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pH-controlled silicon nanowires fluorescence switch

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ABSTRACT

Covalently immobilizing photoinduced electronic transfer (PET) fluorophore 3-[N, N-bis(9-anthrylmethyl)amino]-propyltriethoxysilane (DiAN) on the surface of silicon nanowires (SiNWs) resulted a SiNWs-based fluorescence switch. This fluorescence switch is operated by adjustment of the acidity of the environment and exhibits sensitive response to pH at the range from 8 to 10. Such response is attributed to the effect of pH on the PET process. The successful combination of logic switch and SiNWs provides a rational approach to assemble different logic molecules on SiNWs for realization of miniaturization and modularization of switches and logic devices.

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As a prerequisite of developing molecular computer, molecular logic switches have to be extensively studied. Accordingly, various logic switches have been simulated and the operation of NOT, AND, INH, half-adder, etc. have been realized [1–15]. However, most of present molecular logic switches were operated in solution system. In order to better control the devices, it is essential to graft the logic molecules on a solid surface [16–22]. In recent years, some logic molecules have been immobilized onto the surface of Au, glass and nanoparticles and exhibited some interesting and exciting properties [23–27]. While, the miniaturization and modularization design of logic devices are significant to realize molecular computer. One-dimension nanowires are an ideal substrate to graft logic molecule to realize the miniaturization and modularization of design due to its small volume and easy integration. Among these 1D nanomaterials, silicon nanowires (SiNWs) are particularly favorable to be utilized as building block to fabricate switch based on its compatibility with conventional silicon technology [28,29]. Up to now, the majority of the SiNWs-based switches are based on electronic or electrochemical method [30,31]. It is well known that the optical signal is very convenient for transfer of logic signal without the need of wires and two-electrode configuration [32,33], which facilitates the communication between various logic switches and increases the integration. Therefore, it is worthwhile to design and configure the optical logic switch with SiNWs. In this letter, photoinduced electronic-transfer fluorophore 3-[N,N-bis(9-anthrylmethyl)amino]-propyltriethoxysilane (DiAN) is covalently bonded onto the surface of SiNWs and the fluorescence of the DiAN-modified SiNWs is employed as the output signal to form fluorescence logic switch. The results suggest that quenching and emission of the fluorescence from DiAN-modified SiNWs can be well controlled by the pH of the system. This research opens a window to assemble various logic switches on a SiNWs to form logic device with higher integration.

SiNWs were prepared by simple thermal evaporation of silicon monoxide powder as the single source [34]. SiO powder (Aldrich, 99%) in an alumina boat was placed at the center of a horizontal alumina tube mounted inside a high-temperature tube furnace. The system was evacuated to 10^{-3} Pa. Ar (95%) and H_2 (5%) as carrier gas was introduced at the flow rate of 50 sccm and system pressure was maintained at 400 Torr. The temperature was then heated to 1350 °C and maintained for 6 h then cooled to room temperature naturally. The brown cotton-like product was collected at downstream of the airflow. As prepared SiNWs have a diameter range of 15–28 nm (Fig. 1). These SiNWs were cleaned by first immersed in piranha solution (concentrated H_2SO_4: 30% H_2O_2 70:30, v/v) at 90 °C for 1 h and then left to cool down to room temperature. After repeatedly washed with doubly distilled water, the SiNWs were immersed in a H_2O:30% H_2O_2:$NH_3 \cdot H_2O$ 5:1:1 (v/v/v) mixture at room temperature for 3 h. Then, the SiNWs were copious rinsed with doubly distilled water and dried under vacuum at 50 °C before chemical modification.

The process for modifying SiNWs is similar to that described previously [22,35]. In a round bottomed flask connected to a Dean-Stark apparatus under nitrogen atmosphere, 30 mg dried SiNWs were suspended in 40 mL of anhydrous toluene. The mixture was heated at 140 °C to remove traces of water by azeotropic distillation. After 30 mL of toluene was evaporated, the suspension was cooled to 90 °C and 280 mg (0.47 mmol) of DiAN was added (DiAN was prepared according to the published procedure [36]). The mixture was refluxed for 48 h at 90 °C, then cooled down to room temper-

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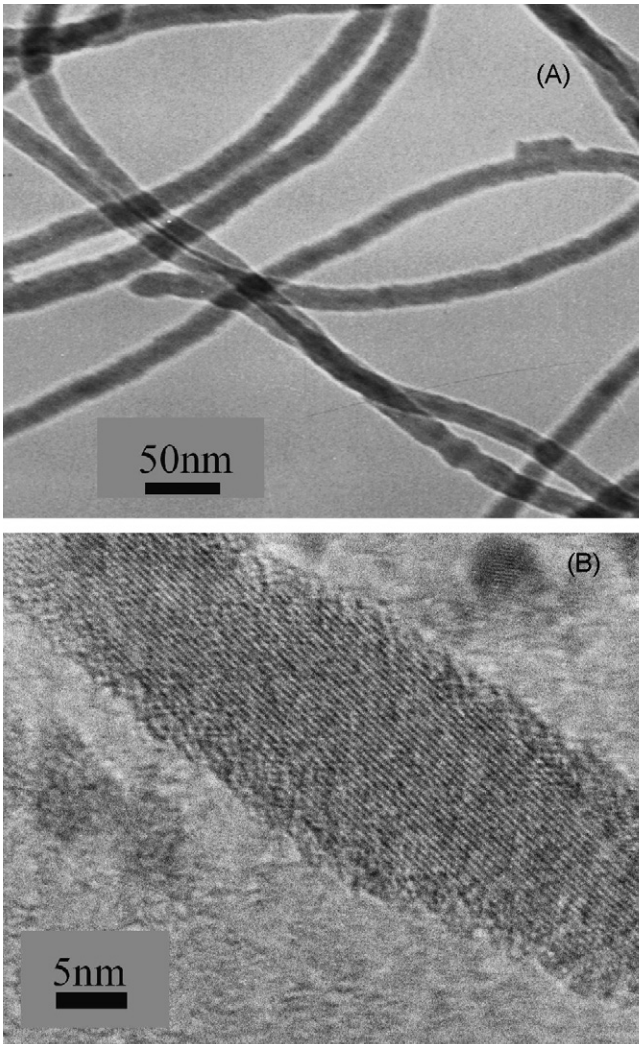

Fig. 1. TEM (A) and HRTEM (B) of as prepared SiNWs.

ature again. The modified SiNWs were collected by filtration and subsequently cleaned by ultrasonic stirring with anhydrous toluene for three times, dichloromethane for three times and ethanol for two times. Through these processes, the residual unreacted DiAN was removed completely and assured by monitoring the fluorescence of the washing liquid. Surface elemental characterization was performed with a MIQ-156 XPS spectrometer (RIBER, France). The IR spectra were recorded on a PerkinElmer Fourier transform infrared spectrometer as KBr pellets. The pH values of suspension were adjusted with HCl or NaOH solution. Fluorescence spectra were recorded on a Hitachi F-4500 fluorescence spectrofluorometer.

The attachment of DiAN on the SiNWs was investigated by IR and XPS spectra at ambient temperature. The results are shown in Figs. 2 and 3, respectively. Comparing with the IR spectra of the bare SiNWs, the DiAN-modified SiNWs has additional peaks at 2917, 2951 and 1560 cm^{-1} that correspond to the C–H vibration and skeletal vibration of the aromatic ring in the structure of the DiAN. Moreover, it can be found from the XPS characterization that the DiAN-modified SiNWs render additional peaks at 399.2 eV that can be attributed to N(1s) of DiAN. These results demonstrate that the DiAN was indeed covalently bonded on the surface of the SiNWs.

The loading density of DiAN on the surface of SiNWs may be determined according to literatures [35,37]. Assuming DiAN and

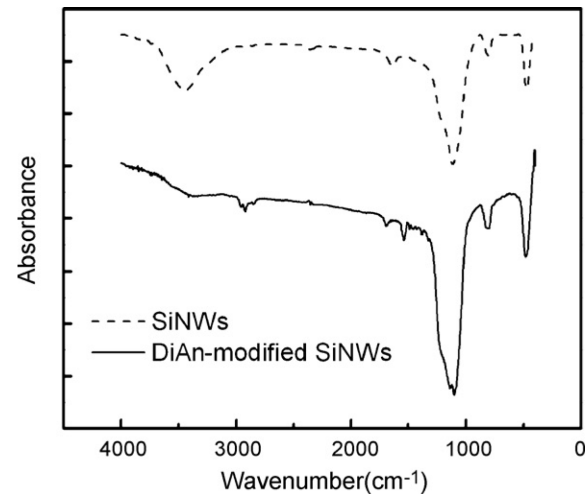

Fig. 2. IR spectra of SiNWs and DiAN-modified SiNWs.

DiAN-modified SiNWs with the maximum fluorescence intensity have equal quantum efficiency under acid condition, the following formula ((1) and (2)) was used to compute the average coverage of DiAn on SiNWs:

$$\eta_s = \eta_R \times \frac{I_s \times A_R}{I_R \times A_s} \quad (1)$$

$$A = \varepsilon bc \quad (2)$$

Here η is quantum efficiency, S, R represents sample and standard substances, respectively, I is integral intensity of fluorescence, and A is absorbance at excited wavelength. Eq. (2) is the absorption formula.

Based on these equations and experimental data, the molecules of DiAN on a microgram of SiNWs could be calculated to be 7.6×10^{13}. That is to say, 60 μg/mL DiAN-modified SiNWs used in our experiments correspond to 7.4×10^{-6} mol/L. If further assuming that the diameter of SiNWs is 20 nm, 1.48 molecules per 100 $Å^2$ could be obtained, which is almost 15.3% of the theoretical value [38].

In order to investigate the response of the DiAN-modified SiNWs to pH value, the modified nanowires was dispersed in aqueous solution containing 8% ethanol. The fluorescence spectra of DiAN-modified SiNWs under different pH were recorded and shown

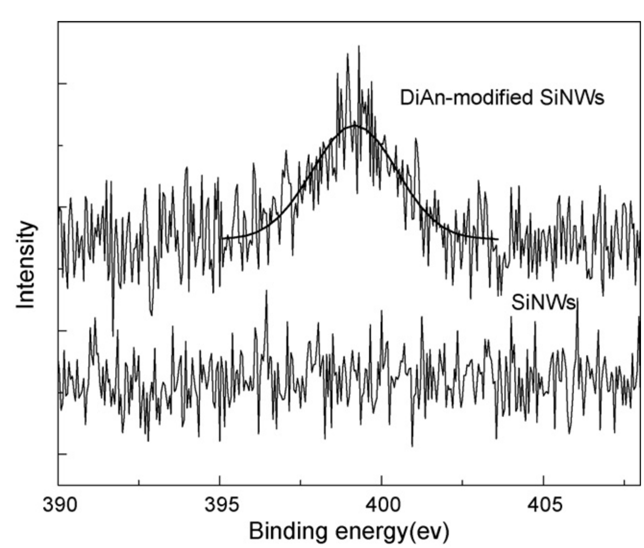

Fig. 3. XPS spectra of SiNWs and DiAN-modified SiNWs: close-up survey at N_{1s}.

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