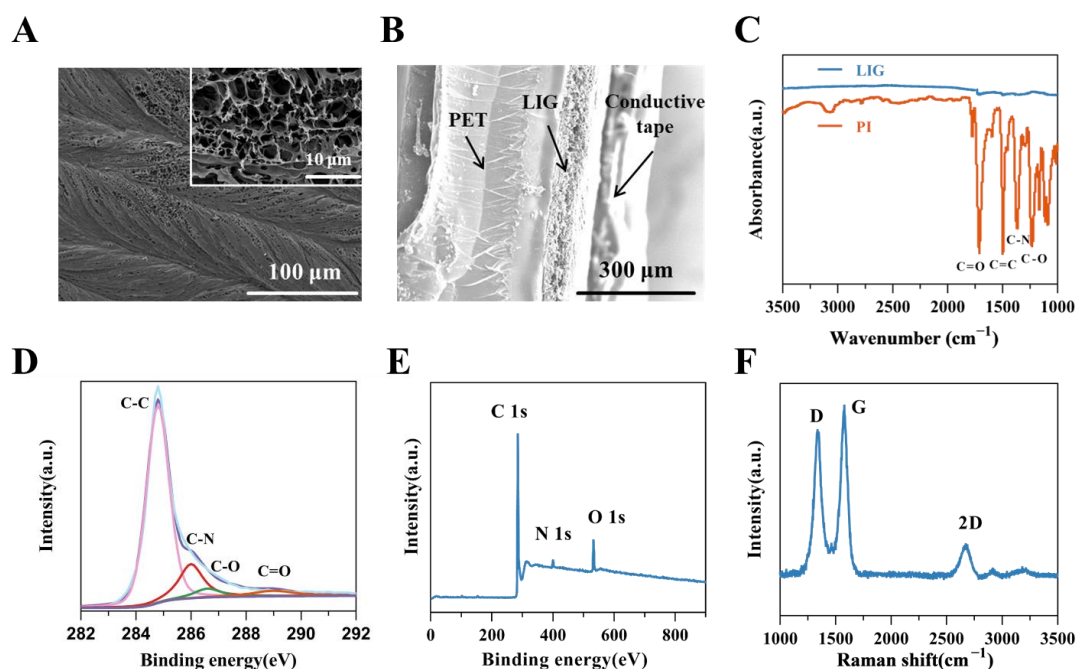


Figure 2. Characterization results of LIG electrode. (A) SEM image of the LIG; (B) SEM image of the cross-section of the LIG; (C) the infrared spectra of LIG and PI; and (D,E) are the XPS characterization diagrams of LIG; (F) Raman spectrum of LIG.

3.2. PEDOT:PSS Film and Bimetallic Nanocomposites Characterization

The structures and morphology of PEDOT:PSS films and Au-Pt nanocomposites were characterized by SEM. Figure 3A shows the surface morphology of the electrode after plating the PEDOT:PSS film, and it can be seen that the PEDOT:PSS film is uniformly decorated on the porous graphene surface. Figure 3B shows that after electrochemical deposition, the Au spikes are uniformly modified on the electrode surface, and the dendritic structure can be clearly seen from the enlarged part of the figure. In Figure 3C, it can be seen that the surface of the Au spikes is coated with spherical Pt, forming the Au-Pt nanocomposites. The coating of Pt on the surface of Au spikes may be due to the uneven distribution of current generated by the morphology of Au spikes, which provides a nucleation site for the subsequent deposition of Pt.

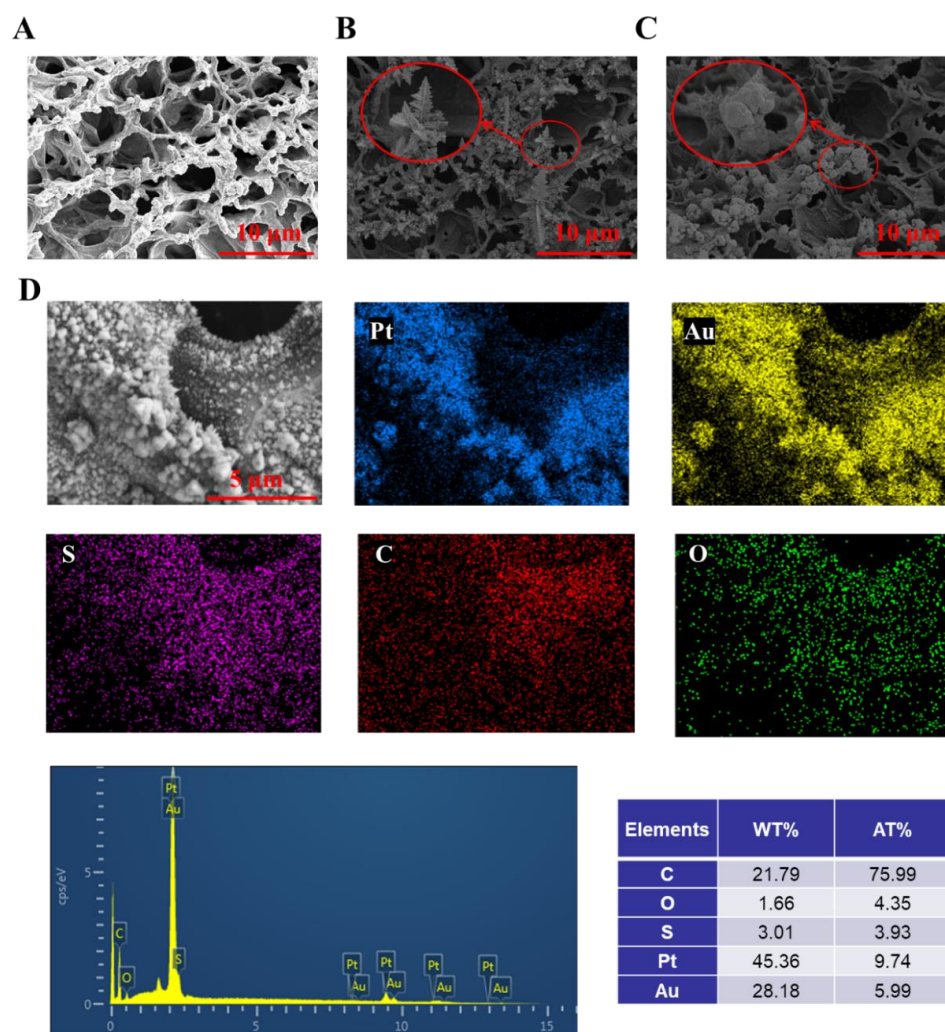


Figure 3. Characterization results of modified materials. (A) SEM image of PEDOT:PSS film; (B) SEM image of Au spikes; (C) SEM image of Au-Pt nanocomposites; (D) EDS spectrum and the corresponding elemental mapping of Pt, Au, S, C, and O.

The modified electrode was also analyzed by EDS, as shown in Figure 3D, and the characteristic peaks of Au, Pt, S, C, and O were observed. The C and O elements are derived from the graphene substrate and PEDOT:PSS film, and the S elements are derived from the PEDOT:PSS film. Focusing mainly on the Au and Pt elements, it can be seen that the distribution area of Pt (blue) and Au (yellow) is almost the same, which also confirms that Pt is wrapped in the surface of Au spikes. All in all, SEM and EDS analysis confirm the successful formation of PEDOT:PSS films and Au-Pt nanocomposites. The PEDOT:PSS

film is uniformly modified on the surface of the graphene substrate, which can increase the specific surface area and provide more active sites. Furthermore, the Au spikes, as the substrate, provide a large surface area of non-uniform current distribution for the subsequent growth of Pt and increase the loading capacity of Pt.

Figure 4A shows the XPS spectrum of PEDOT:PSS/Au-Pt nanocomposites/LIG, which confirms the presence of Pt, Au, and S elements. This is also consistent with the EDS results. Figure 4B,C are the survey scans of Pt 4f and Au 4f, respectively. It can be seen from the figure that the binding energy of Pt 4f_{7/2}, Pt 4f_{5/2}, Au 4f_{7/2}, and Au 4f_{5/2} is about 71.48 eV, 74.68 eV, 84.28 eV, and 87.98 eV, respectively. For HRTEM analysis, the PEDOT:PSS/Au-Pt nanocomposites modified on the electrode were scraped off, dispersed in 50% ethanol solution, then drip-coated on the surface of the copper mesh, and the samples were characterized by HRTEM. As shown in Figure 4D, it can be seen that the lattice spacing is about 0.23 nm, which corresponds to the (111) plane of the Au-Pt nanocomposites.

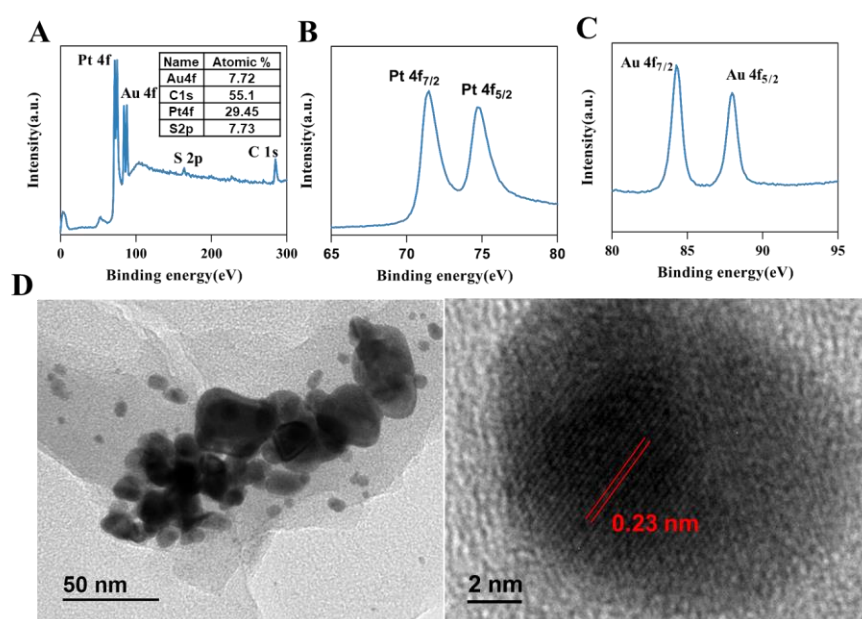


Figure 4. (A) XPS spectra of PEDOT:PSS/Au-Pt nanocomposites/LIG; (B) the survey scans of Pt 4f; (C) the survey scans of Au 4f; (D) HRTEM image of PEDOT:PSS/Au-Pt nanocomposites.

3.3. Gas Detection Performance of Methane Sensor

Cyclic Voltammetry (CV) scanning was performed on each modified electrode in potassium ferricyanide solution to investigate the electrochemical performance of the modified PEDOT:PSS film and Au-Pt nanocomposites on the electrode surface, as shown in Figure 5A. It can be seen that the current is significantly improved after modification of the PEDOT:PSS film and Au-Pt nanocomposites. To determine the applied voltage in the chronoamperometry method, CV scanning was performed on the electrode when standard air and methane were passed, respectively. The results are shown in Figure 5B. It can be seen that there is an oxidation peak of methane at 1 V. The applied voltage of 1 V is used for subsequent chronoamperometry. The influence of gas flow rate on the gas signal response is also explored. As shown in Figure 5C, too large or too small of a flow rate will lead to a smaller signal response, so it is necessary to set an appropriate gas flow rate to ensure efficient gas contact with the electrode. An excessive flow rate may cause the residence time of gas on the sensor surface to be too short, and the contact time between the gas and the sensing surface is not enough to allow the gas to fully diffuse into the ionic liquid, thus affecting the reaction and sensor response. At the same time, the high flow rate of gas may cause dynamic changes in the ionic liquid, affecting the sensing response. Too

small a flow rate will cause the gas mass transfer rate to slow down, and the low-speed gas cannot overcome the viscous resistance of the electrolyte, thus limiting the sensor response. According to the results in the figure, the gas flow rate of 200 sccm was used for subsequent measurement in the experiment. The gas response after modification of the PEDOT:PSS film was also compared, as shown in Figure 5D. It can be seen that the sensor response after modifying the PEDOT:PSS film was significantly enhanced, which may be due to the PEDOT:PSS film providing more active sites, improving the detection sensitivity, and reducing the electrochemical impedance so that the sensor has excellent conductance and electron transfer characteristics.

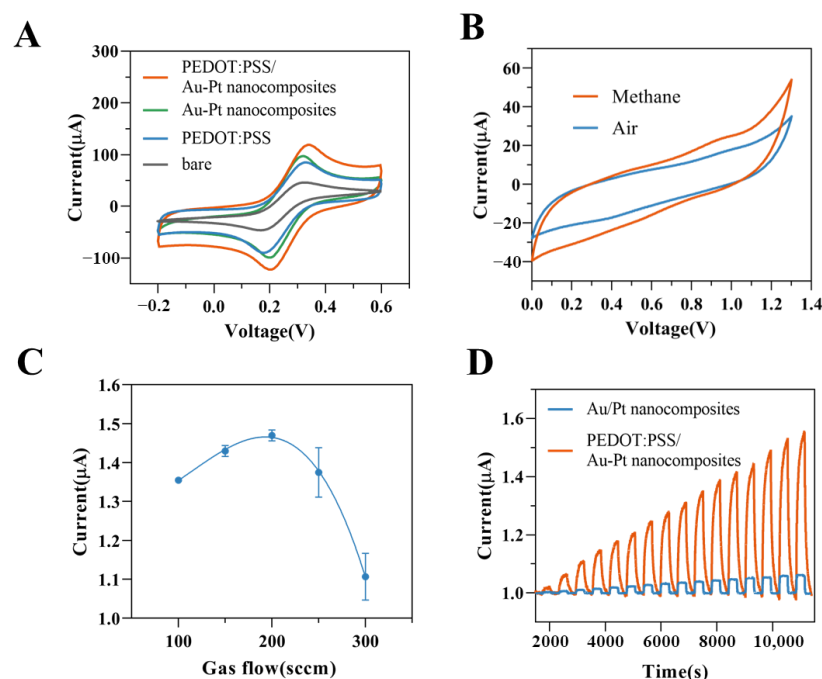


Figure 5. (A) CV test curve after different modification materials; (B) CV curve of the electrode in standard air or methane; (C) effect of gas flow rate on sensor response; (D) comparison diagram of the electrodes with or without the PEDOT:PSS film.

Before testing, the sensor was placed in standard air, and 1 V voltage was applied to stabilize the baseline. Then, different concentrations of methane gas and standard air are alternately fed into the gas chamber. Figure 6A,B show the actual results of the detection of methane at 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 200, 300, 400, and 500 ppm concentrations as well as the linear fitting diagram. It can be seen that the sensor has a good linear range. Due to the low concentration of methane in human exhaled gas, a further gradient detection was made for the low concentration of methane gas, and the gradient was set to 2, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, and 80 ppm, as shown in Figure 6C,D. It can be seen from the fitting result diagram that the R^2 value is 0.9994, which shows good linearity, and the sensitivity is 6.658 nA/ppm. From these results, it can be seen that the sensors have good linearity in the range of 2–500 ppm and have a low detection limit (LOD). The LOD was evaluated using the blank standard deviation method. Before exposure to CH_4 , take the average value of 10 consecutive data points and calculate the standard deviation according to Formula (1). The 10 consecutive data points used to calculate LOD are shown in the inner illustration of Figure 5D. According to Formula (1), $\sigma = 0.00079$.

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (x_i - \mu)^2}{N}} \quad (1)$$

where σ is the standard deviation, x_i is the i -th data point, μ is the mean of the data, and N is the number of data points. The LOD is defined as Formula (2):

$$LOD = \frac{3\sigma}{k} \quad (2)$$

where k is the slope of the standard curve. And the LOD was calculated to be about 0.36 ppm, according to Formula (2).

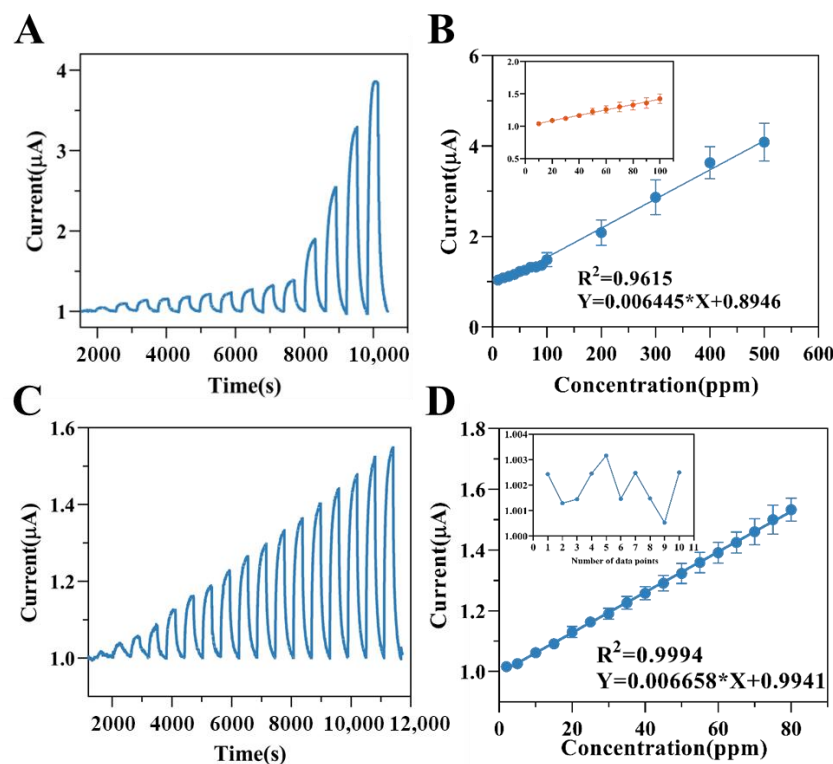


Figure 6. Gas detection results (A,B) show the actual results and fitting graphs of methane detection at 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 200, 300, 400, and 500 ppm, respectively. (C,D) show the actual results and fitting graphs of methane detection at 2, 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, and 80 ppm, respectively. The inner illustration of (D) shows the 10 consecutive data points of the blank baseline.

Comparing our work with the performance of some electrochemical methane sensors reported in the previous literature, the results are shown in Table 1. It can be seen that our LIG methane sensor has advantages in LOD and room-temperature sensing.

Table 1. Performance comparison of electrochemical methane sensors.

Sensing Platform	Operating Temperature (°C)	Sensitivity	LOD (ppm)	Reference
PbNPs/LIG	RT ¹	0.55 μ A/ppm	10	[42]
ITO/Pt/YSZ	550	0.675 mV/ppm	25	[43]
Pt	RT ¹	-	3000	[44]
Four electrodes: La _{0.87} Sr _{0.13} CrO ₃ , ITO, Au, Pt	450–600	-	1000	[45]
PEDOT:PSS	RT ¹	0.5 pA/ppm	600	[46]
PEDOT:PSS/Au-Pt nanocomposite/LIG	RT ¹	6.658 nA/ppm	0.36	This work

¹ RT: Room temperature.

The repeatability, response time and recovery time, and difference in electrochemical gas sensors are also key parameters that characterize the performance of the sensor. In this study, the repeatability of the sensor was assessed by alternating pumping of the standard air and 50 or 200 ppm of methane for six cycles, and each cycle lasted for 20 min, and the current response is shown in Figure 7A. As can be seen from the figure, at a concentration of 50 ppm or 200 ppm, the response signals of the sensor change little in these six cycles, which indicates that the LIG sensor has good stability. We calculated the response time and recovery time of the sensor, and the results are shown in Figure 7B. The time required for the sensor signal to rise from baseline to 90% of the stable state was taken as the response time of the sensor, and the time required for the sensor to drop from the stable state to 10% of the stable state was taken as the recovery time of the sensor. It can be seen from the figure that the response time is about 200 s, and the recovery time is about 160 s. The difference between the electrodes was evaluated by measuring the response of different electrodes to the same gradient of methane gas, and the results are shown in Figure 7C. Figure 7C is the fitting diagram of the response results of four different batches of sensors, and it can be seen that the difference between the electrodes is small. In order to evaluate the time stability of the prepared RTIL-LIG electrode system, the internal resistance and charge transfer impedance of the electrode system were measured by electrochemical impedance spectroscopy (EIS) at regular intervals, and the results are shown in Figure 7D. The original EIS data graph is shown in Figure S4. The time interval in the area with the blue background is 1 h, and the time interval in the area with the yellow background is 6 h. It can be seen that the internal resistance of the electrode system is small, and the resistance of the electrode system is relatively stable within 48 h. In summary, the LIG electrode has good repeatability, a fast response time and recovery time, and there is little difference between different electrodes.

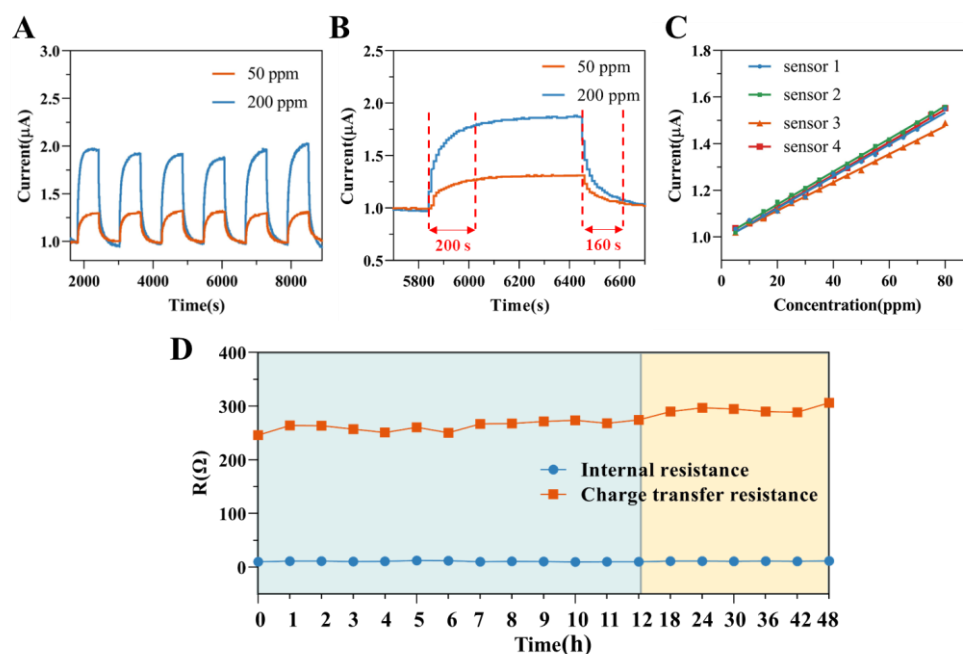


Figure 7. Sensor performance. (A) Sensor detection stability; (B) response time and recovery time; (C) differences between sensors; (D) resistance versus time (the time interval in the area with the blue background is 1 h, and in the area with the yellow background it is 6 h).

4. Conclusions

In this paper, LIG-based sensors modified with PEDOT:PSS films and Au-Pt nanocomposites for high-sensitivity methane gas detection are presented for the first time. LIG

technology enables low-cost batch manufacturing of electrodes. PEDOT:PSS films greatly reduce electrode conductance and improve ion transfer characteristics. At the same time, the synergistic effect of Au-Pt nanocomposites further increases the catalytic performance of the sensor and improves the sensitivity of the sensor. The sensor has a wide linear range, low LOD, and good repeatability. The sensor is easy to manufacture, low cost, operates at room temperature, has high sensitivity, has a low detection limit, and is expected to be used for methane breath detection in the future.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/chemosensors13010003/s1>: Figure S1: Electrode and gas chamber diagram. (A) Figure of LIG electrode; (B) figure of LIG electrode pasted to PCB board; (C) physical diagram and design drawing of gas chamber.; Figure S2: The connection diagram of each part.; Figure S3: Effect of different laser power levels on LIG electrode surface: (A) underpower; (B) over-power; (C) suitable power.; Figure S4: The original EIS data graph.

Author Contributions: Conceptualization, J.S., Z.H., S.M. and H.X.; methodology, J.S., X.Z., W.Z. and W.Y. validation, X.Z., H.X., S.M. and W.Y.; formal analysis, J.S., S.M. and W.Z.; investigation, J.S., Z.H. and W.Z.; resources, X.Z., W.Y. and H.X.; data curation, J.S. and Z.H.; writing—original draft preparation, J.S.; writing—review and editing, J.S., Z.H. and H.W.; supervision, H.W. All authors have read and agreed to the published version of this manuscript.

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Data Availability Statement: The data presented in this study are available on request from the corresponding author.

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Conflicts of Interest: Author W.Z. was employed by the Evaluation Center of Medical Device of Zhejiang Province. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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