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The Effect of carbon nanoparticles doping of active layer on the Electrical properties of P3HT/PVA Organic Field Effect Transistor.

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ABSTRACT:

In this work, we are studied the effect of carbon nano-particles (CNP) doping of an active layer on the electrical properties of P3HT/PVA organic field effect transistor (OFET). A top gate-bottom contact (TG-BC) configuration was used to fabricated an organic field effect transistor with Poly(3-hexylthiophene-2,5-diyl (P3HT)) (as pure and doped with CNP) as an active layer, and polyvinyl alcohol (PVA) as a dielectric layer the gate electrode. OFET was made with AL/PVA/(P3HT:%CNP)/Au structure, by using masks and required instrument which were manufactured in lab, in a constant conductance channel width and length (W=1mm, L= 60 µm). The dielectric layer of polyvinyl alcohol (PVA) was deposited by spin coating method, which its electrical capacity per unit area at AL/PVA/Au sandwich structure is (Ci=223 nF/cm2). While the semiconducting polymer was deposited by spin coating method to (203nm) as active layer thickness.

The Atomic Force Microscopy-AFM was used to investigate the surface properties of thin films of the pure and doped P3HT with CNPs. The surface nature and the size and distribution of grains for thin films were examined for pure and doped P3HT polymeric with two ratios (1%, 5%) of carbon nanoparticles (CNPs), to see the effect of conformation, using an atomic force microscope device. The results shows affects the surface structural properties of synthetic thin films, as they reduce crystal volumes, increase the number of crystals formed and redistribute the formed crystals, In general, the pure and doped seems a semi-crystalline films, and each less surface roughness and Rq at additive ratio 1% and increase at 5% CNPs, according to the results of the AFM.

The results shown that all devices were operating in enhancement (accumulation) mode. The calculation of OFET some parameters was done after measurement of output and transfer characterizations. The results of the saturation mobility and the ratio I_{on}/I_{off} and the threshold voltage were be influenced with doping of semiconductor layer, due to the changing of the resistance of the conduction channel. The best characteristic of the organic field effect transistor is when the doping ratio of an active layer 3% of CNP, which has the highest saturation mobility value ($1.41x10^{-1}$ cm^2/Vs) and (I_{on}/I_{off}= 199), and threshold voltage (-1.2V).
Keywords: Organic field effect transistor OFET, Poly(3-hexylthiophene-2,5-diyl) (P3HT), Polyvinyl alcohol (PVA), CNP, Thin film, Mobility, AFM, Threshold voltage, Channel resistant.

Introduction

The materials which basically made of plenty carbon-based components called an organic materials (organic semiconductors). These materials were used as the active layer of the organic electronics, which can be manufacturing as a thin films at a low temperature processes and for a flexible, environmentally friendly and large area of applications of electronic devices that would be impossible using traditional materials.[1] Therefore, the intrinsic characteristics of organic materials are engaged of the basic processes of charge carriers which are controlling and limiting the efficiency and performance the organic electronic devices.[2-3]

The organic semiconductors were used to manufacturing a many of an unique and modern organic applications like flexible displays, touchable screens, sensors, Biochemical Detectors, solar cells, radio frequency identification tags (RFID), textiles electronics, thin films batteries and processors & memories devices. The most important devices which capacitate to made these organic technologies are light emitting diodes (LED) and organic field effect transistors (OFET).[4-9]

Organic field effect transistors (OFETs) are field effect transistors in which the semiconductor layer is made from organic semiconductors. Today, thin-film transistors are made with different substrates and even the flexible ones. Transistors are the fundamental building block of modern integrated circuits, which are used as an amplifier or a switch. As transistors enter into the manufacture of many devices such as video devices, flexible displays, small screens on the traditional glass substrates of the mobile phone and other applications growing rapidly such as liquid crystal displays, smart cards, memory cards, wearable electronics and others. OFET can be an ideal for the infrastructure of these applications and other applications due to the close compatibility of the materials that make both OLED and OFET. OFET is a two-dimensional structure of the fact that the semiconductor and dielectric layers are formed in two-dimensional thin films, due to the linear structure of many semiconductor organic materials used. In addition to using organic semiconductors and organic insulation, all transistor components can be replaced with organic materials. The field effect is the phenomenon in which the electrical conductivity of semiconductors is changed by applying an electric field to its surface, as the electric field is applied through the gate electrode in the device.[10-12]

The performance of an organic field effect transistor is determined by finding the parameters associated with this device which are the mobility (μ) of the charge carrier in conductive channel, the (I_{on} / I_{off}) ratio and the threshold voltage (V_{th}) and others, whose value can be determined from the typical properties obtained when measuring the electrical properties of these devices (output and transfer characteristics).

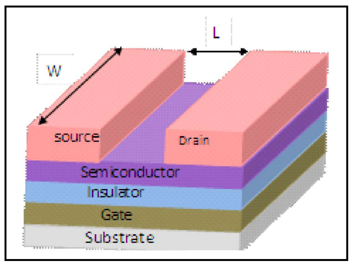


Figure (1) the general structure of the field effect transistor.

OFET are continuously need to improve their properties, such as high operating voltage and low performance compared to inorganic transistors, the reason for this fact due to the low charge carriers mobility of organic semiconductors, because of the high resistance of these materials and the nature of the interfaces in the hardware architecture. Many different methods have been taken in an attempt to improve the performance of organic transistors not only in terms of electrical properties of organic semiconductors but also in terms of improving the structures of the device, the simplest method is doping method.[13]

There is hardly to find a completely pure conjugated polymer, often due to the presence of some residue impurities in the material resulting from the polymerization process, so the Fermi energy level of a polymer is not an intrinsic property of the polymer, but rather depends on the amount of impurities present. Usually, there is a small concentration of positive impurities in the material, which transports Fermi energy down compared to the original pure polymer. [14]

Manufacture of doped materials by adding dopant to the polymer matrix is a traditional method to improving its optical, mechanical and electrical characteristics, and to obtain a new material that differs from the pure polymer and additive materials. Studies in the past two decades have focused on the use of nano-additives, including carbon nanoparticles (CNP). Carbon nanostructures have emerged from among many types of advanced nano-materials in the past years, as they have been used in a wide variety of application, due to their unique physical and chemical properties. Carbon nano-materials showed excellent optical and electrical properties which made it useful in optical and biological and chemical sensing. The ability of carbon nanoparticles to form a suspension enabled to manufacture of printing inks for printed electronics. Carbon nano-materials that are simply formed by carbon are relatively safe compared to many other inorganic nano-materials like quantum dots (QDs), which usually are a heavy metals.[15-19]

The choose of the materials as a gate insulator and active layers is very critical for the performance of the manufactured device. One of important organic semiconductor is poly 3-hexylthiophene (P3HT), is a component of the [poly (3-alkylthiophene)] family, and it has a major conjugate solid axis and flexible side-alkyl chains, which give this polymer great solubility P3HT is a hydrophobic polymer. The ability of P3HT to form a self-assemble layer in the case of the solution and the solid state enables to obtain a uniform installation polymeric chains, which leads to increased mobility of charge carriers, as it has a good charge carrier mobility up to (0.1 cm^2/Vs). P3HT is one of the positive polymers including a holes as a charge carriers. There are several factors that mainly effect on the conformation of P3HT thin films, including degree of regioregularity, the deposition method used, the thermal annealing process, the molecular weight and the solvent used.[20-23]

The characteristics of polymeric insulators such as polarity, viscosity, hydrophobic and dielectric constant, each have a role in the performance of the active layer. Several polymers may be used to form a gate insolating layer, which can improve the insulation strength or capacity and reduce the leakage currents and provide a better surface for in the device (e.g. PMMA, PVC, PS and PVA). Some experts in the field of microelectronics expect that the condition for dielectric layer thickness is approximately (d < (L / 10), and others estimated it to be (d < (L / 4)), where L is the conductive channel length.[24-26]

In organic field effect transistor for saturation region, the charge carrier mobility (μsat) is:[27]

$$\mu_{Sat} = \frac{2L}{WC_i}\left(\frac{\partial\sqrt{I_{ds}}}{\partial V_{gs}}\right)^2 \quad \ldots\ldots\ldots (1)$$

Where L, W are the conductance channel length and width, Ids is channel current, V_{gs} is the gate-source voltage and C_i is the electrical capacitor per unit area of gate dielectric layer, V_{th} is the threshold voltage.

In this study, OFET in (AL/PVA/(P3HT+%CNP)/Au) configuration was fabricated by using P3HT as an active layer and Polyvinyl Alcohol (PVA) as the gate insulator, and we investigated the effect of the doping of active layer with various v/v ratio of carbon nanoparticles (1%, 2%, 3%, 4%, 5%) on the electrical properties of OFET.

Experimental:

Regioregular polymer Poly(3-hexylthiophene-2,5-diyl) (P3HT), product batch number M102 electronic grade, was purchased from Ossila Limited Co., UK, average molecular weight Mw 65200 and Mn 29600, >95.7% head-to-tail regioregularity, molecular formula {(C10H14S)n} is a conjugated polymer was used in this research, figure (2) shows its chemical structure. P3HT solution was prepared in 1,2-dichlorobenzene (from Sigma-Aldrich) in 10 mg/ml concentration by stirring at 60°C for 15 minutes. Finally, after cooling to room temperature, the solutions was filtered with syringe filter of size 200 nm.

As a gate insulator layer we used Polyvinyl alcohol (PVA), with an average molecular weight within of (85,000-124,000) g/m, viscosity (4% aqueous solution at $20^{O}C$): 28-32 cps, hydrolysis degree (99%)was obtained from Sigma-Aldrich-India. It has a chemical structure ([CH2-CHOH]n - as chemical structure shown in figure (2)). Aluminum and gold of purity (99.99%) was provided from Sigma-Aldrich for deposition the electrodes. PVA solution was prepared in deionized water at 70 mg/ml concentration by stirring at 90°C for 8 hours. Finally, after slow cooling to room temperature, the solutions were filtered by syringe filter (220 nm).

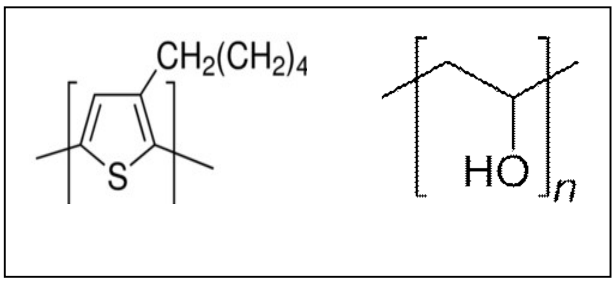


Figure (2): Chemical structures of P3HT & PVA.

The carbon nanoparticles (CNPs) was purchased from (NANOSHEL LLC. – USA) used as a filler material. The characteristics of CNP were shown in table (1). CNPs solution was prepared in 1,2-dichlorobenzene in 1mg/ml concentration by stirring at room temperature for 2 hours. The doping process of P3HT solution was done by (0%, 1%, 2%, 3%, 4%, 5%) v/v from CNPs solution.[28-29]

Table (1): general characteristics of CNPs.

Assay	≥99% trace metals basis
Form	Nanopowder
Particle Size	Less than 50 nm (TEM)
Surface Area	Spc. Surface area >$100m^2/g$ (BET)
Flash Point	1800 Degree
Boiling Point	2640 Degree
Melting Point	3340 Degree
pH	7 pH
Stability	Completely Stable

In cleaning process of a glass substrates, it's received a multistep solvent clean by ultrasonication technique; isopropyl alcohol, acetone and deionized water, 5 min of each, then dried by oven at 90ºC for 1 hour, then cooling to reach room temperature. A very thin aluminum layer than a gold layer as the source and drain electrode have been deposited by thermal evaporation technique on the glass substrates by using a mask with conducting channel constant distance of (W/L=1mm/60µm). The pure and doped P3HT thin films were deposited on electrodes by using the spin coating method in (203 nm thickness), and annealed to 120ºC for 10 minutes in laboratory oven. The gate dielectric layer deposited by using spin coating at constant revolution speed (2000rpm), and annealed at 70ºC for 3 hours. The upper aluminum electrode (gate electrode) was deposited by thermal evaporation method under vacuum (10^{-5} torr), to complete an OFET in Top Gate- Bottom Contact (TG-BC) in (AL/PVA/(P3ht:%CNP)/Au configuration. The samples were kept under vacuum (10^{-2} torr) for two days before get measurement.
The Atomic Force Microscopy device (Model: AA3000), equipped from the American company (Angstrom Advanced Inc.), was used to examine the structural properties of thin films of pure and doped P3HT with (1%, 5% v/v ratio of CNP). The output and transfer characterizations of organic thin film transistor was measured by using couple of Keithley-2400 device which are controlling by a computing system. The measurements done in darkness and at room temperature.

Results & Discussion:

The surface nature and the size and distribution of grains for thin films were tested for pure and doped P3HT polymeric with two ratios (1%, 5%) of carbon nanoparticles (CNPs), to see the effect of conformation, using an atomic force microscope device. The results shows affects the surface structural properties of synthetic thin films, as they reduce crystal volumes, increase the number of crystals formed and redistribute the formed crystals, In general, the pure and doped seems a semi-crystalline films, and each less surface roughness and Rq at additive ratio 1% and increase at 5% CNPs, according to the results of the AFM.

Figures (3), (4) and (5) show AFM results of the prepared samples. Table (2) shows the obtained parameters of results. From these tests. Part A of figures shows the three-dimensional image, and part (b) illustrates the image in two dimensions for the prepared samples. It is clear from these results that the P3HT thin films is a semi-crystalline, and with a simple comparison between the results it was found that the surface roughness and root mean square (Rq) decrease when the polymer is doped by 1% of carbon nanoparticles, and this property of the thin film increases when the ratio of doping is 5% of the dopant material. The number of formed grains also increases and the diameter decreases at a doping ratio of 1% compared to the pure membrane of the P3HT. This behavior is appear by increasing the ratio of the CNT substance to 5%. The effect of the P3HT polymeric additive on the nature of crystallization and the increase in the roughness of the outer surface and Rq compared to the pure film (as additional to other effects such as the nature of the solvent, the cooling process speed and the nature of the polymeric chain propagation), are behavior consistent with previous studies. [30-34]

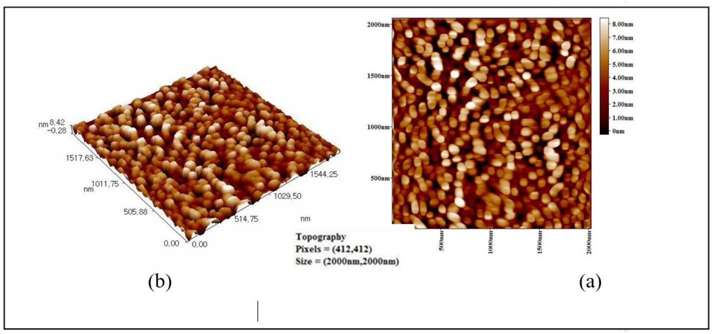


Figure (3): AFM picture of pure P3HT;(a) 3D & (b) 2D.

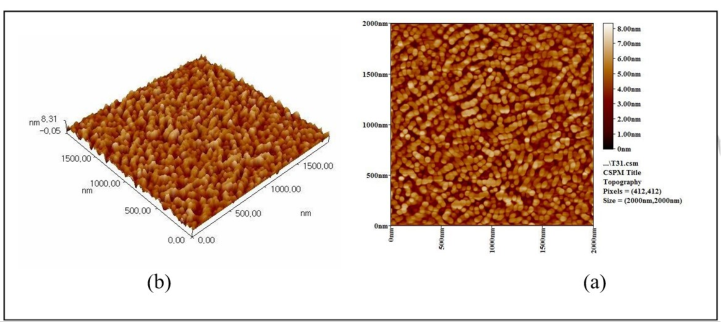
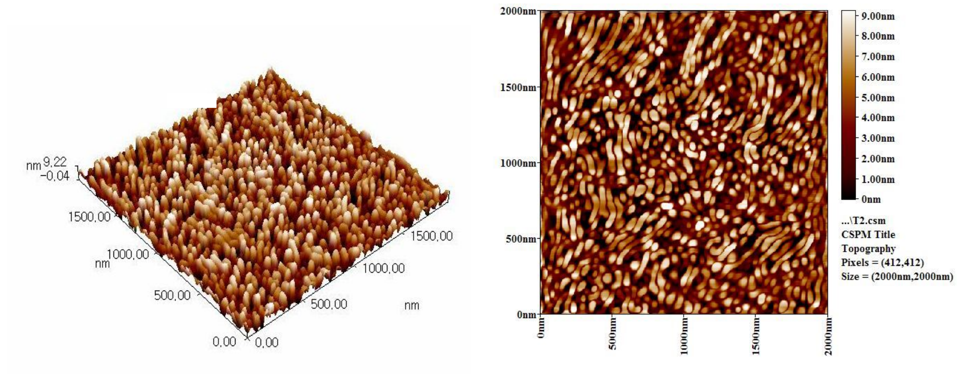


Figure (4): AFM picture of (P3HT:1%CNP);(a) 3D & (b) 2D.



(b) (a)

Figure (5): AFM picture of (P3HT:5%CNP);(a) 3D & (b) 2D.

Tablet (2): The obtained parameters of AFM.

Sample / property	Pure P3HT	P3HT + 1%CNPs	P3HT + 5%CNPs
Rough. Avg. (nm)	1.77	1.02	2.24
$R_{q\ (nm)}$	2.11	1.26	2.6
Grain NO.	173	282	213
Avg. Diameter(nm)	84.93	49.69	57.88

Figures (4) to (8) show the results of the measurement; (a) the output characteristic and (b,c) transfer characteristic of these organic transistors, which shows that the doping of the active layer with various ratio of CNP in the transistor configuration has a significant effect on the electrical properties of this device. Where its electrical capacity per unit area for AL/PVA/Au sandwich structure at constant frequency (1KHz) is (223 nF/cm^2).

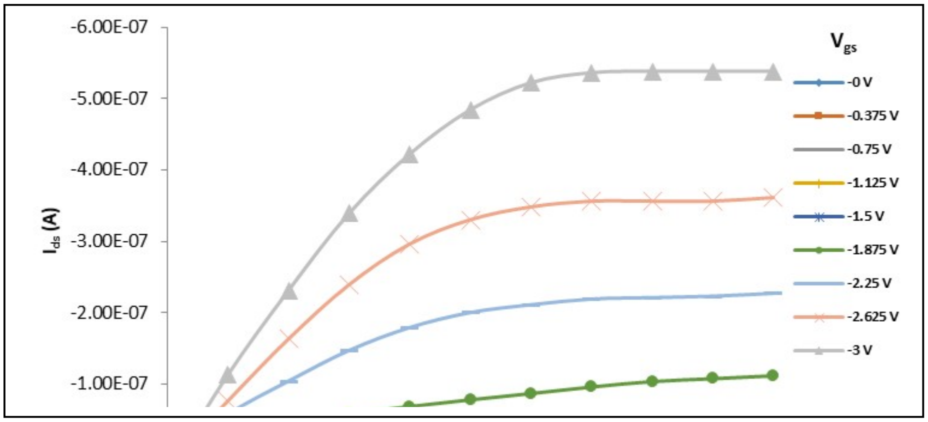


Figure (6a): output characteristic of OFET with pure P3HT.

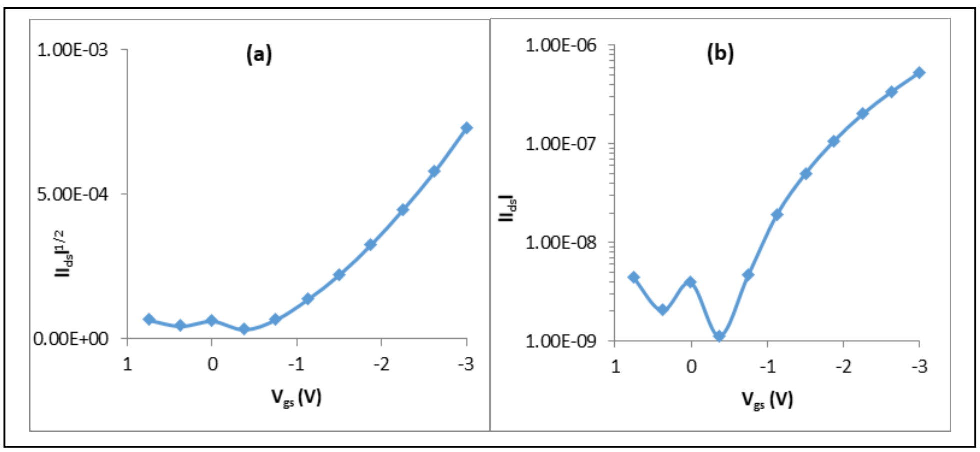


Figure (6b): transfer characteristic of OFET with pure P3HT at (V_{ds}=-2V).

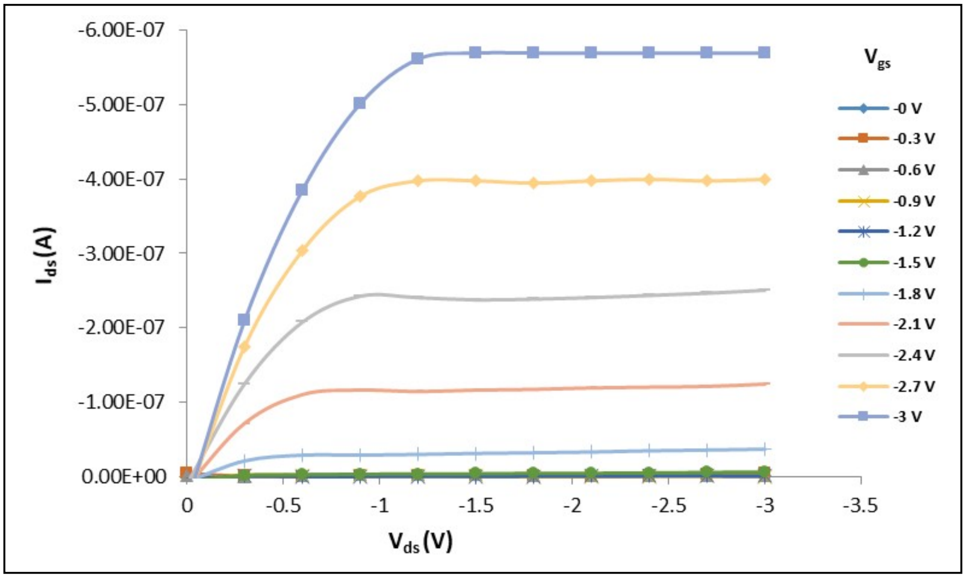


Figure (7a): output characteristic of OFET with P3HT:1%CNP.

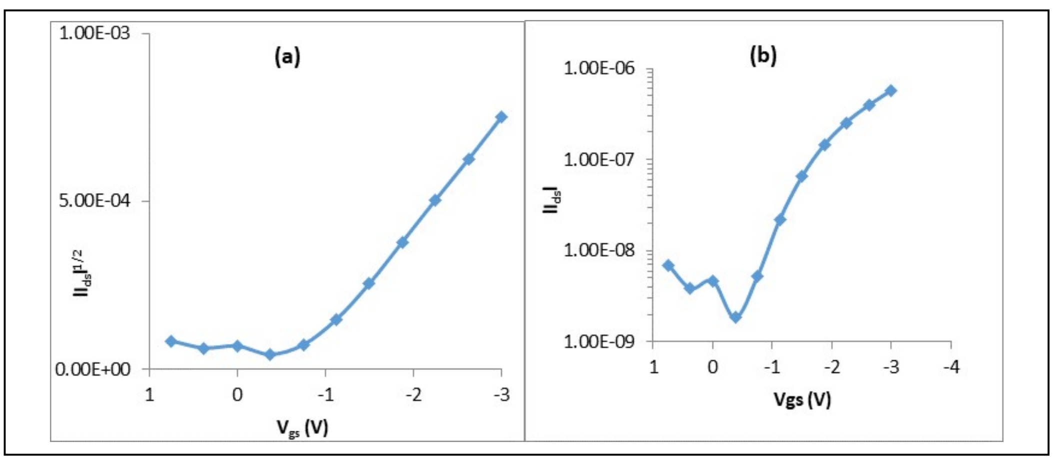


Figure (7b): transfer characteristic of OFET with P3HT:1%CNP at (V_{ds}=-1.2V).

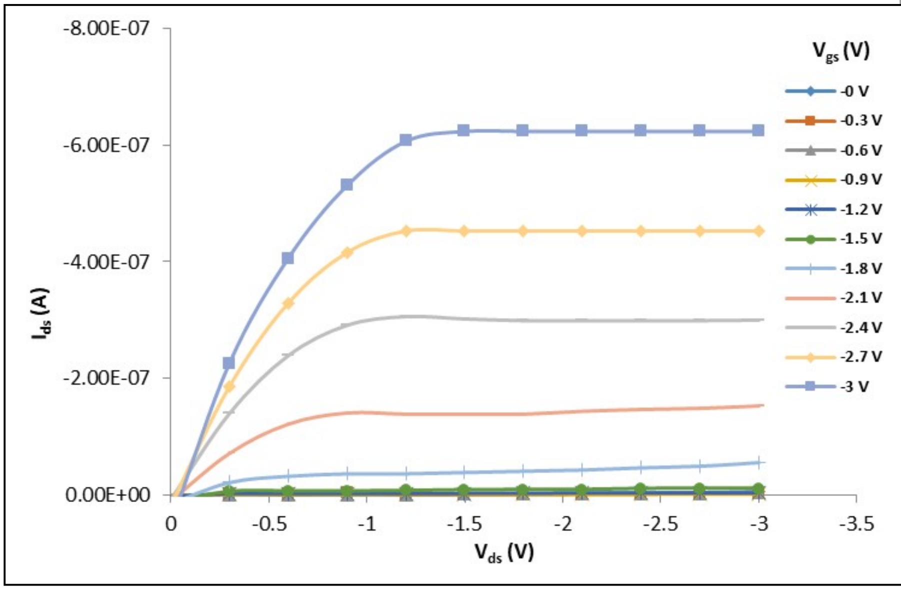


Figure (8a): output characteristic of OFET with P3HT:2%CNP.

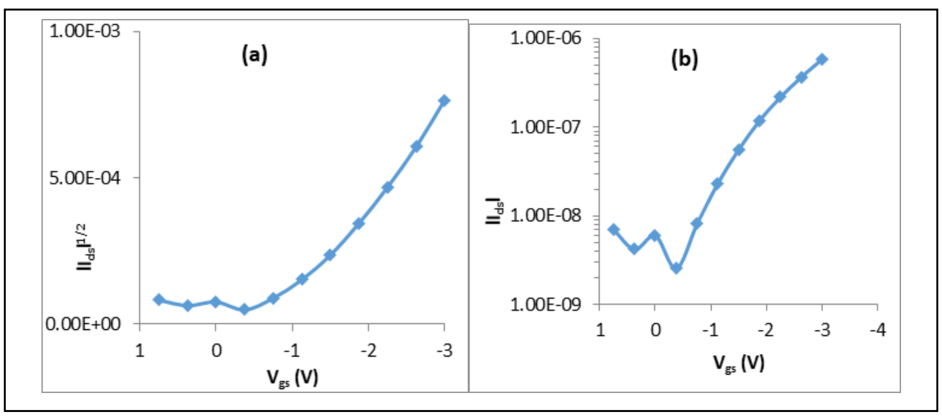


Figure (8b): transfer characteristic of OFET with P3HT:2%CNP at (V_{ds}=-1.5V).

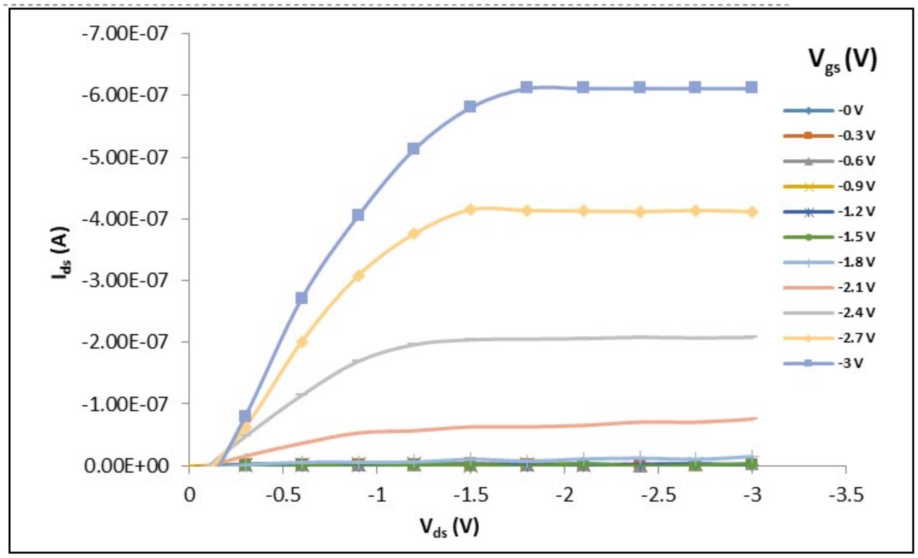


Figure (9a): output characteristic of OFET with P3HT:3%CNP.

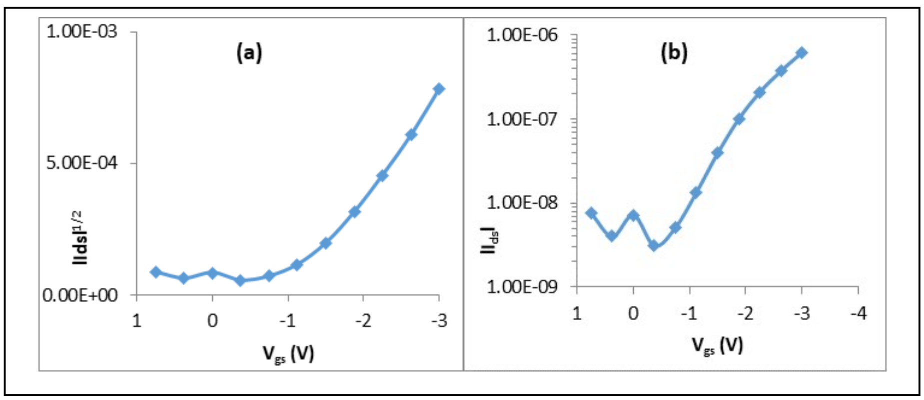
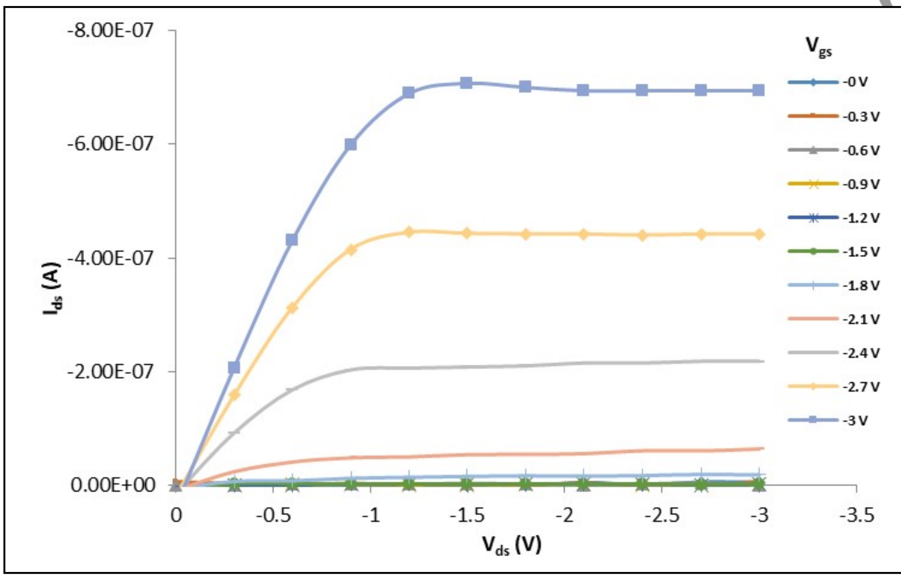


Figure (9b): tranfer characteristic of OFET with P3HT:3%CNP at (V_{ds}=-1.8V).



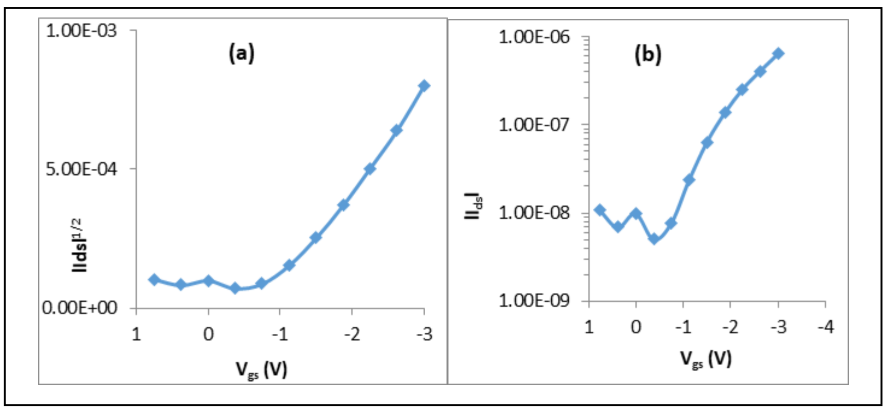


Figure (10b): transfer characteristic of OFET with P3HT:4%CNP at (V_{ds}=-1.2V).

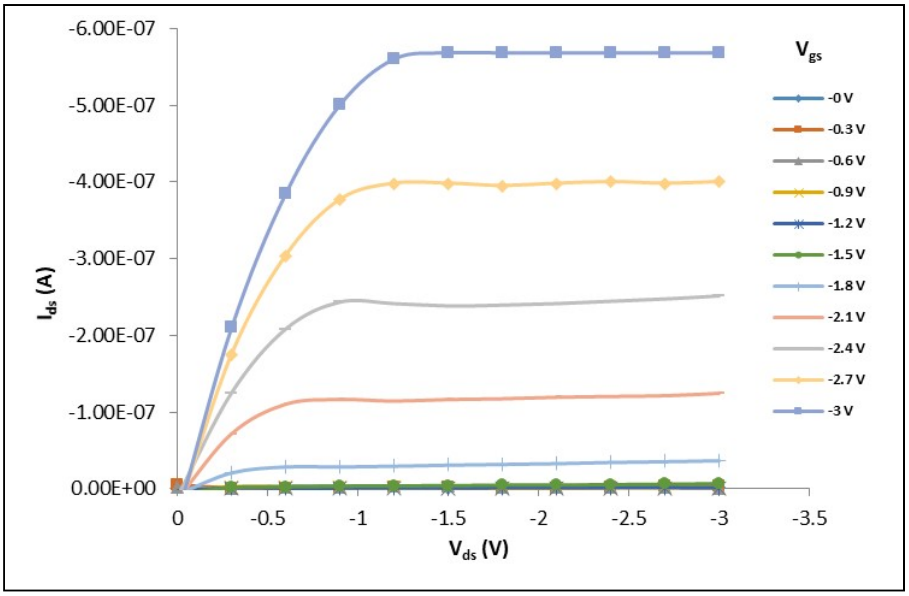


Figure (11a): output characteristic of OFET with P3HT:5% CNP.

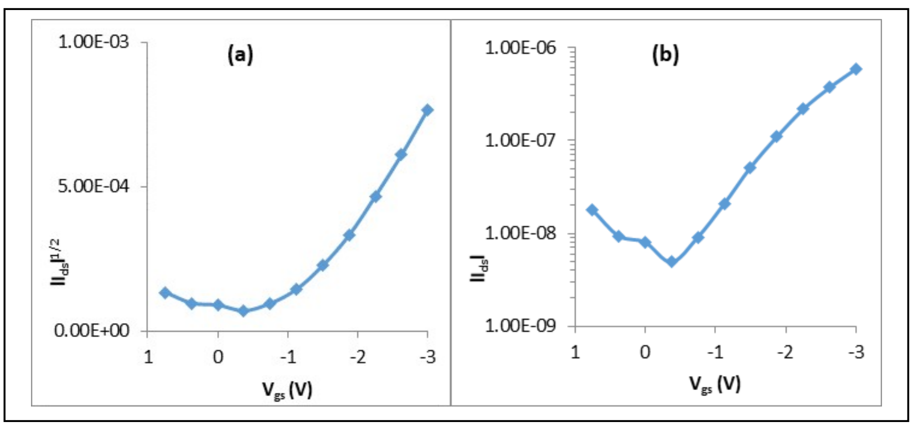


Figure (11b): transfer characteristic of OFET with P3HT:5% CNP at (V_{ds}=-1.2V).

OFET samples was made with a TG-BC configuration to protect the channel of it, by encapsulation it with other layers. The active layer of the P3HT that was deposited in open air and moisture conditions is of course completely unstable in front of these influences due to its low ionization properties (4-4.4 eV), which are considered as a disturbance induces traps. We observed from output characteristics of all OEFT that all these devices operates in enhancement (accumulation) mode. The electrical coefficients of the fabricated organic transistors were calculated from the transfer characteristics of each of them, and by taking advantage of the special relation (1). The mobility of the charge carriers at saturation (μ_{sat}) and the ratio between the operating current (I_{on}) to the extinguishing current (I_{off}) was calculated and the threshold voltage was found directly from the transfer characteristics of the transistors. Table (3) and figures (12) to (14) show the results obtained for this measures.

Tablet (3): the parameters of OFET with doping of active layer.

P3HT:CNPs%	μ_{sat} (cm^2/Vs)	V_{th} (V)	I_{on}/I_{off}
1%	$6.68x10^{-2}$	-0.7	306
2%	$1.09x10^{-1}$	-1.1	229
3%	$1.41x10^{-1}$	-1.2	199
4%	$1.11x10^{-1}$	-1	127
5%	$1.07x10^{-1}$	-0.9	120

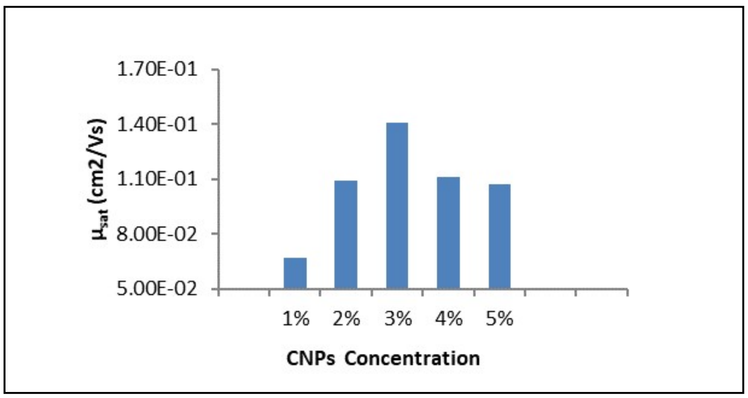


Figure (12): the relationship between mobility at saturation and CNP ratio.

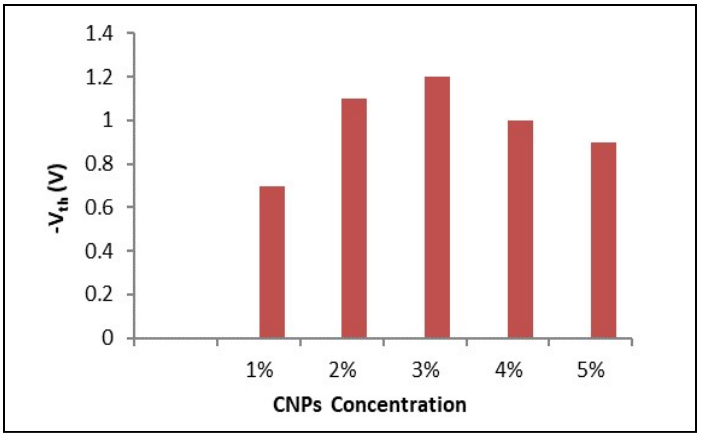


Figure (13): the relationship between threshold votage and CNP ratio.

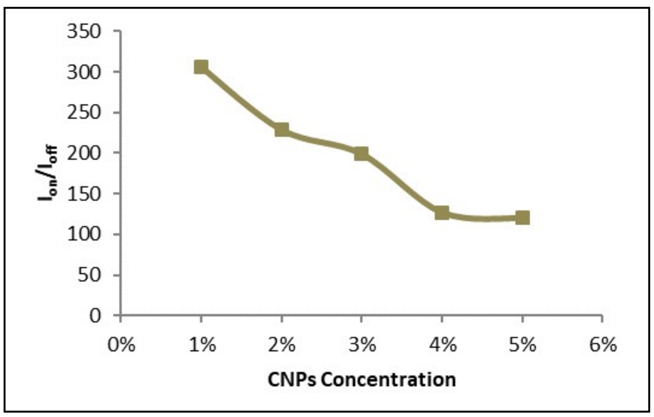


Figure (14): the relationship between I_{on}/I_{off} ratio and CNP ratio.

Conclusion:

The process of conformation of the P3HT thin films with CNPs in two different ratio (1%, 5%) affects the structural characteristics of the synthetic thin films, as it reduces the crystalline volumes, increases the number of crystals formed and redistributes the crystals formed, and in pure and doped samples form semi-crystalline films, The roughness of the surface and Rq decreases at the additive ratio 1% and increases at 5% CNPs, according to the results of the AFM.

The active layer doping with carbon nanoparticles led to reduce the channel resistance, which led to an increase in the mobility of the charge carriers in the conduction channel of the transistors, which is also a natural result of

increasing the electrical conductivity of the active layer as a result of deformation through the formation of the metal islands of the added minutes. Some changes in behavior may be due to the difference in the crystal formation of the gate insulator thin films by changing the characteristics of the active layer which is considered to be the deposition substrate.[35-38]
The decreasing of I_{on}/I_{off} ratio with the increasing of doping ratio of P3HT with CNP possibly due to the changing of the configuration of active layer, thus increasing the contact resistance between the active layer and each of source and drain electrodes.
It is clear from Figure (5-49) that the highest transistors' efficiency was at the T16 transistor, which has a 3% deformation rate in carbon nanoparticles because it has the highest mobility of charge carriers at saturation (1.41x10-1 cm2 / Vs). The threshold voltage change is related to the crystal defects resulting from deformation, which constitute traps for the charge.
Previous studies have shown the possibility of increased mobility between ($2x10^{-4}$-10^{-} cm^2/Vs) by deformation of the active layer P3HT, and in another study increased mobility between ($5x10^{-3}$- $2.5x10^{-2}$ cm^2/Vs) and increase the threshold voltage between ((-8)-(-13)V) and change the negative threshold voltage to positive ((-12.5) - (+3)) through the effective P3HT deformation process with carbon nanoparticles in another study. The researchers found that channel resistance decreased by CNPs by ($5.53x10^8$-$2.08x10^8\Omega$).[39-42]

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