



Investigation of structural, optical and electrical properties of a new cobalt phthalocyanine thin films with potential applications in perchlorate sensor



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ARTICLE INFO

Article history:

Received 17 January 2015

Received in revised form 27 June 2015

Accepted 7 July 2015

Available online 29 July 2015

Keywords:

I–V characteristics

Cobalt(II) phthalocyanine derivative

Raman spectroscopy

Conductance

Impedance spectroscopy

ABSTRACT

Optical, structural and electrical properties of new cobalt phthalocyanine derivatives (Co(II)Pc-AP) thin films have been investigated. The Raman spectroscopy study shows the different vibrations bands corresponding to the metallophthalocyanine. The cobalt-phthalocyanine derivative films exhibit absorption spectra with a resolved electronic structure in the UV–vis range and the energy gap was determined by the Tauc method. Electrical properties of the ITO/Co(II)Pc-AP/Al structure have been investigated by I–V characteristics and impedance spectroscopy measurements. The conduction is governed by space-charge-limited current (SCLC) mechanism. The impedance spectroscopy study showed a hopping transport process, which is a typical behavior of disordered materials. The electrical characteristics of the devices were modeled by a single parallel resistor and capacitor network in series with a resistance.

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1. Introduction

Organic materials have recently attracted a great interest as semi conducting materials [1,2] and have been successfully incorporated into solar cells [3] and chemical sensors [4]. Metallo-phthalocyanines and their polymers gained importance because of their unique photoconducting and semiconducting properties [5]. Metallo-phthalocyanine attracted interest because of their interesting properties like excellent thermal stability, chemical inertness, photoconductivity and semiconductivity. Their useful properties are attributed to their efficient electron transfer ability. Because of these importance properties the metallo-phthalocyanine was used in many fields including solar energy conversion [6,7], chemical sensors [8], photosensitizers [9], gas sensors [10], liquid crystals [11], and nonlinear optics [12].

In other side, Metal-phthalocyanine has already found applications in the design of chemical sensors, in particularity to detect

toxic anions because of the importance of the metal center. El-Nemma et al. [8] have used cobalt-phthalocyanine derivative as a sensing membrane to detect anionic surfactants and have shown that the MPC can be used as an important anions sensing molecule. The anions coordinate as an axial ligand, to the metal center of the carrier molecule. The electrical and optical properties of the molecule have an important effect on their sensing properties. Then it is necessary to investigate these properties to facility the development of a chemical sensor using this molecule.

In this work, we have studied the structural, optical and electrical properties of new cobalt phthalocyanine derivatives, Cobalt phthalocyanine-C-mono amido-butyl acrylate carboxyl acid (Co(II)Pc-AP). Using Raman spectroscopy technique, we have observed the different bands characteristics of metallo-phthalocyanine. The optical properties were studied and we have observed a high absorptivity in the UV–vis region and we have determined the band gap energy of our molecule. The ITO/Co(II)Pc-AP/Al structure was characterized using current–voltage (I(V)) measurements. The transport mechanism was discussed in the different regimes of electronic conduction and the effective mobility in the material was determined. Impedance spectroscopy measurement was determined to investigate the ac behavior and then we have

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modeled the impedance characteristics by an idealized electrical circuit.

2. Materials and methods

2.1. Materials

All the chemicals used were of analytical reagent grade. Deionized (DI) water was used throughout, Tetrahydrofuran (THF) is purchased from Aldrich. The phthalocyanine molecule (Fig. 1) used in this work was synthesized and purified according to the synthesis route reported earlier in our previous works [13].

2.2. Substrate preparation

ITO/Co(II)Pc-AP/Al device structures were fabricated according to the following route: ITO-coated glass substrates (ITO-thickness 100 nm, sheet resistance 20 Ω /square) were used as anodes, for electrical measurements, and were beforehand cleaned in an acetone (CHROMASOLV[®] Plus, for HPLC, \geq 99.9%, Aldrich) and ethanol (ACS reagent, \geq 99.5%, Sigma–Aldrich) bath at 100 °C, rinsed with isopropyl alcohol (CHROMASOLV[®] Plus, for HPLC, 99.9%, Aldrich), and dried under nitrogen flow.

2.3. Device fabrication

Co(II)Pc-AP was dissolved in THF solvent, with a concentration of 4 mg/mL and spin-coated at 2000 rpm for 60 s on the ITO electrode. An aluminum top electrode was evaporated at pressure below 10^{-6} Torr for the study of the transport properties. The active area of the diodes is limited by the overlapping section of the electrodes, which is approximately 7 mm². The developed structure is shown in Fig. 2.

2.4. Instrumentation

- Raman spectroscopy measurements have been realized by a triple spectrometer Raman system T64000 HORIBA, using krypton/argon laser with 632 nm excitation wavelength.
- UV–vis absorption spectra were recorded with a DR 500 HACH LANGE UV–vis spectrophotometer.
- The DC measurements (I – V characteristics) were made with a Keithley 236 source measure unit.
- The AC measurements were performed with a HP 4192A LF impedance analyser. In general, for the $G(\omega)$ characteristics, the excitation potential for AC measurements is given by:

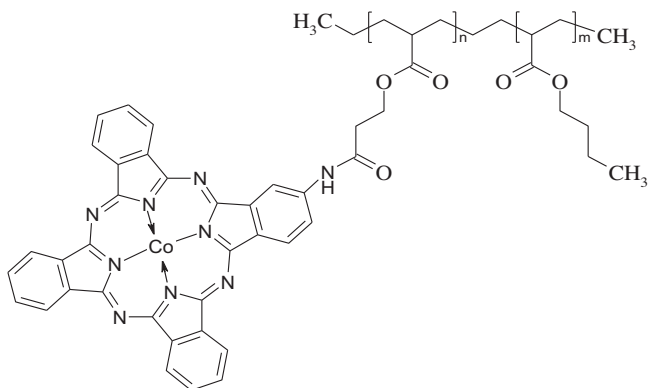


Fig. 1. Cobalt phthalocyanine-C-mono amido-butyl acrylate carboxyl acid (Co(II)Pc-AP).

$$V = V_0 + V_{mod} \cos(\omega t)$$

with V_0 is the DC bias and V_{mod} is the oscillation level and $\omega/2\pi$ is the frequency. In our case, the measurements were performed in the following conditions: $V_0=0V$ and V_{mod} of 20 mV over a frequency range of 100Hz–10 MHz. All electrical measurements were performed in dark and at room temperature.

3. Results and discussion

3.1. Raman spectroscopy study

Fig. 3 shows a very rich Raman spectrum of the cobalt phthalocyanine derivatives (Co(II)Pc-AP) thin layer. As it can be seen from this figure, we observe main bands corresponding to the vibrations of macrocycle deformation C–N (1542.57 cm^{-1} – B_{1g} mode) and CC (686 cm^{-1} – A_{1g} and 751.7 cm^{-1} – B_{1g} mode) [14–16].

The bands at 1542.57 cm^{-1} corresponding to heavy atom–nitrogen (M–nitrogen) in-plane stretching and bending vibrations as well as displacement on the C–N–C bridge bond, which is characteristic for metallophthalocyanine structures [16,17]. Moreover, we recorded a band of A_{1g} symmetry at 592.50 cm^{-1} corresponding to benzene ring deformation, the position of the B_{1g} band at 686 cm^{-1} connected with pyrrole stretch is closely linked to metal ion adapted to phthalocyanine molecule.

The 1542.57 cm^{-1} , 1342.97 cm^{-1} and 686 cm^{-1} bands involve N_α motions and the 751.70 cm^{-1} mode involves both N_α and N_β vibrations. In Table 1 we summarized the different observed bands for the Co(II)Pc-AP derivatives with these assignments.

3.2. Optical study

Fig. 4 shows the UV–vis spectrum of Co(II)Pc-AP thin film. The absorption spectrum shows two absorption bands noted B and Q the first one located in the UV region at about 270 nm and the second one in the visible region at about 657 nm. The Q band is corresponding to the π – π^* electronic transition and the B band corresponding to the deeper π – π^* transition, but contains a small contribution from n to π^* transition [18]. In the Q band, we notice the presence of a vibronic peak separated by about 66 nm to the principal peak for the two molecules which is cloud attributed to aggregated species [18,19], as shown in the inset of Fig. 4. The intensity of the higher energy peak is larger than that of the second one. This behavior is typical of the α -phase of CoPc [19].

The optical band gap energy can be determined by the Tauc method [20]. The absorption coefficient α is related to the photon energy $h\nu$ and gap energy E_g by the following equation:

$$(\alpha h\nu)^2 \propto (E - E_g) \quad (1)$$

the extrapolation of the onset of the curve $(\alpha E)^2$ versus $(E - E_g)$, to energy axis provides the energy gap E_g , Fig. 4. The optical band gap calculated is about 3.38 eV, which is confirmed in the literature by Seoudi et al. [18], Joseph et al. [19] and Wahab et al. [21] for cobalt phthalocyanine derivatives as indicated in Table 2. Also, we notice the presence of trapping levels at 1.36 eV and 1.95 eV.

4. Static and dynamic study of ITO/Co(II)Pc-AP/Al diodes

4.1. dc study: transport mechanism

The I – V characteristics of the ITO/Co(II)Pc-AP/Al multilayer devices shown in the Fig. 5 exhibit typical diode behavior under both forward and reverse biases. The electrical characteristics (I – V) indicate a symmetric behavior for low-voltages. This can be explained by the localized state theory with defects providing the

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