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Preparation of a Nanowire-Structured Polyaniline Composite and Gas Sensitivity Studies

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Abstract: To obtain organic nanowire sensors with high sensitivity and rapid response times, based on the inducement effect of surfactants during in situ polymerization, nanostructured polyaniline composites are obtained by using a chemical oxidation method by adding a small amount of surfactant. A casting method is employed on interdigitated carbon electrodes. The gas sensitivity to a series of chemical vapors is examined at room temperature. The results indicate that polyaniline with regular nanowire structure is obtained when succinic acid is added. The gas sensitivity and response rates of a film with nanowire structure are much better than those of conventional polyaniline films produced by means of organic solution spin coating methods. The film described in this work shows good selectivity to trimethylamine and other related gases and, the reaction being reversible with the ues of high-purity nitrogen.

Keywords: inducement effect · nanostructures · polyaniline · sensors · surfactants

Introduction

At present study on controlling the nanostructure of conductive polymers is important for developing nanomaterials and nanoelectronic devices. Template, nontemplate, and nanofiber-seeding approaches are employed to prepare nanostructured conductive polymers.[1–6] Previously, inorganic aluminum oxide, zeolites with channels, and porous polymer membranes have been used as templates. Although the most consistent results were obtained by utilizing templates, such methods are still difficult to apply for the manufacture of sensors. The complicated procedures involved in forming and removing the templates leads to poor reproducibility and low yields. Furthermore, it is rather difficult to retain the ordered nanostructure after removing the templates, so it is meaningful to develop some nontemplate methods to obtain nanostructures for sensor applications.

In nontemplate methods, either polymerization takes place at the interface, or surfactants or polyelectrolytes are added for structural direction. By utilizing "nanofiber seeding", a polyaniline nanofiber has recently been obtained and characterized.[2] The experimental results indicate that nanostructured conductive polymers have potential applications in very sensitive sensors, such as gas sensors, as a result of their highly specific surface area and excellent channels for charge transmission.

Although a large number of studies on nanostructured conductive polymers have been reported, most of them still focus on the preparation of the materials and morphology characterization.[7–12] Only a few studies have dealt with functional properties of the devices; most devices that have been reported have poor sensitivity and slow response times.[13–16] Many researchers believe[1,2,13] that more sophisticated technologies should be invented to synthesize the materials and to fabricate devices to solve the aforementioned problems. Additionally, it is not easy to fully transfer functional properties of the materials to sensing devices, since the procedures are affected by many factors, such as device structure, electrode material, preparation approach, and transferral and the reappearance efficiency of polymers with ordered nanostructures.

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Recently, Huang et al.[17–19] reported research results on the properties of sensors based on nanostructured polyaniline prepared by using interface polymerization on interdigitated gold electrodes. Liu et al.[20] studied templateless assembly of molecularly aligned, conductive polymer nanowires. The promising results obtained encouraged us to develop an organic sensor, with high sensitivity and rapid response times, by using simple methods.

Gold electrodes have been widely used owing to the catalytic properties of the metal and its good contact to semiconductor substrates. However, besides the high cost, gold adheres poorly to many substrates; therefore, additional transition layers are needed. Considering potential commercialization, we employed screen-printed carbon electrodes. We prepared nanowire-structured polyaniline composite films by using the inducement effects of surfactants over the carbon electrodes to form gas sensors. The gas sensitivity to a series of vapors was investigated.

Results and Discussion

Preparation of a nanowire-structured polyaniline composite: Due to their highly specific surface area and excellent channels for charge transmission, nanostructured conductive polymers have potential applications in the production of gas sensors and other types of sensors. To obtain an organic nanowire sensor with high sensitivity and a rapid response time, we examined the effects of surfactants on the morphology of polyaniline composite films. The results are shown in Figure 1A–E.

Figure 1 shows that the morphologies of polyaniline composite films prepared in different media are quite different under similar synthesis conditions. This observation illustrates that surfactants have an important role in controlling the morphology of the films. The polyaniline composite films have relatively uniform nanowire structure when succinic acid is added. The diameter of the wires is about 50 nm, and the lengths are relatively heterogeneous, the longest wire is about 2 μm, while the shortest is a few hundred nanometers. Therefore, the inducement effect of succinic acid is notable and may be due to the two terminal carboxyl groups of the succinic acid molecule. In dilute solution, the succinic acid molecule unwinds from its random curly shape. Consequently, polyaniline tends to grow along the molecular chain of succinic acid in the inducement process of polyaniline in situ polymerization, thus forming polyaniline with a relatively regular nanowire structure. In comparison to the succinic acid molecule, the tartaric acid molecule has a similar structure but with a hydroxyl group. With the hydroxyl group it is very easy for this acid to form hydrogen bonds with the two terminal carboxyl groups, making it difficult for polyaniline to grow along the molecular chain of tartaric acid in the inducement process during polyaniline in situ polymerization. A distinct morphology of polyaniline is formed (see A and C in Figure 1).

The Fourier transform infrared (FTIR) spectra of the polyaniline composite film containing succinic acid are shown in Figure 2. The peaks at 3248, 1594, 1318, and 1171 $\mathrm{cm^{-1}}$ are attributed to N–H stretching, C=C stretching in the quinoid ring of the emeraldine base and emeraldine salt, C–N, and C=N stretching, respectively. These results il-

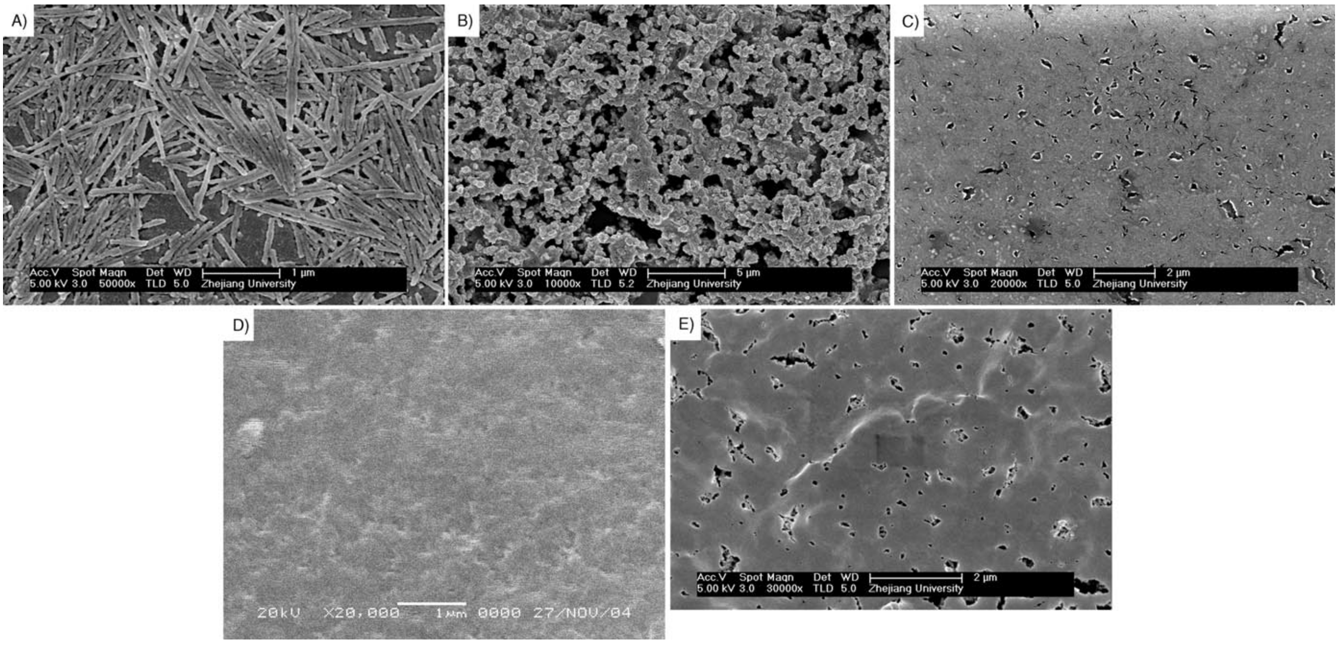


Figure 1. The morphology of polyaniline composite films prepared in different media under similar conditions: A) polyaniline–succinic acid; B) polyaniline–naphthalene sulfonic acid; C) polyaniline–tartaric acid; D) polyaniline–poly(*p*-styrenesulfonic acid), and E) polyaniline in hydrochloric acid media.

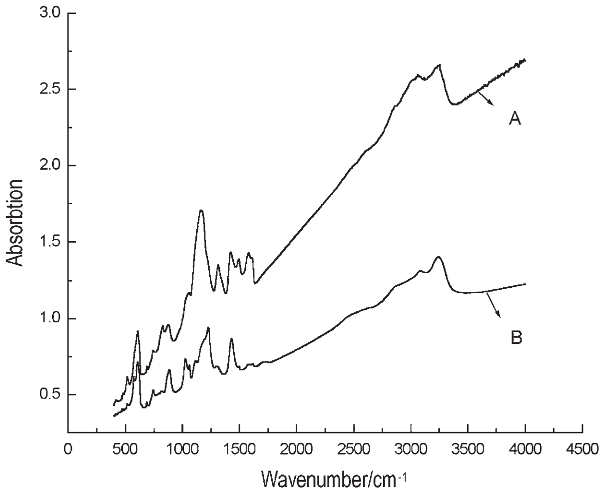


Figure 2. The FTIR spectra of polyaniline (A) and a polyaniline composite film containing succinic acid (B).

lustrate that this composite film contained polyaniline. Because the amount of succinic acid in our experiment was very small, only a slight difference can be observed in the FTIR spectra of polyaniline composite films with and without succinic acid.

Polyaniline can be easily prepared by using a chemical oxide method. Because a lot of papers have already reported the synthesis and characterization of polyaniline, the details are not discussed here.

The UV-visible spectrum of a polyaniline composite film containing succinic acid is shown in Figure 3B. Figure 3A shows the spectrum of a polyaniline film without succinic acid with peaks appearing at 211, 324, and 594 nm; these peaks are consistent with results from previous publications. From our experimental results, we observed peaks at 200, 385, and 757 nm in the UV-visible spectrum of a polyaniline composite film containing a small amount of succinic acid with a regular nanowire structure. A small blue shift and large red shifts are observed in comparison to the UV-visible spectrum of polyaniline without succinic acid. These shifts are possibly the result of the increase in the degree of orientation and the interaction between polyaniline and succinic acid.

The increase of orientation in polyaniline films containing succinic acid can also be seen from the results of X-ray diffraction (XRD) studies (see Figure 4). Figure 4 shows that the polyaniline film containing succinic acid has strong diffraction peaks at $2\theta = 6.08$, 16.74, 18.32, 33.72, and 51.90°. The polyaniline film formed from in situ polymerization without succinic acid has no diffraction peaks; it only has a wide, noncrystal peak at $2\theta = 25.62°$. This illustrates that the degree of orientation of a polyaniline composite film containing a small amount of succinic acid is much higher than that of a film without succinic acid.

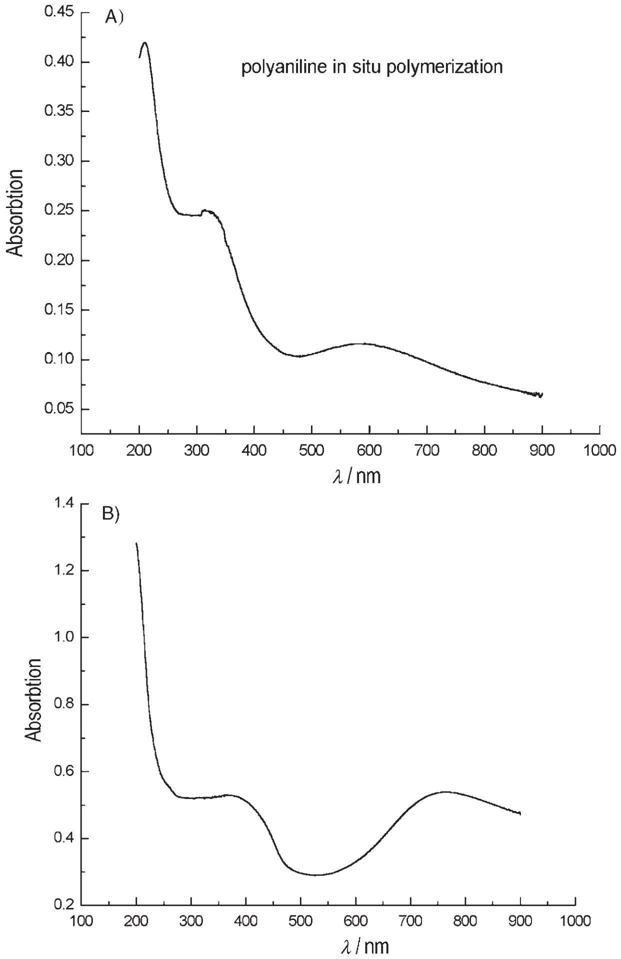


Figure 3. The UV/Vis spectra of polyaniline composite films A) without and B) with succinic acid.

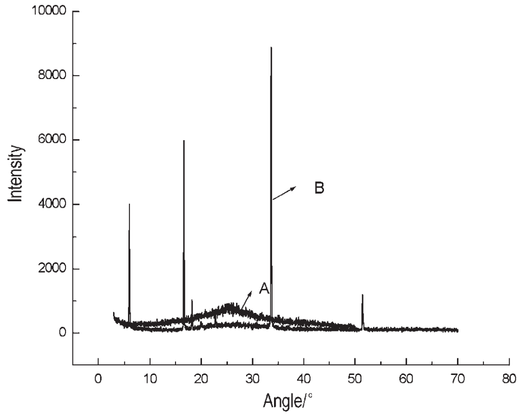
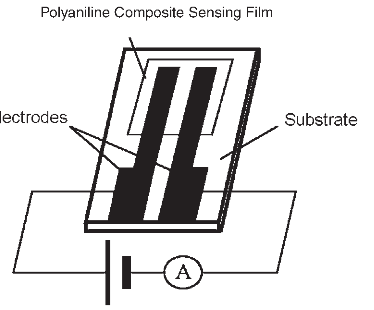


Figure 4. The XRD results of polyaniline (A) and a polyaniline composite film containing succinic acid (B).

Gas sensitivity of the polyaniline composite film with nanowire structure to chemical vapors: A diagram of the sensor used in our experiments is shown in Scheme 1.[19] It is known that the conductivity of polyaniline strongly depends



Scheme 1. The structure of the sensor used in the experiment.

on the doping and dedoping. Preparation of polyaniline is generally carried out in an acid media, so the product obtained is usually emeraldine salt, which is highly conductive. In the process of deprotonation, several redox forms of polyaniline are observed, for example leucoemeraldine base (fully reduced form), emeraldine base (half-oxidized form), conducting emeraldine salt (half-oxidized and protonated form), and pernigraniline base (fully oxidized form).[21] Moreover, polyaniline is an important electroactive material. When a voltage is applied, the absorption ability of the polyaniline film to vapors significantly increases. Therefore, the mechanism of its gas sensitivity to vapors is complex,[18] this mechanism includes doping, dedoping, reduction, swelling, conformational changes of the polymer chain and so on. In addition, crystal boundary effects may also contribute to the sensing mechanism.[22] The polyaniline composite has high resistance after deprotonation and is a multicrystal composite film. When it absorbs chemical vapors, the potential barrier between intercrystallite boundaries is overcome, and leads to an increase in the film conductivity. The actual sensing mechanism is still under investigation.

The main reasons for the gas sensitivity of the chemical sensors can be attributed to the interaction between the adsorbed gas molecules and the sensitive film, including both strong interactions (chemical bonds) and weak interactions (such as hydrogen bonding, van der Waals force, etc.). For a strongly interacting system, the recovery is generally very difficult. For a weakly interacting system, the recovery is relatively easier at room temperature with high-purity N_2. As long as the absorption ability of the sensitive film is improved from the viewpoint of materials design, the interaction between the adsorbed gas molecules and the sensitive film would be enhanced to improve the gas sensitivity. Accordingly, the polyaniline composite film with regular nanowire structure obtained in our experiments is anticipated to have excellent gas sensitivity owing to its morphology.

It is known from previous publications that most sensors made of polyaniline are based on the reversible reaction of acid/base, that is, the conductivity of polyaniline increases with an increasing amount of acid, and accordingly decreases with an increasing amount of base. In our experiments, we observed an interesting and unusual phenomenum; when the film of protonated polyaniline is exposed to a basic atmosphere, the response current of the film decreases sharply at the beginning, and then increases greatly. At this stage, the film can be easily desorbed with high-purity N_2, and can be utilized repeatedly. This film is not only sensitive to the amount of acid in the atmosphere (indicated by the current increase), but it is also sensitive to the ambience of the base (indicated by the current increase also). The essential difference is that the film cannot be desorbed with N_2 upon exposure to acid, it must be recovered in a basic atmosphere. This provided us with a new idea that the conductivity of the deprotonated polyaniline film increases with the quantity of absorbed basic gas. Based on this feature, a novel kind of gas sensor, which has good reproducibility, convenient operation, and could be recovered with N_2, may be developed. In this way, we believe that polyaniline can be fabricated into two kinds of sensors for basic atmospheres. The first kind of sensor is based on the reversible reaction of acid/base (the response current of the film increases on exposure to an acidic atmosphere, and decreases on exposure to a basic atmosphere); the other kind of sensor is based on a reversible process of polar vapors/N_2 (the response current of the film increases on exposure to a basic atmosphere, and decreases on exposure to a N_2 atmosphere). It has also been also found that the sensitivity of the latter sensor is much higher than that of the former one.

In our work, the first exposure of polyaniline to a basic atmosphere was performed as a pretreatment of the sensor. The conductivity of polyaniline increased with increasing exposure to the basic atmosphere, and the reversibility and reproducibility were very good.

Trimethylamine is an important organic amine that is produced in the metabolism process of organs of animal and protein. It is also an important toxic gas in the biological field and food industry. At present, it is usually determined by using sensors made of metal oxide. To improve its sensitivity and response speed, the polyaniline composite film with a regular nanowire structure was exposed to trimethylamine, and the dependence of the electrical response of the film on the exposure time is shown in Figure 5. These results show that the polyaniline composite film has a high sensitiv-

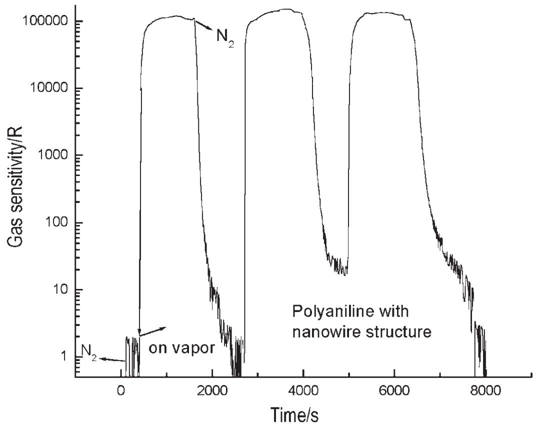


Figure 5. Electrical response versus exposure time of a polyaniline composite film containing succinic acid to trimethylamine (5.14×10^{-7} mol mL^{-1}).

ity and rapid response time to trimethylamine. The initial current of the film tested was about 10^{-10} A. When the film was exposed to the trimethylamine atmosphere (5.14×10^{-7} mol mL^{-1}), it only took about 100 s for the current to increase by 4–5 orders of magnitude.

Another important property of the sensor is its reversibility at room temperature. It has been found that absorbed vapors of the device can be recovered completely with high-purity N_2 at room temperature so the sensor can be used repeatedly. It can also be seen from the plot in Figure 5 that the response in the first and third circles returns to the original baseline. This result illustrates that the film sensor has a reasonable reproducibility.

Comparison tests were carried out with a polyaniline composite film containing a regular nanowire structure and one prepared by using a conventional spin-coating method (polyaniline dissolved in *N*-methylpyrrolidone). The response of both films to trimethylamine with a concentration of 5.14×10^{-7} mol mL^{-1} is shown in Figure 6. It can be seen that the organic film with a regular nanowire structure has a higher gas sensitivity and more rapid response time to trimethylamine than the polyaniline film prepared by using conventional spin-coating methods. This result illustrates that our simple approach is effective in obtaining an organic film that has a high gas sensitivity and rapid response time.

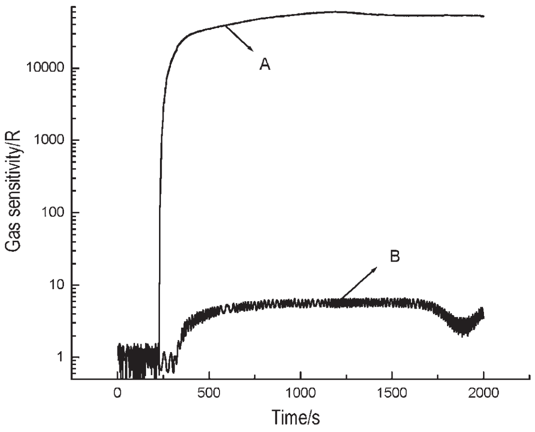


Figure 6. Electrical response versus exposure time of a polyaniline composite film with regular nanowire structure (A) and that prepared by using a conventional spin-coating method (polyaniline was dissolved in *N*-methylpyrrolidone) (B) to trimethylamine (5.14×10^{-7} mol mL^{-1}).

The effect of varying the concentration of trimethylamine on the response of the sensor was also examined. The results are shown in Figure 7. It was found that the magnitude of the response and the rate decreased with a decreasing concentration of trimethylamine. These results may offer some valuable information about quantitative analysis.

It is known that among the organic sensitive materials, phthalocyanine compounds, porphyrin compounds, and their derivatives are important sensitive materials that show good sensitivity to many gases.[22] It is believed that chemical modification of these compounds is also important to enhance their sensitivity and selectivity over a long period of time. In this work we obtained an organic-film device with good sensitivity by means of a simple preparation method instead of by chemical modification. This is a new preparation method and hence the devices needed testing. A series of tests were carried out utilizing some common organic films to make comparisons under the same conditions (response to trimethylamine with a concentration of 5.14×10^{-7} mol mL^{-1}). When we use zinc–phthalocyanine, –porphyin, –perylene, and their fluorinated derivative films instead of polyaniline, we observed results similar to those reported in previous studies.[23] The results showed relatively low sensitivity and slow response rates. This illustrates that it is feasible to improve the response rate and sensitivity of a sensor by using a simple method. The response of different sensing materials to trimethylamine are compared in Figure 8.

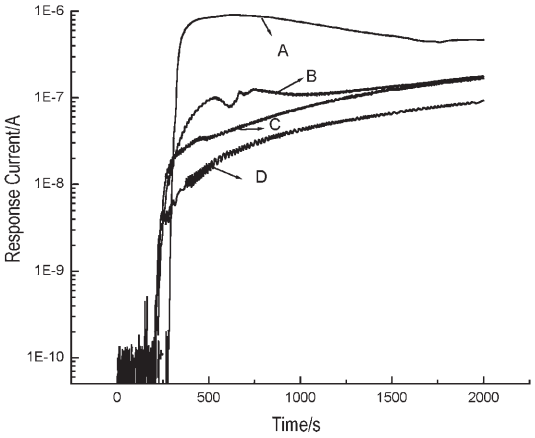


Figure 7. Effect of trimethylamine concentration (A: 5.14×10^{-7}; B: 2.57×10^{-7}; C: 1.28×10^{-7}; and D: 6.42×10^{-8} mol mL^{-1}) on the gas-sensing response.

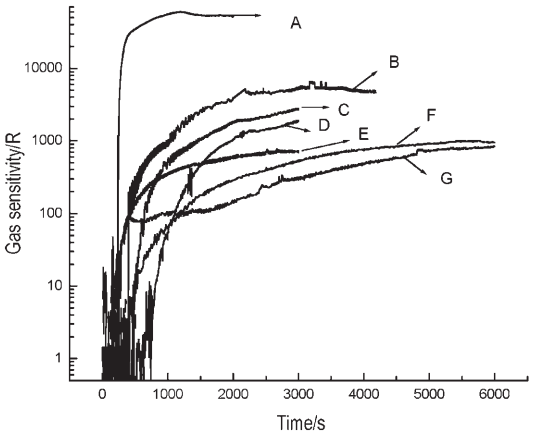


Figure 8. Electrical response versus exposure time for different common sensitive materials (A: polyaniline with nanowire structure; B: zinc–phthalocyanine; C: fluorinated biphenylperylenediimide; D: biphenylperylenediimide; E: fluorinated zinc–phthalocyanine; F: fluorinated zinc–tetraphenylporphyin; and G: zinc–tetraphenylporphyin).

The results illustrated here show that the gas sensitivity and response rate of the polyaniline composite film with a regular nanowire structure obtained in our experiments are much better than those of common organic sensitive materials and their modified compounds. This is probably due to the fact that this polyaniline composite film has a high specific surface area, which allows for easy adsorption of vapors. This material could also be used in other sensors, such as biosensors.

Selectivity of the polyaniline composite film with nanowire structure to chemical vapors: Unlike enzyme sensors, chemical sensors do not have particular sensitivities to certain gases. The identification of different gases may require sensor arrays and an artificial neural network. For this reason, the gas sensitivity of the polyaniline composite film to a series of vapors (trimethylamine, triethylamine, ammonia, and toluene) was examined. A blank comparison test was also carried out by injection of H_2O instead of trimethylamine to determine the effect of moisture on the sensor response. The results are shown in Figure 9. A slight difference in the gas sensitivity and response rate to trimethylamine, triethylamine, and ammonia is evident, but it is difficult to distinguish the three gases from each other. However, the gas sensitivity and response rates to trimethylamine, triethylamine, and ammonia are much better than those to toluene. The effect of moisture on the gas sensitivity is very small though the baseline shifts a little. The influence of moisture in the response of trimethylamine is negligible.

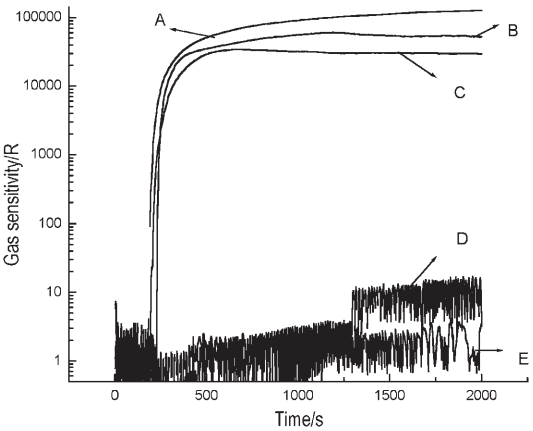


Figure 9. The gas sensitivity of a polyaniline composite film with nanowire structure to a series of vapors (A: triethylamine; B: trimethylamine; C: ammonia; D: H_2O; and E: toluene) of concentration 5.14×10^{-7} mol mL^{-1}.

The polyaniline composite film with a regular nanowire structure has good gas sensitivity and can operate at room temperature repeatedly; therefore, it is potentially useful for a number of applications. Owing to the length of the wires, the regular nanowire structure may be used as a probe for wound healing, biosensors, electromagnetic shielding devices, and stealth materials.

Conclusion

In summary, a polyaniline film with regular nanowire structure has been prepared based on an inducement effect of surfactants during in situ polymerization. The film shows good sensitivity and a rapid response to trimethylamine. It is easily recovered with N_2. The film has good potential applications in the area of sensor development. The method used is simple, feasible, and suitable for mass production at low cost.

Experimental Section

Materials: Aniline (AR), ammonium peroxydisulfate (AR), succinic acid (CP), tartaric acid (CP), poly(*p*-styrenesulfonic acid), naphthalene sulfonic acid (CP), and hydrochloric acid (AR) are commercially available. Aniline was freshly distilled in vacuum prior to polymerization. Deionized filtered water was used in all studies.

The screen-printed electrode was deposited on polypropylene with a thickness of 0.5 mm. The gap and length of the interdigital carbon electrodes was 0.5 and 1 mm, respectively.

Preparation of the sensing film: The coating mixture was prepared by adding an equal amount of aniline and ammonium peroxydisulfate to the hydrochloride solution, and a certain amount of the succinic acid (CP), tartaric acid (CP), poly(p-styrenesulfonic acid), naphthalene sulfonic acid (CP) (about 0.4 % weight in our experiments) was added to the mixture, which was then dispersed with ultrasonification. The mixture was then coated on the interdigital carbon electrodes, and finally dried for 96 h at room temperature (20–25 °). The color of the film changed from colorless to dark blue.

Pretreatment of the polyaniline film: The device was put into an airproof test box (2.5 L), and desorbed with high-purity N_2. A certain amount of trimethylamine or other base volatile solvent (about 0.2 mL) was injected into the test chamber with a syringe for deprotonation, and then desorbed with high-purity N_2.

Gas sensitivity characterization of the sensor: The characterization of the sensor's gas sensitivity to vapors was the same as with the reference sample.[23,24] The details of the procedure are as follows: the device was put into an airproof test box (2.5 L) that was connected to a vacuum and high-purity N_2 system with three-way valves. While 10 V DC voltage was applied on the interdigital electrodes, the DC response was measured by using a Keithley 236 Source Measure Unit. The DC current was measured continuously under a N_2 atmosphere with arbitrary time intervals, such as 1 s, to evaluate the programmed detection time. The current value was recorded automatically by an IBM compatible computer. The test box was flushed with high-purity N_2 repeatedly until the current slowly reached a steady value. At this moment, a certain amount of trimethylamine or other volatile solvent (about 0.2 mL) was injected into the test chamber with a syringe. A definition of the gas sensitivity (R) of the film is the ratio of I_{gas}/I_{N2}, in which, I_{gas} and I_{N2} represent the responsive currents of the sensor on exposure to measured gas and N_2, respectively.

Morphology observations with scanning electron microscopy (SEM): SEM was performed with SIRION field-emission and JSM-5510 microscopes. The polyaniline composite film was polymerized through an in situ method on a glass plate, dried at room temperature, then washed with deionized filtered water repeatedly. The surface of the film was then dried, and sputtered with thin layers of gold for SEM analysis.

X-ray diffraction (XRD) characterization: XRD patterns were recorded on a Rigaku D/max 2550 Pe diffractometer, with a rotating anode X-ray generator at 40 kV and 300 mA, and with $Cu_{K\alpha}$ monochromatic radiation. The polyaniline composite film was polymerized through an in situ method on a glass plate, dried at room temperature, washed with deionized filtered water repeatedly, and then dried for characterization.

FTIR spectra measurement: The FTIR spectra were carried out on silicon disks, and recorded on an IFS 66 VS^{-1} FTIR spectrometer (Bruker). Polyaniline was coated on the silicon disks through in situ polymerization, dried at room temperature, washed with deionized filtered water repeatedly, and then dried for measurement.

UV/Vis absorption measurement: UV/Vis absorption was recorded by using a CARY Bio100 spectrophotometer. Polyaniline was coated on the quartz glass through in situ polymerization, dried at room temperature, washed with deionized filtered water repeatedly, and then dried for measurement.

Acknowledgements

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