Toward Sustainability in Organic Electronics: A Performance Check for DNA Complexes as Dielectric Layer in P3HT based OFET

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ABSTRACT--- ***-Notwithstanding its manifold advantages the fast emerging area of organic electronics can invariably lead to the production of huge amounts of plastic waste and therefore developing ecofriendly organic electronic components to make it sustainable has become a priority task. Here an effort has been made in this direction where in a well-known bio material complex of deoxyribo nucleic acid and cetyltrimethylammonium chloride (DNA_CTMA) is applied as dielectric layer in a top-gate organic field effect transistor (OFET) with Poly 3-hexylthiophene (P3HT) as channel material. The DNA used was isolated from the leaf of Asteracea species plant by the biosynthesis mechanism. The device returned modest but promising operating parameters.***

Keywords--- Organic electronics; organic field effect transistors; DNA, dielectric layer; P3HT.

1. INTRODUCTION

Organic field effect transistors are fast emerging [1-2] as one of the most realizable organic electronic component due to their potential applications, especially in active matrix displays, electronicpaper, and radio frequency tags. One of the most important advantages of these transistors is that OFETs offer printability on large areas of various substrates starting from paper to plastic rolls. Ironically this large area roll-to-roll production possibility itself eventually lead to a major problem of huge amount of plastic waste generation. Plastic solid waste management and disposal has already reached a global issue [3] that poses grave threat to the environment. This makes it imperative to find new materials which arerobust, cost effective as well as easily processable and at the same time environmentally sustainable. With this as the motivating factor an attempt has been made here to fabricate an OFET with a biodegradable gate dielectric as an earnest attempt to identify possible sustainable components for organic electronics devices.

It has been now established [4-7] that P3HT is an effective organic semiconductor as channel material in OFETs. Therefore it is selected here as the channel and aluminum (Al) as source- drain electrodes. A bio complex, DNA_CTMA dielectric layer is employed to form the gate dielectric for the fabricated OFET with top gate architecture. Simple vacuum thermal evaporation and spin coating techniques were used for device fabrication. All fabrication procedures are carried out in air only. DNA was isolated from the Asteracea leaf by natural bio synthesis methods [8-9].

2. EXPERIMENTAL

Poly (3-hexyllthiophene) (P3HT) is from Sigma Aldrich and used as received. P3HT solution was prepared in chloroform (4mg/ml) solvent. DNA_CTMA is synthesised from the young leaves of the species Asteracea (Chromolaela Odorata) [10] as follows: The plant leaves are grounded and mixed in a mortar along with the extraction buffer containing EDTA(Ethylenediaminetetraacetic acid), CTAB (Cetyltrimethylammonium bromide), HCl and a pinch of NaCl, after soaking it in chilled ethanol for 2 hours. The mixture is then incubated in a 65^{o}C water bath for 1 hour followed by filteration. The filtrate is poured into a microcentrifuge tube and the top aqueous layer is extracted out by centrifuging for 15 minute at 10,000 rpm. An equal amount of chloroform:Isoamylalcohol (24:1 v/v) is added to this aqueous layer and stirredgently. This mixture is again centrifuged for 15 minutes at 12,000 rpm at room temperature and the supernatant is carefully removed discarding the organic residue to another tube using a wide tip micropipette so that DNA is not damaged. This step is repeated 3-4 times till a transparent solution is obtained. To precipitate DNA, 1.5 times by volume chilled absolute isopropanol is added to the above supernatant solution, inverted several times for complete mixing of solutions followed by incubation at 273 K and then left for overnight. This is then followed by centrifuging these tubes for 10 minutes at 14,000 rpm to collect the DNA pellets. These pellets are washed 2-3 times with 70%

ethanol. Ethanol is poured out and the pellets are stored at -20^{o}C followed with drying at room temperature. Both DNA and surfactant solutions are then mixed together at 1:1 stoichiometric ratio to facilitate spontaneous formation of hydrophobic DNA-CTMA complex that precipitated in water. The white DNA-CTMA precipitate is then collected and centrifuged to remove excess surfactant. Isolated DNA properties are confirmed by the gel electrophoreses, UV/Visible absorption, SEM, and FTIR. Fig.1 shows the gel electrophoresis of isolated DNA. Bands in electrophoresis [11] can be seen and each contains DNA molecules of a particular size.

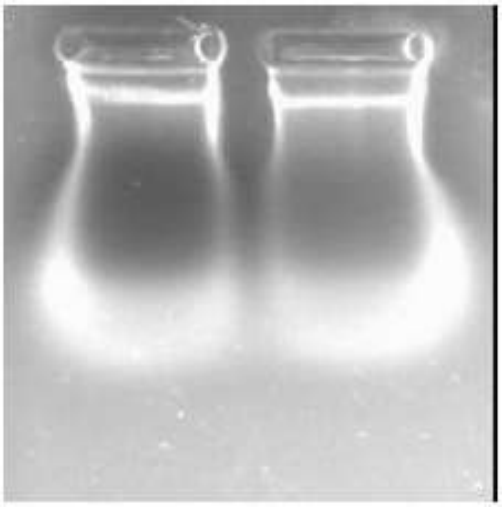

Figure 1: Gel electrophoreses of DNA

For UV Visabsorption study of DNA_CTMA three different combination of samplesare tried. In Fig.2 dark pink color spectrumindicated the DNA_CTMA absorption while blue color denotes absorption of DNA in water. The absorption band from 270 nm to 340 nm corresponds to the π-π* stacking of nucleon band [12] of the DNA. These absorption bands are present in both spectra but in the DNA_CTMA solution the absorption intensity is much better than DNA alone. Such increase in the absorption intensity produces [13] film surfactant effect in DNA solution and this surfactant effect has seen to produce [14] better film formation during the device fabrication stage.

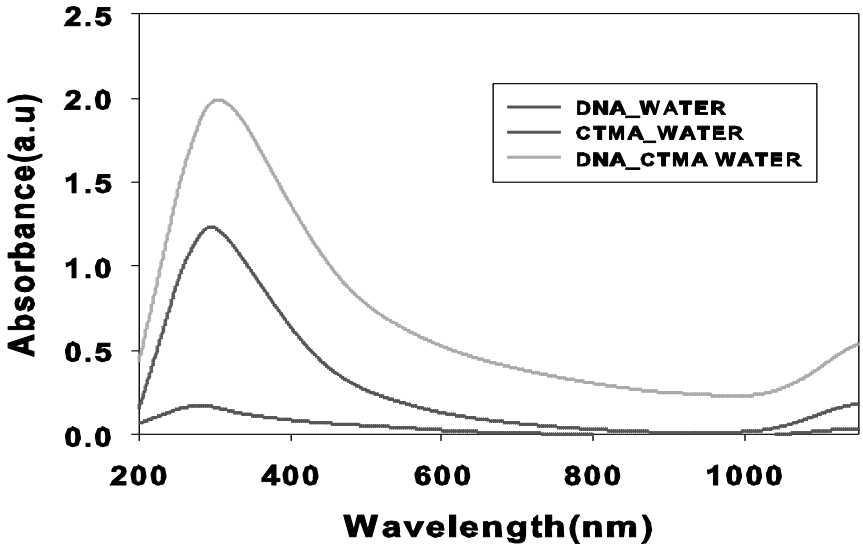

Figure 2: UV/Visible spectrum of DNA_CTMA complex

The FTIR spectrum has been taken in the mid IR region of 500 - 4000 cm^{-1}. The experimental sample is directly placed on the potassium bromide crystals and the spectrum was recorded in transmittance mode. FTIR measurement is carried out to identify the possible biomolecules [15-16] in DNA. In Fig. 3 the dark red line indicate the DNA_CTMA and pink color indicate the DNA. Wide Peak at 3329 representsthe O-H strech and thoseat 2928 and 2852 indicate C-H strech in alkanes. The small wide stretch in 2107 gives confirmation of triple bond in C and N. C-H bend and rock represent the peaks at 1466 and 1375 respectively. The peaks at 1250,1065, 966 and 953 [17] confirmed the presence of C-H and O-H wag and strech. These over all information confirm the presence of isolated DNA. The presence of

DNA_CTMA is also confirmed by FTIR which was indicated by the red solid line. CTMA addition to DNA reduces the C-H strech and bending, but it keeps the C-H wag [18] at 1061. Addition of CTMA in DNA compound did no alter its basic material properties but returned better film propeties.

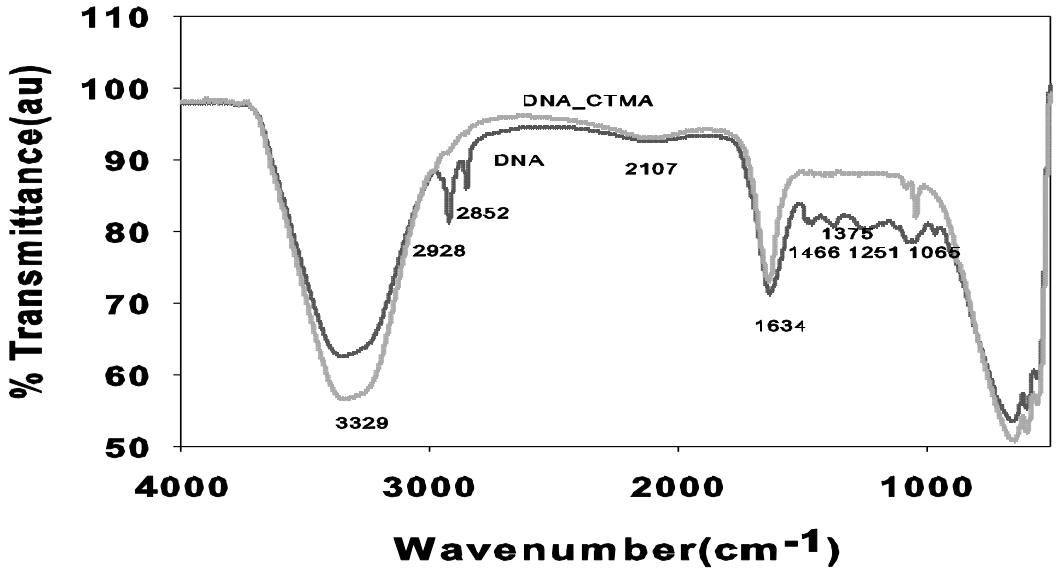

Figure 3: FTIR Spectrum of DNA and DNA_CTMA complex.

The surfactant effect in DNA isalso confirmed by the SEM. Fig. 4(a) represents the SEM of the DNA coated steel plate and 4(b) indicates the DNA_CTMA coated steel plate. From the micrographs itisclear that without surfactant, DNA shows cloud structure in the film and shows more roughness as compared to that in figure 4(b). In Fig 4(a) and (b) it is clear that cloud structure of the DNA decresed due to the addition of CTMA.

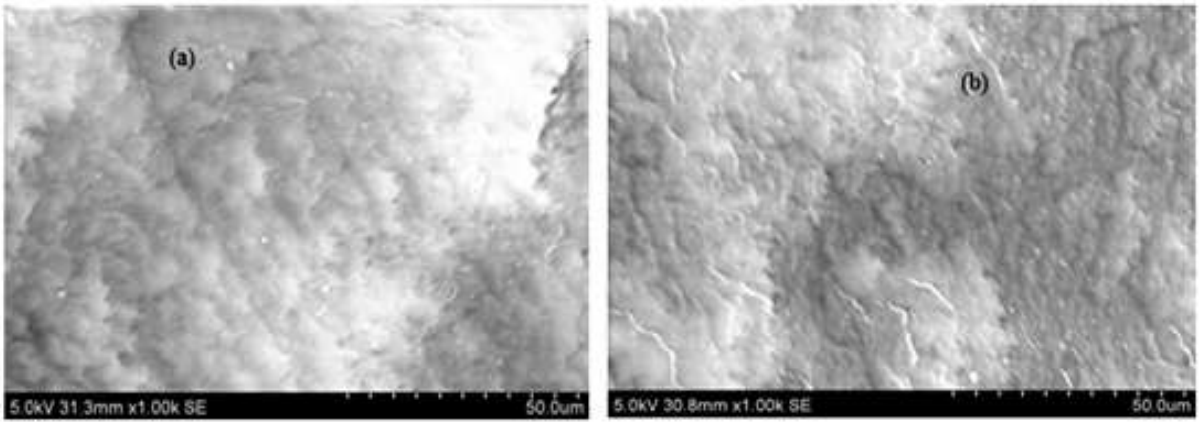

Figure 4: SEM of DNA coated on steel. (a) DNA Alone (b) DNA_CTMA complex.

3. DEVICE FABRICATION

A bottom contact top gate OFET is fabricated with P3HT as active layer, DNA_CTMA as gate insulating layer and Aluminium (Al) as S-D electrodes. Device is fabricated as follows: Micro pattern (60µm) of the source drain electrodes is typically made using evaporation mask in a high vacuum chamber. After channel preparation, the channel material (P3HT) solution was drop casted on the patterned electrodes and was kept in room temperature for drying (30 min) to form the channel (~80nm thick). The dielectric layer DNA_CTMA (~120 µm thick) is then spin coated on the top of the active layer at 400 rpm. Finally aluminum gate electrodes were coated on the dielectric layer by thermal evaporation (~150nm thick). Fig.5 represents the structure of fabricated device.

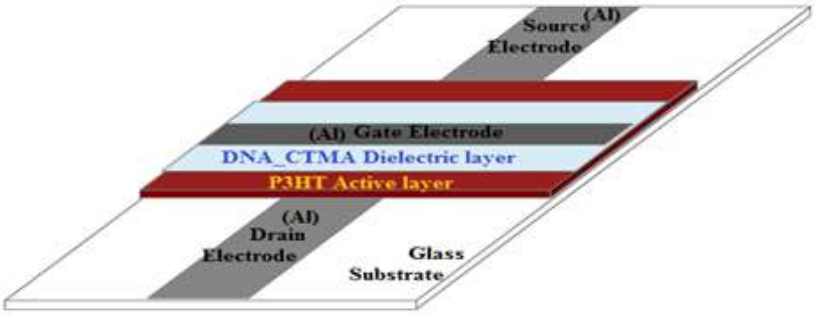

Figure 5: Fabricated device structure on glass plate

4. DEVICE CHARACTERIZATION

Fabricated devices are characterized by Keithely2400 source meter and 6517B high resistance meter. Keithely2400 source meter is connected to the source drain electrode and Keithely 6517B electrometer to the gate source electrodes. The experimental setup is controlled by a Labview program for characterization of the device. From the data obtained OFET parameters like mobility of the channel, I_{on}/I_{off} ratio and threshold voltage (V_{TH}) are evaluated.

5. RESULTS AND DISCUSSION

Fig. 6 and 7 show the electrical characteristics of the fabricated device. It can be seen from fig. 6(a) that the device exhibits P-type OFET operation [19-21] in the depletion mode. This is due to the negative charge induced in the active channel with the help of positive gate bias voltages. Source-Drain voltage (V_{SD}) in the range 0 to +20 V is applied with -5 to +3 V gate bias with a step delay of 1V, and the source _drain current (I_{SD}) is found to be in the ohmic region. It is clear that at +3V gate bias voltage device isturned off. The I_{SD} in the source drain voltage region of 10 to 15 V is pulled to inverse direction due to [22] the S/D electrode interfacing effect. The work function of the Al electrode used is 4.1 eV and the work function mismatch between the P3HT active layer and these Al S/D electrodes led to the observed inversing effect in source drain current. Fig. 7 shows the transconductance curve of the fabricated device. From this the turn off gate voltage (V_g) and threshold voltage (V_{TH}) are evaluated [23] using the standard procedure.

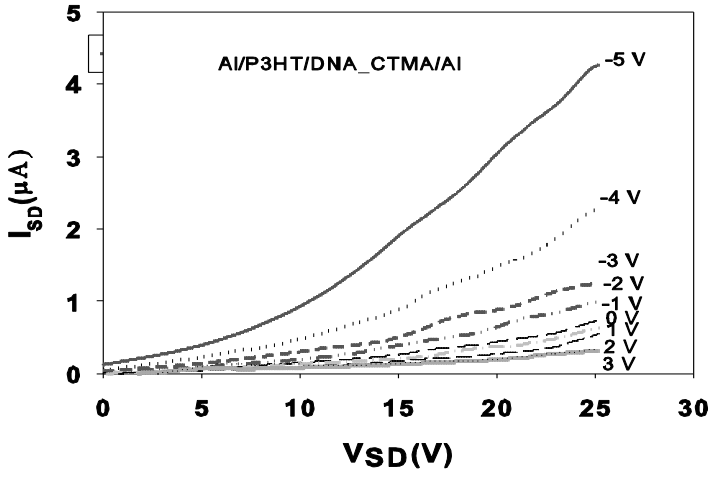

Figure 6: V_{SD} versus I_{SD} at different gate voltages.

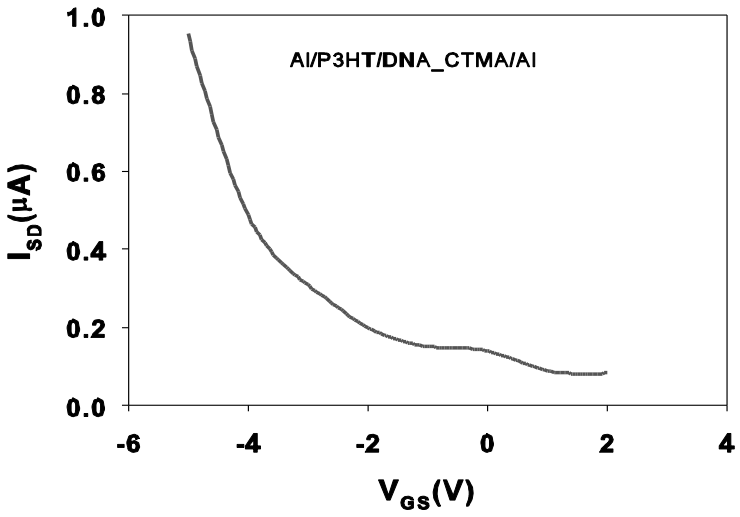

Figure 7: Transconductance curve at V_{SD}=10V

Extrapolation in the linear region (ELR) method was used for calculating the threshold voltage. The field effect mobility (μ) is calculated [24-25] using the linear regime equations. As the I_{SD}-V_{SD} characteristic is found follow a linear regime operation in the entire voltage range parameter extraction is fully carried out using the linear regime equations and methods. The extracted parameters are given in Table 1. Fig. 8 represents the plate capacitance versus frequency response of the fabricated device. Using this unit plate capacitance of the device is evaluated. For plate capacitance measurement the plate capacitance variation of the device with frequency up to 1 MHz is examined using an LCR meter. Unit plate capacitance was considered as the constant region [26] of the graph at higher frequency. From the response curve it is clear that plate capacitance of fabricated device decreases with increase in frequency and comes to constant region at high frequency. This constant region is considered as the device plate capacitance [27] for parameter calculation. Calculations in this line returned the plate capacitance of OFET with DNA_CTMA dielectric as 2.05 x 10^{-10}F/cm^2.

Table 1: Evaluated Device Parameters

Mobility(cm^2/Vs)	I_{ON}/I_{OFF}	Threshold Voltage(V_{TH})	Plate Capacitance (F/cm^2)
0.0469	10	-1	2.059×10^{-10}

From values of device parameters given in table 1 it is clear that the performance of the P3HT device with the green and bio dielectric material (DNA) gives comparable performance to reported [28-29] P3HT OFET devices with plastic dielectric layers like PMMA and PVA.

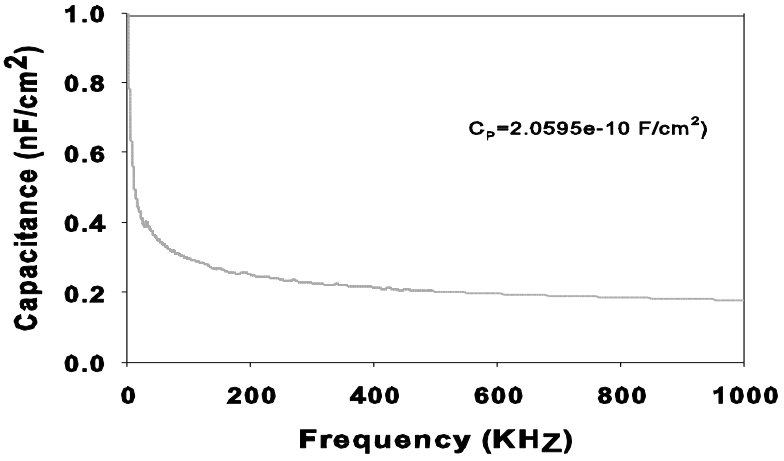

Figure 8: Device frequency versus capacitance curve

For impedance measurement [30] the layer structure of the device is considered. The impedance across the gate and drain electrode is measured in the frequency range of 100 to 10000 Hz. The impedance values at higher and lower frequency for the layers are measured. Fig. 9 represents the frequency versus impedance response of DNA_CTMA

OFET. It is clear that the impedance of the device exponentially decreases with frequencyandboth devicesshowimpedance at mega ohms range at lower frequencies. At higher frequency range the impedance got decreased by one order before finally treading an almost constant path. From the plot it is clear that there do have a capacitance effect [30] between the electrodes, and this capacitance effect must have produced the gate voltage dependency in the device operation.

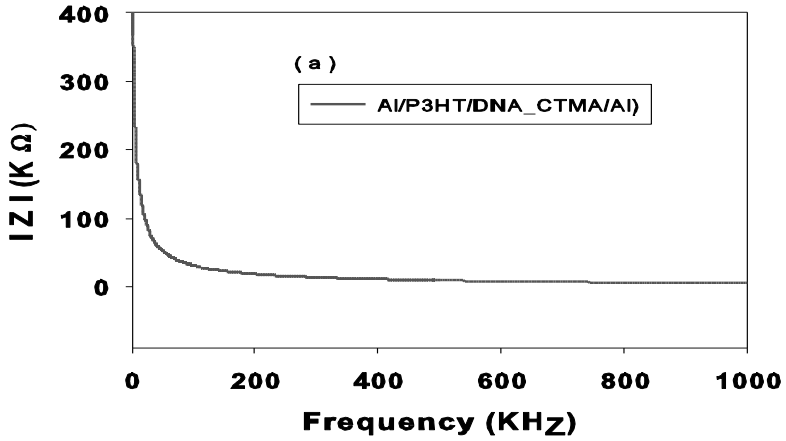

Figure 9: Device impedance versus frequency curve

6. CONCLUSIONS

It has been demonstrated that P3HT based OFETs, made with biodegradable DNA synthesized from plant extracts through a low cost method returned comparable performance with reported P3HT devices using plastic dielectrics indicating that sustainability of organic electronics devices can be achieved by employing eco - friendly natural produces in place of pure plastics.

7. ACKNOWLEDGEMENTS

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