



Fabrication and Evaluation of Electrical Properties of Poly(1,8-diaminonaphthalene) Based Schottky Diode

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ABSTRACT

Poly (1,8-diaminonaphthalene) films as p-type semiconductors were easily synthesized potentiostatically on the Au or indium-tin oxide vacuum deposited glass substrates. Optical transmissions of doped and dedoped films were measured within 250-1100 nm wavelength range and the corresponding optical band gaps were estimated to be 2.84 and 3.62 eV. The polymer was used for the construction of schottky barriers with In, Cu, and Ag metals. The current-voltage and capacitance-voltage characteristics of the diodes were studied to derive information on junction parameters. The electrical characteristics of the junctions at low range potential were analyzed based on the standard thermionic emission theory. The value of ideality factor (η) for In/polymer junction was found to be 9.93 which decreased to 5.23 and 6.30 for Cu and Ag electrodes, respectively. This can be attributed to the formation of complex between Cu and Ag during vacuum deposition of these two metals onto the polymer layer which reduces the thickness of oxide insulator interfacial layer. The capacitance-voltage characteristics of the diode were also studied and the built-in voltage (V_c), the charge carrier concentration (N_s), depletion width, and the work function of poly(1,8-DAN) were estimated. The value of 4.45 eV was obtained for polymer work function.

Key Words:

1,8-diaminonaphthalene;
schottky diode;
band gap;
junction properties;
conducting polymers.

INTRODUCTION

Nowadays, π -conjugated polymers have various applications in different scientific and industrial fields. This is due to their adoptable electrical properties, ease of device fabrication, diversity and also their potentially low cost [1]. Since their discovery, conducting polymers have been extensively studied and used as active components in semiconducting devices. These materials have been considered as electrode active materials in photovoltaic cells, light emitting diodes, electrochromic devices,

and sensors [1].

During the last two decades, there has been considerable interest to replace conventional inorganic semiconductors with organic types in electronic and optical devices. Hence conducting polymers like polypyrrole, polythiophene, polyaniline, and their derivatives have been used for the fabrication of microelectronic devices, particularly Schottky barrier diodes [2-7]. These diodes and other devices have usually been used as power rectifiers, low-level detectors,

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mixers, fast switching circuits, voltage dependent capacitors and field effect transistors. While making electronic devices, metals possessing low work functions are used to make rectifying contacts with semiconductor layers [8-14].

The rectifying quality originates mainly from the Fermi level of the semiconductor. The Fermi level is strongly influenced by the structure, the dopant type, and doping level. Hence, the junction characteristics of the metal-semiconductor Schottky diodes are strongly influenced by the condition of the preparation and nature of the semiconductor layer [13]. Also, the conductivity of the polymer is of major importance in constructing a Schottky barrier. A low doping of polymer is associated with a low number of charge carriers and low conductivity, leading to an extended depletion layer in the junction. On the other hand, high charge carrier densities in the polymer may give rise to a thin barrier with a high tunneling probability [13]. It has been recognized that the conductivity in the order of $10^{-3} \text{ ohm}^{-1}\text{cm}^{-1}$ is suitable for junction formation [13].

Various metal/polymer junctions with electrochemically or chemically synthesized semi-conducting polymers have been used to make Schottky barrier diodes [15-18]. Table 1 compares the reported performance of metal/polymer junctions. Among the conjugated polymers, polyaniline has been considered because of its interesting electrochemical and optical properties, high conductivity, and moderate environmental stability [14]. Various parameters such as the ideality factor (or diode quality factor), barrier height, work function, and saturation current density have been estimated from current-voltage (I-V) and capacitance-voltage (C-V) measurements.

On the other hand, poly(*p*-phenylene) has the advantage of good thermal and environmental stability in comparison with above mentioned polymers [19]. Since, poly(1,8-diaminonaphthalene) (poly(1,8-DAN)) can be considered as a hybrid material of polyaniline and poly(*p*-phenylene), it is less conductive and more stable than polyaniline yet, more conductive than poly(*p*-phenylene). Hence, it possesses the electrical conductivity of the polyaniline together with the stability of the poly(*p*-phenylene), making it a good candidate for junction

fabrication.

Chemical and electrochemical polymerization, applications, and properties of 1,8-DAN in organic and aqueous solutions have been reported in the literature [20-25]. It has been demonstrated that the presence of free amine groups enables poly(1,8-DAN) to form strong complexes with heavy metal cations (Ag^+ , Cu^{2+} , and Hg^{2+}) and this feature makes it useful as metal sensor or metal removing filter for waste water treatment [24]. To our knowledge, no other application of this polymer has been reported up to now.

The aim of this work is to fabricate Schottky diodes based on poly(1,8-DAN) electrochemically prepared in organic solvent. The junction properties of poly(1,8-DAN)/metal devices were studied and compared with those reported in the literature. It is for the first time that this polymer is applied as a semiconductor layer for Schottky diode fabrication. We have reported here the junction characteristics between p-type (anion-doped) poly(1,8-DAN) and various low work function metals such as In, Cu, and Ag. The current-voltage and capacitance-voltage data are analyzed by applying the thermionic emission theory.

EXPERIMENTAL

Materials

Reagent grade 1,8-DAN, tetramethyl ammonium tetrafluoroborate $(\text{CH}_3)_4\text{NBF}_4$, and acetonitrile, all purchased from Merck. Aqueous solutions were prepared from doubly distilled water.

Electrochemical Deposition and Characterization of Poly(1,8-DAN)

Poly(1,8-DAN) were synthesized electrochemically on gold thin film vacuum deposited glass, or indium-tin oxide (ITO) coated glass electrode by constant potential coulometry (CPC) at 900 mV in a simple one compartment glass cell. CPC method was performed with a Metrohm voltammetric analyzer model VA 746. All the potentials are referred to the double junction Ag/AgCl/ Cl^- reference electrode. Poly(1,8-DAN) films were synthesized in acetonitrile solution containing $(\text{CH}_3)_4\text{NBF}_4$ (0.1 M) as

Table 1. Reported performance of metal/polymer junctions.

No.	Device configuration	Ideality factor (η)	Barrier height ϕ (eV)	Reverse saturation current (A/cm^2)	Carrier density (cm^{-3})	V_c (V)	Reference No.		
1	Cu/POT ¹ /PM ² T/Cu	5.7	0.81	3.40×10^{-8}	-	-	6		
2	W/P3MT/Pt	1.85	0.45	-	-	-	7		
3	Al/P3cHT ³ /SnO ₂	9.13	0.80	-	1.36×10^{14}	0.83	8		
	In/P3cHT/SnO ₂	12.3	0.78	-	0.78×10^{14}	1.39			
	Sn/P3cHT/SnO ₂	4.6	0.84	-	0.63×10^{14}	0.69			
	Ag/P3cHT/SnO ₂	ohmic	-	-	-	-			
	Al/P3nHT ⁴ /SnO ₂	20.58	0.46	-	32.5×10^{14}	0.57			
	In/P3nHT/SnO ₂	21.89	0.49	-	18.2×10^{14}	0.49			
	Sn/P3nHT/SnO ₂	25.10	0.48	-	12.4×10^{14}	0.38			
	Ag/P3nHT/SnO ₂	ohmic	-	-	-	-			
	Al/copolymer ⁵ /SnO ₂	18.56	0.50	-	3.86×10^{14}	0.48			
	In/copolymer/SnO ₂	20.57	0.48	-	4.11×10^{14}	0.29			
	Sn/copolymer/SnO ₂	24.57	0.46	-	3.23×10^{14}	0.43			
	Ag/copolymer/SnO ₂	ohmic	-	-	-	-			
	4	Al/PEDT ⁶ /Si	1.42	0.97	-	2.09×10^{18}		0.88	9
	5	Pt/PPy-CuPcTS ⁷ /Au	2.25	-	8.0×10^{-4}	-		-	11
Pt/PPy-NiPcTS ⁸ /Au		3.17	-	2.50×10^{-2}	-	-			
Pt/PPy-TOS ⁹ /Au		4.98	-	1.50×10^{-1}	-	-			
6	Al/PDB ¹⁰ /ITO	2.8	1.08	7.0×10^{-12}	-	-	10		
7	Au/MPP ¹¹ /In-InSe(:Er)	2.45	0.82	7.50×10^{-10}	1.24×10^{18}	0.49	12		
8	Al/PPy ¹² /Au	1.2	0.97	3.30×10^{-10}	2.59×10^{18}	-	13		
	In/PPy/Au	2.1	0.89	8.70×10^{-9}	1.29×10^{19}	-			
9	ITO/PDMA ¹³ /Al	2.4	0.27	2.40×10^{-4}	-	-	17		
	ITO/POMA ¹⁴ /Al	3.7	0.28	2.20×10^{-4}	-	-			
10	Pt/ PAn-PVC ¹⁵ /In	2.61	0.884	2.29×10^{-8}	2.70×10^{16}	0.74	15		
11	In/PAn ¹⁶ /Pt	3.12	0.803	4.90×10^{-7}	3.06×10^{17}	0.69	16		
	In/PAnPS ¹⁷ /Pt	2.69	0.895	1.48×10^{-8}	3.30×10^{19}	0.78			
12	Al/PNmA ¹⁸ /Au	1.83	0.89	7.43×10^{-9}	1.03×10^{19}	0.84	18		
	In/PNmA/Au	1.39	0.79	3.85×10^{-7}	5.19×10^{18}	0.89			
	Sb/PNmA/Au	2.70	0.95	9.10×10^{-10}	5.19×10^{18}	0.76			
	Sn/PNmA/Au	2.13	0.96	4.91×10^{-10}	1.73×10^{14}	0.72			

(1) poly(3-octyl thiophene); (2) poly(3-methyl thiophene); (3) poly(3-cyclohexylthiophene); (4) poly(3-n-hexylthiophene); (5) copolymer of P3nHT and P3cHT; (6) poly(3,4-ethylenedioxythiophene); (7) copper phthalocyanine toluenesulphonate; (8) nickel phthalocyanine toluenesulphonate; (9) tetraethylammonium toluenesulphonate; (10) poly[3-(2',5"-diheptyloxyphenyl)-2,2'-bithiophene]; (11) metallic polypyrrole; (12) polypyrrole; (13) poly(2,5-dimethoxyaniline); (14) poly(o-methoxyaniline); (15) polyaniline with polyvinyl chloride; (16) pure polyaniline; (17) polyaniline with polystyrene; (18) poly(N-methylaniline).

supporting electrolyte and 1,8-DAN (0.1 M) as monomer.

The morphology and thickness of the films were investigated by a Philips XL (series XL30) scanning electron microscope.

The optical transmission of the poly(1,8-DAN)

thin film (186 nm) deposited on ITO electrode was recorded using Agilent 8453 single beam spectrophotometer in the wavelength range of 250-1100 nm. The transmission spectrum was corrected relative to that of the ITO glass. The energy band gap was calculated from the absorbance data.

The Au/poly(1,8-DAN)/metal (In, Cu, and Ag) devices were fabricated by deposition of In, Cu, and Ag (0.03 cm^2) by vacuum evaporation technique under $\sim 1.0 \times 10^{-6}$ millibar using VAS BUC 78535-FRANCE instrument. Conductive silver paste was used to take contact leads from polymer and metal.

I-V characteristics of the diodes were recorded at 298 K with DS-1080C 250 MHz oscilloscope and a function generator (TG 230, 2 MHz) and C-V measurements were made using digital RCL meter (Fluke PM 6306, 1 MHz).

RESULTS AND DISCUSSION

Morphological Characteristics and Optical Properties of Poly(1,8-DAN)

Surface morphology of two poly(1,8-DAN) films are shown in Figures 1a and 1b. Low thickness film in Figure 1a is very uniform and smooth without any pinhole. Defect free semiconductor polymers are

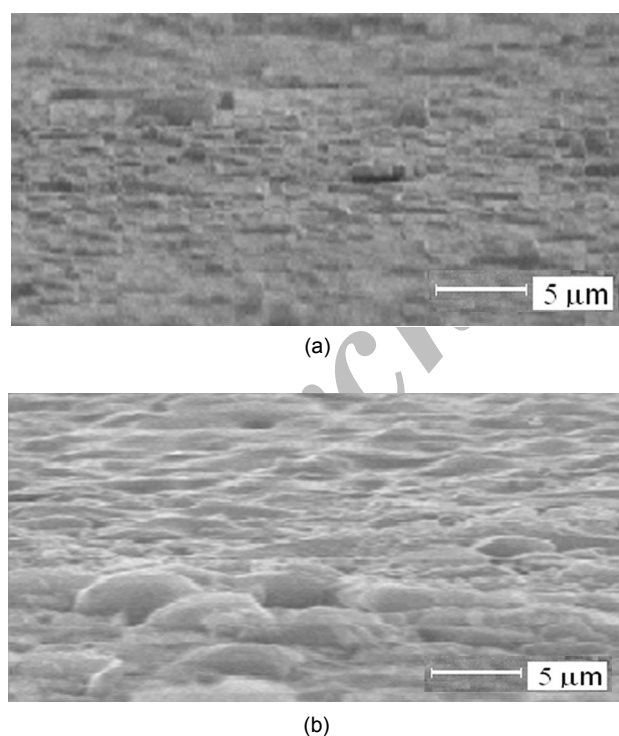


Figure 1. SEM micrographs of poly(1,8-DAN) prepared in acetonitrile solution containing 0.1 M monomer and 0.1 M BF_4^- as dopant ion by constant potential method at 900 mV and different polymerization time: (a) thickness = 186 nm and (b) thickness = 25 μm .

very suitable for creating homogeneous contact with metals during coating process.

Typical absorption spectra of poly(1,8-DAN) at the doped and dedoped states, coated potentiostatically on an optical transparent ITO substrate, are presented as insets in Figure 2. The energy gap of the polymer is estimated using the fundamental law:

$$\alpha = A(h\nu - E_g)^n \quad (1)$$

where, α is the absorption coefficient, h is the photon energy, A is a proportionality constant, $n = 1/2$ for direct transition, and $n = 2$ for indirect allowed transitions.

From the slopes of the graphs of $\log \alpha$ against $\log h\nu$ taking by a linear fit (not shown here) a value

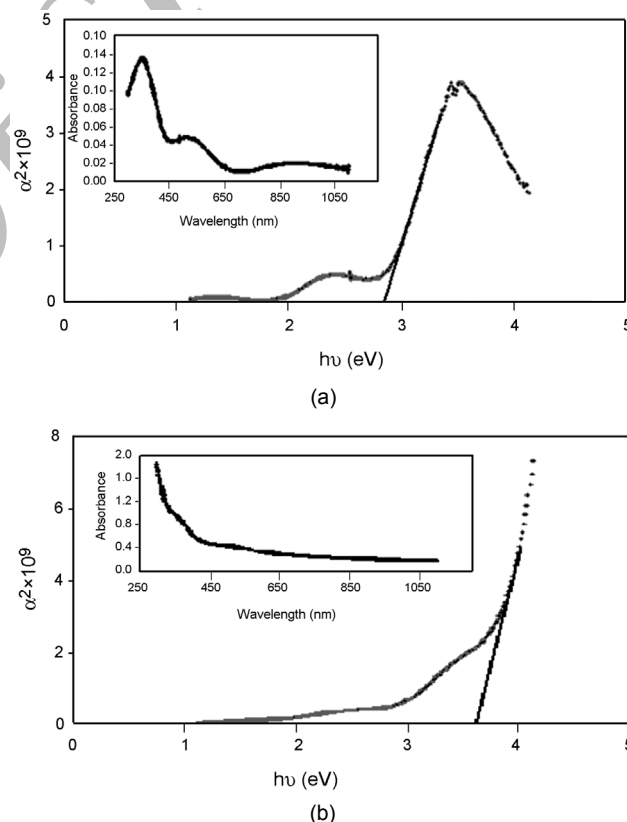


Figure 2. Plots of absorption constant (α^2) vs. $h\nu$ used for the evaluation of band gap of: (a) $(\text{CH}_3)_4\text{BF}_4^-$ doped poly(1,8-DAN) film coated on an indium-tin-oxide (ITO) substrate (thickness = 186 nm) and (b) dedoped poly(1,8-DAN) film using concentrated ammonia solution (30%) for 1.0 h. Insets: UV-vis optical absorption spectra (absorbance vs. wavelength) of films (a) and (b) coated on ITO used for the evaluation of band gap energy (E_g) of poly(1,8-DAN).

of $n = 1/2$ was obtained, indicating the direct transition of the electron during light absorption. Graphs of α^2 versus $h\nu$ (Figure 2) are straight lines, whose intercepts on the energy axis are the energy band gaps. The E_g values of the polymer in the doped and dedoped states are estimated to be 2.84 and 3.62 eV, respectively. Dedoping process leads to elimination of the energy levels distributed near the edges of conduction and valence bands, hence increases the band gap interval of the polymer.

Current-voltage Characteristics

Figure 3 Shows typical I-V characteristics of Au/poly(1,8-DAN)/metal (In, Cu, and Ag) at room temperature. The current which passes across the polymer/metal junctions is usually accounted for the processes on the basis of thermionic emission, space-charge-limited current (SCLC), or Poole-Frenkel emission [18]. SCLC and Poole-Frenkel mechanisms are followed by a system whose graphs of $\log I$ vs. $\log V$ and $\ln(I/V)$ vs. $V^{1/2}$ are linear. These two mechanisms are not applicable to our diodes in the low voltage range (<0.5 V), although they cannot be neglected at higher voltages.

The Schottky effect involves the emission of the electrons by thermal activation from metal electrode into the conduction band of a semiconductor polymer over a potential barrier between the metal/polymer interfaces. For the thermionic emission model, the I-V relationship is expressed by:

$$I = I_0[\exp(qV/\eta kT)] \quad (2)$$

where, I_0 is saturation current, q is elementary charge, V is applied voltage, η is ideality factor, k is Boltzman constant, and T is absolute temperature. The saturation current is given by

$$I_0 = SA^*T^2 \exp(-q\Phi_B/kT) \quad (3)$$

where, S is the surface area of the diode, A^* is Richardson constant ($120 \text{ A/cm}^2\text{K}^2$ for free electron), and Φ_B is barrier height. According to eqn (3) as $qV/kT \gg 1$, $\ln(I)$ versus V should be linear with an intercept corresponding to I_0 .

The ideality factor can be obtained from the slope of the logarithmic graph of I vs. V (Figure 4). From I_0 , barrier height, Φ_B , can be deduced. At a forward bias

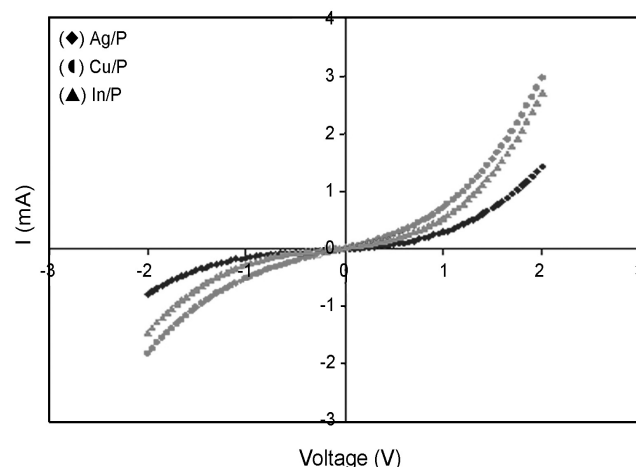


Figure 3. Current versus applied voltage (I-V) for Au/poly(1,8-DAN)/metal.

voltage lower than 0.5 V the graph of $\ln I$ vs. V is linear and the diode follows the thermionic emission mechanism. The parameters of the metal/polymer junctions given in Table 2 are calculated using the above equations.

A system is called a rectifier of electrical current through which electrons flow asymmetrically. In the formation of a rectifying barrier at the interface, the work function of the metal must be smaller than that of the p-type semiconductor. If the work functions are of the reverse order, an ohmic contact would be formed. Rectifying ratio can be defined as the ratio of

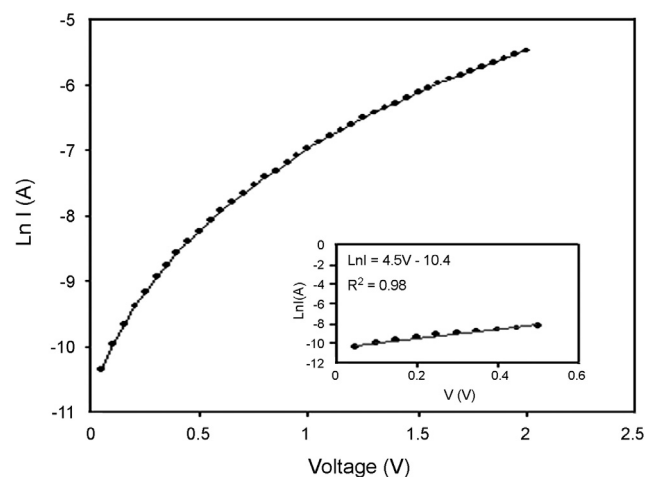


Figure 4. Plot of $\ln I$ vs. applied voltage of the junction for Au/poly(1,8-DAN)/In Schottky diode. Inset shows the linear part of the plot lower than 0.5 V.

Table 2. Performance characteristics of metal/poly(1,8-DAN) junctions.

Device configuration	Ideality factor (η)	Reverse saturation current $I_0(A) \times 10^{-5}$	Contact potential (V_c) (V)	Carrier concentration (cm^{-3})	Depletion width (D) (nm)	Barrier height ϕ_B (eV)	Rectification ratio
Au/poly(1,8-DAN)/Cu	5.23	5.55	0.75	-	-	0.46	1.5
Au/poly(1,8-DAN)/Ag	6.30	2.67	1.25	-	-	0.42	2.3
Au/poly(1,8-DAN)/In	9.93	4.07	0.95	5.23×10^{19}	1.5	0.36	2.0

the current passed through the diode in the reverse and direct bias at a particular voltage. From Table 2 it can be seen that Cu, Ag, and In metals make rectifying junctions with polymer. However, Ag/polymer and Cu/polymer interfaces exhibit more rectifying properties than In/polymer interface.

The value of ideality factor (η) (an indication of the diode quality) for In/polymer junction is 9.93 which decreased to 5.23 and 6.30 for Cu and Ag electrodes, respectively. Better rectification ratios and ideality factors can be assigned to the formation of complex between polymer and Cu or Ag during vacuum deposition of these two metals onto polymer layer [24] which reduces the thickness of oxide insulator interfacial layer. The thicker the oxide layer, the larger would be the value of η .

In overall, deviation from ideal value ($\eta = 1.02$) is a sign of the presence of an interfacial layer between metal and semi-conductor polymer layer which introduces the interface states at the contact area [14]. The bulk resistance and Poole-Frenkel effects may also be responsible for the deviation of the ideality factor from unity [14]. Also the height of the barrier is increased with decreasing the interfacial layer thickness and results in falling saturation current of eqns (2) and (3). A comparison of the reported values for the various parameters of different polymer/metal junctions (Table 1) with the present data (Table 2) indicates superiority of 1,8-DAN than the other investigated polymer (Table 1 rows No. 1 to 7) with respect to the ideality factor, carrier charge density, reverse saturation current, and barrier height for the diode construction.

Capacitance-voltage Characteristics

According to Schottky theory, the capacitance of depletion layer per unit area varies as a function of the

reverse bias voltage:

$$C^{-2} = [2(V_c - V) / S^2 q \epsilon \epsilon_0 N_s] \quad (4)$$

where, V_c is built-in or contact potential at zero bias, N_s is the carrier concentration, ϵ is the semiconductor dielectric constant, and S is the effective area of the metal contact. A graph of $1/C^2$ vs. V is a straight line (Figure 5) unless interface states across the band gap and deep donor levels exist in the bulk of the semiconductor exist. The intercept of the line with the voltage abscissa determines the built-in voltage (V_c) and the slope gives the carrier concentration (N_s). The carrier mobility μ can be determined from the expression for conductivity $\sigma = N_s q \mu$. Depletion width also can be evaluated by the following equation:

$$D = [2\epsilon \epsilon_0 (V_c + V) / (q N_s)]^{1/2} \quad (5)$$

The calculated potential, depletion width, and carrier concentration are given in Table 2. The work function of poly(1,8-DAN) can be obtained using eqn (6) [18].

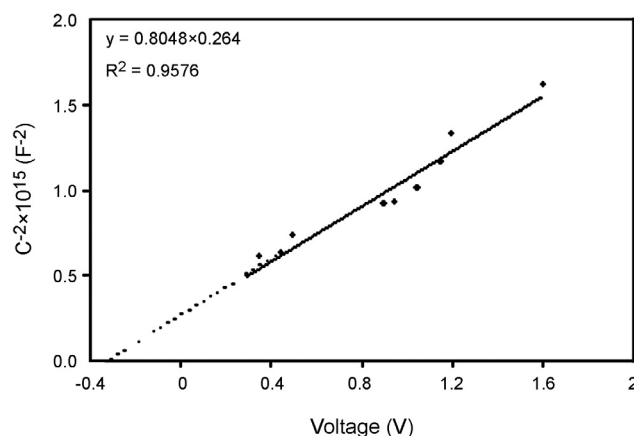


Figure 5. A plot of C^{-2} versus applied voltage for Au/poly(1,8-DAN)/In at 50 Hz.

$$\phi_{sp} = qV_c + \phi_m \quad (6)$$

where, ϕ_{sp} and ϕ_m are the work functions of the semi-conducting polymer and the metal, respectively. By substituting the values of the work function of the metal ($\phi_{In} = 4.12$ eV) and the contact potential, the work function of the polymer is found to be 4.45 eV. The work function of the gold electrode is 5.1 eV and those of Al and Ag are 3.74 eV and 4.28 eV, respectively. Hence, the work function of the polymer lies between the metals and gold and it can form ohmic and rectifying contacts with gold and other metals, respectively.

From barrier height and built-in voltage values, the Fermi level E_F is obtained about 0.59 eV above the valance band edge.

CONCLUSION

We have prepared a new Schottky diode with poly(1,8-DAN) as a semiconductor, where Au and In, Cu and Ag were used as supporting contact metals. Thermoionic emission model appears to be dominant at low bias voltage range. However, Poole-Frenkel and SCLC effect should be considered at high bias voltage along with Schottky emission. All the junctions showed rectification, but the estimated values of saturation current and ideality factor are high. However, it was essential to optimize the synthesis conditions for the preparation of the polymer film. For example by the regioselective synthesis methods, one type of polymer chains can be achieved. It has been demonstrated that 1,8-DAN monomers link to each other from different active sites leading to the formation of polymer chains with different structures. Meanwhile the conductivity should be improved and the thickness of the polymer to be decreased. The work function of the polymer is found to be about 4.45 eV.

SYMBOLS AND ABBREVIATIONS

1,8-DAN : 1,8-Diaminonaphthalene
 I-V : Current-voltage
 C-V : Capacitance-voltage

ITO : Indium-tin oxide
 CPC : Constant potential coulometry
 α : Absorption coefficient
 A : Absorbance
 E_g : Band gap
 A^* : Richardson constant
 SCLC : Space-charge-limited current
 I_0 : Saturation current
 Q : Elementary charge
 V : Applied voltage
 η : Ideality factor,
 K : Boltzman constant
 T : Absolute temperature
 ϕ_B : Barrier height
 V_c : Built-in or contact potential at zero bias
 N_s : Carrier concentration
 ϵ : Semiconductor dielectric constant
 S : Effective area
 D : Depletion width
 ϕ_{sp} : Work function of the semiconducting polymer
 ϕ_m : Work function of the metal
 E_F : Fermi level

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