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Effect of substrate to filament distance on formation and photoluminescence properties of indium catalyzed silicon nanowires using hot-wire chemical vapor deposition

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ABSTRACT

Si nanowires have been synthesized by hot-wire chemical vapor deposition technique, with Indium nanocones employed as catalysts with different substrate to filament distances ranging from 6 to 3 cm. Reducing the substrate to filament distance resulted in the retention of more atomic H radicals on the growth sites. The atomic H radicals acted to induce the catalytic growth and enhance the crystallinity of the Si nanowires. The Si nanowires showed tapering structures due to the radial growth of columnar Si nanocrystallites on the middle and base walls of the nanowires. The oxide-related defects on the outer layer of the Indium nanocones and Si nanowires, as well as the Si nanocrystallites on walls of the Si nanowires, contributed to the visible orange and red photoluminescence.

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1. Introduction

Si nanowires (SiNWs) have been extensively investigated due to their potential in a wide range of energy related applications [1]. Among the variety of techniques, chemical vapor deposition (CVD) remains one of the preferred fabrication methods for SiNWs owing to the controllable growth and size of the NWs [2]. Currently, the preparation of SiNWs using low temperature CVD techniques such as plasma enhanced CVD and hot-wire (HW) CVD is being intensively studied [3,4]. Efficient gas usage and fast decomposition of silane (SiH_4) by HWCVD for SiNWs growth have made this technique very attractive for implementation in various industries. In HWCVD, SiH_4 can be decomposed into Si and H atomic radicals using a heated tungsten filament whose temperature is above 1500 °C. The SiH_4 decomposition rate can be increased by increasing the filament temperature [5]. However, the growth mechanism of SiNWs involves not only the SiH_4 decomposition process, but also the gas phase reactions and the surface reactions, thus leading to the growth of SiNWs. The quality of sample is highly influenced by the gas phase (SiH_4 molecules and radicals) reactions [6]. The deposition pressure and substrate to filament distance ($d_{s\text{–}f}$) play an important role in controlling the gas phase reactions [7]. The key to the growth of NWs is mainly dependent on the optimization of these two parameters.

Indium (In) is currently preferred as a catalyst candidate alternative to gold due to its low melting temperature (~157 °C). The growth of SiNWs from In catalyst at substrate temperature as low as 240 °C was achieved [8]. In addition, In is attractive from an electronics viewpoint as it can induce p-type doping in the SiNWs [9]. However, In forms eutectic with Si at almost zero concentration (smaller than 0.01 atom %) due to its low Si solubility [9]. Thus, large quantities of Si precursors are required in order to initiate nucleation and growth of SiNWs from In catalyst. In this case, HWCVD serves as a promising technique for In catalytic growth of SiNWs due to its high SiH_4 decomposition rate [4].

In this work, the effect of $d_{s\text{–}f}$ on growth of the In-catalyzed SiNWs using HWCVD was studied. The morphologies as well as structural and photoluminescence (PL) properties of the as-grown samples were characterized using field emission scanning electron microscopy (FESEM), Fourier transform infra-red (FTIR) spectroscopy, X-ray diffraction (XRD), micro-Raman spectroscopy, high resolution transmission electron microscopy (HRTEM) and PL spectroscopy.

2. Experimental details

Samples were prepared on p-type Si(111) substrates using a home-built plasma assisted HWCVD system. A coiled tungsten filament of 99.95% purity was employed to evaporate the In wire placed on it as well as to decompose the SiH_4 and hydrogen (H_2) gasses. Plasma was generated through a power electrode by a radio frequency generator (13.56 MHz). The experimental setup was presented in more detail in our earlier work [10]. The reactor was evacuated to a base pressure of ~2×10^{-3} Pa. In wire (diameter of 0.5 mm and length of 0.5–1 mm) with purity of 99.999% was evaporated using the tungsten filament heated at a filament temperature of ~1200 °C. In order to form nanosized In islands, the evaporation was carried out in a H_2 plasma

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environment [11]. The H_2 flow rate and radio frequency power were fixed at 150 sccm and 40 W respectively during this process. The evaporated In vapor was held by a heated Si(111) substrate at temperature of 400 °C. A substrate temperature higher than the melting point of In was applied to melt the In islands for initiation of the vapor–liquid–solid growth process. Following this, SiH_4 gas diluted in H_2 was dissociated at filament temperature of 1700 ± 20 °C, which was measured using a Reytek, Raynger 3i pyrometer. The SiH_4 and H_2 flow rates were fixed at 5 to 150 sccm respectively during this process. Four sets of samples were prepared at different d_{s-f} of 6 to 3 cm, with a decrement of 1 cm. The substrate temperature, deposition pressure and deposition time for this process were fixed at 400 °C, 70 ± 2 Pa and 10 min respectively for each deposition. The substrate temperature, which was measured by a K-type thermocouple touching the bottom of the substrate, would have a temperature increment of approximately 19–32 °C on top of the 400 °C due to the irradiation from the HW.

The surface morphologies of the samples were recorded using a FEI Quanta 200 FESEM at an operating voltage of 20 kV. Si–oxygen and Si–hydrogen bonding properties of samples were investigated using a Perkin-Elmer System 2000 FTIR spectroscopy. The FTIR spectra were recorded with a spectral resolution of 4 cm^{-1} in the range of 400–1600 cm^{-1}. XRD measurements were carried out using a SIEMENS D5000 X-ray diffractometer (Cu K_{α} X-ray radiation wavelength, $\lambda = 1.5418$ Å) at a grazing angle of 5°. The Raman spectroscopy measurements were carried out using a Horiba Jobin Yvon 800 UV Micro-Raman spectrometer with an Ar^{+} laser source operating at an excitation λ of 514.5 nm. These measurements were taken at a low laser power of ~1.5 mW to prevent the laser heating effect.

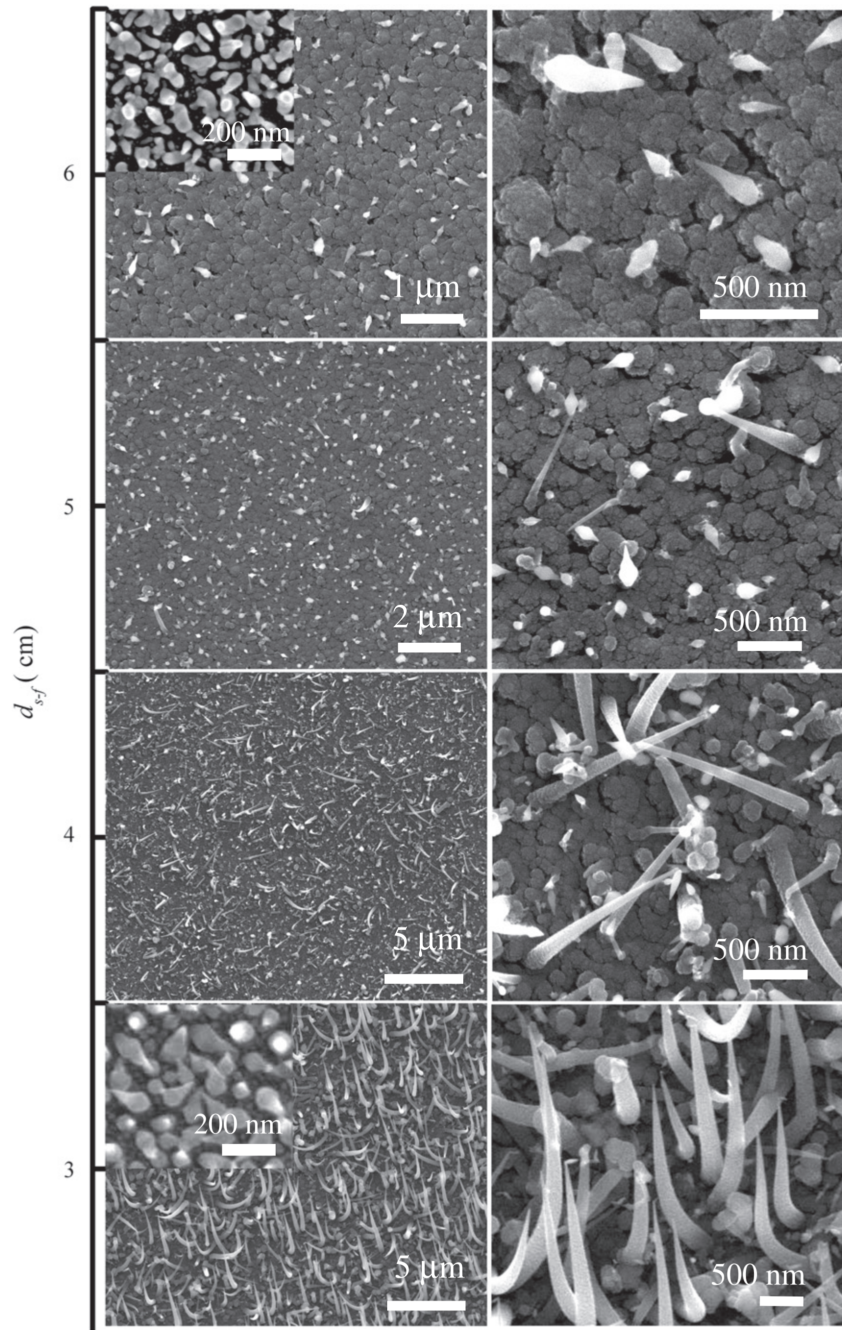

Fig. 1. FESEM images of the samples prepared by HWCVD at d_{s-f} of 6 cm, 5 cm, 4 cm and 3 cm. The right hand side presents the magnified FESEM images of the respective samples. Inset in FESEM images of samples prepared at d_{s-f} of 6 cm and 3 cm are the FESEM images of the as evaporated In nanocones formed at the respective d_{s-f}.

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