# Novel, Flexible and Low-power Driven AC Electroluminescent Lamp from Carbon Nanotube Embedded Phosphor Material

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## ABSTRACT

We present a novel methodology to design a hybrid electroluminescent (EL) lamp by embedding carbon nanotubes (CNTs) inside the ZnS:Mn phosphor particles. By doing so, the phosphor particles exhibited increase in EL brightness and efficiency at low operating voltages (<100  $V_{AC}$ ). Interestingly shorter the length of CNTs used, greater was the field enhancement effect observed in the EL lamps. The role of CNTs have been identified to form conductive paths inside the ZnS particle thereby triggering EL due to electron injection to active centres at nominal voltges. In addition, a detailed electrical characterization of the novel EL lamp along with its spectral energy distribution studies are presented.

*Keywords*: electroluminescence, phosphor, carbon nanotube, hybrid material, electro-optical studies

# **1 INTRODUCTION**

There are innumerable potential applications of carbon nanotubes (CNTs) in electronics, composites, energy and life sciences. Due to their high electrical conductivity and the unbeatable sharpness of their tip they are the best field emitters of any known material and similarly due to the high current density, low turn-on and operating voltage, steady and long-lived behaviour make CNTs ideal field emitters for illumination and display applications [1–3]. Thare are few reports on enhanced electroluminescence (EL) at reduced threshold and operating voltages when CNTs are used as a field enhancer before the phosphor layer [4, 5].

The electroluminescence (EL) phenomenon has been strong potential for display applications due to many advantages such as non-thermal light source, high energy conversion efficiency, better brightness and contrast, wide viewing angle and ease of fabrication without vacuum requirement [6]. Alternating current driven EL lamps already occupy a segment of the high-resolution, flat panel display market. AC-EL displays in thin-film form are robust, possess long lifetimes, and offer high luminance with relatively low power consumption [7-9]. However, some constraints originating from the use of high driving voltages (>1000  $V_{AC}$ ) and/or short life times (few hundred hours) limit its commercial use [10-12].

Hence, in this paper a novel, cost-effective and facile technique is suggested, in which CNTs are deliberately embedded into a ZnS:Mn phosphor material forming a hybrid system which exhibits efficient EL at low voltages (<100  $V_{AC}$ ).

# **2 EXPERIMENTAL**

#### **2.1 Preparation of the Sample**

Orange emitting ZnS:Mn phosphor has been synthesized by sol- gel method and was used as a starting material. The multi-walled carbon nanotubes (CNTs) were grown by chemical vapor deposition technique and procured from M/s J. K. Impex, India. Since, the weighing of lower amounts of CNTs with precision is a tedious job, we have dispersed a known weight of CNT in ethyl alcohol and ultra-sonicated rigorously for 2 h to make a suspension. This was further added to ZnS:Mn phosphor and allowed to dry at room temperature (25°C). After complete drying the mixture was blended mechanically and annealed at 450°C under an inert N2 atmosphere. The amount of CNT addition is found to be highly critical in our experiments. An admissible range to exhibit EL was only 0.005-0.03 wt%. However, ~0:01 wt% of CNT was found to be optimum and showed stable and bright EL from the ZnS/CNT hybrid system.

#### 2.2 EL Lamp Fabrication



Figure 1: Schematic diagram of inorganic ZnS:Mn phosphor powder EL lamp. The bar indicates a scale of 10 µm

A  $25\text{mm}^2$  lamp was fabricated at room temperature by doctor's blade technique. The CNT embedded phosphor powder mixed with minimum amount of dielectric (dielectric constant,  $r\sim5$ ) was taken and spread between the two electrodes, out of which one is silver coated glass substrate to reflect the light and the other is ITO to offer transparency. To drive this circuit a low-cost portable power supply was developed indigeneously as shown in figure 2.



Figure 2: Block diagram of EL lamp driving circuit

The circuit consists of a 555 astable multivibrator as square wave oscillator. A potentiometer to vary the input voltage to the current booster, which feeds a high voltage amplifier to get the desired voltage. To reduce the cost and better portability, the circuit was developed using 9 V DC battery. The output voltage can be varied from 12-300 V with the help of the potentiometer used in the circuit.

## 2.3 EL Lamp Characterization

For phase identification, the structural characterization was performed using x-ray diffraction (XRD; Rigaku: MiniFlex, Cu K ; = 1:54 Å). The surface morphology and microstructural characterization was carried out by scanning electron microscopy (SEM, model number LEO 440). Room temperature steady-state luminescence characterization was done using a Perkin Elmer Luminescence Spectrometer (Model No. LS-55). The electro-optical characterizations of the samples have been performed using an indigeneous setup described above.

## **3 RESULTS AND DISCUSSION**



Figure 3: XRD patterns of pure ZnS:Mn and CNT embedded ZnS:Mn

The XRD patterns of pure ZnS:Mn and CNT embedded ZnS:Mn are shown in figure 3. Both the patterns match very well with the JCPDS card number 05-0566 of Zn blende (sphalerite) structure conforming the phase purity of ZnS. It can be seen from figure 3 that the addition of CNTs does not effect the crystallinity of the ZnS lattice.



Figure 4 shows the photoluminescence (PL) spectrum of ZnS:Mn phosphor having a bright orange-red emission peaking at 582 nm, when observed under an UV excitation of 346 nm. The inset shows the energy level diagram for the  $Mn^{2+}$  ion the ZnS lattice. It suggests that the orange-red PL emission originates from a transition between the  ${}^{4}T_{1}$  excited state and  ${}^{6}A_{1}$  ground state of the  $Mn^{2+}$  ion within the ZnS host. The PL emission at ~582 nm is due to an intra-configurational 3d<sup>5</sup> transition on the  $Mn^{2+}$  ion located within the band gap of ZnS lattice.



Figure 5: Raman Spectra of long (~15 µm) and short (~2 µm) CNTs

Figure 5 shows the Raman spectra of long (~  $15\mu$ m) and short (~2  $\mu$ m) CNTs procured from M/s JK Impex, India. The Raman spectral peak at 1340 cm<sup>-1</sup> represents the disorder state of carbon atoms in the graphitic structure known as the D- band. The ratio of areas under D and G band of long CNT is approximately 1, where as this ratio for short CNT is less than one which indicates that short CNTs have better graphitic stucture as compared to long

CNTs. Hence, in the current study short CNTs were employed for embedding in the ZnS:Mn phosphor system and a prototype of the EL lamp has been made as described above.



Figure 6: V-I characteristics of ACEL lamp

Figure 6 shows the V-I characteristics of the EL lamp operated in the voltage range 0-240  $V_{AC}$ . The profile clearly indicates that the EL lamp is ohmic up to 240 V. However, it has been observed that the lamp profile changes from linear to exponential beyond 240 V (not shown). This may be due to the fact that the EL lamp acts as a capacitor up to 240 V and thereafter it starts behaving like a semiconductor due to accumulation of large number of charge carriers in the phosphor/CNT hybrid material at higher voltages. The reciprocal of the slope of the above curve indicates the resistance of the EL lamp, which is ~200 k . A similar trend has been observed for the brightness of the EL lamp as shown in figure 7. It is important to note that the luminance increases with an increase in applied voltage up to a certain value before attaining a saturation.



Figure 7: Luminance - Voltage curve of the developed EL lamp

The threshold for the EL lamp to exhibit orange-red emission was found to be as low as ~80  $V_{AC}$ . In general, the EL brightness of the lamp increases with an increase in the applied voltage and attains a saturation value. A dependence of brightness on voltage could be explained by the process of tunneling of electrons as well as injection of hot electrons in the ZnS crystal [13]. However, the presence of CNTs seems to have modified the B-V relationship, which may be due to efficient intra-CNT impact excitation by hot carriers inside the ZnS lattice [14]. The primary role of CNT has been identified as formation of conductive *hot spots* inside the ZnS grain to enhance the local electric field, which allows electron injection to the active centre of phosphor at nominal (<100  $V_{AC}$ ) operating voltages [15].



Figure 8: EL Intensity variations with voltage

# 4 CONCLUSIONS

An alternating current driven EL lamp has been designed by a novel methodology of embedding carbon nanotubes (CNTs) inside the ZnS:Mn phosphor particles. The presence of CNTs have introduced efficient electron transport inside the ZnS:Mn phosphor exhibiting bright orange-red EL brightness at low operating voltages (<100  $V_{AC}$ ). Also, the length of CNTs used for the lamp fabrication has a major influence on the field enhancement effect. The purpose of CNTs was to form conductive paths inside the ZnS particles for hot electron injection to active centres at nominal voltges.

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