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Fabrication of CNT-ZnO based Nanostructures for the Enhancement of Field Emission Properties

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Abstract

CNTs are the material of 21st century which possess excellent mechanical, chemical, electrical, thermal and field emission properties. SWCNT's unique tip type geometry when aligned vertically gives an excellent field emission which makes this material suitable candidate for designing high quality field emission based display devices. Present research work includes synthesis and optimisation of CNT-ZnO nanostructures for the enhancement of field emission performance. CNTs were successfully synthesized for this purpose using Plasma enhanced chemical vapor deposition (PECVD) method. The surface morphology was observed through field emission scanning electron microscopy (FESEM) and internal structures were observed through high resolution transmission electron microscopy (HRTEM). The diameter distribution was confirmed through Raman Spectroscopic analysis which fairly fell in the nanometer range of 1-2nm. Nanostructures were synthesized using varied ZnO wt% starting from 0%, 2.0%, and 6.0% to 8.0%. Field emission analysis of as synthesized nanostructures was carried out under the framework of well-known Fowler-Northeim theorem. Field Emission study revealed that maximum current density was obtained up to 15 mA/cm^2 at a turn on field of 1 V/ µm with field-enhancement factor 3.6×10^4 was achieved for VA-SWCNT-ZnO with 8.0 wt% ZnO.

Keywords: CNT-ZnO Nanostructures; RF-Sputtering, PECVD; RTCVD, Field Emitter.

1. Introduction

Carbon nanotubes are the material of 21th century and are the demanding material as per the application point of view [1-5]. They possess unique structure and are used widely in different applications. They are seamless cylinders having various forms like single wall, double wall and multi wall carbon nanotubes. Every structure is unique in its domain of applications. The main applications covered by CNTs are field emitters, gas sensors, super capacitors etc. [6-10]. The vertically aligned carbon nanotubes (VA-CNTS) are emerging material in field emission device applications. The tip type geometry of vertically aligned carbon nanotubes (VA-CNTS) is responsible for excellent field emission [11-16]. It is easy to emit electrons from the tip type geometry of CNTs. The CNTs provide excellent field emission on low turn on voltage [17-20]. Many scientific communities try to attach many other nano particles for high quality of field emission [21-26]. The nanoparticles include copper (Cu), silver (Ag), ZnO and gold (Au). These particles are attached for the enhancement of field emission and gas sensor properties. The nanoparticles lower the turn on voltage which enhances the field emission [17-30].

Vertically aligned single wall carbon nanotubes (VA-SWCNTs) are highly efficient for field emission device applications [31-36]. Mostly vertically aligned single wall carbon nanotubes (VA-SWCNTs) prepared through plasma enhanced chemical vapor deposition are excellent candidates for these suitable applications. Excellent vertical structures are being fabricated through this technique. This technique is considered as the low temperature technique for the growth of VA-SWCNTs [37-38]. Through this technique, a good optimization is possible for the proper growth of VA-SWCNTs [39-42]. However to enhance field emission properties of VA-SWCNTs, ZnO nanoparticles have been attached through rapid thermal evaporation technique (RTCVD) [43-48]. High quality and uniform distribution of ZnO particles have been obtained through this technique [49-56]. As per the literature survey, pure/ pristine VA-SWCNTs are less effective in field emission as compared to ZnO decorated VA-SWCNTs 57-60].

Here an excellent approach has been applied to increase the field emission of as grown VA-SWCNTs. Hybrid VA-SWCNT-ZnO nanostructure were fabricated and analyzed for field emission applications. Excellent structure of VA-SWCNT-ZnO nanostructures was observed through FESEM and HRTEM. The diameter distributions of VA-SWCNT-ZnO nanostructures were around 1-2nm which was confirmed through Raman Spectroscopy [61-70]. The varying wt % of ZnO was 0.0%, 2.0%, 6.0% and 8.0%. Field emission properties were investigated from the as grown samples. An excellent enhancement was observed through ZnO wt % (8.0%) [71-80].

2. Experimental

The single wall carbon nanotube was fabricated by plasma enhanced chemical vapor deposition technique (PECVD). This technique involves two steps. First step involves the deposition of Fe (iron) catalyst on the Silicon (Si) substrate through RF- sputtering technique and then it follows the growth of SWCNT by PECVD technique. The SWCNTs were grown by PECVD technique which is considered as low temperature growth of CNTs. The CNTs grown through this technique are highly efficient for various applications like gas sensors and field emission devices.

2.1. Iron (Fe) Catalyst Deposition

Fe particles on Si substrate were deposited through R-F Sputtering Technique. Suitable sizes of Si substrate were ultrasonically cleaned with acetone for 50 minutes and dried. These suitable sizes of Si substrate were loaded in R-F sputtering chamber for the deposition of Fe catalyst particles. A uniform thin film of Fe was deposited on Si substrate through this technique. The technique has been shown in fig. 1.

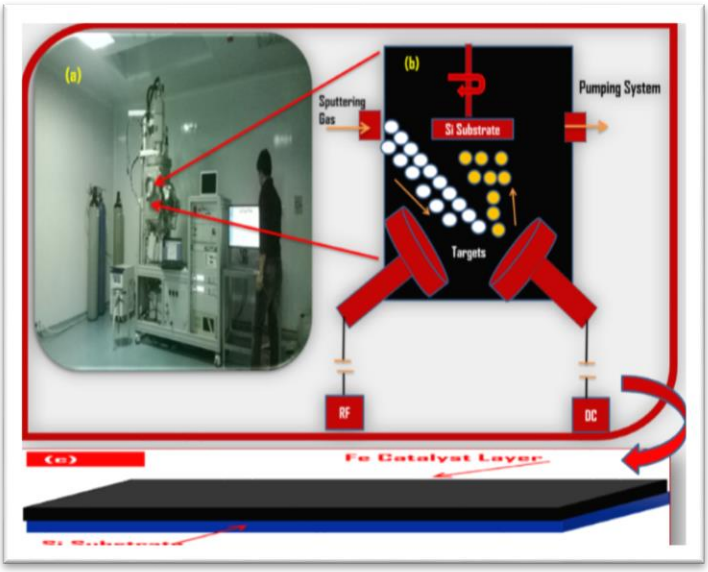

Fig.1. **(a)** RF-Sputtering Unit

2.2. Growth of SWCNTs through PECVD technique

The Fe Si deposited substrate was subjected into the chamber of PECVD for the growth of SWCNTs. A good vacuum of the order of 10^{-3} torr has been raised in the chamber of PECVD and the temperature was maintained about 600^0C. At this temperature the Fe catalyst brakes down in Nano Island and creates the sites for the growth of SWCNTs. The acetylene C_2H_2 was inserted into the ball jar with the flow rate of 20 sccm through the shower head.

The source gas C_2H_2 gas dissociate into its components and the carbon atoms are being nucleated from the Fe based surface of Si substrate. From these Fe based Nano-island, SWCNT growth take place. The growth time was maintained about 10min. The System was cool downed and the grown SWCNTs were achieved as shown in fig.2.

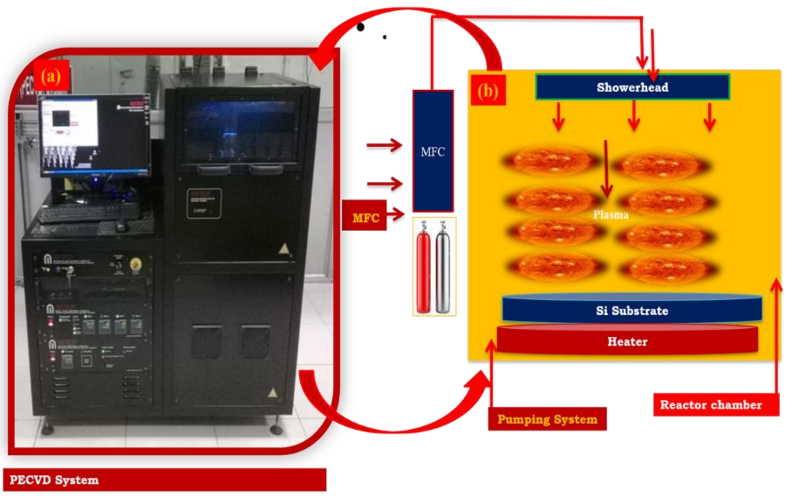

Fig.2. **(a)** PECVD unit **(b)** Mechanism of PECVD unit

2.3. Growth of VA-SWCNT-ZnO Nanostructures

The surface of as grown VA-SWCNTs were decorated with ZnO nanoparticles and VA-SWCNT-ZnO hybrid nanostructures were obtained. The ZnO nanoparticles were decorated through rapid thermal evaporation technique (RTCVD). The as grown SWCNTs were subjected to RTCVD chamber a uniform thin film of ZnO particles was obtained. The vacuum in the chamber was about 10^{-6} torr.

From the as grown VA-SWCNTs, VA-SWCNT-ZnO hybrid nanostructures were grown through for further applications. The RTCVD chamber used has been shown in fig.3.

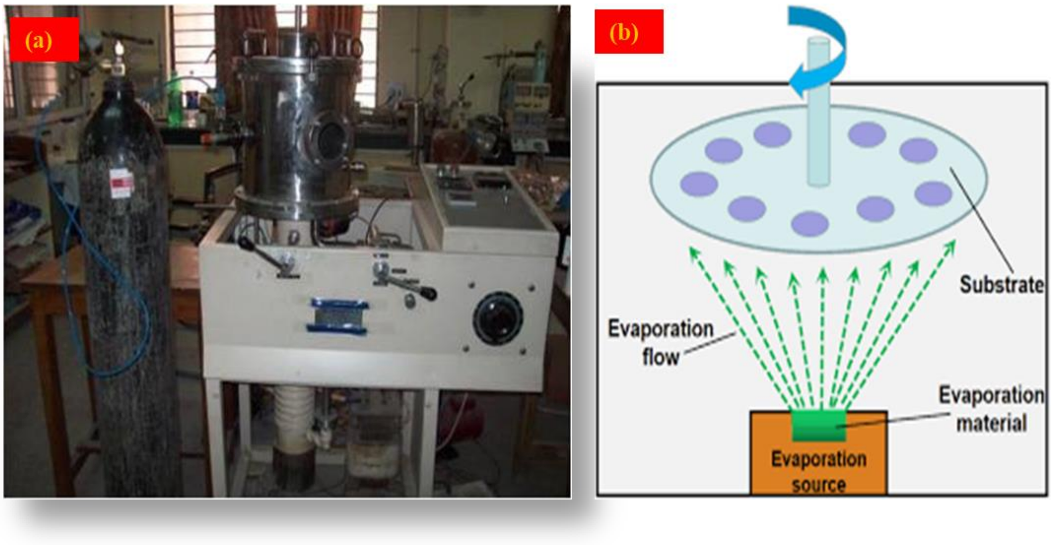

Fig.3. **(a)** RTCVD unit **(b)** Mechanism of RTCVD

The ZnO was deposited in a varying wt% and that wt% was of ZnO was 0.0 wt%, 2.0 wt%, 6.0 wt% and 8.0 wt%. Excellent VA-SWCNT-ZnO nanohybrid structures were obtained for field emission applications.

2.4. The flow chat of VA-SWCNT-ZnO

The flow chart of VA-SWCNT-ZnO nanohybrid structures has been shown in fig. Every step of this flow chat has been explained in previse steps. The steps growing steps has been shown in fig. 4.

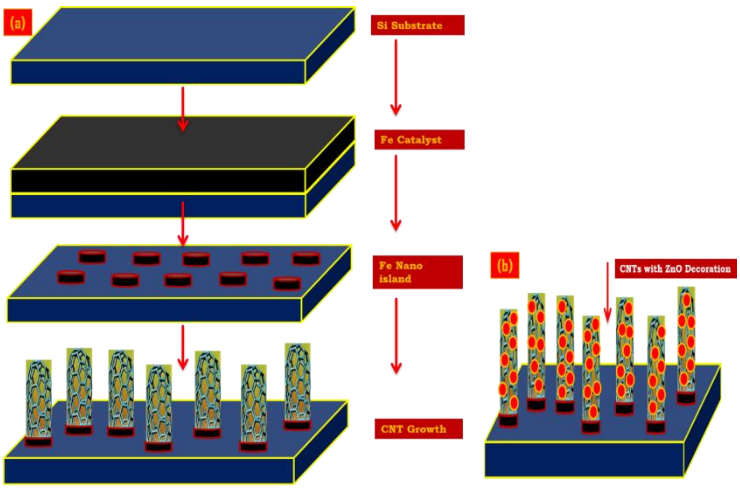

Fig.4. Flow chart VA-SWCNT-ZnO structures.

2.7. Field Emission Mechanism of VA-SWCNT-ZnO Nano hybrid Structures

Field emission is the processes of ejecting of electrons from the emitting material which largely depends on the work function and geometry of the material. In case of VA-SWCNTs, the high current density can be obtained from the tip of the VA-SWCNTs at low turn on voltage. So, VA-SWCNTs are getting catchy attention by the scientific community for field emission electronic device applications.

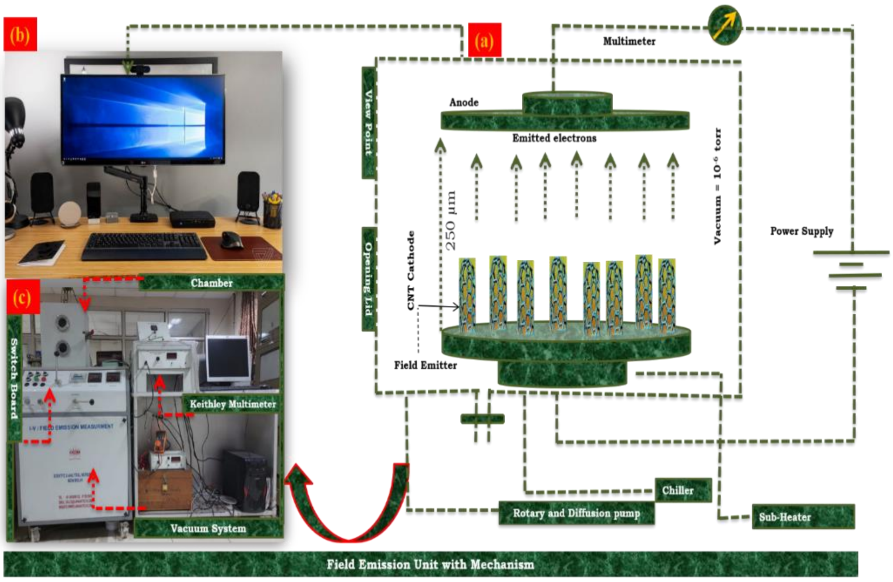

Fig.5. Field emission schematic mechanism with unit used.

Low work function leads higher current density. The mechanism of field emission by SWCNTs and unit has been shown in fig.5.

3. Results and Discussions

3.1. FESEM Study

The VA-SWCNT-ZnO hybrid nanostructures were investigated through field emission scanning electron microscope. This study reveals that high quality VA-SWCNT-ZnO hybrid nanostructures were obtained. The ZnO % in FESEM micrographs was not uniform as shown in fig.9.

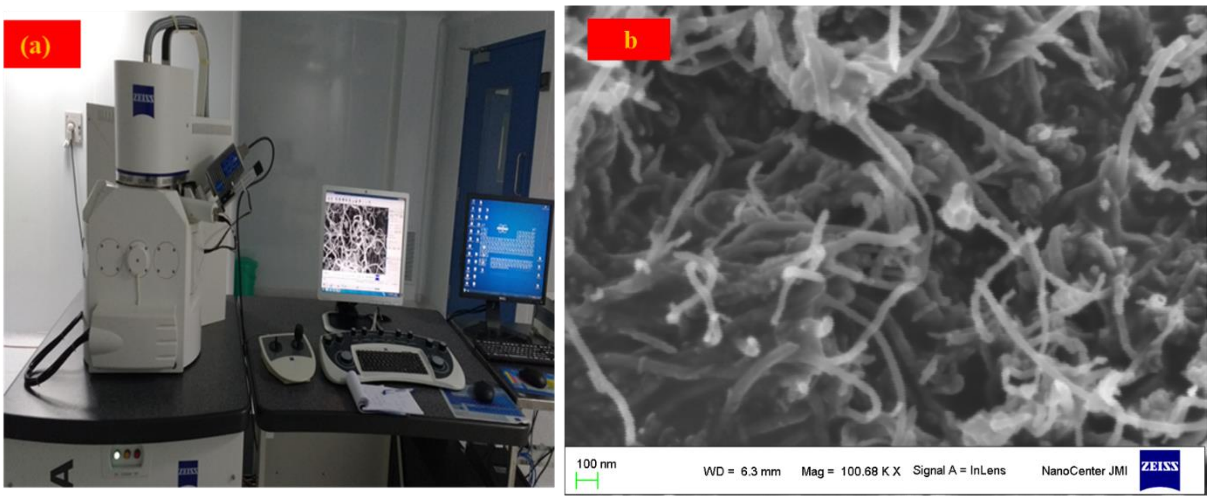

Fig.9. FESEM micrographs

3.2. HRTEM Study

The internal structural observations of VA-SWCNT-ZnO monohybrids were investigated through high resolution transmission electron microscopy (HRTEM). The study of HRTEM micrographs showed that high quality VA-SWCNT-ZnO nanohybrids were obtained. The nonuniform distribution of ZnO nanoparticles is clearly visible in the HRTEM micrographs as shown in fig. 10.

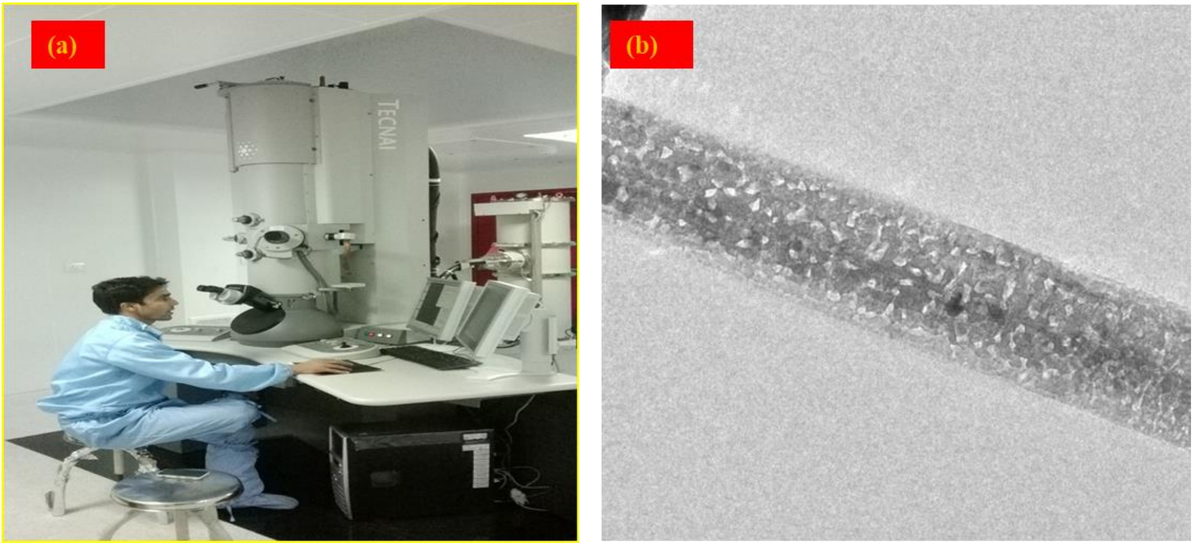

Fig.10. HRTEM micrographs

3.3. Raman Spectroscopic Study

The diameter of as grown CNTs was studied through Raman spectroscopy. The calculated diameter distribution was in the range of 1-2nm. The RBM peak was achieved at 110cm^{-1} respectively. This peak represents the successful growth of SWCNTs. The calculated diameter range of these sample was obtained by using the correlation d = 248/v as shown in fig.11.

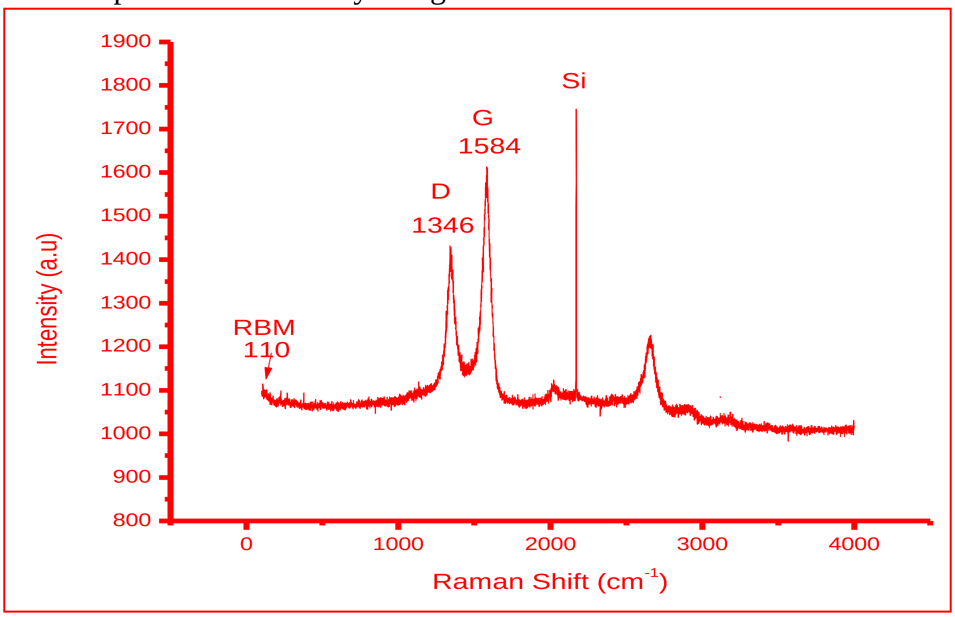

Fig.11. Raman Spectra

The values of D and G-bands were obtained at around (1345, 1585). Calculated I_D/I_G ratio for these CNTs was 0.82 respectively, which reveals that maximum defects have been produced on the surface of VA-SWCNTs-ZnO through the production process.

5. Field Emission Study

5.1. Field Emission Investigation

Field emission behavior of samples was analyzed in a diode arrangement under high vacuum environment of 10^{-6} torr. The obtained Field Emission results were interpreted under the frame work of Fowler-Northeim theory which correlates current density J to the applied electric field E as-

$$J = AE^2 \exp(-B\Phi^{3/2}/E)/\phi \ldots\ldots\ldots (2)$$

Where Φ stands for the work function of the material and A and B are constants, Where A = $1.54\text{x } 10^{-6}$ $AevV^{-2}$, B = $6.83\text{x}10^7$ $ev^{3/2}V\ cm^{-1}$.

5.2. JE Plots

The FE-measurements were performed in a complete voltage range of 50 V-750 V for all the sets of VA-SWCNT-ZnO nanohybrid structures. The maximum current density of VA-SWCNTs with ZnO 0.0 wt%, 2.0 wt%, 6.0 wt% and 8.0 wt% were 2 mA/cm^2, 5.1 mA/cm^2, 11 mA/cm^2, 15 mA/cm^2 achieved at a low turn on field of 2.5 V/µm, 2 V/µm, 1.5 V/µm, 1 V/µm respectively as shown in fig. 12 (a). An excellent enhancement in current density was achieved by VA-SWCNT-ZnO wt 8.0% at a low turn on field 1 V/µm.

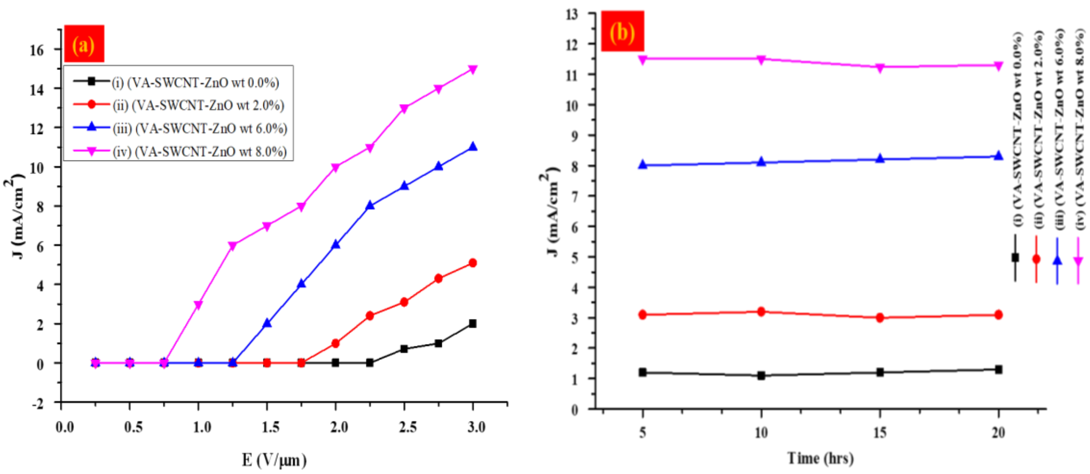

Fig.12. (a) Field emission measurements. **(b)** Stability measurements.

The attachment of ZnO nanoparticles reduces the work-function of VA-SWCNTs and thereby lowers-down the turn on field of theses field emitters to give the maximum current density with utmost stability.

VA-SWCNT-ZnO based nanohybrid structures are regarded better field emitters as compared to other metal oxides nanostructures. This approach will be a way better for the development of field emission display devices. Field Emission Stability measurements were performed at a constant voltage supply of 2.5 V/µm for consecutive 20 h and it was noticed that all the as-synthesized VA-SWCNT-ZnO nanohybrid structures possess good stability as shown in fig.12 (b).

5.3. FN-Plot

The FN plots of VA-SWCNT-ZnO nanostructures with ZnO 0.0 wt%, 2.0 wt%, 6.0 wt% and 8.0 wt% has been shown in fig. 13 (a), (b), (c) and (d). Using correlation $\beta = \frac{B\phi^{\frac{3}{2}}d}{m}$, where m is the slope of these plots value for Field enhancement factor were also calculated and the highest value of 3.6×10^4 is achieved for VA-SWCNTs-ZnO nanohybrid with 8.0wt% ZnO.

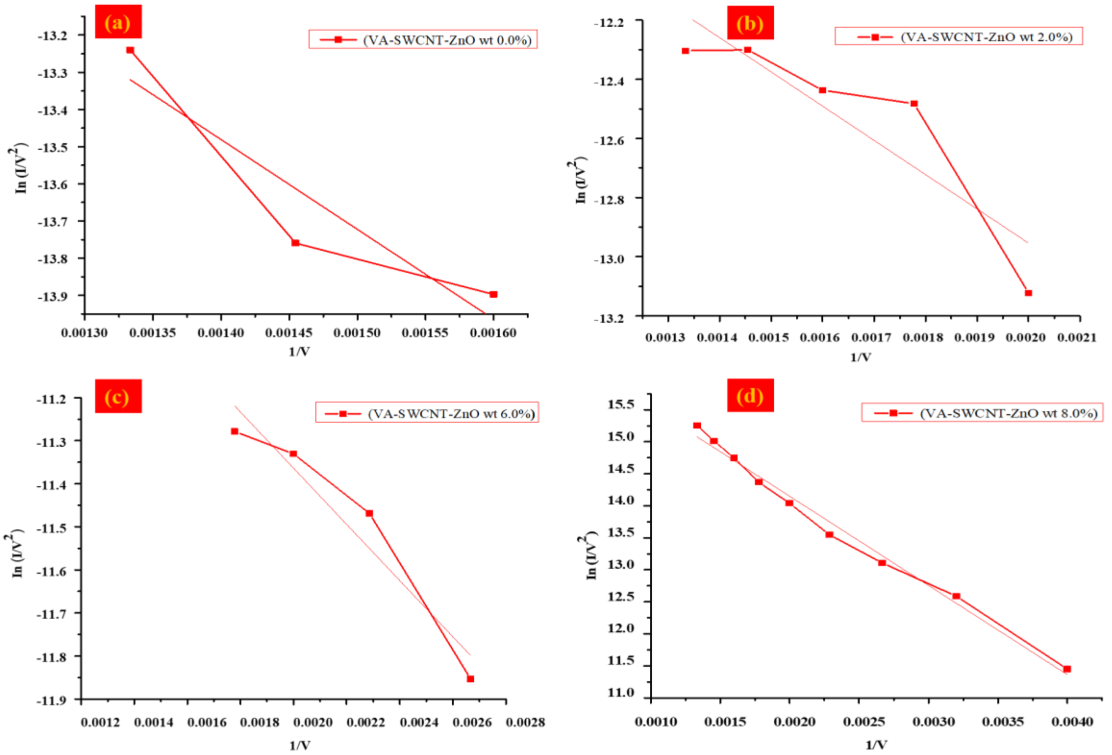

Fig.13. FN Plots.

Table. 1. (a) Experimentally Observed data for Field Emission

S. No	**Parameters**	**VA-SWCNT with ZnO 0.0 wt%**	**VA-SWCNT with ZnO 2.0 wt%**	**VA-SWCNT with ZnO 6.0 wt%**	**VA-SWCNT with ZnO 8.0 wt%**
03	Turn on field E_{turn}(V/μm)	2.5 V/μm	2 V/μm	1.5 V/μm	1 V/μm
04	Max. Current density (mA/cm^2)	2 mA/cm^2	5.1 mA/cm^2	11 mA/cm^2	15 mA/cm^2
05	Field enhancement factor (β)	0.79×10^4	1.4×10^4	2.58×10^4	3.6×10^4
06	Field emission Stability	(1-2) mA/cm^2	(3-4) mA/cm^2	(7-8) mA/cm^2	(11-12) mA/cm^2

A well systematical pattern of the data has been observed for all calculated parameters which has been summarized in table (1) and (2). All the characteristics observed are in good agreement with reported work.

6. Conclusion

High quality SWCNTs were grown by plasma enhanced chemical vapour deposition technique (PECVD). The as grown SWCNTs were investigated through field emission scanning electron microscope (FESEM) and high resolution transmission electron microscope (HRTEM). Excellent SWCNT structures were grown and confirmed by FESEM and HRTEM. A good surface morphology with high quality vertical aligned SWCNT structures were observed. Through HRTEM, a uniform distribution of ZnO particles was observed. The diameter distribution of as grown SWCNTs was about 1-2nm which was confirmed by Raman spectroscopy. All the sets of VA-SWCNT-ZnO nanostructures were fabricated for the field emission properties. The varying wt % of ZnO was 0.0%, 2.0%, 6.0% and 8.0%. All the fabricated VA-SWCNT-ZnO nanostructures were tested for field emission properties. Field Emission study revealed that maximum current density 15 mA/cm^2 at a turn on field of 1 V/ μm with field-enhancement factor 3.6×10^4 was achieved for VA-SWCNT-ZnO 8.0 wt%. The study revealed that by attaching ZnO nanoparticles with SWCNTs will lead the enhancement in field emission properties.

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8. References

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