The use of zinc metal as a rectifying contact for polyaniline Schottky devices

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Films of polyaniline (PANI) doped with various acids, e.g., hydrochloric acid (HCl), dodecylbenzensulphonic acid (DBSA) and p-toluene sulphonic acid (p-TSA) were prepared. The effects of different dopants on the asymmetric of the current-voltage characteristics of the ITO/PANI/Zinc metal electrode herterostructure had been investigated. The electronic parameters that controlled the device performance such as rectifying ratio, barrier height, ideality factor, built-in potential and concentration of carriers were calculated. Ideality factors were obtained in the range between 1.5 and 3.93 and the barrier heights of polyaniline-metal heterojunctions was about 0.75 V. It was found that the dopant type affects the conduction mechanism of these devices. The use of HCl as a dopant for PANI results in better rectification behavior than those obtained with either dodecylbnezen sulphonic acid or p-toluene sulphonic acid. The obtained C-V characteristics showed that the concentration of charge carriers is in the range of $10^{16}/cm^3$.

هذا البحث يصف طريقة لتحضير غشاء رقيق من مادة البولى أنيلين الموصلة المطعمة بأحماض مختلفة . أيضا فإن الوصلة الثنائية والتى تم تحضيرها بتبخير عنصر الخارصين تحت التفريغ قد تمت دراستها واختبارها عن طريق دراسة الخواص الإلكترونية لهذه النبيطة مثل (Rectifying Ratio, Barrier Height, Ideality Factor, Built in potential and Carrier Concentration) النتائج التى تم الحصول عليها تؤكد أن هذه النبيطة ذات خواص جيدة ويمكن استخدامها فى تطبيقات عديدة .

Keywords: Conducting polymers, Polyaniline, Schottky devices, Zn contact

1. Introduction

Intrinsic conducting polymers are an exciting new class of electronic materials which have attracted increasing interest since their discovery in 1970's [1, 2]. These conducting polymers have the potential of combining both high conductivities of pure metals and the ~~processability~~processability and low density.

——Polyaniline (PANI) receives great attention as a conducting polymer due to its good environmental stability, ease of preparation and moderate conductivity [3]. ~~Polyaniline has been known to be p-type semiconductors [ ].~~ PANI is applied for many industrial applications such as Schottky devices, light emitting diodes, sensors, smart windows, photoelectrochemical solar cells and rechargeable batteries [3--9–].

— Schottky diode consists of a metal and p-type polyaniline films where metals possessing low work function provide the rectifying contact. Pandy et al. [7] have fabricated junctions by depositing different metals such as Al, Ag, Sn and In on films of polyaniline [7]. Chanudhari et al. [10] studied metals-polyaniline Schottky junctions, where polyaniline was doped with various dopants such as hydrochloric acid, formic acid, iodine and methylene blue. In addition, Schottky junctions based on conjugated polymers have been investigated in the dark and under illumination with white light [11]. Angappane et al. prepared Schottky diode using polyaniline and insulating polymers like polyaniline-polymethylmethacrylate blends, where Schottky barriers were prepared by thermal evaporation of gold metals electrode onto free standing films of the blends [12].

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Schottky devices fabricated using polyaniline with different doping levels and Sn and Al contacts were also investigated [13, 14]. Schottky barrier diode based on composite of polyaniline with polyvinylchloride has been fabricated using Al as Schottky contact and platinum as an ohmic contact [15, 16]. Poly (O-methoxyaniline) and poly (2, 5-dimethoxyaniline) were used for the fabrication of Schottky diode devices ~~with sandwich structure~~ [17]. Schottky barrier diode of polytoludine with different metal contacts such as Al, Cu and Zn has been fabricated by Singh et al. [18].

The work function of polyaniline was found to be in the range 4.1 to 4.38 eV. and an ohmic contact can be realized using a metallic electrode having large work functions such as Au or Ag or Pt or ITO. In the case of a metallic electrode having a small work function, such as Al, Sn, In or Pb Schottky barrier is formed at the interface between the metal and the polymer. The properties of these devices are strongly influenced by the nature and the structure of the interface between the polymer and the metals. This interface is expected to play a prominent role in the context of electronic devices because the interactions at the metal/polymer interface affect, not only the mechanical and thermal stability, but also the transport properties of the junction [19].

Although, Zn has a low work function (4.11 eV), which makes it suitable for a rectifying contact, there is no available studies using Zn as rectifying contact with PANI. The aim of this work is to study Zn metal as a rectifying contact with PANI films and the study the performance of the formed diodes. In addition, the effect of dopant type of PANI films on electronic parameters of Schottky diode was also investigated.

~~Conductive polymers like polypyrrole, polythiophene, polyaniline, polycarbazole , polyazulene and their derivatives have been known to have defined electronic properties which are suitable to make electronic devices [1-3]. Fabrication of solid state heterojunction/ Schottky devices using semiconducting polymer is one of the most important application for such materials [4-6].~~

~~The new generation of polymer solar cells and Schottky devices are cheap and easyand easy to fabricate. Polyaniline receives greater attention as an organic semiconduting material due to its good environmental stability, easy of preparation and moderate conductivity. Polyaniline has been known to be p-type semiconductors [4].~~

~~First report on such a Schottky diode explored the use of polyacetylene as the semiconducting polymer [7]. The electronic parameters controlling the devices performance, such as rectifying ratio, barrier height, ideality factor, concentration of carriers and built in potential were evaluated using current density (J) versus applies voltage (V) and capacitance versus voltage (C-V) measurements.~~

~~S.S Pandy et al [8] have fabricated junctions by depositing different metals such as Al, Ag, Sn and In on pellet of polyaniline [8]. Metals-polyaniline Schottky junctions , where polyaniline was doped with various dopants such as hydrochloric acid, formic acid, iodine and methylene blue were studied by Chanudhari et al [5]. In addition, Schottky junctions based on conjugated polymers have been investigated in the dark and under white light illumination [9]. Angappane et al [10] prepared the fabrication of Schottky barrier using polyaniline and insulating polymer like polyaniline-polymethylmethacrylate blends, where the Schottky barriers were reported by thermal evaporation of gold metals electrode onto free standing films of the blends [10]. Schottky barrier diode based on composite of polyaniline with polyvinylchloride has been fabricated using Al as Schottky contact and platinum as an Ohmic contact [11-12]. Poly (O-methoxyaniline) and poly (2,5-dimethoxyaniline) were used for fabrication Schottky diode devices with sandwich structure [13]. Schottky barrier diode of polytoludine with different metal contacts such as Al, Cu and Zn has been fabricated by Singh et al [14]. There are very little studies using SnZn metals as Schottky contact with PANI films. The aim of this work is devoted to study Sn Zn metal contact with PANI films~~

~~prepared electrochemically with comparison of conventional contact (AL and Sn). Also the effect of dopant of polyaniline on the devices performance will be investigated.~~

2. Experimental work

2.1. Preparation of polyaniline

Aniline was purified by double distillation under reduced pressure. The electrochemical polymerization of aniline was performed using a potentiostat (Wenking M87) and a voltage scanner (Wenking MVS 87). A three electrode single-compartment electrochemical cell was used for the electrochemical experiments. The working electrode was an indium-tin oxide (ITO) coated glass plate with ~~(~~sheet resistance of~~=~~ 10 Ω/ڨ~~)~~ and~~with~~ ~~a~~ area of 1x2 cm^2, a platinum flag was used as the auxiliary electrode, and a saturated calomel electrode (SCE) as the reference electrode. The electropolymerization was performed using an aqueous solution containing 0.5 M HCl and 0.12 M of aniline. The films of PANI were deposited on the working electrode using cyclic voltammetry technique. The electrode potential was cycled between -0.2 V and 1.3 V vs. SCE for one hour, with a potential sweep rate of 5 mV/s. The polymeric films were prepared at 25 ^{0}C. After the synthesis; the films were dedoped by dipping~~ed~~ them in ammonia solution (8 %) for 12 hrs. Redoping these films was carried out either in 0.1 M HCl, 0.1 M dodecylbenzensulfonic acid (DBSA), 0.1 M p-toluene sulphonic acid (p-TSA) for 7 hrs. After the redoping process these films were dried. A piece of Zn metal of 99.998 % purity was evaporated on the prepared PANI films to form the Schottky electrode using Edward E306A vacuum coating machine at pressure of 10^{-5} mbar.

2.2. ~~*After the redoping process these films were dried. On the other side of the PANI films Sn were deposited by vacuum evaporation technique using E306A coating system at pressure of the order of 10 $^{-5}$ torr to fabricate the Schottky devices.*~~

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Characterization of PANI schottky devices

The characterizations of different Schottky devices with PANI films doped with the different acids were carried out by measuring the current-voltage curves and measuring the capacitance-voltage-frequency characteristics using 4277A LCZ meter. The estimated parameters of the device, namely, saturation current, barrier height, ideality factor, built in potential and the rectification ratio which is defined as the ratio between the forward current and reverse current at the same voltage (1V) were calculated from I-V and C-V characteristics.

3. Results and discussion

~~*According to the theory of Schottky barrier, the work function of the metal must be smaller than that of the p-type semiconductor and a rectifying barrier would be formed at the interface. If the work functions were in reverse order, an Ohmic contact would exist rather than the rectifying barrier [15].*~~

~~*It has been reported that the work function of polyaniline lies between 4.1 and 4.38 eV. As generally known, in the case of p-type semiconductors (such as polyaniline) an Ohmic contact can be realized in the junction using a metallic electrode having large work function such as Au or Ag or Pt or ITO. In the case of a metallic electrode having a small work function, such as Al, Sn, In, Pb Schottky barrier is formed at the interface between the metal and the polymer. The properties of these devices are strongly influenced by the nature and the structure of the interface between the polymer and the metals [16].*~~

3.1. *I-V characteristic curves*

~~All the I-V characteristics curves are asymmetrical and show a rectifying behavior. It is found that the type of dopant affect I-V characteristic curves significantly.~~Fig. 1 shows the current vs. voltage characteristics of Schottky diodes, ITO/ PANI-based film/Zn, using different doping acids; HCl, DBSA, p-TSA~~Sn~~. All the I-V characteristics curves show a rectifying behavior. These curves indicate that in the forward bias conditions; relative blocking of charge carriers occurs at the interface up to a certain voltage, after which the junction become conducting. It is observed that for diode where HCl was used as a dopant, the current pass in the diode is higher than those where using DBSA and P-TSA dopants as shown in fig. 1. The dopant type is found not only to affect conductivity of PANI films but also to induce further change the I-V characteristics of the ITO/PANI/Zn heterostructure. This may be explained to be the interaction between dopant and metal contact which leads to the formation of macromolecular complexes [20]. So, chemical interactions of dopant and metal electrode at the film/metal interface can be anticipated, which affects charge transport through the heterojunction substantially.

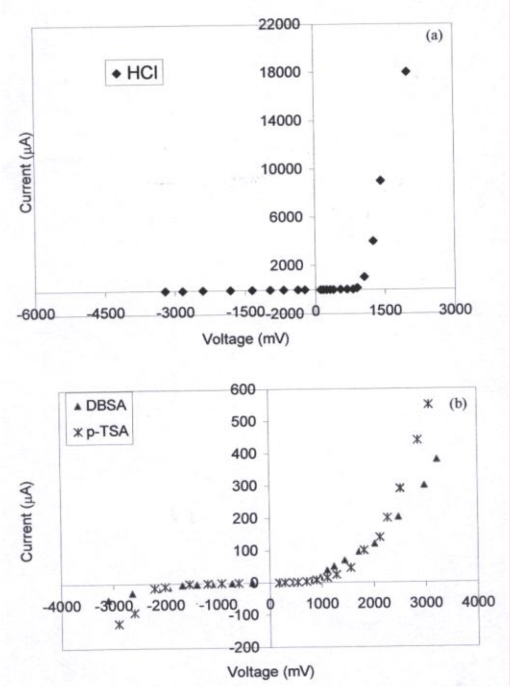

Fig. 1. Current vs voltage for different Schottky devices of Zn/PANI/ITO using different dopant for PANI; curve a) HCl and Curve b) DBSA and p-TSA.

3.2. Mechanisms of current transports in An/ PANI-based film/ITO heterostructure

Up till now there are debates concerning the mechanism of conduction in polyaniline Schottky diodes. It is known that the carrier transport in metal semiconductor contacts is mainly due to majority carriers, in contrast to the case for p-n junction [21~~6~~]. Generally, the non-linear I-V characteristics of the PANI/Zn junction could be due to the space charge limited conduction (SCLC), Poole-Frenkel~~emission~~ emission,~~or~~ thermoionic emission –theory, ~~or~~ diffusion theory or trap charge and discharge phenomena [15, 16, 20, 22].

The non-linearity of Ln~~n~~ J versus Ln V curves of Schottky diodes of PANI doped with HCl or DBSA as shown in fig. 2 eliminates the possibility of SCLC process [19, 21]. But the linear relation between Ln J against Ln V for Schottky diode of PANI doped with p-TSA indicates that the dominant mechanism of this diode is SCLC [20, 22]. The Ln (J/V) versus $V^{1/2}$ plot is also non-linear as shown in fig. 3, which eliminates the possibility of Poole-Frenkel emission [22~~11~~].

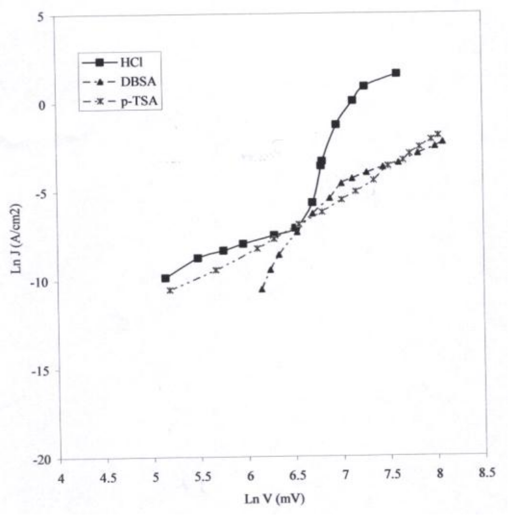

Fig. 2.Ln J vs. Ln V for different Schottky devices of ITO/PANI doped with different acid/Zn.

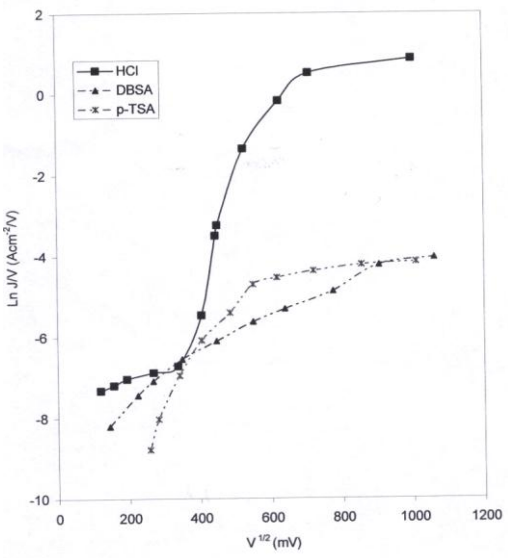

Fig.3. Ln J/V vs. $V^{1/2}$ for different Schottky devices of ITO/PANI doped with different acid/Zn.

Fig. 4 shows the reverse current versus the square root of the applied reverse voltage of Schottky diodes used PANI films doped with HCl, DBSA or p-TSA and Zn metal as a rectifying contact. It is noted that the reverse current increases sharply at large values of reverse voltage for the diode used PANI films doped p-TSA. This indicates that the conduction mechanism in this diode is controlled by both SCLC and diffusion theory [21].

The nonlinear relationship between Ln J and V as given in fig. 5 for different Schottky diodes with PANI doped using different acids (HCl, DBSA and p-TSA) indicates that thermoionic emission mechanism is dominanat in these devices.

The electrical transport described by thermoionic emission can be written as shown in the following equation:

$$J = J_s \exp\,(qV/nKT-1). \qquad (1)$$

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Where J_s is the saturation current density, V is the applied voltage and n is the diode ideality factor. Saturation current densities have been calculated from the intercept of Ln J versus V plots and are given by the following equation:

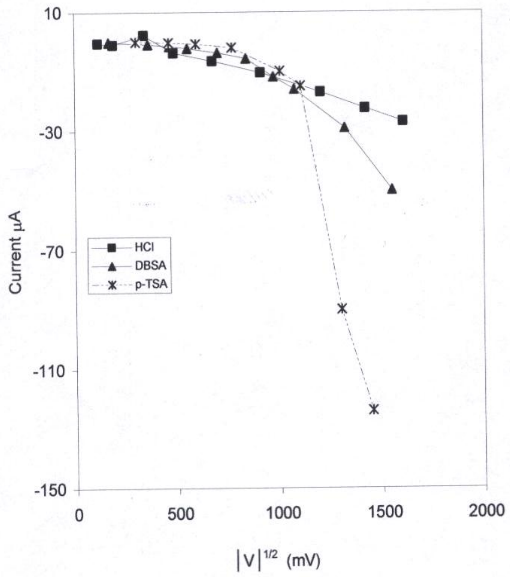

Fig. 4. The reverse current versus absolute value of $V^{1/2}$ for different Schottky devices used PANI films doped with different acids and *Zn* as rectifying.

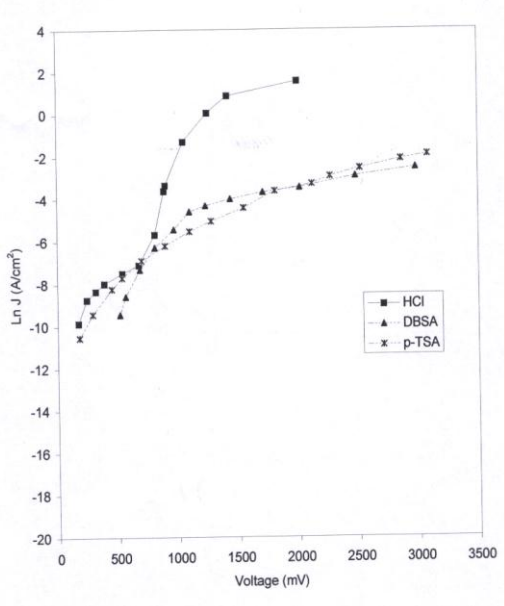

Fig. 5. Ln J vs voltage for different Schottky devices of ITO/ PANI doped with different acids/*Zn* as.

$$J_s = AT^2 \exp(-q\Phi_B / KT). \qquad (2)$$

Where A is the Richradson constant, T is the temperature and Φ_B is the barrier height of the metal semiconductor barrier. Making use of the free electron value for A as 120 A/cm^2 K^2, Φ_B for different devices can be evaluated from J_s equation [21].

Table 1 shows the junction parameters of PANI/Zn diodes for PANI doped with either HCl, DBSA, or p-TSA. It is observed that changing the dopant type significantly affects these parameters. The values of ideality factor (n) is found to be equal to 1.673 ~~1.644~~, 1.623~~3.39~~ and 1.652~~2.66~~ for devices of PANI doped with HCl, DBSA, and p-TSA, respectively. The relatively large values of n may be due to the presence of the interface state density [9].

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The rectification ratios of PANI/Zn diodes for PANI doped with DBSA or p-TSA have values of 15.2 and 50, respectively. These low values may be due to involvement of diffusion process or SCLC in the transport mechanisms. On the other hand, ~~This was explaining that the dopant is changing the effective mass of charge carrier's changes, giving different values for these electronic parameters [5]. This was also explaining that change of the dopant used was found to affect conductivity of PANI films and to induce further changes in I-V characteristics. In addition, the change of dopant will change interaction with metal electrode and form another complex with PANI films [18-19]~~Schottky diodes of PANI doped with HCl have the best rectification ratio and its value is around 257~~781~~.

The values of barrier height for PANI/Zn diodes using PANI doped with HCl, DBSA, or p-TSA are 0.782, 0.808 and 0.73 V, respectively. These values are not correlated with the values of ideality factor or concentrations of carriers. This could be due to the participation of the diffusion process or SCLC with thermoionic emission process in PANI/Zn Schottky diodes used PANI doped with, HCl, DBSA or p-TSA.

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~~**Table (1)** Junction parameters obtained by I-V and C-V characteristics of polyaniline systems with different dopants at room temperature~~

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~~Junction parameters~~ ~~Schottky systems~~	~~J_s (A/cm^2)~~	~~Φ_B (V)~~	~~Rectification ratio~~	~~n~~	~~V_{bi} (V)~~
~~ITO/PANI-HCl/ Sn~~	~~(-10) - 4.543.35x10-54~~	~~0.626~~	~~200781~~	~~1.642.464~~	~~0.4165~~
~~ITO/PANI-DBSA/ Sn~~	~~(-12) 6.18.3X10-67~~	~~0.858~~	~~1027.5~~	~~1.623.39~~	~~0.517~~
~~ITO/PANI-p-TSA/ Sn~~	~~(-14)8.34.54x10-75~~	~~0.678~~	~~13175~~	~~1.92.66~~	~~0.448~~

3.3. C-V characteristic curves

In order to gain further insight into the junction properties, the variations of capacitance as a function of the reverse bias were investigated. Fig. 6 depicts $1/C^2$ vs. reverse bias voltage for Schottky devices, ITO/PANI doped with HCl, DBSA, and p-TSA/ Zn.

The built-in potential of the barrier can be determined from the depletion layer capacitance using Mott-Schottky relationship [20, 22, and 23~~[12, 20-21~~]:

$$C^{-2} = 2\,[V_{bi} + V_R - KT/q]\ /\ q\varepsilon_o\varepsilon_s N_d\, S. \qquad (3)$$

Where C is the capacitance, V_R is the applied reverse bias, S is the active device area, N_d is the hole carrier concentrations, KT/q is the thermal voltage, V_{bi} is built-in potential, ε_o is the permittivity of vacuum and ε_s is the polymer relative dielectric constant. The carriers concentration can be determined from the slope of $1/C^2$ vs. V_R relation.~~plot by the relation:~~

~~N = (2 / q ε ε_s S) dV_R / d (1/ C^2) 15~~

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The values of V_{bi} were obtained by extrapolating 1/ C^2 to zero voltage.

Plots of 1/C^2 vs. reverse bias voltage are linear which indicates the formation of Schottky junction [16~~2~~] as shown in fig. 6. This also showed that the diode characteristics of these devices and charge density within the depletion region are uniform [184]. Calculated values of N_d and V_{bi} for different Schottky devices are given in Table (1).

From table 1, it is found that the order of charge carrier concentration of these devices is in the range of 10^{16}~~7~~ cm^{-3}. These values are comparable to the values reported earlier [15~~1~~].

Table 1
Junction parameters obtained by I-V and C-V characteristics of polyaniline systems with different dopants at room temperature

Junction parameters schottky systems	*Js* (A/cm^2)	*ΦB* (V)	Rectifying Ratio	*N*	*Vbi* (v)	*Nd* (/cm3)
ITO/PANI-CHl/Zn	8 × 10^{-7}	0.782	257	1.673	0.9	4.2 ×10^{16}
	3.1 × 10^{-7}	0.808	15.2	1.621.	0.6	4.2 ×10^{16}
	6.1 × 10^{-6}	0.73	50	65	0.9	0.4 ×10^{16}

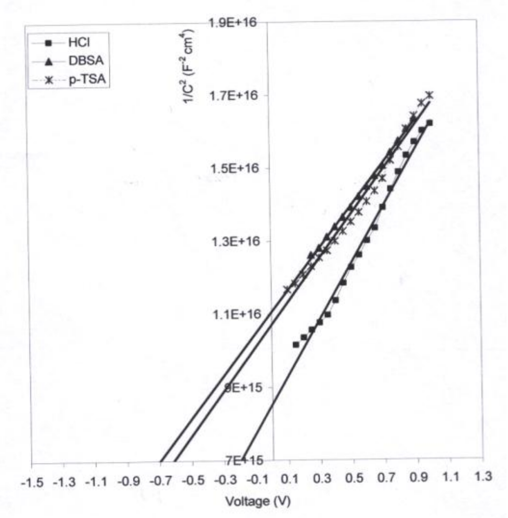

Fig. 6. The relation between the inverse square capacitance and voltage for ITO/ PANI doped with different acids/*Zn* at 60 k Hz.

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4. Conclusions

Schottky barrier diodes of polyaniline doped with different dopants (HCl, DBSA or p-TSA) have been fabricated using zinc as a new Schottky contact. The conduction mechanism in these devices depends on the type of the dopant where dopant interacts with metal electrode to form complex compound. It has been observed that the polyaniline doped with HCl and using Zn~~Sn~~ as rectifying contact has better Schottky diode quality compared to the diodes with PANI doped with DBSA or p-TSA.

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