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Research Paper

Portable device for acetone detection based on cataluminescence sensor utilizing wireless communication technique



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ABSTRACT

Portable sensor system was constructed for detection of acetone vapor based on the cataluminescence (CTL) theory. Excellent microspheres were prepared successfully which increased the contact probability with vapors. The instrument architecture, optimal experiment conditions and measurement characteristics were discussed in detail. The results showed the CTL sensor instrument exhibited excellent CTL properties including visible intensity, high signal/noise (S/N) values, short response time (within 2 s) and recovery time (within 3 s). The sensor instrument covered a linear test range from 5 ppm to 2500 ppm, while the sensor showed non-linearity when concentration ranged from 2500 ppm to 8000 ppm. Furthermore, the sensor system showed outstanding selectivity to acetone compared with other ten kinds of common vapors. Moreover, the CTL sensor also showed an excellent response for H₂S, the CTL performance of H₂S under different wavelength and temperature was also discussed. The excellent sensing and online detecting properties proved this sensor instrument to be an excellent candidate for real-time gas contaminant detection. Finally, the sensing control processes and monitoring software interface was also presented.

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1. Introduction

Harmful gas commonly exists in the air which can lead to short or long term physical injury [1]. Long term exposure to this kind of gas may cause serious physical injury, such as a wide range of feeling irritation or chronic injury. Harmful gas detection and identification is one of the most important topics in gas detection area, and it is still a huge challenge. Acetone, which is a typical harmful gas, has serious harm to our health and environment. The intermediate products (such as small organic matter) formation during the gas sensing process make it difficult to develop long-term stable gas sensors for detection of acetone [2–9]. In addition, increased demand for using of acetone in industrial leads to gaseous and liquid environmental pollution. Therefore, there is an urgent need to discover excellent method and apparatus for acetone determination.

After Breysse et al. [10] reported that the catalytic oxidation of carbon monoxide on the surface of thoria could produce a weak chemiluminescence (CL) emission, the concept of "catalumines-

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cence (CTL)" was established for the first time. The widely accepted CTL working principle is the formation of highly reactive peroxides in intermediates. When the CTL-based gas sensor is exposed to reducing gases, some chemical reactions between oxygen species and gas analytes will occur, resulting in the release of electrons back to materials and thereby increasing the response. During the past years, excellent performance CTL sensor with visible sensitivity, excellent selectivity, fast response and recovery had been developed [10–15]. For example, sensor device with 5×5 array, along with principal component analysis and hierarchical cluster analysis had been developed for detecting methane, ethane, propane, and butane hydrogen sulfide patterns [16], while the sensor system can not realized remote control and test data transmission, which is not appropriate for real time and remote detection situation. Wen Sha and co-workers [17] proposed a sensitive CTL sensor for the determination and recognition of benzene and toluene vapors. Although this technique is effective and sensitive for vapor detection, it still has inherent disadvantages for remote controlling and data transmission. The analytical intelligent CTL sensors for real time detection and identification of multiple harmful vapors are still very rare. With the development of wireless communication and wireless sensor network (WSN) technique, several sensor systems had been used for remote monitoring and control applications, such as meteorological monitoring [18], human health monitoring [19],

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road tunnel monitoring [20], phased array structural health monitoring [21], gas leak [22] and fire detection [23]. The excitement for wireless communication is motivated by the following advantages: long distance monitoring, energy saving [24], rapid respond, self organization and flexibility layout. As described above, portable CTL sensor system combined with wireless communication technique may be an excellent method for harmful gas monitoring.

As we know, sensing materials were one of the most important factors for the design of CTL sensor. Many efforts were devoted to synthesis of decorated materials with excellent CTL performance. The secondary nanomaterial deposited on the raw material may enhance the total porosity and surface area, which may provide additional locations for the interaction with vapor. For example, Weng et al. [25] have developed Mg-doped SnO₂ which used as CTL material for determination of acetone vapor. Song et al. [26] have used SnO₂/rGO film as the sensing material to detect the CTL performance of diethyl ether. Among various oxide semiconductor materials, TiO₂ [27,28] had been proved as one of the most suitable material for sensor fabrication because of its high reaction stability, low toxicity, and excellent degradation capacity for harmful substance. TiO₂ with high reaction surface areas and excellent catalytic properties had been synthesized successfully [29]. Recently, the immobilization of catalyst on the surface of magnetic materials can be easily separated from the solution with the help of an external magnet. The process can not only prevent agglomeration of the catalyst materials during recovery but also increase the durability of the catalyst. Deposition of noble metals such as Ag, Au and Pt on the surface of TiO₂ enhances the catalytic activity of TiO₂. The precious metal act as an electronic trap, can promote the interface of the charge transfer process [30].

In our study, CTL sensor system was selected as powerful tool for harmful gas detection. Ag-coated Fe₃O₄@TiO₂ microspheres were prepared successfully which increased the contact probability with harmful gas. Excellent material used as CTL sensor material for acetone vapor was studied during our experiment. High electronic emission generated during oxidization of acetone vapor was detected, which was the so-called cataluminescence (CTL) reaction. Embedded ARM11 was used for controlling the flow rate controller, temperature controller, optical filter and so on. Data transfer unit (DTU) was used for transferring sensing data to remote server. Due to the stable development of cloud computing and cloud storage, such as semantic security search [31], auditing protocols [32] and encryption technique [33], all collected data was outsourced into cloud for further analysis. Finally, the sensing control processes and monitoring software interface was also presented. The performance of this device was discussed meticulously in terms of short

response/recovery times, selectivity and stability for acetone vapor. The excellent reaction properties illustrated this CTL sensor system to be a promising candidate for detection and determination of gas contaminants.

2. Experimental and methods

2.1. Experiment reagents

All reagents used in our experiments were with high purity (>98.5%) analytical standard. Acetone, *n*-hexane, ethyl acetate, propane, carbon tetrachloride, H_2S , chloroform, methanol, ethanol, formaldehyde and propionaldehyde were purchased from Hefei Huilian chemical reagent company (Hefei, Anhui, China). Other chemicals were obtained from Aladdin Chemical Co., Ltd. (Shanghai, China). These reagents used in our experiments were without further purification.

2.2. Preparation of sensor materials

In our study, an efficient method for the preparation of Ag-coated $Fe_3O_4@TiO_2$ microspheres was presented. Tetra-butyl ortho-titanate (TBOT) was selected as the titanium source for $Fe_3O_4@TiO_2$ core/shell microstructure preparation by using vapor-thermal method [34–36]. During typical procedure, before transferred into a 20 mL beaker, Fe_3O_4 microstructure (30 mg) was dispersed in ethanol and TBOT. Then, it was placed into a 80 mL stainless steel automatic grinding machine with a Teflon liner. Distilled water was filled into the free space between Teflon liner and 20 mL beaker. The high pressure reactor was heated to 200 °C for more than 12 h after sealing. The distilled water evaporated and hydrolyzed the TBOT during the experiment. At last, when the high pressure reactor was washed with ethanol for three times and dried at 60 °C for 4 h.

 $Fe_3O_4@TiO_2$ microstructure (30 mg) were dispersed in a 0.2 m $Ag(NH_3)_2{}^+$ solution (40 mL), which stirred continuously for 2 h to ensure sufficiently adsorb of $Ag(NH_3)_2{}^+$ microstructure. The solution was exposed to UV high-pressure radiation (300 W) at 20 °C for 5 h. Finally, the materials were separated magnetically, washed, and dried at 60 °C for 5 h.

Fig. 1 (a) showed scanning electron microscopy (SEM) photograph of the Ag-coated $Fe_3O_4@TiO_2$ microspheres, it can be seen that Ag microparticles conglutinated to the surface of $Fe_3O_4@TiO_2$ microstructures. In order to get more information of the sample structure, further investigated by high-resolution (HR) transmis-

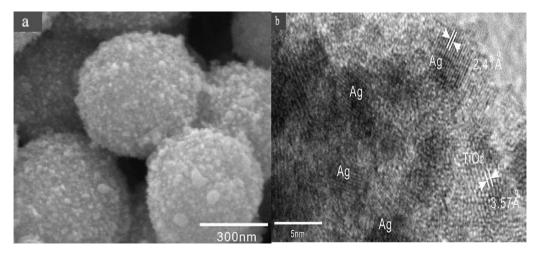


Fig. 1. (a) The SEM photograph of the Ag-coated Fe₃O₄@TiO₂ microspheres; (b) the high-resolution (HR) TEM photograph of the Ag-coated Fe₃O₄@TiO₂ microspheres.

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