

Organic light-emitting diodes based on PVK and Zn(II) salicylidene composites



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ARTICLE INFO

Keywords:

Luminescent Zn(II) coordination compounds

OLEDs

Polymeric composites

Solution-processed diodes

Charge mobility

ABSTRACT

The photo- and electroluminescence properties of solution-processed organic light-emitting diodes (OLEDs) consisting of composites of PVK as a host with [Zn(salophen)] or [Zn(sal-3,4-ben)] as guests were studied. Devices consisted of a single spin-coated active layer. The electro-optical properties (voltage turn-on (V_{on}), brightness (L_{max}) and current efficiency (η_{curr}) of the coordination compounds were better than those of the ligands. This improvement can be explained by the increase of the hole mobility (μ_p) compared to the PVK and PVK:salicylidene composites. The L_{max} and η_{curr} were three times higher for the PVK:[Zn(sal-3,4-ben)] ($V_{on} = 6.5$ V; $L_{max} = 131$ cd m⁻²; $\eta_{curr} = 36$ mcd A⁻¹) diode than the PVK:[Zn(salophen)] ($V_{on} = 9.5$ V; $L_{max} = 52$ cd m⁻²; $\eta_{curr} = 11$ mcd A⁻¹) due to the increase of the hole mobilities of μ_p ([Zn(sal-3,4-ben)]) $\sim 10^2 \times \mu_p$ ([Zn(salophen)]). Moreover, the PVK:[Zn(sal-3,4-ben)] diode presented L_{max} and η_{curr} comparable to the PVK:8-hydroxyquinoline compounds (Alq₃ and Znq₂) prepared using the same procedure which can be explained by the hole mobilities of these composites and photoluminescence quantum yields (PLQY) of the coordination compounds.

1. Introduction

There is global interest in the development of environmentally friendly materials for applications in opto-electronic devices, including light-emitting diodes, operating with high efficiency, easy processability and low environmental impact [1–3]. Light-emitting diodes are one particular type of potential device, in particular those fabricated using organic materials, the organic light-emitting diodes (OLEDs), which can be more easily processed and recycled than the inorganic-based conventional systems [1,4,5].

Among the materials employed in OLEDs, conjugated polymers are materials whose performance should be improved via their formation into composites with coordination compounds [6–12]. One example of a coordination compound successfully used in OLEDs is tris(8-hydroxyquinolate)aluminum(III) (Alq₃) evaporated on an ITO substrate (anode) with Al as the cathode [13]. Several other examples have been reported, such as Ir(III) [14], Pt(II) [15], Os(II) [16], Ru(II) [17], Eu(III) [18], Cu(I) [19], Zn(II) [20–22], Be(II) [23], and Li(I) [24] compounds. However, some of these ions are very expensive, with low natural

abundance [25–28]. Therefore, attempts to synthesize new coordination compounds with high electroluminescence (EL) efficiency and lower costs using materials of greater abundance is an important task.

In several coordination compounds, the ligand coordinated to the central ion has been 8-hydroxyquinoline [23,24,29]. Nevertheless, other ligands have been used, such as salicylidene and N,O-electron-donor molecules, which are interesting due to the presence of a -N-M-O-chelating cycle. These groups enable a charge-transfer bridge between the ligands and the metal [30], which may increase the ligand planarity and π -charge conjugation, decreasing the band gap (E_{gap}) [31,32]. In addition, salicylidene are interesting molecules that undergo excited-state proton transfer reactions (ESIPT) and exhibit more than one emission band [33,34]. The presence of these two emissions might be useful for applications as white emission materials [35].

There are several applications of light emitting diodes where the emissive layers are formed by coordination compounds using the vapor deposition process. In such diodes, the active layer is formed by solid state molecules. On the other hand, OLEDs can be produced by spin-coating technique using solutions of the coordination compounds and a

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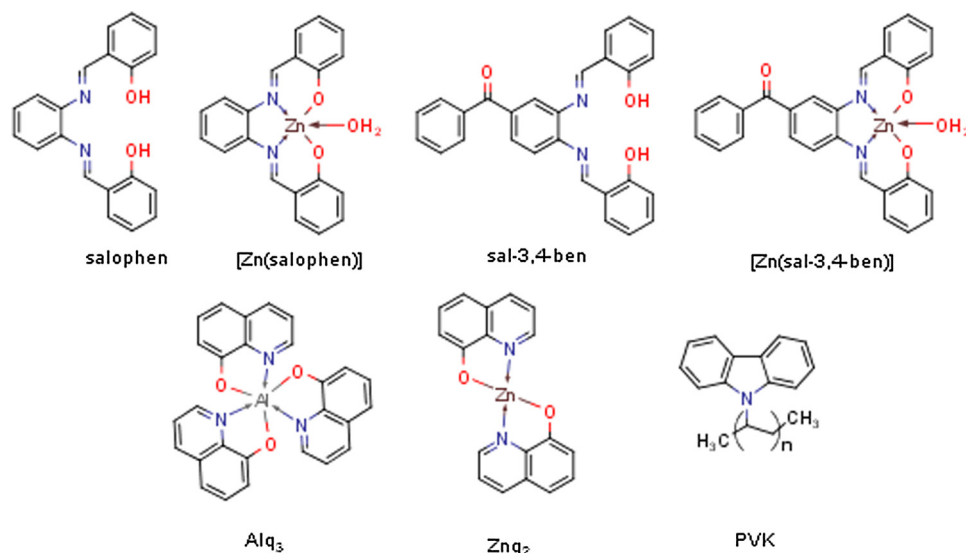


Fig. 1. Chemical structures of Alq₃, Znq₂, PVK, salophen, [Zn(salophen)], sal-3,4-ben and [Zn(sal-3,4-ben)].

host polymer forming a host/guest composite [18,21,22,36,37,38]. These are named solution-processed diodes. In addition, composites of electroluminescent polymers with coordination compounds might be an interesting strategy to improve diode performances because they can combine the good properties of both materials. In the diodes with composites there is a bulk heterojunction and both the charge transport and injection process might improve optical-electronic properties of the OLEDs [39–41]. In general, Zn(II) coordination compounds are poorly electroluminescent (EL) in the solid state and consequently the vapor deposition might give an inefficient emitting diode. Nevertheless, we recently demonstrated that the EL emission can be enhanced if the compound is incorporated as a guest in a conjugated polymer host [12].

In order to extend the possibility of preparing solution-processed OLEDs with better performances, we present here a comprehensive study of diodes using poly(9-vinylcarbazole) (PVK):aquo[*N,N'*-bis(salicylidene)-1,2-phenylenediamine]zinc(II) ([Zn(salophen)]) or aquo[*N,N'*-bis(salicylidene)-3,4-diaminobenzophenone]zinc(II) ([Zn(sal-3,4-ben)]) (Fig. 1) composites in a host:guest systems. The efficiencies of these two diodes were compared with the Alq₃ and bis(8-hydroxyquinolate)zinc(II), Znq₂ materials prepared using the same procedure. Furthermore, their performances were compared with diodes containing the respective free ligands, *N,N'*-bis(salicylidene)-1,2-phenylenediamine (salophen) and *N,N'*-bis(salicylidene)-3,4-diaminobenzophenone (sal-3,4-ben). The diode structure was ITO|PEDOT:PSS|PVK:X|Ca|Al (Fig. 2), where X is the coordination compound or the free ligand. The concentration of the guests in the PVK matrix was 2.5 % mol/mol. Although PVK is known as a poor EL material, often it has been used due to its hole transport and electron blocking properties [42,43] and may be used as a host for small molecules [26]. To gain good understanding about the nature of the diode emission, we compared the electro-optical properties (steady-state

photoluminescence (PL) and dynamic photoluminescence) of the compounds (Fig. 1) in the PVK matrix. Also, the current density vs. voltage (*JxV*) curves were adjusted by trapped-charge limited current (TCLC) [44–46] model to explain the diode performances based on the hole injection mechanism and hole mobility (μ_p).

2. Experimental

2.1. Materials

Poly(9-vinylcarbazole) (PVK) ($M_w = 1,100,000 \text{ g mol}^{-1}$), 3,4-diaminobenzophenone, salicylaldehyde, Alq₃ and Znq₂ were purchased from Sigma-Aldrich, São Paulo, Brazil. Zinc acetate was purchased from Fisher Scientific Company (Brazil). THF solvent was purchased from Sigma-Aldrich, São Paulo, Brazil as anhydrous ($[\text{H}_2\text{O}] < 99.9\%$) inhibitor-free grade. Poly(ethylene dioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS) was purchased from Baytron P CH 8000, Bayer AG, Germany. The salicylidene derivatives and their Zn(II) coordination compounds were synthesized and characterized according to the literature [31].

2.2. Methods

2.2.1. Synthesis and characterization of sal-3,4-ben and [Zn(sal-3,4-ben)]

The synthesis of the sal-3,4-ben ligand was described in a previous work [33]. The [Zn(sal-3,4-ben)] was produced by dissolving 133 mg (0.316 mmol) of sal-3,4-ben in 20 mL of methanol under constant stirring. A methanolic solution of zinc(II) acetate dihydrate (69 mg; 0.316 mmol) was slowly dropped into the ligand mixture. The precipitate was filtered and washed with hot deionized water and ethanol. The product was an orange polycrystalline powder, which was obtained with 83 % yield. Elemental analysis (%) of the C, H and N atoms was performed using a Perkin-Elmer microanalyzer model PE 2400: calculated, C 64.62, H 4.02 and N 5.58; found, C 64.60, H 4.05 and N 5.56. ¹H and ¹³C NMR 1D spectra (Figures S1 and S2) were measured using a DMSO-d₆ solution (33 mg mL⁻¹) in a Varian Mercury 300 MHz spectrometer. The results are: ¹H NMR (300 MHz, DMSO-d₆) δ (ppm): 6.50 (*m*, 2H), 6.69 (*dd*, $J = 8.55 \text{ Hz}$, 2H), 7.25 (*m*, 2H), 7.42 (*d*, $J = 8.4 \text{ Hz}$, 2H), 7.63 (*m*, 2H), 7.80 (*m*, 2H), 8.00 (*d*, $J = 8.7 \text{ Hz}$, 2H), 8.22 (*d*, $J = 1.5 \text{ Hz}$, 2H), 9.03 (*s*, 1H) and 9.06 (*s*, 1H); ¹³C NMR (75 MHz, DMSO-d₆) δ (ppm): 113.83, 114.01, 117.22, 118.19, 120.07, 120.11, 123.81, 124.02, 129.42, 130.43, 133.60, 135.37, 135.79, 136.00, 137.19, 137.26, 137.62, 140.15, 143.59, 164.53, 165.07,

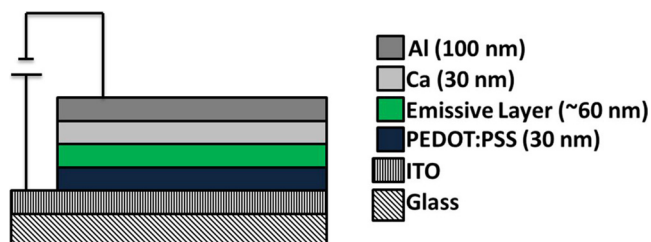


Fig. 2. Device architecture: ITO|PEDOT:PSS|PVK:X|Ca|Al where X = Alq₃, Znq₂, salophen, [Zn(salophen)], sal-3,4-ben or [Zn(sal-3,4-ben)].

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