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Tailoring transport and dielectric properties by surface passivation of silicon nanowires with Polyacrylic acid/TiO_2 nanoparticles composite

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ABSTRACT

We have presented the composite device including silicon nanowires (SiNWs), Polyacrylic acid (PAA) and Titanium dioxide nanoparticles (TiO_2 NPs). SiNWs and TiO_2 NPs were synthesized by metal assisted electroless chemical etching (MACE) and co-precipitation method respectively. Solution containing PAA and TiO_2 NPs in DI water was spun on already grown vertical SiNW arrays. We have investigated the transport and dielectric properties of *p*-type SiNWs/PAA/TiO_2 NPs (*p*-SPT) and *n*-type SiNWs/PAA/TiO_2 NPs (*n*-SPT) composite devices. Presence of PAA/TiO_2 NPs on the surface of SiNWs have increased electrical current in *p*-SPT device than that of *n*-SPT device. Ohmic like conduction was dominant at lower bias voltages followed by space charge limited current (SCLC) with traps at intermediate voltages. The calculated values of trap densities (H_t) were 7.73×10^{11} cm^{-3} and 5.34×10^{11} cm^{-3} for *p*-SPT device and *n*-SPT device respectively. Similarly *p*-SPT device shows higher real part of dielectric constant (ε') and AC conductivity (σ_{ac}) ~15 times and ~85 times respectively than that of *n*-SPT device. Increment in electrical and dielectric properties can be attributed to the presence of hydrophilic materials (PAA/TiO_2 NPs) which may results in enhancement of acceptor like states.

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1. Introduction

Silicon nanowires (SiNWs) are highly efficient for various applications in thermoelectric, photodetection and highly scaled information processing devices [1–3]. Different types of organic, inorganic and composite materials have been used for surface modification of nanostructures to tailor various properties e.g. sensing, electronic and optoelectronic [4–7].

Similarly carrier transport properties of SiNWs were modified using organic and inorganic materials [8–10]. Polyacrylic acid (PAA) is the key polymer for surface passivation because of its ionic nature. PAA can be easily diluted up to any extent in polar solvents e.g. acetone, isopropanol and DI water. PAA can absorb oxygen and sense gases even from environment [11].

TiO_2 NPs is one of important oxide in semiconducting oxide family. Hybrid materials containing TiO_2 NPs are significant for enhanced dielectric and photovoltaic properties [12–13]. Dielectric properties of TiO_2 NPs can be enhanced drastically via surface capping with polymer [14]. PAA covered SiNWs [15] and TiO_2 NPs decorated SiNWs [9] has been proved efficient for enhanced electrical transport and photodetection properties.

Here we present composite devices including PAA, TiO_2 NPs and SiNWs arrays. We have mixed TiO_2 NPs in highly diluted PAA and spun on vertically grown SiNWs arrays. We have investigated the carrier transport properties of fabricated composite devices. Incorporation of PAA/TiO_2 NPs had improved electrical and dielectric properties of *p*-SPT composite device in comparison to *n*-SPT composite device. The electrical and dielectric properties of SiNWs, ionic polymer and inorganic NPs based composite devices has not been investigated before according to best of our knowledge.

2. Experimental

SiNWs were prepared by metal assisted electroless chemical etching (MACE) which is an anisotropic wet etching technique. In this technique immersion of silicon substrate in hydrofluoric acid (HF) and silver nitrate ($AgNO_3$) solution forms silver (Ag) dendrites on the substrate. The substrate is then immersed in the solution containing HF and hydrogen peroxide (H_2O_2) for etching [15]. As a result long SiNWs are formed and length of SiNWs is highly dependent on etching time. Increase in etching time resulted in

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longer SiNWs. TiO_2 NPs are prepared by co-precipitation method [9,16] by using titanium tetrachloride ($TiCl_4$), hydrochloric acid (HCl) and sodium carbonate (Na_2CO_3). Prepared precipitates of TiO_2 NPs were cleaned with DI water. PAA having molecular weight 450 k and 15.8 wt.% in DI water was used as starting material. Highly diluted PAA ~3.3 wt.% in DI water was mixed with TiO_2 NPs under continuous stirring. Furthermore, ultrasonication was performed for 5 h. This solution containing PAA/TiO_2 NPs in DI water was spun at 1000 rpm for 60 s on already grown SiNWs. This step was followed by heating the sample at 50 °C in air to dry PAA/TiO_2 NPs for 20 min. This process was repeated two more times. With this process whole area between SiNWs was not fully covered with PAA/TiO_2 NPs and some pits and empty spaces might exist in the PAA/TiO_2 NPs filling. However the empty spaces may not be down to the bottom of the substrate. The addition of PAA/TiO_2 NPs on SiNWs can act as a source of interfacial doping by creating donor like or acceptor like states at the SiNW surface.

In order to make better electrical contact on the top of the SiNWs, composite devices were exposed to oxygen plasma for etching the PAA left on the top of SiNWs. Parameters were set as Ar (2 sccm), O_2 (98 sccm), pressure (50 Torr), temperature of chamber was 20 °C and power 200 W. The samples were etched for 60 min. The etching rate of polymer depends on the molecular weight and related functional groups. Oxygen containing polymer like PAA is more degradable as compared to other polymers (e.g. polystyrene and polyethylene) [15]. After oxygen plasma etching, the tips of SiNWs were clearly seen.

Finally 10 nm Cr followed by 300 nm Au was sputtered to form top (SiNWs) and bottom (Substrate) metal contacts in two separate steps. All experimental conditions including etching time for SiNWs fabrication, spinning, oxygen plasma etching and metal contacts are identical for both *p*-SPT and *n*-SPT composite devices for comparison. Degradation and other properties of PAA can be influenced by its PH value and temperature. That's why we have avoided further heating because temperature can influence the ionic character of PAA.

3. Results and discussion

Fig. 1(a) shows the cross-sectional scanning electron microscope (SEM) image of composite device. Energy dispersive X-ray spectroscopy (EDS) was performed to confirm the presence of PAA and TiO_2 NPs on the surface of SiNWs shown in the inset of Fig. 1(a). The length of SiNWs and diameter of TiO_2 NPs were ~40 μm and ~50 nm respectively. Fig. 1(b) shows the planer SEM image of composite device and inset shows planer SEM image after plasma etching. Similar results were observed for both *n* and *p*-SPT composite devices. Schematic diagram of SiNWs/PAA/TiO_2 NPs composite device with top and bottom metal contact is shown in Fig. 1(c). Synthesis of SiNWs and device fabrication process is approximately same as described elsewhere in case of PAA and TiO_2 [9,15]. Further, we have performed the temperature dependent *IV* characteristics (290–77 K) to investigate and compare the transport properties of composite devices. Fig. 1(d) shows the comparison of electrical current in *p*-SPT and *n*-SPT composite devices at 290 K and 77 K. PAA is ionic polymer and can easily dissolve in ionic solvents like water, acetone etc. PAA shows high sensitivity to humid environment and gases that's why, PAA can be used in humidity and gas sensing devices. SiNWs are of two types, hydrophilic and hydrophobic. Instant HF treatment on the surface of SiNWs resulted in hydrophobic groups but aged SiNWs are mostly hydrophilic in nature. Usually presence of Si–OH group makes SiNWs hydrophilic. The addition of highly diluted PAA/TiO_2 NPs on the surface of SiNWs results in the enhancement of hydrophilic groups on SiNWs surface [17].

For comparison, we have compared the values of electrical current. The *p*-SPT composite device shows ~94 and ~10^3 times enhanced conductivity at 290 K and 77 K respectively than that

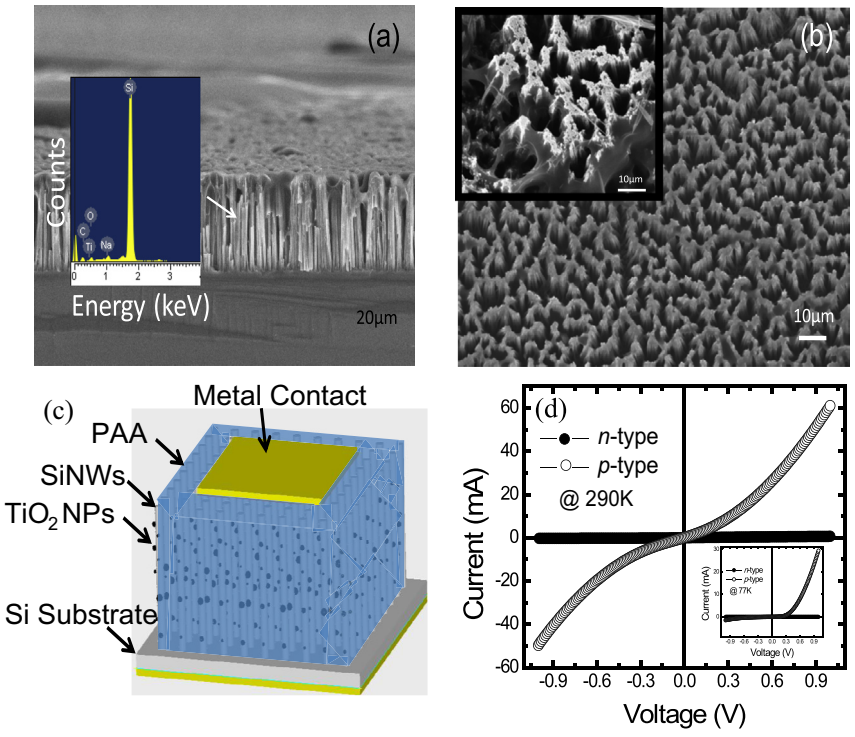

Fig. 1. (a) Cross-sectional SEM image of SiNWs/PAA/TiO_2 NPs composite device with EDS spectra (inset). (b) Planer SEM image of composite device and inset shows planer SEM image after plasma etching (similar images were seen in both *n* and *p*-SPT composite devices). (c) Schematic diagram of fabricated composite device. (d) Comparison of *IV* characteristics at 290 K and 77 K for *n*-SPT and *p*-SPT composite devices.

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