

Transport mechanism and trap distribution in ITO/azo-calix[4]arene derivative/Al diode structure

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Abstract

Electrical measurements have been performed on spin-coated azo-calix[4]arene derivative on pre-cleaned indium thin oxide (ITO) substrates using current versus voltage, capacitance versus voltage and impedance spectroscopy measurements. The nature of trap states in single layer ITO/azo-calix[4]arene derivative/Al organic diodes has also been investigated. The energy band gap of the thin film containing calixarene derivative has been measured by UV–Vis absorption spectroscopy and is about 2.77 eV. The current–voltage characteristics have shown ohmic behavior at low voltages. At high applied bias the I – V characteristics can be successfully modeled by space-charge limited current (SCLC) theory. The impedance dependence of bias and frequency is discussed in terms of the presence of a depletion layer in the bulk. The device is accurately modeled, in a range of frequency between 100 Hz and 1 MHz, as a single parallel resistor and capacitor network placed in series with a resistance. Their values deduced from fitting experimental data to the model have given a dielectric relaxation time in the ms range and an exponential trap distribution.

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1. Introduction

In the last decade organic materials have been of great interest for their low-cost manufacturing, easy processing and interesting sensing properties or high luminescence efficiencies, which make them promising for many electronic and optoelectronic applications, such as chemical sensors [1,2], light emitting diodes [3–5], thin film transistors [6,7] and solar cells [8]. For these purposes, three important families of organic materials were used, namely, small molecules (Alq₃, phthalocyanines) [9], conjugated polymers [3,10] and macromolecules like ionophores such as cyclodextrin [11,12] and calix[n]arene derivatives [2,13–15]. The latter attracted more attention in recent years due to its better preparation and purification

as well as their deposition process in thin film form, by thermal evaporation [14] or spin-coating [15,16]. Moreover, for light emitting diodes or chemical sensors applications, good charge carrier transport and tunable electronic properties are crucial points.

Knowing that Al is a stable cathode commonly used for organic electronic applications and spin-coating is a suitable technique for industrial processes, we have to focus on the active material. Thus, recently we have adopted several approaches to enhance the above-mentioned criterions for soluble organic materials. The hybrid nanocomposites [17], MEH-PPV/calix[4]arene composites [18] and chemically modified calix[4]arene structures [2,16] have the interesting properties. Indeed, in previous works [16,19,20], we have presented experimental details of synthesis, optical and electrical results on several azo-calix[4]arene derivatives-based diodes with different chromophores on the upper rim and substituents at the lower one. These materials presented energy band gaps in the range of 1.7–3 eV characteristic of semiconductors [16].

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In addition, they showed good stability under ambient conditions. These proprieties make the azo-calix[4]arene family a good candidate for physical investigations with potential application opportunities. This gives a great stimulus to the deeper study of their electrical and dielectric properties. Impedance spectroscopy (IS) has proved to be a powerful tool for studying the transport mechanisms and relaxation processes in organic devices [21,22].

In this study, we report the results of electrical measurements performed on thin films of new chromogenic calix[4]arene derivative sandwiched between anodic (indium thin oxide:ITO) and cathodic (aluminum:Al) electrodes. Current–voltage characteristics were studied. Moreover, a detailed study of these organic diodes by IS in the frequency range from 100 Hz to 1 MHz varying bias voltage from 0 to 10 V is presented. After the determination of the diagram energy of our diodes in Section 3.1, we have concentrated on transport mechanism in Section 3.2, discussing different regimes of electronic conduction and the description of traps present in the diode structure. In Section 3.3, we will determine the nature of traps distribution from fitting the IS results, for different biases, with an equivalent electrical circuit.

2. Experiment details

2.1. Materials

This work is based on new chromogenic material, azo-calix[4]arene derivative, whose synthesis procedure was reported elsewhere [20,23]. It is characterized by the presence of chromophores at the upper rim and an ester group at the lower rim. The studied material is represented in Fig. 1.

2.2. Device fabrication and experimental techniques

The devices in our study consist of a single organic layer sandwiched between two electrodes on top of a glass

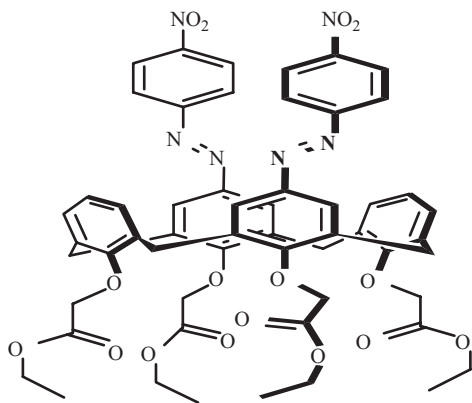


Fig. 1. Chemical structure of azo-calix[4]arene derivative used in this work (5,17-bis(4-nitrophenylazo)-25,26,27,28 tetra(ethoxycarbonyl methoxy)-calix[4]arene).

substrate. They were fabricated on ITO-coated glass substrates (ITO thickness 100 nm, sheet resistance $20 \Omega/\square$) used as anodes. The ITO substrates were supplied by Merck Display Technologies (MDT). Before the etching process, the ITO substrates were successively pre-cleaned for 20 min in acetone and isopropyl alcohol in an ultrasonic bath and finally dried by a nitrogen gas flow. Then azo-calix[4]arene derivative was spin-coated from CHCl_3 solution onto ITO electrode to obtain 200 nm thickness. Organic thin films were dried for 30–60 min at 80°C to evaporate organic solvents. In order to study the transport properties an aluminum contact was evaporated as a top electrode at pressure below 10^{-6} Torr. The active areas of the diodes were confined within the overlap of the electrodes which is approximately 5 mm^2 . Gold wires were glued with silver paint to the substrate and the top electrode of the device.

The UV–Vis absorption spectra were recorded with a Perkin-Elmer UV–Vis spectrophotometer (Lambda 35). For UV–Vis study, azo-calixarenes thin films were spin-coated on spectroil glass substrates to avoid strong absorption of ITO in the UV region.

The DC measurements were performed with a Keithley 236 source measure unit and the AC measurements were performed with an HP 4192A LF impedance analyzer. All these measurements were performed in dark and at room temperature.

3. Results and discussion

3.1. Electronic properties of ITO/azo-calix[4]arene/Al diode

UV–Vis spectra of new azo-calix[4]arene derivative performed in solution and on thin film samples exhibit one significant absorption band as shown in Fig. 2. Indeed, we observed a large band, attributed to the $\pi-\pi^*$ transition of azo-calix[4]arene and a tender shoulder about 450 nm assigned to the electronic transition related to the $\text{N}=\text{N}$ bond of the azo-group. The absorption maxima and the estimated optical band gap were collected in Table 1, from which we note optical band gaps in the range of semiconducting materials.

3.2. Transport mechanism

In Fig. 3(a) the $I-V$ characteristic of ITO/azo-calix[4]arene/Al device is presented showing typical diode behavior with a threshold bias voltage of 4.8 V in dark. Nevertheless, this curve looks rather symmetric for a device that is supposed to behave as a diode. This is explained by the formation of a dipolar layer at the interfaces. Kushmerick [24] has observed this kind of characteristics, for either metal/molecule/metal or metal 1/molecule/ metal 2 structures. Indeed, the chemical effect (interface polarization due to polar molecules) has got the upper hand over the

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