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Lateral growth of aligned mutilwalled carbon nanotubes under electric field

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Abstract

In this work we report laterally aligned multi-walled carbon nanotube (MWNT) by an electric field during growth. The MWNTs were selectively grown between lateral sides of the catalytic metals on predefined electrodes by chemical-vapor deposition. The electric field distribution for various geometries was simulated using *Maxwell 2D* simulation in order to realize better alignment of laterally grown carbon nanotubes (CNTs). The experimental results show that the electric field direction at the vicinity of catalyst and nanotubes–substrate interactions are principal factor in aligning CNTs laterally.

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1. Introduction

Carbon nanotubes (CNTs) as molecular-scale device elements and nanocomponents for nanomachines were intensively studied [1–7]. Recently, a molecular-scale device element based on a suspended, crossed nanotube geometry that leads to bistable, electrostatically switchable on/off states was introduced [2]. The device elements are naturally addressable in large arrays by the CNT molecular wires making up the devices. These reversible, bistable device elements could be used to construct nonvolatile random access memory and logic function tables at an integration level approaching 10^{12} elements per square centimeter and an element operation frequency in excess of 100 GHz. Also, various possibilities of CNTs for electronic and spintronic applications such as field effect transistors and resonant tunnel transistors were reported [3–6]. However, for electronic applications an additional effort was necessary to remove CNTs from the templates and disperse them onto prepatterned electrodes and, further, an electric field was applied to form a low-resistance contact to the electrodes. Dai et al. showed aligned single-walled CNTs grown in electric fields established across patterned metallic electrodes at 900 °C. Electric field has been used to manipulate the growth direction of multi-walled carbon nanotubes (MWNTs) previously, but not for directly lateral growth as shown by Dai et al [8–12].

In this work, we experimentally present a direct nanowiring of MWNT between two parallel patterned electrodes under electric fields using the chemical vapor deposition (CVD) method at 750 °C [13,14]. The exact growth mechanisms not addressed in this work may be similar to the possible mechanism reported for the electric field assisted aligned growth of single walled CNTs [8,9].

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2. Experimental details

In our experiment, the layer structure was prepared by stacking SiO_2–Nb–Co–SiO_2 layers on Si substrate. Though process steps varied according to the electrode structures, the basic sequences of our process are as follows. (i) The SiO_2 layer was grown on the n-type heavily doped Si wafer by a conventional wet oxidation process at 1100 °C (Fig. 1(a)). (ii) The 100 nm-thick Nb electrodes were deposited by sputtering using Ar plasma, and then patterned by photolithography and plasma etching. The spacing of the two electrode pattern were ≈2 μm and width of electrode was ≈20 μm (Fig. 1(b)). (iii) We patterned a catalyst region on the electrodes using a second photolithography step aligned to the electrodes. Catalyst and oxide were then deposited by sputtering into patterned region followed by liftoff in acetone, where top oxide layer is introduced as a barrier layer for vertical growth, covering the top of the Co catalytic layer (Fig. 1(c)). (iv) Aligned MWNTs across the gap between the electrodes were grown in thermal CVD system with electrical feedthroughs (Fig. 1(d)). The CVD chamber temperature was rapidly raised to the process temperature of 750 °C within 10 min by halogen lamps after evacuation down to 10^{-2} torr. During the growth, a DC V bias (0–10 V) was applied across the Nb electrodes and the total pressure of the chamber was kept constant at 50 torr, while the total flow rate of process gases was maintained at 200 sccm. For instance, the flow rate of the hydrocarbon source, C_2H_2 (acetylene), was 5 sccm and the remaining 195 sccm was for the carrier gases. After the CNT growth, the chamber was purged continuously with a mixture of H_2 and Ar until the chamber temperature reached room temperature. The samples were examined by field emission scanning electron microscope (SEM, Hitachi S4300).

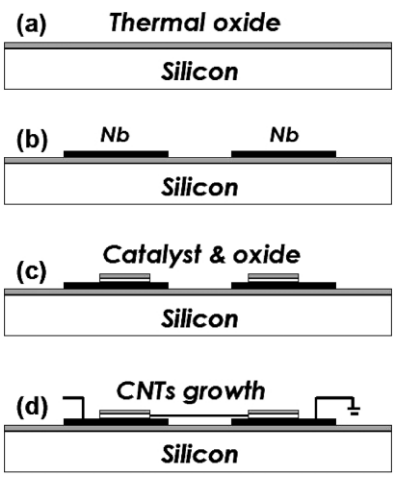

Fig. 1. Schematic diagram of process flow for lateral growth of aligned MWNTs by an electric field.

3. Results and discussion

First of all, the electric field distribution for various geometries was simulated using *Maxwell 2D* simulation program (EM simulation tool, Ansoft corp.). Fig. 2 shows a cross-sectional view of electric field vectors and equipotential lines for three type structure; (a) the Co catalyst was formed underneath the Nb electrode, (b) the catalyst was formed on the electrode, and (c) the catalyst region was patterned on the elevated electrode. Fig. 2(a) shows the electric field directions at catalyst are towards the substrate. On the other hand, In structure of catalyst on electrode, the electric field vector at the catalyst is nearly perpendicular to the substrate and upward as shown in Fig. 2(b) and (c). The electric field vectors are parallel to the substrate gradually in the middle of gap between the electrodes.

We formed a 100 nm thick catalyst island on SiO_2 layer, followed by deposition of a 100 nm thick electrode film of Nb by sputtering. Then Nb electrodes were patterned by photolithography and plasma etching before CNTs growth. Fig. 3(a) shows a SEM image of CNTs grown under 4 V applied across the Nb electrodes. The CNTs appear dark since they are electrically conducting, whereas the SiO_2 surface appears bright due to charging because metal was not coated on the sample. Most of CNTs are located at the edge of each electrode. The long noodle-shaped CNTs are lying down and nonaligned perpendicular to edge of the electrode. It was found that the alignment of CNTs was not

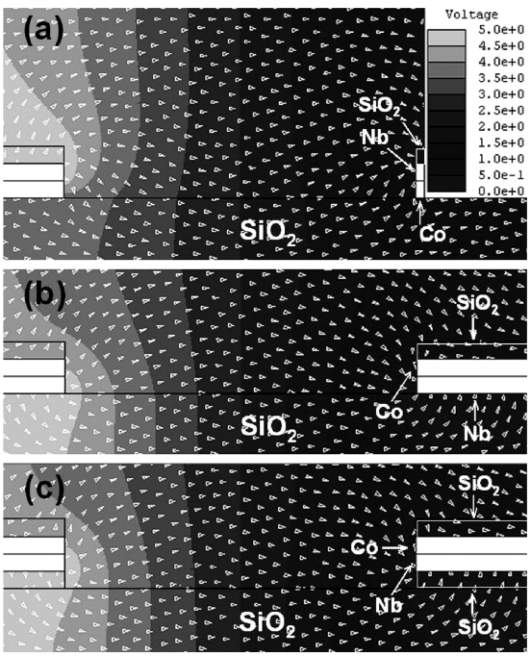

Fig. 2. Electric field vector and equipotential lines calculated for the left and right electrodes biased at 5 and 0 V, respectively. The layer structure was prepared by stacking (a) electrode on catalyst, (b) catalyst on electrode, and (c) catalyst on elevated electrode.

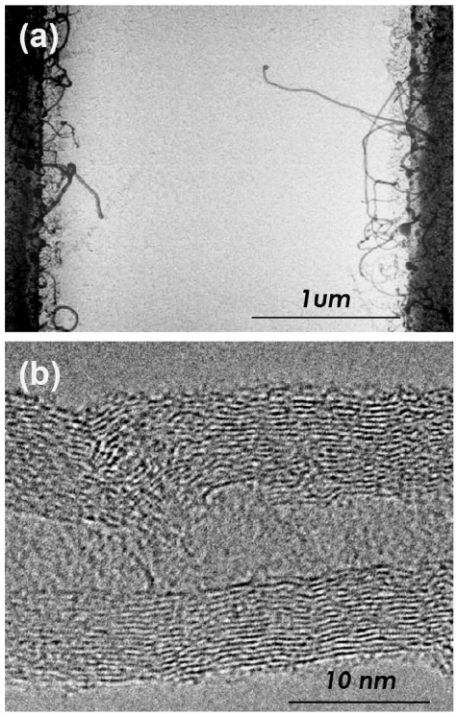

Fig. 3. (a) SEM image of CNTs grown on catalyst beneath the Nb electrode in applied voltage of 4 V. (b) TEM image of CNT corresponding to (a).

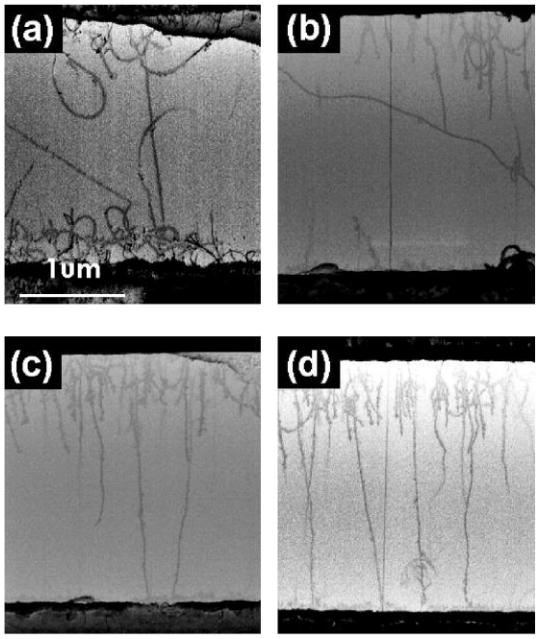

Fig. 4. SEM images of MWNTs grown in various electric fields. The spacing between the electrodes is 2 μm. Applied voltage is 0 V in (a), 1 V in (b), 2.5 V in (c), and 4 V in (d).

improved in spite of increasing the applied voltage from 0 to 10 V. We inferred from Maxwell 2D simulation (Fig. 2(a)) that the MWNTs initially pinned on substrate by electric field direction and van der Waals forces. Fig. 3(b) shows TEM image of the laterally grown MWNTs s between Nb electrodes. The CNTs are multiwalled CNTs with outer diameter range from 15 to 20 nm and consists of hollow compartments, looking like a bamboo structure that is well known in the vertical growth of CNTs on a catalytic metal deposited substrate. From these results we confirm that the bamboo growth mechanism can be applied to our selective lateral growth of CNTs by thermal CVD.

To prevent MWNTs from pinning on substrate at early stage for growth, the catalyst region was placed on electrode as simulated in Fig. 2(b). We have carried out nanotube growth under applied voltages in the range of 0, 1, 2.5, 4 V, respectively. Fig. 4(a) shows a SEM images of randomly oriented MWNTs in the gap region between the Nb electrodes when no electric field is applied. In contrast, the degree of alignment increases as applied voltage goes up as shown in Fig. 4(b)–(d), respectively. The MWNTs are clearly aligned perpendicular to the edges of the electrodes, in the direction of electric field and were grown on surface of SiO_2. No obvious improvement is observed in alignment of MWNTs when the voltage is increased to 4 V. At early state, the pinning effect decreased due to an upward electric field direction. But many of MWNTs did not go across the gap between the electrodes. We have carried out with several samples, and observed a reproducible though there is small disparity in the density of CNTs. At relatively high temperature, above 950 °C, CNTs showed good linearity but there was the problem of damaging the metal electrode and insulator during CNT growth.

To grow the suspended MWNTs, we modified the electrode structures to prevent MWNT from easily contacting the substrate. The 100 nm-thick SiO_2 layer was introduced beneath the electrode metal. Fig. 5(a) shows a SEM images of MWNTs grown under 4 V applied across the electrodes. It was observed that MWNTs were well aligned along the electric filed but most of the MWNTs grown from catalyst were grown on the surface and went over the gap between the electrodes. The structure with elevated electrode has weakened interactions between of nanotubes and substrate. The suspended MWNTs are very straight with little bending across the whole length as shown in Fig. 5(b). On the other hand, MWNTs landed on the bottom between the electrodes have a slightly wavy shape as shown in Fig. 5(c). When we changed the deposition procedures of the electrode and catalyst, aligned CNTs were not observed at elevated structure. Maxwell 2D simulation also showed the electric field vectors at catalyst were dramatically towards the substrate.

In case of lateral growth of CNTs, we have not yet exactly controlled the density of CNTs and the proportion of suspended CNTs to landed CNTs. In order to realize better controls of the lateral grown CNTs, the modification of

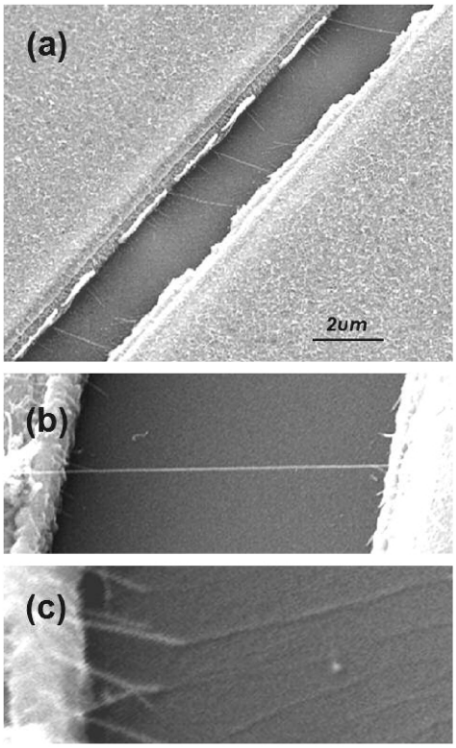

Fig. 5. (a) SEM images of MWNTs grown in electric field of 2 V/μm. (b) Magnified SEM image of suspended MWNT. (c) Magnified SEM image of landed MWNT.

electrode structure and nanotubes–substrate interactions should be investigated more in future study.

4. Summary

We have experimentally demonstrated electric field directed growth of MWNTs. Maxwell 2D simulation and experimental results show that electric field direction at the vicinity of catalyst and nanotubes–substrate interactions are principal factor in aligning CNTs laterally. These CNT bridges can be used as an active component nanowire for the molecular-scale electronic devices with high integration density, or a key block for the nanomachines.

Acknowledgements

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