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Correlation between the photoluminescence and chemical bonding in silicon nitride nanowires deposited by chemical vapour deposition

Umesh Rizal [a], Bhabani S. Swain [b], Bibhu P. Swain [a, *]

[a] Nano-Processing Laboratory, Centre for Material Science and Nanotechnology, Sikkim Manipal Institute of Technology, Majitar, Rangpo, East Sikkim, 737136, India

[b] School of Advanced Material Engineering, Koomin University, Jeongnung-dong, Sungbuk-gu, Seoul, Republic of Korea

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ABSTRACT

We have fabricated needle-shaped silicon nitride (Si_3N_4) nanowires (NWs) on p-type Si substrate by chemical vapour deposition reactor using Si powder and (H_2 and NH_3) as precursor gases. The NWs were characterized by fourier transform infrared (FTIR) spectroscopy, photoluminescence (PL) spectroscopy and X-ray photoelectron spectroscopy (XPS) in order to investigate structural and electronic properties. The vibrational signature at 467, 799 and 1085 cm^{-1} confirmed the Si–N stretching vibration bands. The qualitative of Si_3N_4-NWs network were analysed by curve fitting of FTIR signature. The room temperature photoluminescence spectra of Si_3N_4-NWs reveals several emission bands centred at 3.219, 3.265, 3.319, 3.373, 3.432, 3.504, 3.577 and 3.631 eV, which are associated with electronic transitions between the conduction band and defect states. The valence band of appeared p, sp, sp^2, sp^3 and 2s orbitals at 3.36, 6.42, 8.11, 9.73 and 12.36 eV respectively and shifted to higher binding energy with increasing of ammonia flow rate.

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1. Introduction

Silicon nitride nanowires (Si_3N_4-NWs) is an wide band gap (5.3 eV) semiconductor with fascinating properties, such as high strength at high temperature, good resistance to corrosion, wear, thermal shock and creep, excellent chemical stability, and so on [1–3]. Above mentioned properties favour Si_3N_4-NWs as a suitable material for a wide range of applications in high-temperature nanoscale electronics devices [4], optoelectronics devices [5] and sensors [6]. In the recent years, tremendous effort has been made to develop a variety of techniques to control the morphology of the synthesized Si_3N_4-NWs. Many of synthesised process use simple vapour phase reaction approach without using catalyst [7] thermal evaporation of SiO_2 and Si powder in presence of NH_3 [8], silica nano powder [9], polymer precursor [10], polysilazane [11], and using SiH_4 and NH_3 precursor by PECVD [12]. Cui et al. synthesised Si_3N_4-NWs by using microwave plasma enhanced chemical vapor deposition (MWPECVD) process using NH_3 gas [13]. Huo et al. synthesised of single-crystalline α-Si_3N_4 nanobelts has been developed, consisting of nitridation of a high-Si-content Fe–Si 'catalyst' by ammonia at 1300 °C [14]. Tang et al. synthesised Si_3N_4 nanorods by nitriding borosilicate glass with ammonia [15]. NH_3 preferred as a source of atomic nitrogen than N_2 which decomposition temperature is lower than that of molecular N_2. Ultra-high purity NH_3 is important for the growth of defect-free epitaxial nitride. Si_3N_4-NWs thin films were synthesised by using NH_3, N_2 and H_2 precursor gases. However, none of the above research group analyzes details of chemical network of Si_3N_4-NWs. Therefore, chemical network of Si_3N_4-NWs with optical properties and valance band study of Si_3N_4-NWs at different ammonia flow rate is an important attempt for fundamental research.

2. Experimental details

Si_3N_4-NWs samples were prepared on RCA cleaned p-type Si (100) substrate by using thermal CVD reactor. Silver (Ag) thin films of 5 nm was coated on p-type Si substrate by sputtering technique followed by annealing 700 °C in H_2 ambient environment for 1 h to form separate Ag-NPs. Si powder (99.999%) of 10 g was taken in a ceramic boat and placed at the middle (heating zone) of horizontal furnace and the Si substrates were placed at 6 cm from the ceramic boat containing Si powder. After the ceramic boat and Si substrate were placed inside the reactor, the reactor was pumped to sub-

* Corresponding author.
E-mail address: bibhu.s@smit.smu.edu.in (B.P. Swain).

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atmospheric pressure. N_2 was flowed into the reactor for 2 h in order to overcome the residual gases in the reactor. The temperature of the furnace was increased to 1050 °C at a rate of at 5 °C/min. A typical condition was used in which N_2, H_2 flow rate were fixed at 120 and 80 sccm respectively, while NH_3 flow rate was varied from 2 to 12 sccm. The growth duration was fixed for 2 h. After deposition, furnace was cooled down gradually at a rate of 3 °C/min. The morphology and chemical network of Si_3N_4- NWs were studied using field emission scanning electron microscope (FESEM: JEOL-JEM-3000F), and Fourier transform infrared spectroscopy (FTIR). FTIR spectra were collected with a Perkin Elmer spectrometer (Model: Spectrum 2) operating in transmission mode in the range 450–1500 cm^{-1} with a resolution of 1 cm^{-1}. Photoluminescence studies were conducted with a Perkin Elmer LS-45 fluorescence spectrometer with a Xe discharge lamp operating at room temperature. X-ray Photoelectron Spectroscopy (XPS) was characterised by SIGMA PROBE, ThermoVG, UK for chemical analysis of Si_3N_4-NWs. In particular, the valence band was analysed with a step size of 0.1 eV to examine electronic network of Si_3N_4-NWs.

3. Results and discussion

3.1. Microstructure

Fig. 1(a–d) shows FESEM images of Si_3N_4-NWs deposited on Si substrate using Ag-NPs. The as-synthesized Si_3N_4-NWs possess needle shaped morphology with the diameter varies in between 20 and 120 nm. As can be seen from Fig. 1(b–d), bunch of straight Si_3N_4 NWs with smooth morphology are distributed randomly on the surface of a silicon substrate. Larger diameter NWs was observed for 4 sccm NH_3 flow rate, however the diameter of NWs decreases with increasing of NH_3 flow rate from 4 to 12 sccm. The majority of NWs rooted in clusters, develop into prism like microstructure with a sharp tip. Niu et al. observed similar kinds of microstructure of Si_3N_4-NWs on Au-NPs/Si substrate [16].

3.2. Bonding network of Si_3N_4-NWs

Fig. 2 (a) shows broad scan FTIR spectra of Si_3N_4-NWs ranging from 450 to 1500 cm^{-1} in transmission mode. Moreover, the FTIR spectrum of our Si_3N_4-NWs sample exhibits four well defined vibration signatures centred at 464 cm^{-1}, 622 cm^{-1}, 793 cm^{-1} and 1088 cm^{-1}. The broad vibrational signature at 464 cm^{-1} corresponds to symmetric stretching of Si–N bonding [17]. A weak transmission peak centred at 620–670 cm^{-1} corresponding to Si-H_n wagging [18]. The shifting of Si–H bond towards higher energy side may be due to backing of higher electronegativity nitrogen atom [19]. Inukai et al. observed the vibration signature of Si–N asymmetric TO stretching at 850 cm^{-1} for Si_3N_4 thin films [20]. However, we have observed Si–N asymmetric TO stretching at 793 cm^{-1}. This red shift in Si–N asymmetric TO stretching can attribute to quantum confinement effect brought out by nano dimension material or phonon localization due to presence of defects in nanostructures. The presence of defects in nanostructures was further confirmed by our photoluminescence studies. Wada et al. observed Si–N stretching vibration signature of α-Si_3N_4 at

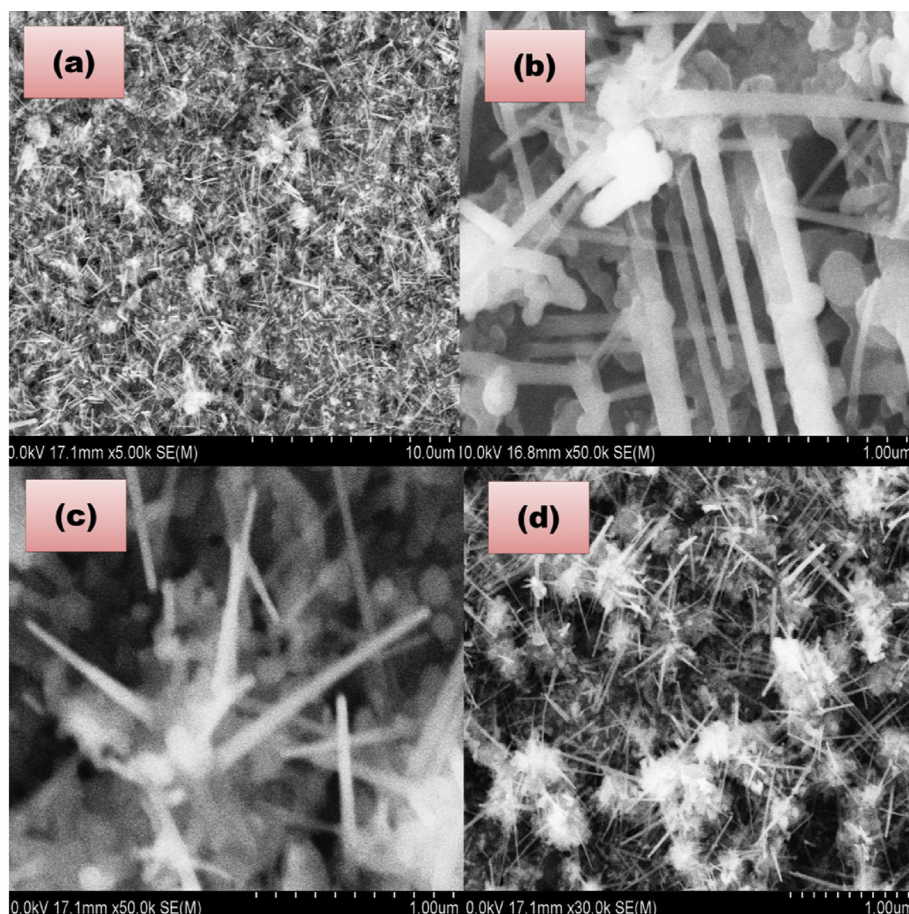

Fig. 1. Broad scan FESEM images of Si_3N_4-NWs (a), FESEM of Si_3N_4-NWs at 4 sccm NH_3 flow rate (b), FESEM of Si_3N_4-NWs at 8 sccm NH_3 flow rate (c), FESEM of Si_3N_4-NWs at 12 sccm NH_3 flow rate (d).

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