

Synthesis of Carbon Nanotube Using Olive Oil and It's Application in Dye Sensitized Solar Cell

Swati Sharma*, Pinaki Ranjan**, Sthitadhi Das**, Sushil Gupta**,
Ravinder Bhati**, Dr. Amitava Majumdar**

*Nanotechnology Department, Amity Institute of Nanotechnology(AINT),Amity University,Sec- 125

**Corporate R&D,Chemical and Solar Energy Lab,Moserbaer India Ltd,Moserbaer66 udyog vihar Greater

‡Corresponding Author; Swati Sharma,Nanotechnology Department, Amity Institute of Nanotechnology(AINT),
Amity University,Sec- 125 Noida, India, sharma9.swati@gmail.com

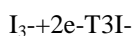
Received: 02.03.2012 Accepted:07.04.2012

Abstract- In this study we report the synthesis of carbon nanotubes using Green chemistry (Olive Oil) for application in dye-sensitized solar cells (DSSCs) as counter electrode materials (cathode). Prior research has resulted in using platinum deposited over transparent conducting oxide (TCO) as a cathode in DSSC to achieve high efficiency cells. However, the degradation of platinum over time poses a significant challenge in identifying the right material to maintain efficiency and operation over time. In this regard, the high conductivity and transparency of CNTs make them a potentially attractive option so for the purpose of increasing the energy conversion efficiency of DSSCs, carbon nanotubes were deposited on a TCO and the efficiency of CNT-counter electrode DSSC was improved .Nanotubes catalyze the reduction of triiodide, a reaction that is important for the DSSC.

Keywords Carbon Nanotube, Olive oil, Carbon Vapour Deposition, Dye-sensitized Solar Cell, Counter Electrode

1. Introduction

In this study, we investigated the performance of DSSCs with the fast electron transfer kinetics and the large surface area of multi-wall CNTs on the counter electrode. It has began to attract considerable interest for electrocatalytic applications. Many efforts have been made to apply nanotubes to the photoanode and counter electrodes[1,2]. The DSSC consists of dye molecules chemically adsorbed on the surface of a TiO₂ nanoparticle network anode and a contacting redox active electrolyte, most commonly a solution containing iodide and triiodide molecules[3,4].The cell anode serves as the current collector for the photogenerated electrons in the nanoparticle network, while the cathode injects charge into the electrolyte and catalyzes the reduction of triiodide:



The cathode is commonly made from a thin (1-5 nm) layer of platinum catalyst deposited on a TCO such as fluorine tin oxide (FTO) on glass or indium tin oxide (ITO) on a glass or polymer substrate. A transparent cathode allows

for straightforward use of lowcost, flexible substrates because the TiO₂ films can be sintered at high temperature on an opaque metallic film. A transparent cathode can also allow for construction of a tandem cell containing two or more separate compartments with dye molecules that absorb different parts of the solar spectrum, to achieve high efficiency cells and lower the cost of generated electricity [5].

Previous work has shown that carbon nanotubes also catalyze the reduction of triiodide and thus may be able to replace platinum in the DSSC [6]. In this context, carbon nanotube films have several advantages. Platinum was found to degrade over time while in contact with an iodide/triiodide liquid electrolyte, reducing the efficiency of a DSSC, whereas carbon nanotubes did not degrade [7] and enhance the efficiency of the cell.

Carbon species such as methane, acetylene, benzene, xylene, toluene,etc., have been used as a carbon feedstock to synthesize CNTs [8-14]. These carbon precursors are non-renewable resources and are depleting with time so it is necessary to develop carbon precursor from natural

resources. The reported natural precursors are : camphor ($C_{10}H_{16}O$), turpentine oil ($C_{10}H_{16}$), eucalyptus oil ($C_{10}H_{18}O$) and palm oil ($C_{67}H_{127}O_8$) for synthesis of CNTs [15-23]. These natural precursors are inexpensive and in near future have no chance of shortage. The technique which we have used is carbon vapour deposition (CVD) which is a simple and economic technique for synthesizing CNTs at low temperature and ambient pressure. In yield and purity, CVD beats the arc and laser methods and when it comes to structure control or CNT architecture, CVD is the only answer. CVD is versatile in the sense that it offers harnessing plenty of hydrocarbons in any state (solid, liquid or gas), and allows CNT growth in a variety of forms, such as powder, thin or thick films, aligned or entangled, straight or coiled nanotubes, or a desired architecture of nanotubes on predefined sites of a patterned substrate. It also offers better control on the growth parameters [24].

Advantages of CNTs such as electrical conductivity, chemical stability, high surface area, and optoelectronic properties that make them excellent candidates for a variety of energy conversion and storage technologies.

In this article, we report the synthesis of aligned CNTs bundles using olive oil as the carbon source (Precursor) using CVD technique. Among the new CNT precursors, olive is highlighted because it is a natural source which is renewable, environment-friendliness and has the potential to be the green alternative for industrial scale production of CNTs.

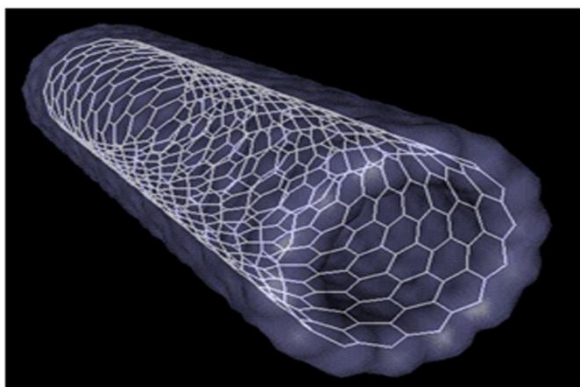


Fig. 1. Carbon Nanotube

2. Experiment

2.1. Synthesis of Carbon Nanotube

For the thermal CVD process, An electric furnace with one meter long quartz tube kept inside the furnace has been used. The quartz tube was flushed with Nitrogen (N_2) gas. The role of Nitrogen was: (a) removing O_2 from the furnace at the first minutes before putting olive oil into the main chamber, (b) carrier gas for Olive oil, and (c) cool the system after the reaction. The mixture of olive oil and ferrocene was heated at $300^\circ C$ in flask and its vapour was passed inside quartz tube with the help of N_2 gas. The flow rate of N_2 was 100 sccm. The experiments were conducted at different temperature ($750-850^\circ C$) at atmospheric pressure, with a typical reaction time of 40min for each deposition. After deposition, the furnace was switched off and allowed to cool

down to room temperature under N_2 gas flow. A uniform black deposition on the inner wall of the quartz tube inside furnace was observed. The black deposition in the form of carbon soot was taken out from the quartz tube. Fig. 2 shows the schematic synthesis setup for the synthesis of CNT:

As-grown carbonaceous materials were characterized by using, Scanning electron microscope (SEM), Atomic force microscope (AFM), Optical microscope. For SEM observation, the black soot-like material was directly mounted to the sample holder (STUB) with Carbon tap which is electrically conductive. The electrons interact with the atoms that make up the sample producing signals that contain information about the sample's surface topography. For AFM observation it consists of a cantilever with a sharp tip (probe) at its end that is used to scan the specimen surface.

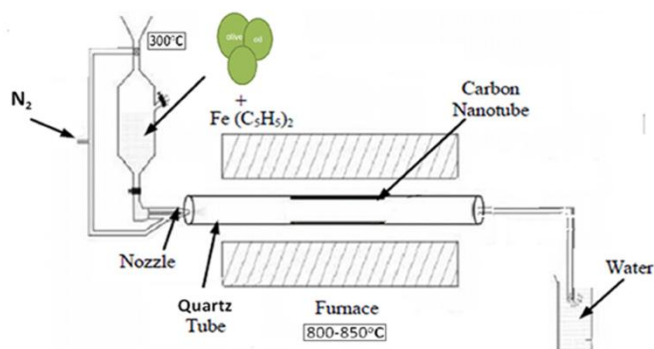


Fig. 2. Schematic synthesis setup for the synthesis of CNT

2.2. Preparation of Carbon Nanotube Paste

For making carbon nanotube paste the procedure is: 9.2 gm of carbohydrate binder dissolved in 50 gm of alpha-Terpineol at $80-85^\circ C$ until clean solution obtained under high speed stirrer and keep it for 30 mins then cool to room temperature.

Now weight 3.2 gm of wetting agent (triton X-100) and pour into above solution as stirrer for 30 mins at room temperature then take 0.5 gm of above mix and 0.05 gm of CNT's and pour part by part and grind by motor pastel for 3-4 Hrs for deagglomeration or deaggregation.

When the paste becomes uniform apply on glass plate for DSSC fabrication.

2.3. Fabrication of Dssc

DSSC is a photo electrochemical cell that consists of two electrodes acting as anode and cathode. On the top is a transparent anode made of fluorine-doped tin dioxide ($SnO_2:F$) deposited on the back of a (typically glass) plate. One coat of TiO_2 paste which shows the particle size range of (30-50) nm and loading of 20 % was applied to the conducting side of the anode glass electrode. On the back of the conductive plate is a thin layer of titanium dioxide (TiO_2), which forms into a highly porous structure with an extremely high surface area. TiO_2 only absorbs a small fraction of the solar photons (those in the UV).

The plate is then immersed in a mixture of a photosensitive ruthenium-polypyridine dye (also called molecular sensitizers) and a solvent. After soaking the film in the dye solution, a thin layer of the dye is left covalently bonded to the surface of the TiO₂. A separate backing is

made with a thin layer of the carbon nanotube spread over a conductive sheet, typically. The front and back parts are then joined and sealed together to prevent the electrolyte from leaking.

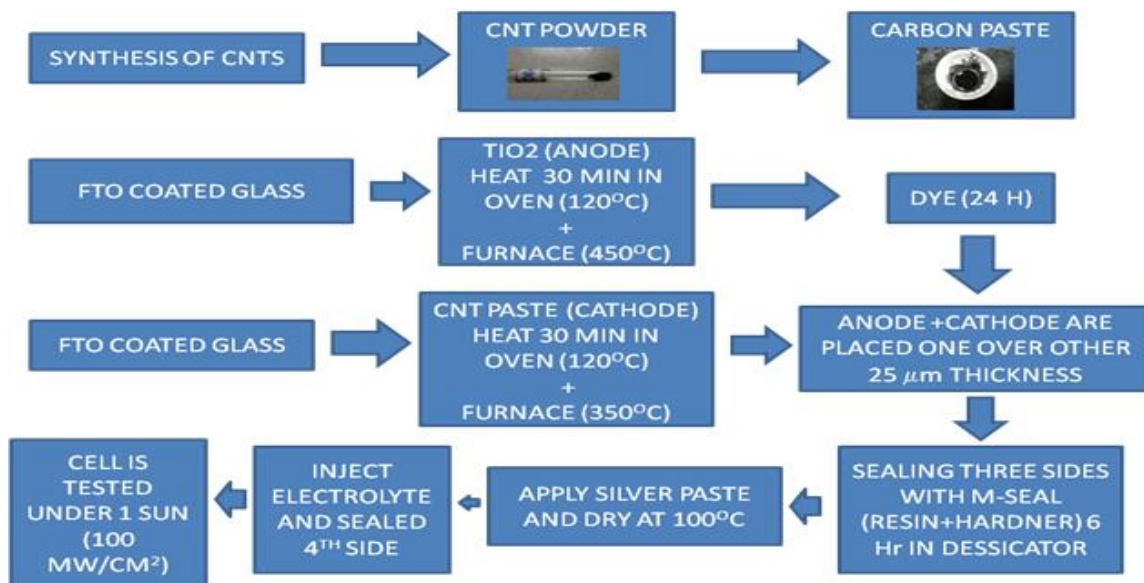


Fig. 3. Flow diagram of whole process

3. Result and Discussion

3.1. Scanning Electron Microscope

Sample: 1

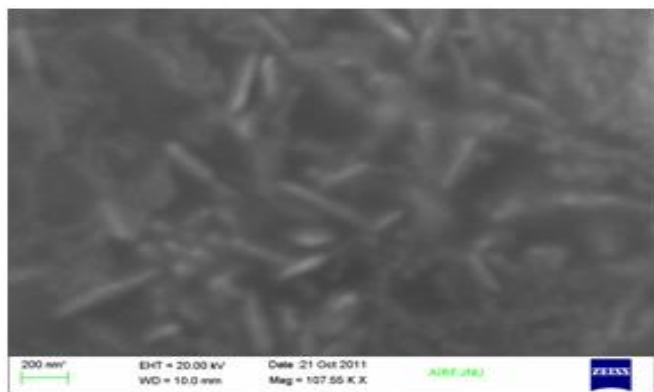


Fig. 4. SEM Image of Sample 1

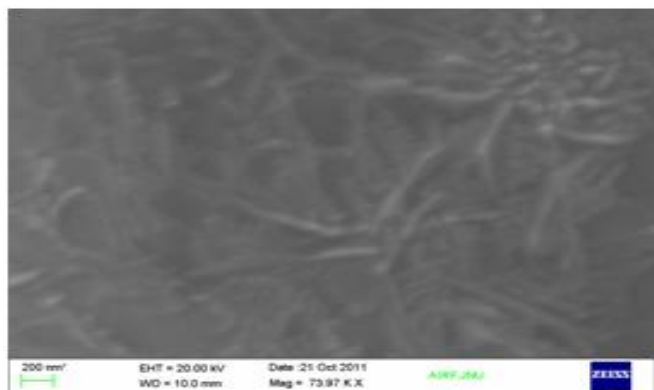


Fig. 5. SEM Image of Sample 1.

Fig. 4, 5. shows the SEM images of carbon nanotube from sample 1 which is of 200 nm.

Sample: 2

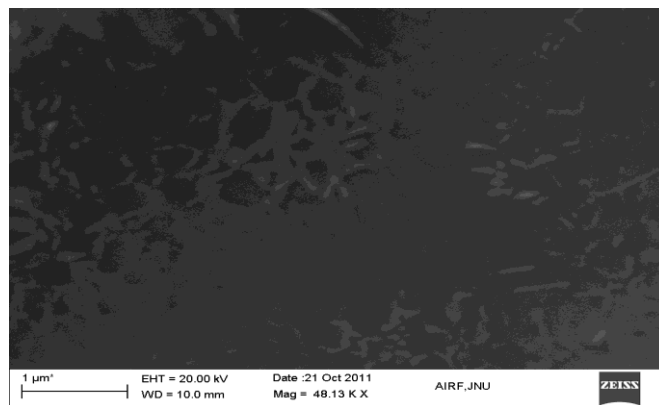


Fig. 6. SEM Image of Sample 2.

Sample: 3

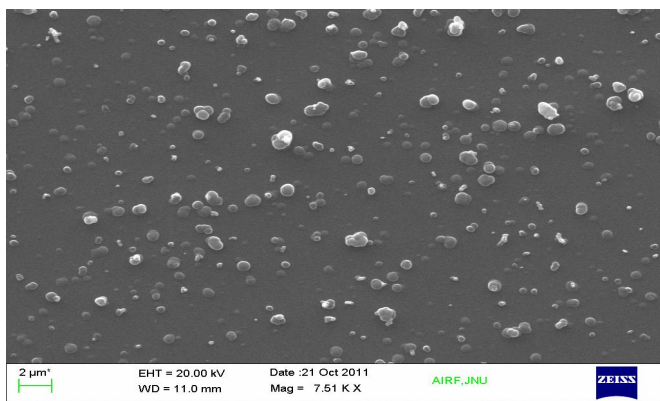


Fig. 7. SEM Image of Sample 3.

Fig. 6,7. shows the SEM images of carbon nanotube from sample 2 and sample 3 which is of 1μm and 2μm

3.2. Atomic Force Microscope

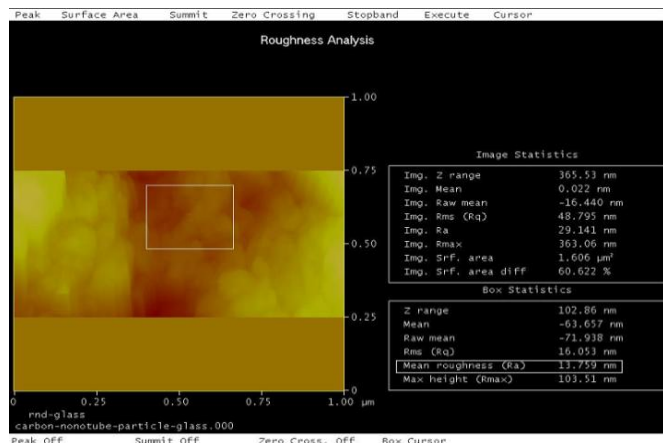


Fig. 8. MIN RA Mean Roughness 13.759 nm

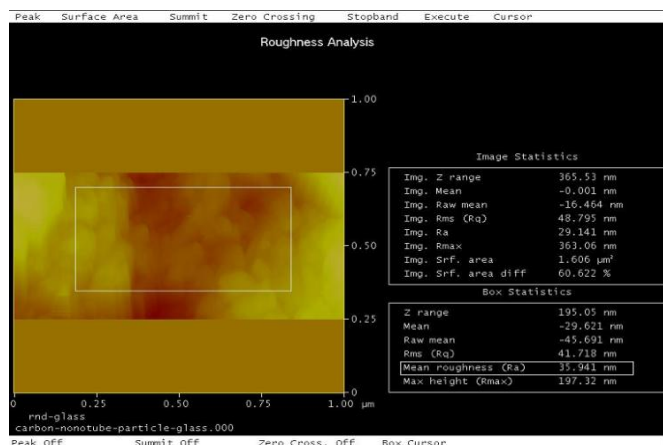


Fig. 9. MAX RA Mean Roughness 35.941 nm

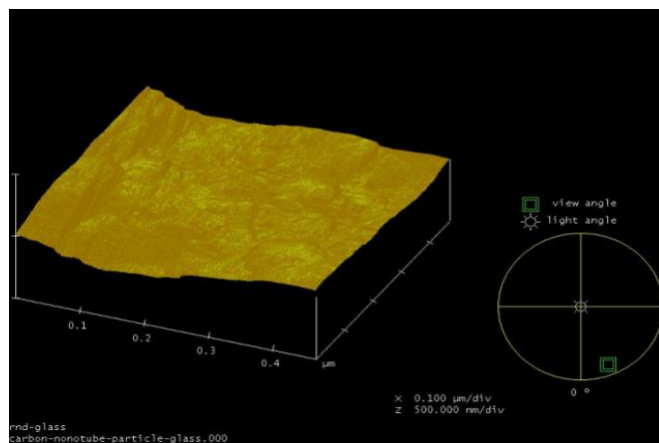


Fig. 10. 3D view

3.3. Optical Microscope

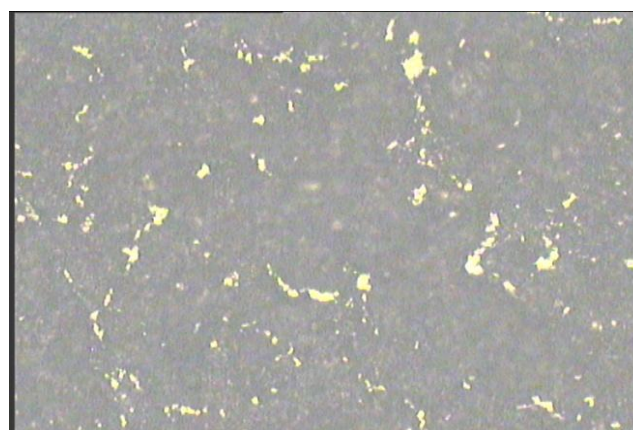


Fig. 11. Optical Microscope Image shows the uniformity of the paste.

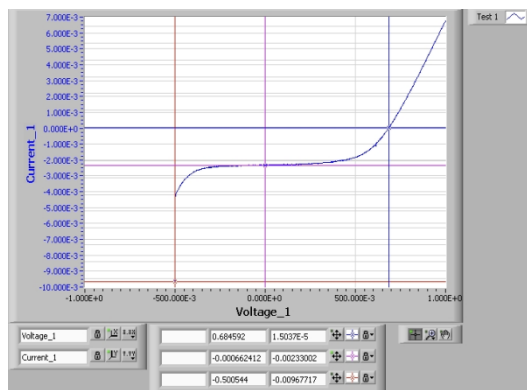
3.4. Results of Dssc

Current-voltage (IV) curves represent the most important and most direct characterization method for DSSCs. The open circuit voltage (V_{oc}), short circuit current (I_{sc}) and the shape of the IV curve determine the efficiency (η) of DSSCs under given light condition. There is one more additional important descriptors for IV curves of solar cells i.e. fill factor which describes how well the area under the IV curve “fills in” the maximum possible rectangle defined by $I_{sc} \times V_{oc}$.

With an addition of CNT, the circuit current increased. It was confirmed that CNT shows the improved material and efficiency as compared to TiO_2 .

3.5. The Test Results of Last Three Samples

Sample: 1

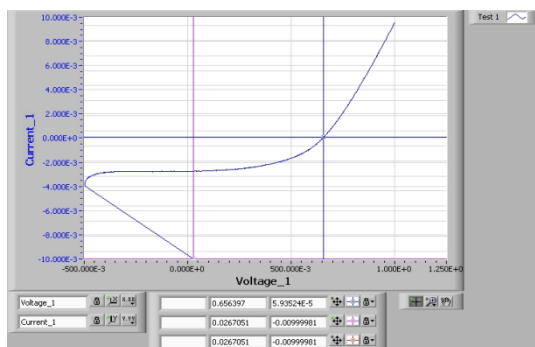


Graph 1. Characteristic of sample 1.

Table 1. Sample 1.

V_{oc} (mV)	0.694	
I_{sc} (mA/cm ²)	0.00194	
FF	0.583916	58.39158
Eff (%)	0.033398	3.339765

Sample: 2

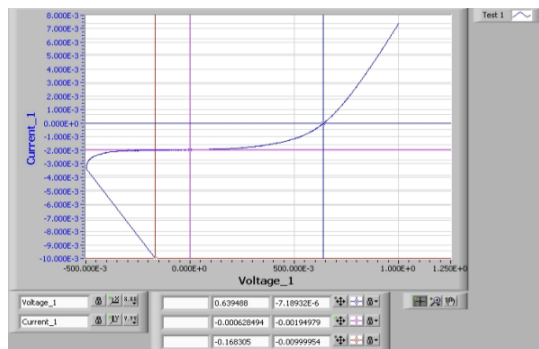


Graph 2. Characteristic of sample 2.

Table 2. Sample 2.

V_{oc} (mV)	0.693	
I_{sc} (am/cm ²)	0.00276	
FF	0.486008	48.60083
Eff (%)	0.035641	3.564148

Sample: 3



Graph 3. Characteristic of sample 3.

Table 3. Sample 3.

V_{oc} (mV)	0.653	
I_{sc} (am/cm ²)	0.00376	
FF	0.496008	49.60083
Eff (%)	0.037641	3.764148

4. Conclusion

In , summary we have shown that carbon nanotube Carbon nanotubes prepared from olive oil may adhere to each other, forming assemblies whose morphology or structure shows catalytic properties. In an electrostatically stabilized dispersion charges generated on the surface of particles prevent or control reagglomeration. Steric stabilization takes place when large molecules adsorb on to the surface of particles thus introducing physical barriers between them. A combination of electrostatic and steric mechanisms produces electrosteric stabilization. All three stabilization mechanisms prevent nanotubes from agglomeration. The micro structural changes of electrodes prepared from different method do not affect the cell efficiency significantly, implying that beside paste composition and layer morphology there are other factors (e.g. the presence of surface-states which leads to trapping), which limit the rate of the overall charge carrier transport process.

The efficiency of solar cell fabricated with CNT's as catalyst showed improved efficiency as compared to Platinum catalyst.

References

- [1] K.Y. Chun, B.W. Paek, Y.M. Sung, D.J. Kwak, Y.T. Hyun, M.W. Park, Thin Solid Films 517 (2009) 4196.
- [2] Y. Ohsaki, N. Masaki, T. Kitamura, Y. Wada, T. Okamoto, T. Sekino, K. Niihara, S. Yanagida, Phys. Chem. Chem Phys. 7(2005) 4157.
- [3] O'Regan, B.; Grätzel, M. Nature 1991, 353, 737.
- [4] Wang, Q.; Ito, S.; Gratzel, M.; Fabregat-Santiago, F.; Mora-Sero, I.; Bisquert, J.; Bessho, T.; Imai, H. J. Phys. Chem. B 2006, 110, 25210.
- [5] Durr, M.; Bamedi, A.; Yasuda, A.; Nelles, G. Appl. Phys. Lett. 2004, 84, 3397.
- [6] Suzuki, K.; Yamaguchi, M.; Kumagai, M.; Yanagida, S. Chem. Lett. 2003, 32, 28.
- [7] Koo, B.-K.; Lee, D.-Y.; Kim, H.-J.; Lee, W.-J.; Song, J.-S.; Kim, H.-J. J. Electroceram. 2006, V17, 79.
- [8] Iijima S: Helical microtubules of graphitic carbon. Nature 1991, 354:56.
- [9] Li WZ, Xie SS, Qian LX, Chang BH, Zou BS, Zhou WY, Zhao RA, Wang G: Large-Scale Synthesis of Aligned Carbon Nanotubes. Science 1996,274:1701.
- [10] Ren ZF, Huang ZP, Xu JW, Wang JH, Bush P, Siegal MP, Provencio PN: Synthesis of Large Arrays of

- Well-Aligned Carbon Nanotubes on Glass. *Science* 1998, 282:1105.
- [11] Sen R, Govindaraj A, Rao CNR: Carbon nanotubes by the metallocen route. *Chem. Phys Lett* 1998, 277:276.
- [12] Zhang ZJ, Wei BQ, Ramanath G, Ajayan PM: Substrate-site selective growth of aligned carbon nanotubes. *Appl Phys Lett* 2000, 77:3764.
- [13] Mayne M, Grobert N, Terrones M, Kamalakaran R, Ruhle M, Kroto HW, Walton DRM: Pyrolytic production of aligned carbon nanotubes from homogeneously dispersed benzene-based aerosols. *Chem Phys Lett* 2001,338:101.
- [14] Singh C, Shaffer MSP, Windle AH: Production of controlled architectures of aligned carbon nanotubes by an injection chemical vapour deposition method. *Carbon* 2003, 41:359.
- [15] Sharon M, Hsu WK, Kroto HW, Walton DRM: Camphor-based carbon nanotubes as an anode in lithium secondary batteries. *J Power Sources* 2002, 104:148.
- [16] Chatterjee AK, Sharon M, Banerjee R, Spallart MN: CVD synthesis of carbon nanotubes using a finely dispersed cobalt catalyst and their use in double layer electrochemical capacitors. *Electrochemics Acta* 2003, 48:3439.
- [17] Afre RA, Soga T, Jimbo T, Kumar M, Ando A, Sharon M: Growth of vertically aligned carbon nanotubes on silicon and quartz substrate by spray pyrolysis of a natural precursor: Turpentine oil. *Chem Phys Lett* 2005, 414:6.
- [18] Afre RA, Soga T, Jimbo T, Kumar M, Ando A, Sharon M, Somani PK, Umen M: Carbon nanotubes by spray pyrolysis of turpentine oil at different temperatures and their studies. *Microp Mesop Mater* 2006,96:184
- [19] Kumar M, Ando Y: A simple method of producing aligned carbon nanotubes from an unconventional precursor - Camphor. *Chem Phys Lett* 2003, 374:521.
- [20] Kumar M, Ando Y: Camphor-a botanical precursor producing garden of carbon nanotubes. *Diamond Relat Mater* 2003, 12:998.
- [21] Kumar M, Ando Y: Controlling the diameter distribution of carbon nanotubes grown from camphor on a zeolite support. *Carbon* 2005, 43:533.
- [22] Ghosh P, Afre RA, Soga T, Jimbo T: A simple method of producing singlewalled carbon nanotubes from a natural precursor: Eucalyptus oil. *Mater Lett* 2007, 61:3768.
- [23] Suriani AB, Azira AA, Nik SF, Nor RM, Rusop M: Synthesis of vertically aligned carbon nanotubes using natural palm oil as carbon precursor. *Mater Lett* 2009, 63:2704.
- [24] Mukul Kumar. and Yoshinori Ando Department of Materials Science and Engineering, Meijo University, Nagoya 468-8502, Japan Vol.10.3739- 3758,2010.