Electrode separation optimization of triple-electrode carbon nanotube sensor

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Abstract: For triple-electrode carbon nanotube sensor, the electrode separation between electrodes is one of the key factors to influence the accuracy of detection. It is very difficult to decide the sensors′ electrode separations when detecting the multi-component gas mixture using sensor array. A electrode separation optimization method for triple-electrode carbon nanotube gas sensor was presented. The method is based on particle swarm optimization (PSO) and includes following procedures: designing electrode separation, constructing sensor array using multi-sensor with different electrode separations, building data base including electrode separation and detection ionic current, creating quantitative analysis model of mixed gas, and optimizing electrode separation. The NO and SO_2 gas mixtures were detected by multi-group sensor array, which were composed of three carbon nanotube sensors with different electrode separations. The electrode separations of three sensors was optimized using above-mentioned method. The experimental results show that the proposed method is able to select the optimal distances between electrodes effectively and the sensor with optimized electrode separation achieve higher detection accuracy.

Key words: carbon nanotube sensor; electrode separation; optimization; particle swarm optimization

三电极碳纳米管传感器的极间距优化

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摘　要： 三电极碳纳米管传感器各电极之间的间距大小是影响检测精度的关键因素之一。在用传感器阵列检测多组分气体混合物时，各传感器的极间距很难确定。为三电极碳纳米管气体传感器提出一种基于粒子群算法(PSO)的极间距优化方法。该方法包括设计极间距、组建由不同极间距的多个传感器组成的传感器阵列、建立包括极间距及检测离子电流的数据库、建立混合气体定量分析模型及极间距优化等步骤。采用多组由不同极间距的三个碳纳米管传感器构成的传感器阵列对 NO 和 SO_2 混

合气体进行测量,其中各传感器的极间距均采用上述方法优化。实验结果显示,上述极间距优化方法能够有效地选择电极之间的最佳间距,优化极间距后的传感器也获得了更高的检测灵敏度。

0 Introduction

In the last decade, with the continuous development of carbon nanotechnology, gas sensor, temperature sensor, and humidity sensors based on carbon nanotube sensing element are emerging[1-5]. A micro-nano triple-electrode carbon nanotube film sensor was publicized in a Chinese patent, which can be used for gas, temperature and humidity detection. The triple-electrode sensor has three electrodes isolated from each other by insulating pillars; the carbon nanotube film is on the surface profile of the first electrode [6]. Meanwhile, concentration detection method for multi-component gas mixture was proposed using the triple-electrode carbon nanotube film sensor array[7].

Because of the unique advantages of low voltage, small overall size and so on, the carbon nanotube sensor has broad application prospects in biological, chemical, mechanical, aerospace and many other fields. However, the existing triple-electrode carbon nanotube sensors are working in particular electrode separations. To different test gases, there is not targeted structure yet to obtain higher detection sensitivity. In addition, it needs to be clarified how to decide the distances between every two electrodes for every sensors in sensor array, which is used for mixed gas detection. Therefore, in order to improve the detection sensitivity, and promote the sensor applications, it is urgent to find a way to optimize the electrode separation for triple-electrode carbon nanotube sensor.

1 Electrode separation optimization procedure

The presented electrode separation optimization method is shown in Fig.1. It includs designing the electrode separations, constructing sensor array, detecting the gas samples, building data base, creating the quantitative analysis model, optimizing the electrode separation.

Designing the electrode separations needs to design the possible separations between cathode and extracting electrode, extracting electrode and collecting electrode. And the combinations of above two separations also needed to be considered. The possible separtions come from previous experimental data and some rational interpolations. All triple-electrode carbon nanotube film sensors uesed here is fabricated according the designed electrode separations. Three triple-electrode carbon nanotube film sensors are selected to construct sensor array. The gas samples are prepared using standard gas and detect using constructed sensor array. The detected experimental data are collected to build data base, used for creating the quantitative analysis model. The optimal electrode separation is determined when the minimal relative error analyzed by the quantitative analysis model appeared.
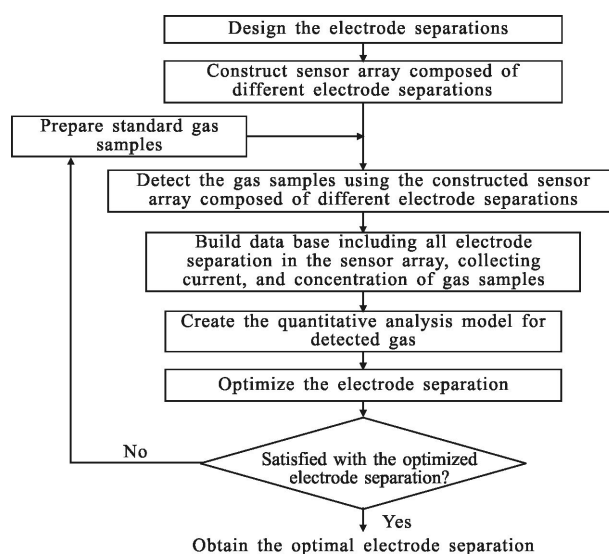

Fig.1 Procedure of optimizing the electrode separation of triple-electrode sensor

2 Experiment details

2.1 Triple-electrode carbon nanotube sensor

The electrode structure of the triple-electrode carbon nanotube sensor is shown in Fig.2. This structure is firstly presented by Zhang Yong[7]. The multiwalled nanotube (MWNT) film is used as the cathode, which is grown by thermal chemical vapor deposition (TCVD) on one side of the Si cathode covered by a Au film; the silicon slices are cut as the substrates of cathode, extrating electrode and collecting electrode respectively; polyester film is cut as the insulating strips between electrodes which corresponds to various electrode separation of the device[8]. The voltage between cathode and extracting electrode, cathode and collecting electrode are U_e and U_c respectively. U_e is higher than U_c and lower than 150 V. Non-self-sustaining discharge current is measured directly to calculate the concentration of detected gas.
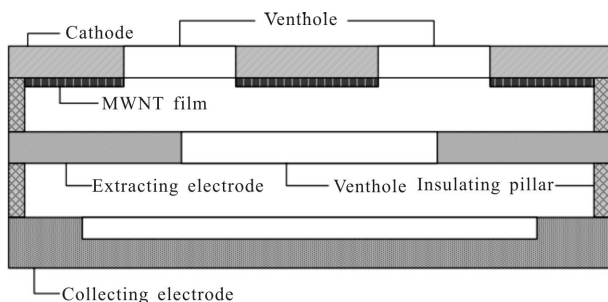
Fig.2 Triple-electrode sensor structure

2.2 Designing electrode separation

The sensor array used for detection is composed of three triple-electrode nanotube sensors named 1#, 2# and 3# sensor. The 1# and 2# sensors are used to measure the concentration of gas, 3# sensor is used for temperature measurement. For 1# sensor, the electrode separation of cathode and extracting electrode, extracting electrode and collecting electrode are d_{11} and d_{12}, respectively. Similarly, for 2# and 3# sensors, the electrode separations are d_{21} and d_{22}, d_{31} and d_{32}, respectively. The two electrode separations of one sensor could be equal or not. Twelve different groups

are designed for constructing sensor arrays (Tab.1). The designed separations are from 50 μm to 200 μm, including various combination of d_{i1} and d_{i2}.

Tab.1 Twelve sensor arrays with different electrode separations

Group no.	d_{11}/μm	d_{12}/μm	d_{21}/μm	d_{22}/μm	d_{31}/μm	d_{32}/μm
1	50	150	150	150	50	130
2	50	180	180	180	90	110
3	100	180	100	150	130	190
4	100	200	180	200	50	90
5	150	180	180	180	90	110
6	150	200	200	200	210	250
7	50	150	150	150	130	130
8	50	180	180	180	50	90
9	100	180	100	150	90	110
10	100	200	180	200	150	150
11	150	180	180	180	170	170
12	150	200	200	200	130	130

2.3 Designing and preparing mixed gas samples

The ratios of eight detected gas mixture samples are designed as Tab.2. All samples are prepared using standard gases by volume flow dynamic distribution method. A 16 −bit digital flow controller, manufactured by United States Alicat Scientific Company with an accuracy of ±1.0% of full scale, is used to control the volume flow of the gas components. All of the component gases are standard gas with 99.99% purity. The environment gas is N_2. The concentration range of component gases is from 500 ppm(1 ppm=10^{-6}) to 1 500 ppm.

The prepared gas samples are injected into clear and dry detection chamber and sealed. A vacuum pump is connected with the outlet of the detection chamber for pumping the detected gas out of it. The gas to be detected will be injected into the detection chamber for next detection when the chamber is pumped to a low vacuum.

Tab.2 Eight gas mixture of SO₂ and NO

Gas	Concentration/ppm							
	1	2	3	4	5	6	7	8
SO_2	500	500	500	800	800	1 000	1 300	1 500
NO	500	800	1 000	800	1 100	800	500	1 500

2.4 Measuring and building the data base

The sealable detection chamber for containing the mixed gas is made of stainless steel and equipped with pressure detection. The detected environment temperature is from 60 ℃ to 80 ℃. The sensor array is placed in the detection chamber. The detection data is transferred to computer through an interface and wire bonded to electrodes. All the 8 gas mixture samples are detected using designed 12 sensor arrays in Tab.1 one by one. The non-self-sustaining discharge current I is measured at $0-150$ V U_e. The component concentrations of 8 mixed gas samples, electrode separations of every sensor in 12 sensor arrays, and the measured ionic current value I are all added into the data base for further research.

2.5 Creating mixed gas quantitative analysis model

Getting rid of the odd samples, and ionic current and couple data (d_{i1}, d_{i2}) as input, test component gas concentration as output, a Kernel Partial Least Square Regression (KPLSR) quantitative analysis model of mixed gas is created. The Gaussian kernel function is employed in the KPLSR. The parameter L and latent variable t are optimized by cross validation[9]. The analysis results of KPLSR model are evaluated by relative error. The detection data of $3^{\#}$ sensor is used to overcome the cross sensitivity of temperature.

2.6 Optimizing the electrode separation

To aim at the minimal relative error, PSO optimization algorithm is employed to calculate the optimal electrode separation. The PSO seeks the optimal solution through information transfer and information sharing among groups of individuals[10]:

$$V_i=\omega V_i+c_1\times rand()\times(pbest_i-X_i)+c_2\times rand()\times(gbest_i-X_i)\quad(1)$$

where i is particle, $i=1, 2, \cdots, M$; ω is the inertia weight factor; V_i is the speed of i-th particle; gbest represents the best position of the population; pbest represents the best position of this particle; rand () is random number between (0, 1); c_1 and c_2 are learning factor, $c_1=c_2$; X_i is the position of i-th particle.

$$X_i=X_i+V_i\quad(2)$$

Each particle is required to calculate the fitness value according to the objective function, and then, pbest and gbest are determined according to the fitness value. The speed and position of particles revise according to the formula (1) and (2).

3 Result and discussion

3.1 Measurement of ionic current value of gas mixture

All the 8 mixed gas samples in Tab.2 are detected respectively using 12 sensor arrays in Tab.1. For every measurement, the ionic current will be continuously collected two times, and the average value of the two collections is used for further study. As an example, the measured positive ionic current (Fig.3) shows the effect of different electrode separation on collecting ionic current. The concentration of gas mixture of SO₂ and NO is equal to 800 ppm, the gas temperature is 60 ℃, and the U_e is 120 V.
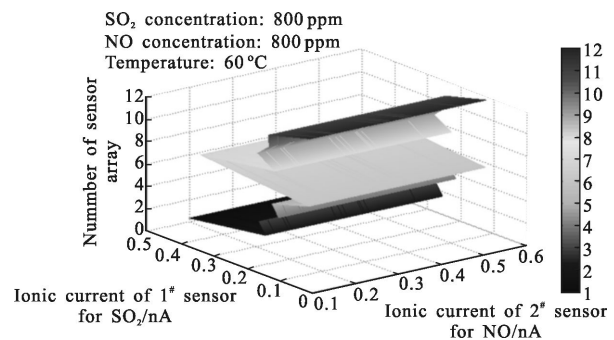

Fig.3 Triple-electrode sensor structure

Figure 3 shows that the collecting ionic current of SO₂ and NO sensor is different when detected use different sensor array at different electrode separation. The collecting ionic current of SO₂ sensor decreases from 0.48 nA to 0.09 nA when the electrode separation

increases from 50 μm to 150 μm. The collecting ionic current of NO sensor decreases from 0.58 nA to 0.16 nA when the electrode separation increase from 150 μm to 200 μm. At the same time, different combination of two sensors with same electrode separation almost does not affect the collecting ionic current.

3.2 Analysis results of KPLSR model

After 12 odd samples are eliminated, there are 84 valid data in total. The relative error of 84 detected data is analyzed by KPLSR model (Fig.4). In Fig.4, all the conducted detections are numbered in sequence. It indicates that the relative errors of some particular samples are smaller when these particular samples are detected by 5#, 6#, 11#, and 12# sensor arrays. The 5# and 11# sensors obtain the minimal relative errors. This reflected from the other side that the optimization of electrode separation is necessary.
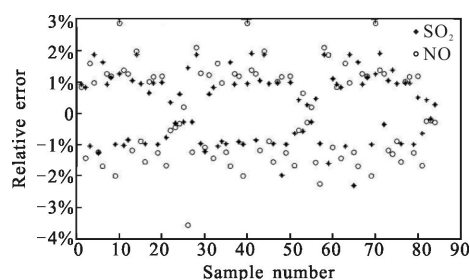

Fig.4 Relative error of detected gases analysis by KPLSR

3.3 Electrode separation optimization result

Set the number of particle swarm as 1, iterations as 12, the initial value and termination value of weight factor ω as 0.95 and 0.1, respectively; threshold value as 50−200; c_1 is 1.59, equal to c_2; iterate 100 times. The relative error of gas concentration decreases with increasing iterations. After 23 and 18 iterations, the optimal d_{11}, d_{12} and d_{21}, d_{22} are found. They are 153 μm, 186 μm and 185 μm, 186 μm respectively.

4　Conclusions

An optimization method for electrode separation of carbon nanotube sensor based on PSO is proposed.

Experimental results show that the collecting ionic current is different when the detection is conducted using different sensor array with different electrode separation. The collecting ionic current shows downward trend with the increase in electrode separation. The PSO is an effective method for electrode separation optimization. For mixed gas detection, it achieves highest detected accuracy at the optimal electrode separation.

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