

DEPENDENCE OF THE ELECTRON EMISSION BEHAVIOUR ON OXIDE LAYER THICKNESS IN THE CASE OF ALLIGNED CARBON NANOTUBES GROWN ON PATTERNED OXIDE LAYERS

B.S.Satyanarayana^{1*}, & Niranjana S²

¹MIT Innovation Center & Electronics & Communication Dept, ²Biomedical Engineering, Manipal Institute of Technology, Manipal, India
*bs.satyanarayana@manipal.edu / satyang1@yahoo.com

Abstract: *The excellent field emission characteristics displayed by carbon nanotubes / nanofibers has led to the continued interest in field emission based displays, besides it's attractiveness for other vacuum microelectronics applications. The usefulness of carbon nanotube based emitters has been enhanced, by the feasibility of selective growth of aligned carbon nanotubes, over patterned metal catalysts. Reported in this paper is the field emission behaviour from carbon nanotubes grown on patterned oxide with out pre deposited patterned catalyst layers. The emission seems to be dependent on the oxide layer thickness. These samples exhibit very field electron emission at fields as low as 2 - 6 V/ μm . Another interesting observation is the possibility of negative differential resistance type behaviour of this field assisted electron emitters under optimum oxide thickness.*

Keywords: Carbon nanotube; patterned silicon dioxide layer; self aligned growth; field assisted electron emission.

Introduction

Carbon nanotubes from the time of it's invention have drawn lot of attention. Further on most counts they seem to have better properties than silicon for electronic applications [1-3]. Field assisted electron emission from carbon based electron emitters seem to be, the first possible carbon nanotube based electronic product to be on the market [4-12]. The key to success of any carbon nanotube based technology would be the development of inexpensive process technology to grow aligned carbon nanotubes on any desired location. Thus continued efforts are on towards developing methodologies to grow controlled and aligned carbon nanotubes at desired locations, a prerequisite for fabricating nanotubes based nanoelectronic devices. Recently there has also been success in growing aligned carbon nanotubes using a combination of growing pre deposited patterned catalyst and then using chemical vapor deposition (CVD) technique to grow carbon nanotubes. The CVD techniques used to grow vary from thermal CVD to RF / DC / Microwave plasma CVD. Growing three-dimensional architectures of carbon nanotubes, which might be integrated into microelectronic circuits, however, still remains a big challenge. Further growing the nanolayers of metal catalyst layers and then growing carbon nanotube calls for greater control and sophistication. The need is for a simple and standard technology for the growth of aligned carbon nanotubes.

Recently Zhang et.al had shown that it could be possible to grow carbon nanotubes selectively on any substrate on patterned oxide layers.[12]. The possibility of field assisted electron emission from aligned carbon nanotubes grown on any desired location is technologically very interesting. Especially if carried out on inexpensive substrate like glass, it would be very useful for quite a few applications based on carbon nanotubes. We had reported some preliminary results emission from carbon nanotubes grown selectively on oxide regions [13]. Presented in this paper, is field assisted electron emission from carbon nanotubes/fibers selectively grown on patterned silicon dioxide, without a pre-deposited metal catalyst layer. The process of growing aligned carbon nanotubes on patterned oxide drastically simplifies the technology as compared to the more conventional practice of patterning metal catalysts to make templates on which nanotubes selectively grown. The only requirement is patterning of the silica layer on silicon substrates, which is an established technology and can be easily extended to the nanometer scale. The key to the success of the emitters is to ensure that the interface between the carbon nanotubes and the back contact. The barrier be as small or negligible as possible, so that the supply of electrons to the emitters is not hindered. Thus the need is to find a balance between the oxide layer needed to grow the aligned carbon nanotubes and also ensure the potential drop at the interface between the emitter and the back contact is as minimal as possible. Hence reported is one of the first study on the influence of varying oxide layer thickness on field emission from aligned carbon nanotubes grown on patterned oxide layer.

Experimental conditions

Highly conducting n++ silicon was chosen as substrates for this experiments. Silicon dioxide layers were then grown on the silicon substrates, using the thermal CVD process. The oxide layer thickness was varied from 15nm to 100nm. The silicon dioxide layers were then subjected to standard lithography techniques to define the area where carbon nanotubes were to be grown on the n++ silicon substrates. The substrates were then supersonically cleaned in acetone, alcohol, and deionized water baths and subsequently, mounted into a tube furnace pumped down to 10^{-2} Torr, backfilled with flowing argon to 1 Torr. After the tube was heated to the deposition temperature of 800 °C, a solution of ferrocene dissolved in Xylene(C_8H_{10}) (the ratio of ferrocene/xylene is variable) was prevaporized at about 150 °C and introduced into the gas stream. Ferrocene

(Fe (C₅H₅)₂) was used as the catalyst precursor for the growth of the aligned carbon nanotubes were then grown on the substrates. The morphology of the carbon nanotubes grown were examined by scanning electron microscopes (SEM). Field emission measurements have been carried out in a parallel plate configuration, where the cathode consists of the carbon nanotube films to be tested and the anode was either an ITO coated glass plate or a metal disc. The I-V measurements were made with an anode –cathode spacing of 200 μm and a vacuum of 10⁻⁵ pascal. No conditioning or forming process was required to initiate the emission.

Result and Discussion

Carbon nanotubes were deposited on silicon substrates with varying thickness from 15 to 100nm. Shown in figure 1(a) is the SEM image of the as deposited growth of Carbon nanotubes on the patterned area over a 50nm thick oxide layer. Shown in figure 1(b) is a closer view o the same carbon nanofibers.

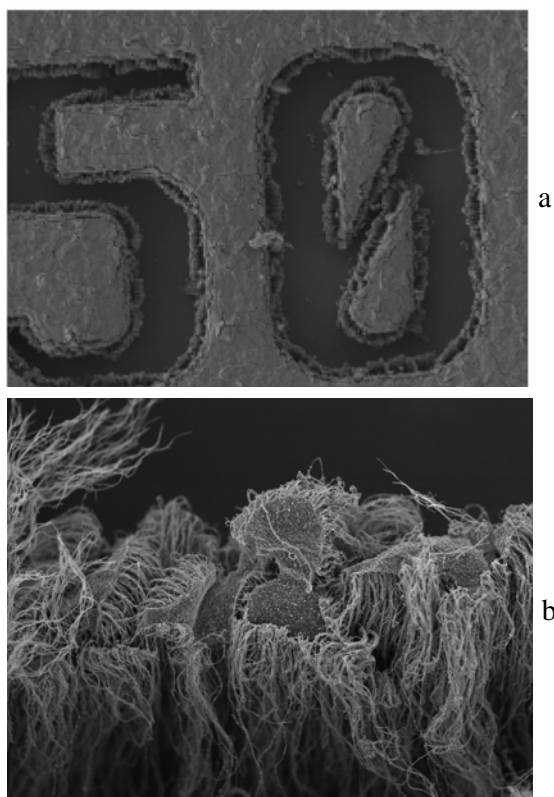


Figure 1. SEM images of the as deposited carbon nanotubes on silicon substrate with a pre deposited oxide layer thickness of 50 nm (a) Image of the nanotubes grown on only the patterned oxide region b) A closer image of the carbon nanotubes

The field emission measurements on the carbon nanotubes grown on oxide layers of varying thickness clearly show that the threshold field is influenced by the

oxide layer. The threshold field is defined as field at which the measured emission current density is 1μA/cm². The threshold field was observed to vary from 1V/μm to 6 V/μm, when the oxide thickness was varied from 15nm to 100nm. Shown in figure 2 is the variation of the emission threshold field with the oxide layer thickness.

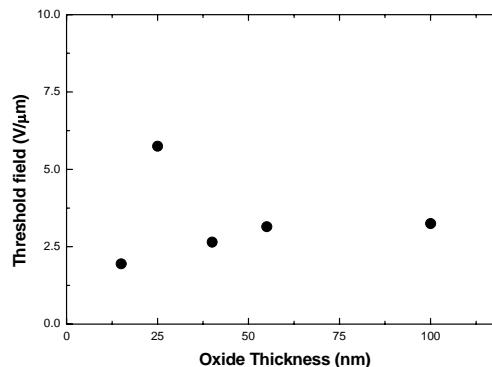


Figure2. Variation of the emission threshold field with varying oxide layer thickness.

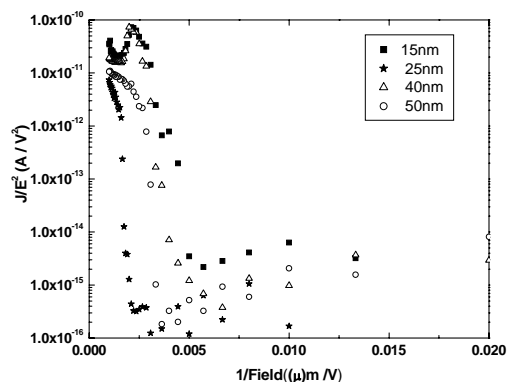


Figure 3. Fowler Nordheim plot of the field assisted electron emission behaviour from the carbon nanotubes grown on patterned silicon dioxide of varying thickness.

Shown in figure 3 is the Fowler Nordheim plot of the field assisted electron emission behaviour from the carbon nanotubes grown on patterned silicon dioxide of varying thickness. It can be seen from the figure that at higher thickness of the oxide the FN plot slope is sharper. In the case of thinner oxide layer there seems to be some space charge phenomenon taking place at higher fields as can be seen from the non linearity at the top. Another interesting feature of the field emission behaviour of these nanocarbons is the observation of a behaviour similar to negative differential resistance in the emission current Vs applied voltage plot, as can be seen from figure 4. We have earlier reported similar behaviour in the case of nanodiamond and nanocluster carbon heterojunctions or multilayers [14].

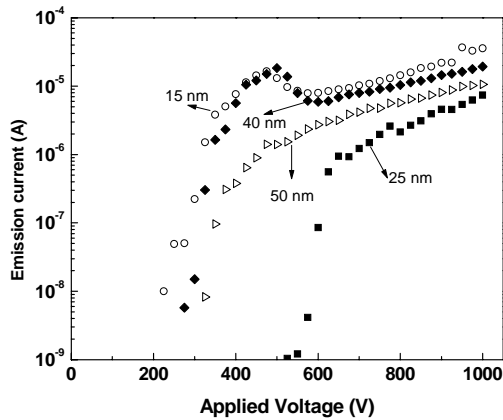


Figure 4. Field assisted electron emission current Vs Applied Voltage plot showing possible negative differential resistance type behaviour in the case of multilayered cathodes consisting of a thin oxide layer and carbon nanotube on the top.

In the earlier case the heterostructure cathode consisting of nanocluster carbon (small band gap – high conductivity) and nanocrystalline diamond (large band gap – low conductivity), with the two nanostructured materials lying side by side laterally or vertically above each other in a sandwich configuration. Under optimum ratio of the two nanostructured material it might act like the quantum well structures consisting of alternate band gap material. In the presence of an high field when the barriers become finite for these small grained materials, the electrons could become quasi-bound or resonant. Thus now the electron may have a very high transmission probability, leading to very high currents at sharply defined values of the external field. We may think of a similar analogy in the case of the 15 to 40 nm silicon dioxide and carbon nanotubes on the top of it. However this needs to be studied further. The observance of this resonant tunneling effect or the negative differential effect at room temperature in this carbon based system, could be very interesting for future nanoelectronics and vacuum nanoelectronics. To our knowledge, this is the one of the first report of observation of such a behaviour in the case of carbon nanotube and silicon dioxide multilayered system. Observance of this resonant tunneling effect or the negative differential resistance effect at room temperature in this carbon based system, could be very interesting for future nanoelectronics and vacuum nanoelectronics. To our knowledge, this is the one of the first report of observation of such a behaviour in the case of carbon nanotube and silicon dioxide multilayered system.

Thus it may be seen that it should be possible to grow aligned carbon naotubes on any substrate as the growth temperature becomes lower, when using the oxide templates to grow carbon nanotubes. The emission behaviour seems to suggest the existence of a linear

relation with the oxide thickness in the measured range of the samples. This process of growth offers a very good opportunity to grow carbon nanotubes on any substrate including glass substrates, on just patterned silicon dioxide. The oxide layer may also act as the current limiting layer under optimum conditions. Thus limiting the probability of carbon nanotubes burning due to high current density. With further optimization the aligned carbon nanotubes grown on patterned oxide layers could be extremely useful in vacuum nanoelectronics. Another interesting observation is the negative differential resistance type behaviour. This also need further analysis.

Acknowledgment

We would like to thank and acknowledge Prof.Ajayan and Dr.Weii from Rensselaer Polytechnic Institute, USA, for kindly providing the samples for this study and for the useful discussions.

Conclusion

Low field electron emission has been reported from carbon nanotubes grown on patterned oxide substrates. The emission threshold varied from 2 V/ μm to 6 V/ μm as the oxide thickness was varied from 15nm to 100nm. The F-N plot slope also suggests that at lower oxide thickness, at higher fields, the emission is non-linear. The oxide layer may also act as the current limiting layer under optimum conditions. With further optimization the aligned carbon nanotubes grown on patterned oxide layers could be extremely useful in vacuum nanoelectronics. The observed behaviour similar to resonant tunnelling in some samples is also very interesting.

Reference

1. M.Terrones,A.Jorio,M.Endo,A.M.Rao,Y.A.Kim,T.Hayashi,H.Terrones,J.C.Charlier,G. Dresselhaus and M.S.Dresselhaus, “ New Directions in Nanotube Science”, *MaterialsToday*, p-30, Oct 2004.
2. NirajSinha,JohnT,W.Yellow,“Carbon Nanotubes for biomedical Applications” :*IEEE Transaction on Nanobioscience*,Vol.4, 180, 2005.
3. Walt A De Heer, “Nanotubes and Pursuit of Applications”,*MRS Bulletin*,April,2004,281.
4. W.A.de Heer, A.Chatelain, and D.Ugrate, “A Carbon nanotube field emission electron source” *Science* **270**, 1179 (1995).
5. Y.Chen, S.Patel, Y.Ye, D.T.Shaw & L.Guo, “field emission from aligned high density graphitic nanofibres” *App. Phys. Lett*, 73, 2119 (1998).
6. Zhu, G P Kochanski & S Jin, “Low field electron emission from undoped nanostructured diamond” *Science*, 282, 1471, (1998).
7. Obratsov, I.Yu.Pavlovsky and A.P.Volkov, E.D. Obratsov, A.L.Chuvilin, V.L.Kuznetsov “Aligned

B.S.Satyanarayana

- carbon nanotubes for cold cathode applications” . J. Vac. Sci. Technol. **B 18** , 2000, 1059 - 1063,
8. B.S.Satyanarayana, J.Robertson, W.I.Milne, “Low Threshold Field Emission From Nanoclustered Carbon Grown by Cathodic Arc”,*J. App. Phys.* **87**, 3126, 2000.
 9. N.S.Xu and S.Ejaz Huq, “Novel cold cathode materials and applications”, *Material Science. & Engg. R* **48**, 47, 2005.
 10. Hiraki.A ,B.S.Satyanarayana, “Field emission from multylayered Carbon films consisting of nanoseeded Diamond and nanocluster carbon Deposited at Room temperature on glass substrate”, *IEICE*,Vol.**E 86-C**, 816,2003.
 11. B.S.Satyanarayana, X.L.Peng, G.Adamapolous, J.Robertson, W.I. Milne, and T.W. Clyne, “Very Low Threshold Field Emission from Microcrystalline Diamond films grown using Hot Filament CVD process” ,*MRSSymp. Proc.*,Vol.621, Q.5.3.1, 2000.
 12. Z. J. Zhang, B. Q. Wei, G. Ramanath, P. M. Ajayan, “Substrate site selective growth of aligned acrbon nanotubes” *Appl. Phys. Lett.* 2000, 77, 3764.
 13. B.S.Satyanarayana,B.Q.Wei,Y.Jung, G.Ramanath and P.M. A.Jayan, “Field Emission from aligned carbon nonotubes grown on patteredned oxide layers”, *16th IVMC Proceedings*, p 209,2003.
 14. B.S.Satyanarayana & A.Hiraki. “Resonant tunnelling behaviour in multilayered cold cathodes made of nanoseeded diamond and nanocluster carbon”. *MRS Symp.Proc.* Vol 676, 2001, pY.8.42.1-5..