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Opto-electronic properties of ITO/CuPc/NiPc/Al and ITO/NiPc/CuPc/Al double junction cells

By

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Abstract:

Thin organic semiconductor films of copper phthalocynanine (CuPc), Nickel Phthalocyanine (NiPc) and a metal (Al) were deposited on a conductive glass (ITO) substrate using vacuum evaporation technique to fabricate ITO/CuPc/NiPc/Al and a ITO/NiPc/CuPc/Al cells. These structures have double junction: heterojunctions (CuPc-NiPc, and NiPc-CuPc) and Schottky barrier (CuPc-Al, and NiPc-Al) junctions. This study investigates the effect of the thickness on the properties of the cells (The thicknesses of each organic semiconductor films in the three cells were equal to 25 nm, 50 nm and 75 nm.). The effective surface area of the samples was equal to 80 mm^2 (8x10 mm^2). The cells were carried through an annealing process at 150^{o}C for 2 hrs and its effect on the properties of the cells was studied. The voltage-current characteristics under dark conditions as well as under filament lamp illumination were investigated. The open-circuit voltage (V_{oc}) and short circuit current (I_{sc}) illumination relationships, the dependences of photo-capacitance (C_{ph}) on illumination and under modulated Red LED light frequency were studied. The absorbance of double CuPc/NiPc layer in the visible-IR spectrum was also examined. It was observed that the I-V characteristics rectification ratios was upto 13; the ratio of the photo- to dark-currents were in the range of 3-6; V_{oc} and I_{sc} reached 400 mV and 0.2 μA respectively and the relative value of C_{ph} increased by 50 % under illumination of 2000 lx. The frequency response of the V_{oc}, I_{sc} and C_{ph} was investigated in the range 0-100 kHz. The V_{oc} (DC) and C_{ph} showed a frequency response up to 100 kHz, whereas V_{oc}(AC)'s response was up to 100 Hz respectively. It was found that the overall performance of the ITO/NiPc/CuPc/Al cells is better than the ITO/CuPc/NiPc/Al cells. Using the experimental data, the charge transport mechanism in the double junction structures are discussed and the energy-band diagram and an equivalent circuit were developed.

Keywords:

Organic heterojunction cell, Schottky barrier, double junctions, opto-electronic properties, I-V characteristics, photo-electric cell, photocapacitance, open-circuit voltage, short-circuit current.

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1. Introduction:

During the past decade, organic semiconductor devices based on conjugated polymers, oligomers and low molecular weight materials have been envisioned as a viable alternative to traditional ones that are based on inorganic materials. Lower material and fabrication cost of organic semiconductor devices are attracting extensive interest for their potential applications in organic devices [1-4].

In order to improve devices' characteristics, heterojunction structures may be optimal for some kinds of applications. Especially, it may be beneficial for the fabrication of photoelectric cells. Phthalocyanines [5,6] and in particular copper phthalocyanine (CuPc) are one of the well-studied organic photosensitive semiconductors [7-10]. It has a high absorption coefficient in a wide spectra and high photo-electromagnetic sensitivity at low intensities of radiation. The deposition of thin CuPc films by vacuum sublimation is easy. Purification of CuPc is simple and economical as the sublimation occurs at relatively low temperatures (400 – 600 ^{o}C). Energy band gap of the CuPc is equal approximately 1.6 eV [7] to 2.0 eV [11].

In recent years nickel phthalocyanine (NiPc) has received increasing attention due to its potential applications in the area of photovoltaics and gas sensing [12-23]. One of the major advantage of NiPc over CuPc is its higher mobilty of charge carriers (0.1 cm^2/V s and 10^{-4} cm^2/ V s respectively) [23]. The energy band gap of the NiPc is equal to 2.24 eV and 3.2 eV for indirect and direct allowed transitions [15].

Many authors have investigated CuPc [9-11,24,25] and NiPc [5,6,8-10] based organic-inorganic semiconductors heterojunctions. It would be interesting to study phthalocyanine based organic-organic semiconductors heterojunctions due to their molecular and structural similarities that were observed in many cases. For example , it was found that the values of for effective density of states in CuPc and NiPc are the same ($N_V = 10^{27}$ m^{-3} , i.e. one electronic state per molecule). There molecular size and crystalline structure are also same [13]. This is important to minimize surface state density and defects concentration in the heterojunction interface. Moreover both CuPc [9, 11] and NiPc [13] form Schottky-type metal-semiconductor heterojunctions with aluminum. In the current study opto-electronic properties of ITO/NiPc/CuPc/Al and ITO/CuPc/NiPc/Al double-junction cells are presented.

2. Experimental:

The *CuPc* and *NiPc* were obtained from Sigma-Aldrich. Fig.1 shows the structure of the CuPc and NiPc molecules [14] used as *p*-type organic semiconductors. It is known that, at least, seven crystalline polymorph states of *CuPc* exist: α, β, γ, *R*, δ, ε etc. [25,26]. The form α-*CuPc* is a metastable one at T = 165 ^{o}C and can be converted thermally or with solution to the β-form. The α and the β-forms are the most frequently encountered states of *CuPc*. X-ray diffraction data in the current investigation showed that the deposited *CuPc* films were in α-form [27]. Scanning electron micrographs showed no particular crystalline orientations in the CuPc films [27]. Obtained by vacuum deposition NiPc films were in short-whisker like α-phase. As the annealing temperature increases to 200 ^{o}C whiskers grow longer. Annealing at 300 ^{o}C converted α-phase into β-phase [16]. Thus we assume that NiPc films in our case were in α-phase.

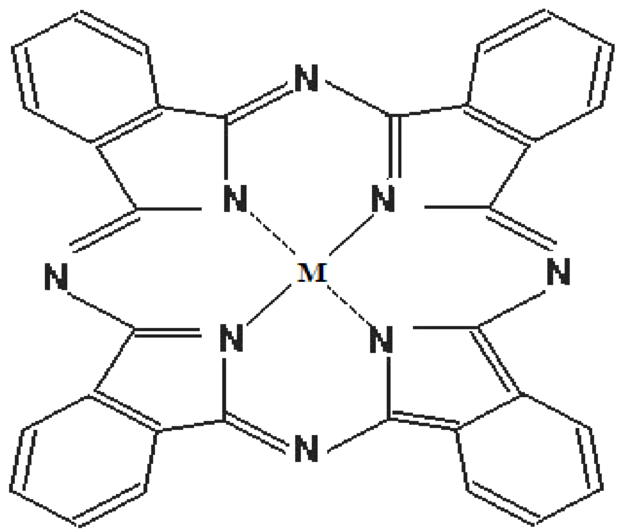

Figure (1): *Molecular structure of CuPc and NiPc : M is Cu or Ni.*

Thin films of CuPc were thermally sublimed onto a conductive glass substrate (ITO) at 400 - 450 ^{o}C and at ~10^{-4} Pa in an Edwards AUTO 306 vacuum coater having a diffusion pumping system. The deposition rate was 2 nm/min. The substrate's temperature in this process was held at ~40 ^{o}C. The CuPc or NiPc films were deposited on the conductive glass (ITO) substrates under the same conditions. A Perkin Elmer Lambda 19 UV/VIS/NIR spectrometer was used for measurements of absorption spectra. Actually the spectrum covers the NIR- UV, i.e., wavelengths between 200-1000 nm [6, 10]. Fig.2 shows absorption spectra of the deposited CuPc and NiPc films in NIR-visible spectra. It shows that maximum of the spectrum corresponds to energy of 1.5 eV.

On CuPc and NiPc films an Al film was deposited. Thickness of the CuPc and NiPc was obtained using an Edwards FTM5 film thickness monitor. The investigations [6] showed that usually pthalocyanines form ohmic contacts with Ag and Schottky-type junction with Al [9,11,22].

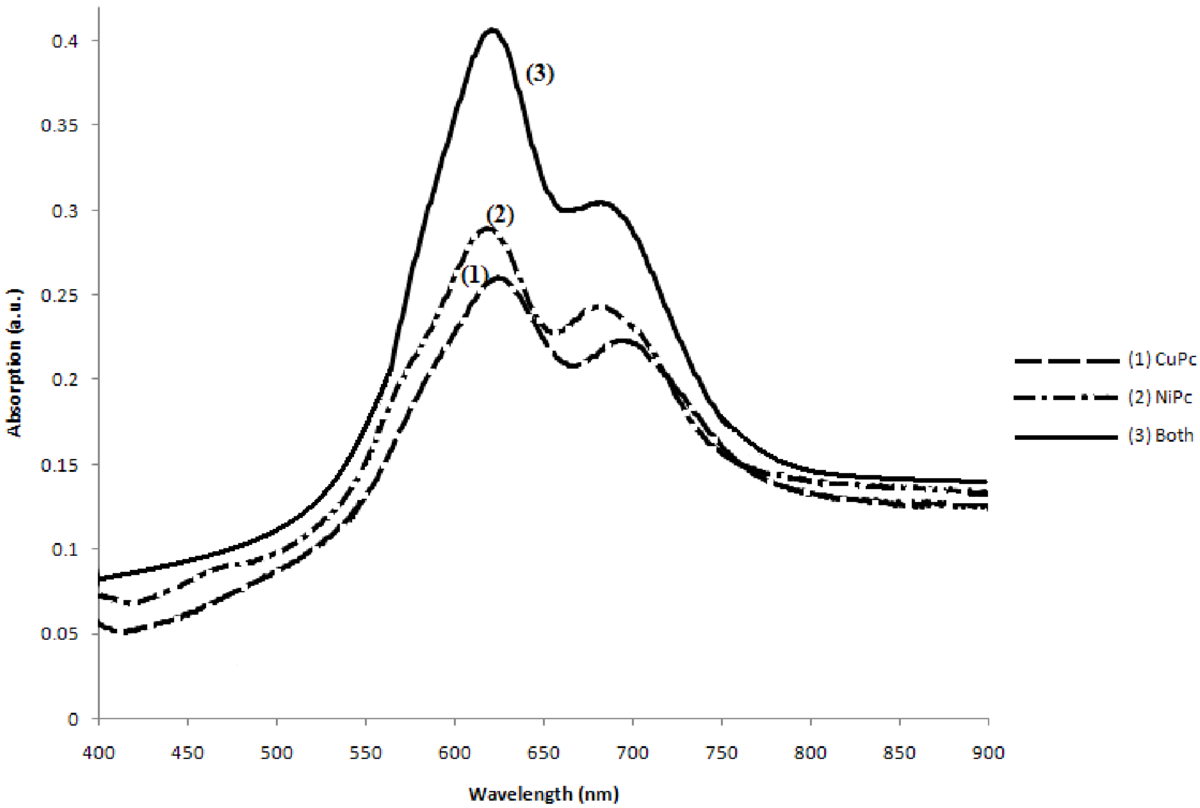

***Figure (2)**: Absorption spectra of the CuPc / NiPc double layer film deposited on conductive glass (ITO) substrate by vacuum evaporation.*

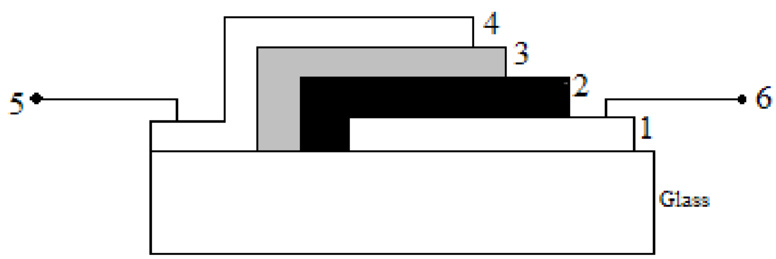

***Figure (3):** Cross sectional view of ITO/NiPc/CuPc/Al and ITO/CuPc/NiPc/Al double junction cells: 1-ITO, 2-CuPc or NiPc, 3-NiPc or CuPc, 4 – Al, 5 and 6 - terminals*

Fig. 3 shows a cross-sectional view of the fabricated ITO/CuPc/NiPc/Al and ITO/NiPc/CuPc/Al cells. The thicknesses of each organic semiconductor films were equal to 25 nm, 50 nm and 75 nm in the three different cells. Effective surface area of the samples was equal to 80 mm^2 (8x10 mm^2). The cells were carried through an annealing process at 150°C for 2 hrs and the effect on the properties of the cells was studied. A filament lamp and Red LED were used as the source of light. The measurements of voltage and current were conducted at room temperature using digital meters.

3. Results and Discussion:

Fig. 4 shows dark and photo *I-V* curves of the ITO/CuPc/NiPc/Al cell at room temperature under illumination (340 lx) by filament lamp. The thickness of CuPc and NiPc layers was equal to 150 nm (2x75 nm). It is seen that reverse saturation current of the cell increases under illumination approximately 5 times, as ratio of photo and dark currents (I_{Ph}/I_D) showing good response in the photo-diode mode of operation. Rectification ratio was around of 2 at an applied voltage of 5 V. The I-V characteristics of the ITO/NiPc/CuPc/Al cells were in principle similar, but the cells showed better photoresponse (I_{Ph}/I_D was upto 5) and higher RR (up to 4). The forward bias I-V characteristics under dark and under illumination were similar for all cells.

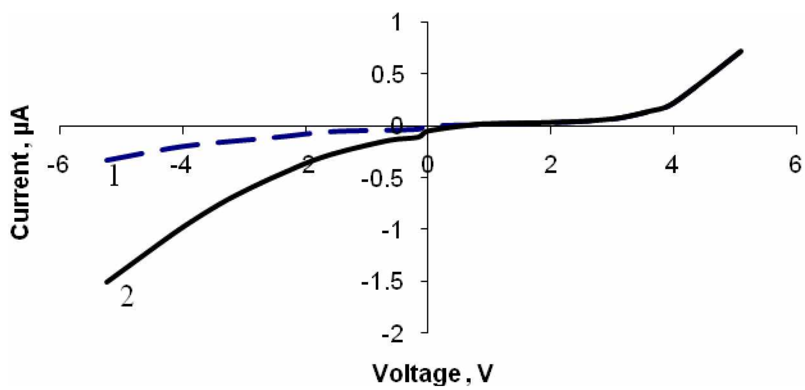

Figure (4): *Dark (1) and photo (2) I-V curve of the ITO/CuPc/NiPc/Al. cell at room temperature.*

For analysis of the dark *I-V* curves in the narrow potential range (0 - 0.5V) the modified Schockley equation may be used [25,28] :

$$I = I_o \left[Exp\, \frac{q(V - I R_s)}{nkT} - 1 \right] + \frac{(V - I R_s)}{R_{sh}} \qquad (1)$$

where R_s and R_{sh} are the device series and shunt resistances, and *I* and *V* are the terminal current and voltage, *n* is the diode ideality factor, *k* is Boltzmann constant, *T* is absolute temperature, and I_o is the reverse saturation current given in ref.[29]:

$$I_o = A^* T^2 Exp\left(-\frac{q\Phi}{kT}\right) \qquad (2)$$

Where A^* is the Richardson constant and $q\Phi$ is the height of contact barrier (Schottky barrier [29]).

Examination of *I-V* characteristics in Fig. 4 shows that the diode junction resistance, $R_j = \frac{\delta V}{\delta I}$, can easily be determined from the given plot. The R_{sh} was obtained from maximum junction resistance (9.1 MΩ), whereas R_s is equal to the minimum junction resistance at forward bias (2.2 MΩ).

If at some approximation the forward bias voltage-current characteristics of the sensor may be described by Eq.1 and processed in a similar manner to that of a heterojunction barrier diode [28,29], i.e. if the current is plotted on a log scale, the value of I_o may be determined and the A^* of Eq.2 may be calculated. Using this method we found the reverse saturation current I_o = 0.007 μA by taking into account $q\Phi$ = 0.96 eV for Au/NiPc/Al cell [22]. Finally from Eq.2 it was found that $A^* = 1.28\ 10^{3}$ $A/K^2\ cm^2$.

Fig. 5 shows an open-circuit voltage – illumination relationships for the ITO/NiPc/CuPc/Al cells at different thickness of CuPc and NiPc films.These characteristics are nonlinear as observed in phthalocyanine cells [28]. It is seen that with increase in thickness V_{OC} decreases that may be assosiated with absorption of light on the way to CuPc-Al Schottky junction.

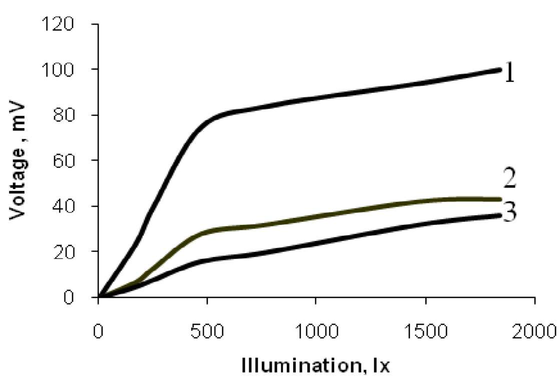

Figure (5):*An open-circuit voltage – illumination relationships for the ITO/NiPc/CuPc/Al cells respectively at different thickness of CuPc and NiPc films : 1-50 nm (2x25 nm), 2- 100 nm (2x50 nm) and 3 – 150 nm(2 x 75 nm).*

The samples were illuminated with a filament lamp. The value of V_{OC} on average for ITO/NiPc/CuPc/Al cells is higher by 20 % in comparison to V_{OC} of ITO/CuPc/NiPc/Al cells. The polarity of the photo-induced voltage was negative on Al and positive on ITO for both ITO/NiPc/CuPc/Al and ITO/CuPc/NiPc/Al cells showing that Schottky barrier metal-semiconductor CuPc/Al or NiPc/Al junction plays a dominant role with respect to organic-organic semiconductor CuPc/NiPc (or NiPc/CuPc) junction . This means a buit-in potential barrier (equal to differences of work functions [29]), of Schottky junction in this case is higher than the barrier of organic-organic semiconductor junction. Therefore we can assume that thermoionic emission theory may be used (Eq. (2)) [29] to describe current mechanism of majority charge carriers over the Schottky barrier. Short-circuit currents – illumination relationships of the same samples shows quasi-linear behavior .Under illumination the current increased up to 0.03 μA and a higher current was observed through the thin films cells (50 nm).

Fig.6. shows dependences of ITO/NiPc/CuPc/Al cells' DC open-circuit voltage (V_{oc} (DC)) on Red LED modulated beam's frequency. It is seen that in the first approximation the DC open-circuit voltage is independent of the frequency of the modulation, whereas V_{oc} (AC)'s response was up to 100 Hz respectively. The short-circuit currents responses were as V_{oc} (DC) and V_{oc} (AC) accordingly. The illumination of the Red LED was equal to 470 lx. The thickness dependence of the voltage is the same as was observed in Fig.5.

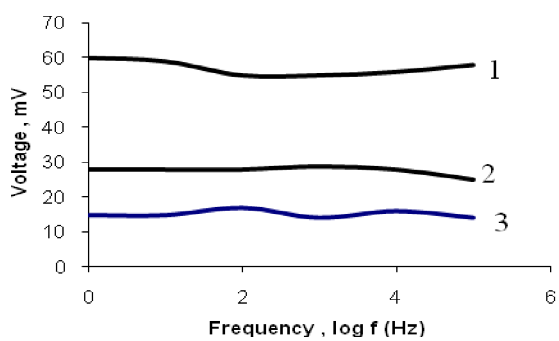

Figure (6):*Dependences of ITO/NiPc/CuPc/Al cells' DC open-circuit voltage on Red LED modulated beam's frequency at different thickness of CuPc and NiPc films : 1-50 nm (2x25 nm), 2- 100 nm (2x50 nm) and 3 – 150 nm (2 x 75 nm).*

Fig.7 shows capacitance-illumination relationship for the ITO/NiPc/CuPc/Al cells having different thickness of the NiPc and CuPc films. It is seen that the relative value of the capacitance C_{ph} increases by 50 % under illumination of 2000 lx, unlike the ITO/CuPc/NiPc/Al cells where the increase was 30 % only. It is seen (Fig.7) that a higher photo-response is shown by thicker film (150 nm) and a lower by the thinner one (50 nm). It may be due to more absorption of light in the former case. In [30-32] the polarizability due to the transfer (α_{td}) of charge carriers as electrons and holes that are present at normal, including dark conditions were investigated. In this case we assume that photo-induced charge carriers are repsponsible for observed photo-capacitive effect in the ITO/NiPc/CuPc/Al and ITO/CuPc/NiPc/Al cells. Detail analysis of this effect based on Clausius-Mosotti retaion [33] with respect to poly-N-epoxypropylcarbazole was done earlier [34].

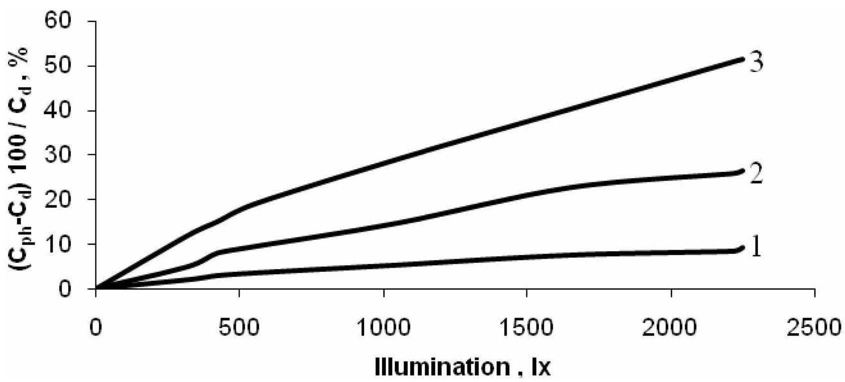

Figure (7):*Capacitance-illumination relationship for the ITO/NiPc/CuPc/Al cells having different thickness of the NiPc and CuPc: 1-50 nm (2x25 nm), 2- 100 nm (2x50 nm) and 3 – 150 nm (2 x 75 nm).*

It was found that annealing increased RR up to 13 ,and V_{OC} upto 400 mV, that may be due to decrease of total concentartion of traps as was observed in Au/NiPc/Al cell [13]. At the same time a visible effect of the annealing to I_{SC} was not observed, that may be caused due to oxidation of Al film in the annealing process and a decrease in its condcuctivity.

Taking into account the data presented above and the concept related to heterojunction properties [29], an energy-band diagram of the ITO/NiPc/CuPc/Al heterojunctions was developed (Fig.8). In this diagram ΔE_c and ΔE_v are the differences between the two conduction bands energies and valence bands energies respectively and show discontinuities in the conduction and valence bands. The eV_{bi1} and eV_{bi2} are built-in potential barriers seen by holes in CuPc/Al and NiPc/CuPc heterojunctions interfaces respectively. In Fig.8 it is shown that $eV_{bi1} > eV_{bi2}$. On the CuPc and NiPc side of the junctions, unlike the delocalized states in Al or ITO sides, there are localized states because in these organic semiconductors the mobility of charge carriers is much less than 1 cm^2 / V s [7,8].

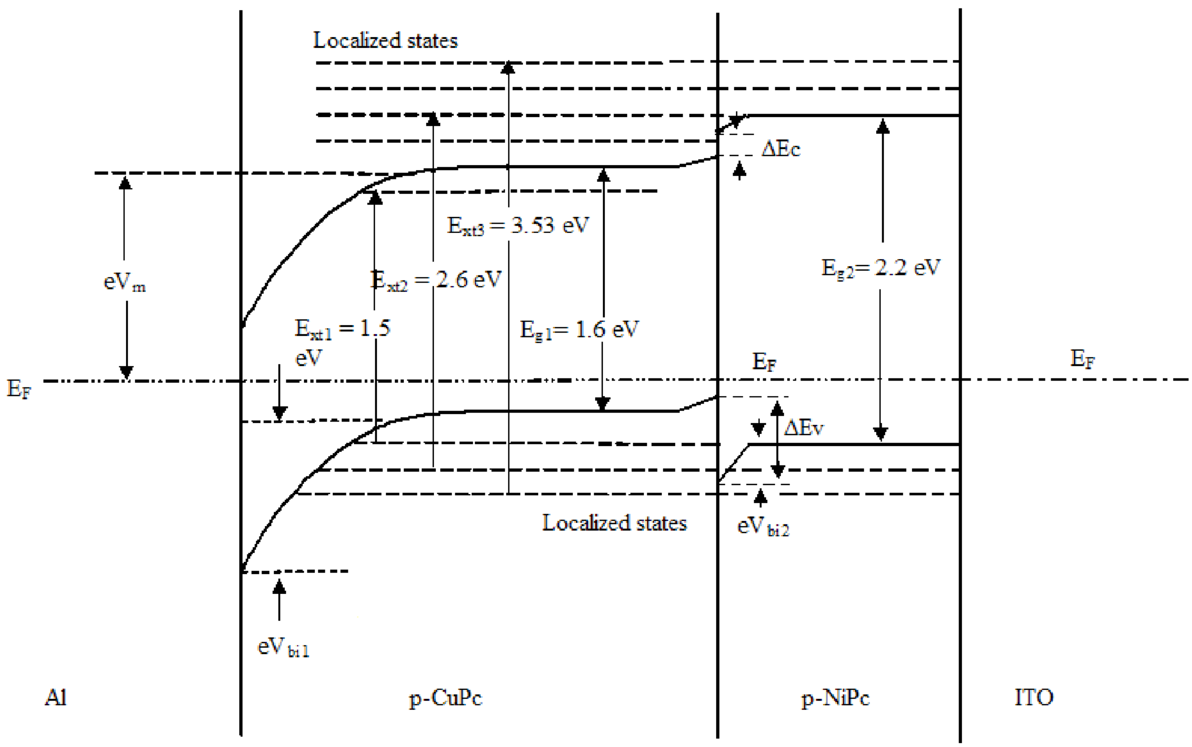

Figure (8): *Energy-band diagram of the ITO/NiPc/CuPc/Al heterojunctions.*

The equivalent circuit of the organic photo-electric cell [25] developed for the ITO/NiPc/CuPc/Al double junction cell is shown in Fig. 9. Current sources I_1 and I_2 represent CuPc/Al and NiPc/CuPc photo-electric cells. For the ITO/CuPc/NiPc/Al cell the equivalent circuit is considered as the same , only direction of I_2 will be opposite The R_{sh} and R_s are the cell's shunt and series resistances determined from the dark *I-V* curves, C_1 is the sum of the transition and diffusion capacitances of the heterojunction, C_2 is inter-terminal capacitance of the cell . It may be assumed as a first approximation that $C_1 = C_2$.

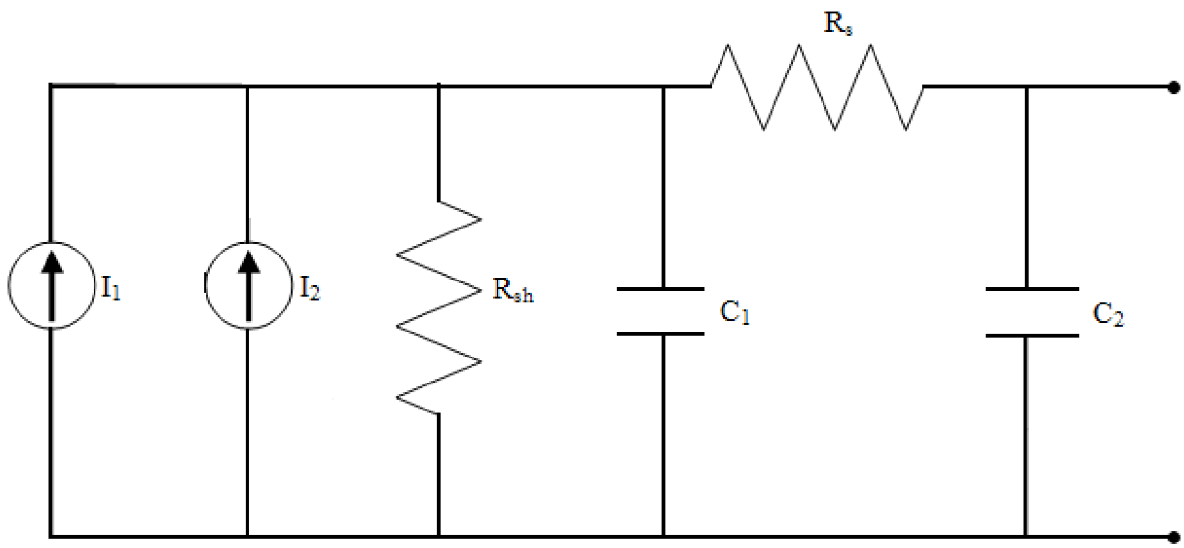

Figure (9): *Equivalent circuit of the ITO/NiPc/CuPc/Al cell.*

4. Conclusions:

ITO/CuPc/NiPc/Al and ITO/NiPc/CuPc/Al double junctin cells were fabricated and investigated. It was found that I-V characteristics show rectification behavior and exhibit an increase in current under illumination under reverse bias. The cells show photo –electric response in photo-voltaic mode of operation: on average the response of ITO/NiPc/CuPc/Al cells is 20 % higher than the ITO/CuPc/NiPc/Al cell, revealing that Schottky barrier metal-semiconductor CuPc/Al or NiPc/Al junction plays a dominant role in comparison to organic-organic semiconductor CuPc/NiPc (or NiPc/CuPc) junctions. The cells show high photocapacitive response: in ITO/NiPc/CuPc/Al cells the capacitance increase under illumination by 50 % under illumination of 2000 lx. The energy-band diagram and an equivalent circuit of the double junction cells were developed that may be used for simulation of their opto-electronic properties.

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