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2003 Nanotechnology 14 497

(http://iopscience.iop.org/0957-4484/14/5/303)

INSTITUTE OF PHYSICS PUBLISHING NANOTECHNOLOGY

Nanotechnology **14** (2003) 497–500 PII: S0957-4484(03)55837-8

Suppression of leakage current via formation of a sidewall protector in the microgated carbon nanotube emitter

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Received 31 October 2002, in final form 16 January 2003
Published 28 March 2003
Online at stacks.iop.org/Nano/14/497

Abstract
In this work, we have fabricated a triode field emitter using directly grown carbon nanotubes (CNTs) as an electron emission source. Vertically aligned CNTs have been grown in the centre of the gate hole, to the size of 1.5 μm in diameter, using thermal chemical vapour deposition. A comparison of the field emission characteristics of two types of microgated nanotube emitter with and without a sidewall protector for the gate hole is made. A sidewall protector formed by the method of tilting the substrate can enhance the electrical characteristics by suppressing the problem of short circuits between the gate and the CNTs. The leakage current of an emitter with a sidewall protector is approximately sevenfold lower than that of an emitter without a sidewall protector at a gate voltage of 100 V.

1. Introduction

Recently, carbon nanotubes (CNTs) have aroused considerable interest because of their unique physical properties and many potential applications [1, 2]. In particular, due to their extreme aspect ratios, CNTs exhibit large local field enhancement and thus yield considerable field emission currents at relatively low applied voltages. Therefore, it is well accepted that CNTs have a high potential for application in field electron emitters [3, 4]. Two main strands in fabrication processes for CNT field emitters are thick-film printing and direct growth. The thick-film printing processes are mainly used for preparation of large-area electron sources and field emission displays [5, 6]. But this has intrinsic limits as regards reducing pixel size and, thus, scaling of a device unit. Therefore, gated field emitters incorporating CNTs directly grown into micrometre-sized holes are advantageous for small-area electron sources such as microwave generators and micro-displays. But the field emission from the gated CNT structure has been an open problem until now. The greatest difficulties in demonstrating successful fabrication of a gated field emitter array (FEA) using CNTs are the quite high gate current and the requirement for precisely controlled selective growth of CNTs within a micro-hole. This includes the height and density control of CNTs, selection of a stable gate electrode material, and fabrication optimization without damaging the predefined structures during the etching process. Hsu *et al* have grown non-aligned CNTs inside an open trench with oxide used to reduce the leakage current. In their work, low-pressure chemical vapour deposition (CVD) and reactive ion etching (RIE) were used to fabricate an oxide spacer lining sidewall of gated aperture and to remove catalyst from the part where CNTs should not be grown [7–11].

In this work, we present the fabrication of a triode structure field emitter using CNTs directly grown by thermal CVD. Two types of microgated nanotube FEA were fabricated: a silicon trench well structure and a silicon trench well structure with a sidewall protector. We easily formed an oxide spacer and a catalytic metal layer inside the emitter holes using a parting layer of a type that is in general use in metal tip process. Furthermore, we have characterized the field electron emission

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characteristics of the triode CNT field emitter with and without a sidewall protector to prevent unwanted growth of CNTs.

2. Experiments

As shown in figure 1, the fabrication flows of microgated nanotube emitter without and with a sidewall protector are (a) → (b) → (c) → (d) → (e) → (f) → (h) → (i) and (a) → (b) → (c) → (d) → (e) → (f) → (g) → (h) → (i), respectively. The details of our process are as follows. After 1 μm thick thermal oxide and 0.3 μm thick sputtered gate metal were sequentially grown on highly doped silicon substrate, a gate aperture of 1–2 μm diameter was defined by photolithography. The gate metal was removed by reactive ion etching, followed by SiO_2 and silicon substrate etching via dipping in buffered oxide etchant and tetramethyl ammonium hydroxide (TMAH) (figures 1(a)–(d)). Then an Al parting layer was deposited by electron beam evaporation, and during the deposition the substrate was rotated and tilted about an axis perpendicular to the substrate surface. The Co catalytic layer, of 15 nm thickness, was evaporated onto the specimen with normal incidence. Then, a sidewall protector was simply deposited by the method of tilting the substrate. The tilting angles were 15°, 9°, and 45° (figures 1(e)–(g), respectively). After the parting layer was removed, the specimens were placed into a quartz reactor to grow the CNTs (figure 1(h)) and it was pumped down to less than $\sim 10^{-3}$ Torr using a mechanical pump. The substrate was then heated to the synthesis temperature while introducing Ar and H_2 with flow rates of 70 and 80 sccm, respectively. The synthesis temperature and pressure were about 750 °C and 50 Torr, respectively. When temperature and pressure were stabilized, NH_3 gas (instead of H_2) was introduced for 10 min. C_2H_2 gas was then introduced with a flow rate of 5 sccm (figure 1(i)). The samples were then cooled in Ar and H_2 flow ambient. To investigate the CNTs and emitter structure, high-resolution scanning electron microscopy (HRSEM) and transmission electron microscopy (TEM) were used. The field emission measurements were carried out in a high-vacuum chamber (base pressure: 10^{-7} Torr) equipped with cathode, gate, and anode probes using a Keithley 237 source measuring unit. The anode plate was placed about 1 mm from the gate metal and an anode voltage of 800 V was used.

3. Results and discussion

For a CNT emitter with a triode structure, the CNTs have to be selectively grown in micro-holes without overgrowth and without contact to the gate metal and insulator. Before describing the gated structure, we describe the planar CNTs grown on the cobalt in a 1 μm pattern. Cobalt with a thickness of 15 nm was evaporated as a catalyst metal. Following repeated trial and error, we successfully grew good free-standing CNTs without disturbing neighbouring CNT bundles at each patterned dot. Figure 2 shows a typical SEM image of vertically aligned CNTs with a growth time of 90 s. The CNT length is about 3 μm. The ends of the CNTs were slightly bent because each CNT is free-standing and not leaning against neighbouring CNT bundles. Though we have not yet fully prevented the ends of the CNTs from bending, we ignore this in this work because the electron emission under a DC field could come from the whole body of the CNTs behaving as a nanowire conductor [12, 13]. The inset of figure 2 shows a TEM image of the vertically grown CNTs which are multiwalled CNTs with outer diameter ranging from 20 to 25 nm and consisting of hollow compartments; the appearance is that of a bamboo structure, which is commonly encountered in the vertical growth of CNTs on a catalytic metal-deposited substrate.

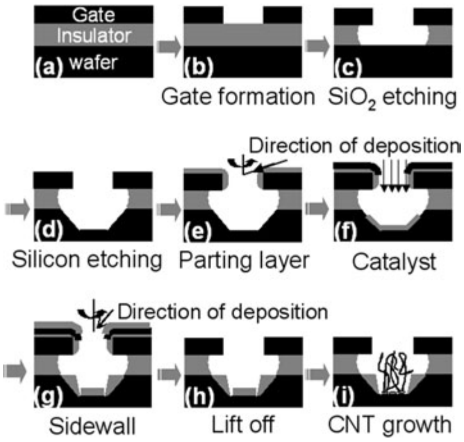

Figure 1. The process flows for fabricating the microgated emitter using CNTs directly grown by thermal CVD.

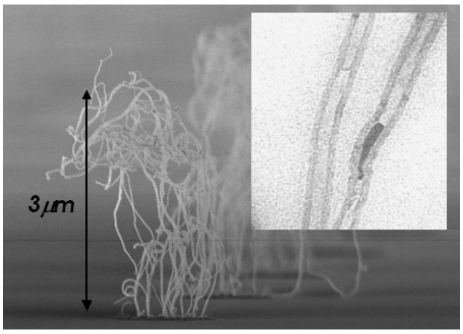

Figure 2. SEM images of vertically aligned CNTs directly grown on Co nanodots of 15 nm size with a growth time of 90 s; the inset is a TEM image of the CNTs.

Figure 3(a) shows a cross-sectional view of the microgated emitter formed using CNTs directly grown into silicon trench wells without and with a sidewall protector, respectively. The gate aperture is about 1.5 μm in diameter and the hole depth is about 2.5 μm. The thickness of the gate insulator is 1 μm. The CNTs are well aligned perpendicular to the substrate and are typically 2.5 μm high. The growth time is about 60 s. Here, one notes the etched Si structure. The reason for the deep etching of the silicon substrate was our inability to reduce the length of the vertically aligned CNTs to below 2 μm. Hence, we had to increase the hole depth by either increasing the thickness of the gate insulator or etching the Si substrate, the preferable choice being to etch the silicon substrate. The inset of figure 3 shows a top view of the gate hole after CNT growth. CNTs were well located at the centres of holes without shorting with the gate.

Figure 3(b) shows a magnified image of the sidewall of the emitter after CNT growth corresponding to figure 3(a).

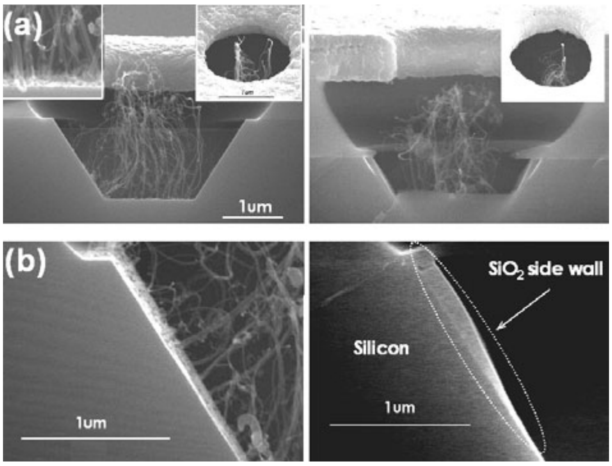

Figure 3. (a) Cross-sectional views of the microgated CNT emitter without and with a sidewall protector, respectively. The inset shows a top view of the gate hole. (b) Magnified images of the sidewall protector after CNT growth corresponding to (a).

In the case of the emitter without a sidewall protector, it was occasionally observed that CNTs grown from catalyst deposited on a sidewall were contacting the insulator and gate. CNTs were randomly grown on the sidewall because we tried to selectively deposit the catalytic metal at the centres of holes but occasionally observed catalytic metal to be deposited on the sidewalls of gate holes. However, the selectively deposited insulator prevents CNTs from being grown on sidewalls and in contact with the insulator and gate.

We measured the field emission properties of the gated CNT emitter after finding by inspection whether or not CNTs had shorted with the gate, using FE-SEM. Figure 4(a) shows the field emission current–gate voltage characteristics of the gated CNT emitter. There is no serious difference in anode current except as regards the threshold voltage of the emission. The anode currents of the CNT emitters with the conventional gate hole structure and with the sidewall protector are 1.9 and 1.7 μA at a gate voltage of 100 V respectively. In spite of the vertical alignment of the CNTs, these values are lower than the emission current of a triode emitter constructed using non-aligned CNTs as shown in our previous report [7] and the report of Hsu [9]. In our study, the vertical alignment of CNTs is not related at all to the improvement of emission properties of the microgated CNT emitter. The inset of figure 4(a) shows a Fowler–Nordheim (F–N) plot, whose linearity indicates that the I–V characteristics are governed by a conventional field emission mechanism, $I \propto V^2 \exp(-6.53 \times 10^7 \phi^{3/2}/\beta V)$, where ϕ and β are the work function and field enhancement factors for CNT emitters, respectively. The field enhancement factors of an emitter with a sidewall and without a sidewall are calculated to be $\beta = 3.09 \times 10^6$ and 2.66×10^6 cm^{-1} from the F–N slopes of -102.7 and -118.2, under the assumption of a work function of 5 eV for CNTs, respectively. A microgated nanotube emitter has a higher β compared with a conventional metal tip emitter.

Figure 4(b) shows the gate current–gate voltage characteristics of a gated CNT emitter. In the case of CNT emitters with sidewall protectors, the gate-to-anode current ratio, I_g/I_a, was 0.38 at a gate voltage of 100 V, while it was 2.4 for the CNT emitters without sidewall protectors.

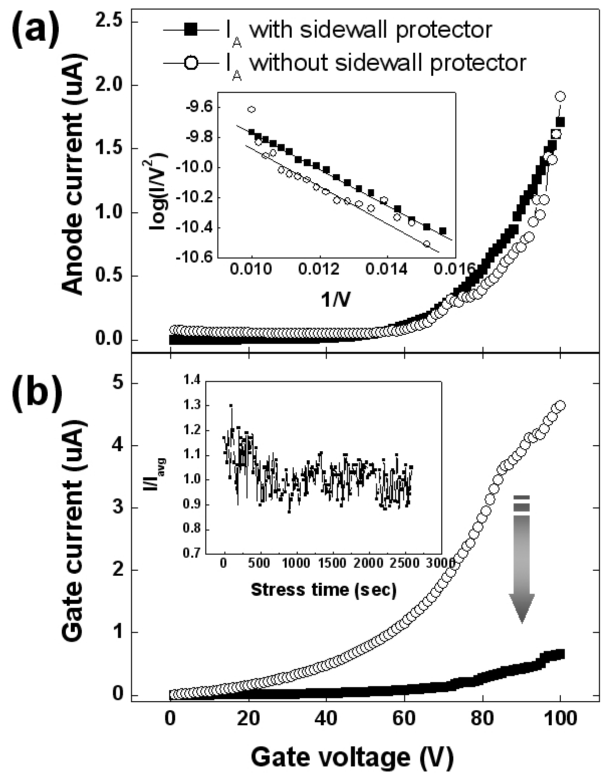

Figure 4. (a) Field electron emission current versus gate voltage for a gated CNT emitter corresponding to figure 3. The inset shows a F–N plot of the anode current. (b) Gate current versus gate voltage characteristics, according to the gate structure.

The reduction in leakage current was about 85.8% for CNT emitters with sidewall protectors as compared with ones without sidewall protectors. The gate current of the emitter is distinctly reduced but the gate-to-anode current ratio has not yet been moderated due to the low emission current. The leakage current measured in this study is not higher than that reported by Hsu. The low emission obtained with our CNT emitter may have several origins. The most important factors would be chemical attack on the interfacial layer between the gate insulator and the gate electrode during wet etching, and degradation of the gate material integrity at the high growth temperature. In order to realize better functioning of the CNT emitter, modification of the gate materials should be investigated in a future study. The inset of figure 4(b) reveals the stability of the emission current of the gated CNT emitter. There was little degradation of the emission current in the early stages. However, the fluctuation was about $\pm 10\%$ over 2600 s when the gate voltage was set at 75 V.

4. Summary

In this study, we have demonstrated the feasibility of a new process for fabrication of a microgated CNT emitter which contains a sidewall protector to prevent CNTs from being grown on sidewalls of gate holes. The sidewall protector, formed by the method of tilting the substrate, can enhance the electrical characteristics by suppressing the problem of short circuits between the gate and the CNTs. The leakage current of an emitter with a sidewall protector is approximately sevenfold lower than that of an emitter without a sidewall protector at a gate voltage of 100 V. Although the emission current is relatively low due to degradation of the gate electrode by the high-temperature process, we expect to improve the emission properties by modifying the gate materials and the structural parameters.

Acknowledgments

This work was partially supported by the ITRC Programme of the Ministry of Information and Communication in Korea and IMT 2000 Nano Technology Development Programme of the Ministry of Commerce, Industry and Energy.

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