

# Effect of single walled carbon nanotubes on the threshold voltage of dye based photovoltaic devices



S. Chakraborty, N.B. Manik\*

Condensed Matter Physics Research Center, Department of Physics, Jadavpur University, Kolkata 700032, India

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## ABSTRACT

Carbon nanotubes are being widely used in organic photovoltaic (OPV) devices as their usage has been reported to enhance the device efficiency along with other related parameters. In this work we have studied the energy ( $E_c$ ) effect of single walled carbon nanotubes (SWCNT) on the threshold voltage ( $V_{th}$ ) and also on the trap states of dye based photovoltaic devices. SWCNT is added in a series of dyes such as Rose Bengal (RB), Methyl Red (MR), Malachite Green (MG) and Crystal Violet (CV). By analysing the steady state dark current–voltage ( $I$ – $V$ ) characteristics  $V_{th}$  and  $E_c$  is estimated for the different devices with and without addition of SWCNT. It is observed that on an average for all the dyes  $V_{th}$  is reduced by about 30% in presence of SWCNT. The trap energy  $E_c$  also reduces in case of all the dyes. The relation between  $V_{th}$ ,  $E_c$  and total trap density is discussed. From the photovoltaic measurements it is seen that the different photovoltaic parameters change with addition of SWCNT to the dye based devices. Both the short circuit current density and fill factor are found to increase for all the dye based devices in presence of SWCNT.

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## 1. Introduction

Since last two decades, organic dye based photovoltaic devices (OPV) have gained much importance due to its light-weight, flexibility and easy fabrication techniques [1–6]. Organic dyes and polymers used in these devices [7,8] have wide possibilities in photovoltaic studies. Over a large area, the dyes act as photosensitizer and also possess excellent film forming properties. Different dyes with different band gaps respond to different frequencies of the solar spectrum. Selection of suitable dyes is an important criterion in order to obtain better efficiency of OPV devices. In our earlier works [9,10] we have observed the electrical and photovoltaic characteristics of several dye based OPV devices. But these devices exhibit poor performance. Presence of traps, recombination of charge carriers at the trap centres and degradation contribute to the low performance in these devices [11,12]. Traps play a crucial role in the conduction process of these organic devices. Due to the presence of traps, majority of the generated charge carriers get immobilized [13,14] during conduction. The traps introduce additional electric fields in these systems and influence the current–voltage ( $I$ – $V$ ) characteristics. Depending on the energy and distribution of trap states the conduction process in these devices may vary. In almost all OPV

devices it is observed that beyond a certain transition voltage the conduction mechanism changes. Below this transition voltage the current follows Ohm's law and remains almost linear, but above it the current shows a non-linear behaviour. This transition voltage can be restated in terms of a threshold voltage ( $V_{th}$ ). This is an important parameter controlling the performance of the devices. Optimization of this voltage is very important for better operation of these devices. Reports suggest that trapping of charge carriers increases  $V_{th}$  [15,16]. As already mentioned during conduction charges get trapped in the trap centres and recombine. The energy of the trap states and the total trap density are important factors in relation to capturing of charges and their recombination. There are many techniques by which these factors can be influenced and the device performance can be altered. Of them, incorporation of new materials in the host system is a very effective way to change the performance of OPV devices. Recently to enhance the performance of these devices, people have started to incorporate different nanoparticles, carbon nanotubes (CNT) and different buffer layers [17–20] into these systems. Addition of carbon nanotubes [21–24] have shown promising results when incorporated in these devices. We have reported earlier [19] that addition of COOH-SWCNT affects the electrical and photovoltaic characteristics of Malachite Green dye based photovoltaic devices. CNTs have also been reported [25,26] to be used in active layers and as counter electrodes of photovoltaic devices. Though a number of works are being done by using CNT in OPV devices, much work is not available regarding the effect of CNT on the trap states of different organic dye based

\* Corresponding author.

E-mail address: [nb\\_manik@yahoo.co.in](mailto:nb_manik@yahoo.co.in) (N.B. Manik).

OPV devices. In the present work we study the effect of addition of single walled carbon nanotubes on the threshold voltage and also on the trap states of dye based photovoltaic devices. We considered a series of dyes such as Rose Bengal (RB), Methyl Red (MR), Malachite Green (MG) and Crystal Violet (CV) dyes for this purpose. Different devices were fabricated by dissolving these dyes in a blend of polyvinyl alcohol (PVA). Subsequently we incorporated SWCNT with each of the above dyes to prepare the OPV devices. Both types of devices were characterized to estimate the transition voltage and also the trap energy of the devices.

## 2. Materials and methods

Fig. 1(a)–(e) shows the structure of Rose Bengal (Loba Chemie Pvt. Ltd., Mumbai), Methyl Red (Finar Chemicals, Ahmedabad), Malachite Green (Finar Chemicals, Ahmedabad), Crystal Violet (BDH, England) and SWCNT (SRL, India). ITO coated glass is used as the front electrode and Aluminium coated with Mylar sheet (Al-M) as the counter electrode. We have previously reported [27,28] that the optical reflectivity of Al-M is quite high. Due to this back reflection more optical energy can be confined in the organic dye layer. The dyes under consideration have been chosen because they have a good optical response but much work is not available with these dyes in solid state photovoltaic film mode.

RB [4,5,6,7-Tetrachloro-3',6'-dihydroxy-2',4'5',7'-tetraiodo-3H-spiro[isobenzofuran;1,9'-xanthen]-3-one] is a polyhalogenated tetracyclic carboxylic acid dye belonging to the fluorescein acid group [24]. RB with molecular formula  $C_{20}H_4Cl_4I_4O_5$  has often been used as a biological stain. It has a high absorption coefficient in the visible region of the spectrum and has a tendency to transfer electrons from its excited triplet state, producing long-lived radicals. MR has been chosen because MR, with molecular formula  $NC_6H_4COOH$  (2-(N,N-Dimethyl-4-aminophenyl) azobenzene-carboxylic acid) is a less reported dye in photovoltaic mode [19]. It is an aromatic azo dye with good optical response. Its colour originates from absorbance in the visible region of the spectrum due to the delocalization of electrons in the benzene and azo groups forming a conjugated system. MG is triarylmethane dye with the

chemical formula  $[C_6H_5C(C_6H_4N(CH_3)_2)_2]Cl$ . The strong green colour comes from the absorption band of the cation at 621 nm in DCM solvent [25,31]. The chloride and oxalate anions have no effect on the color. MG dye has been chosen because it has a good quantum yield. CV [Tris(4-(dimethylamino) phenyl)methylium chloride] is a triarylmethane dye [29]. CV is commonly used as a histological stain and has a molecular formula  $C_{25}N_3H_{30}Cl$ . The absorption spectra of these dyes in distilled water are shown in Fig. 2(a)–(d). For comparison the absorption spectra of the dyes mixed with SWCNT have also been shown in the same figure. It is seen that RB shows a peak at 540 nm while for MR the peak appears at 527 nm. The absorption peak of MG occurs at 613 nm and for CV the peak is at 586 nm. It is interesting to note that for all the dyes, with the addition of SWCNT no change in absorption peak occurs.

In a precleaned beaker 10 ml of distilled water is taken and 1 gm of polyvinyl alcohol (PVA) is added in it. Here PVA acts as an inert binder. The mixture is stirred for an hour in a magnetic stirrer to form a homogeneous solution of PVA. 1 mg of RB dye is added with this solution and again stirred well for around 30 min to get the RB dye solution. This solution is then divided into two parts in two beakers. The first beaker is kept aside and in the other beaker 1 mg of SWCNT is added with the dye. The blend is again stirred in the magnetic stirrer for one hour to form a homogeneous dispersion of SWCNT and RB dye. After preparation of the solution, the RB dye solution is spin coated on a pre-cleaned ITO coated glass electrode at a speed of 1500 rpm and then dried at room temperature at a speed of 3500 rpm. The same solution is spin coated on the Al-M electrode. When the two electrodes are in the semi-dry state, they are sandwiched to form the device. To prepare the SWCNT added RB dye based device, similar technique is applied. Photovoltaic devices were fabricated with MR, MG and CV dyes with and without adding SWCNT. The different devices prepared were kept under vacuum for 12 h before characterization. Fig. 3 shows the schematic diagram of the device prepared.

The absorption spectra of the devices are measured using a Elico scanning minispectrometer. Dark current–voltage ( $I$ – $V$ ) characteristics of the devices have been measured with a Keithley 2400 source measure unit. During measurement, the bias voltage

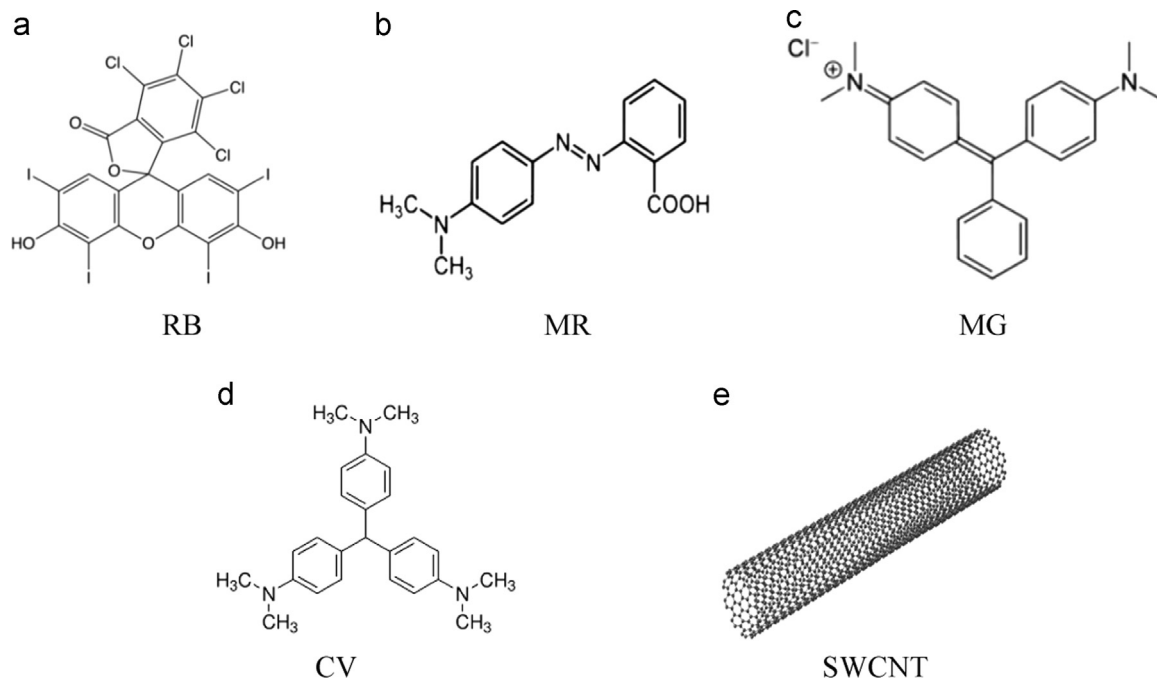


Fig. 1. Structure of (a) Rose Bengal Dye (RB), (b) Methyl Red (MR), (c) Malachite Green (MG), (d) Crystal Violet (CV) and (e) single walled carbon nanotubes (SWCNT).

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