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Novel metal–chelate emitting materials based on polycyclic aromatic ligands for electroluminescent devices

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We have designed and synthesized novel metal–chelate complexes based on polycyclic aromatic ligands for electroluminescent devices. These complexes exhibited strong luminescence with blue and green colors. EL properties of devices using these complexes for an emitting layer have been studied. Several good emitting materials were obtained and the EL properties were found to strongly depend on the ligand structure.

Introduction

Organic electroluminescent (EL) devices are of great interest because of attractive applications such as large-area light emitting displays which are operative at a low drive voltage.[1] Organic EL devices can be easily tuned by choosing a suitable emitting material.[2] The most representative emitting material is Alq3 [tris(8-quinolinolato)aluminium] which was originally reported by Tang *et al.*[3] Alq3 has high luminous efficiency, high electron mobility and also good chemical stability. The chemical structure of Alq3 consists of one aluminium ion and three ligands [8-quinolinolate (quin)] based on a fused ring system with imine and phenolate functionalities as coordination sites.

Recently, metal–chelate materials based on polycyclic aromatic ligands which chelate through a phenolic oxygen and doubly bound nitrogen [=N(*o*-phenol)] have been reported.

These compounds are attractive for tuning emission colors, because such metal–chelate materials with polycyclic aromatic ligands emit various luminescence colors depending on the molecular structure. For example, it has been reported that Zn(BOX)$_2$ {bis[2-(2-hydroxyphenyl)benzoxazolate]zinc}[4] emits blue light and Zn(BTZ)$_2$ {bis[2-(2-hydroxyphenyl)benzothiazolate]zinc}[5] emits whitish light. However, the relationship between the ligand structure and the PL spectrum is, as yet not clear. The hetero atoms (O,S,N) in the aromatic ligands are presumed to affect the photoluminescence (PL) and EL properties by changing the energy level of the molecular orbitals.

Here, we report novel metal–chelate complexes based on polycyclic aromatic ligands containing an =N(*o*-phenol) moiety, in which electron donating (N,S,O) groups are introduced at the 1 position of the imino ring and/or electron withdrawing (—N=) groups are introduced at the 4 position of the imino ring (Fig. 1). The PL and EL properties of the complexes have been studied. Especially, the influence of the hetero group in the polycyclic ligands and metal center in the complexes upon the PL spectrum has been studied systematically. The EL spectra of the EL devices using these materials were almost identical to the PL spectra, and several good blue and green emitting materials were obtained in this study.

Results and Discussions

The novel metal–chelate complexes synthesized are shown in Fig. 1 and the ligands are shown in Fig. 2. ODZ, TDZ, BIZ and PhPy are novel ligands while BTZ, BOX and 8-quinolinol have been reported.[4,5] The novel ligands (Fig. 2) were synthesized according to Scheme 1.

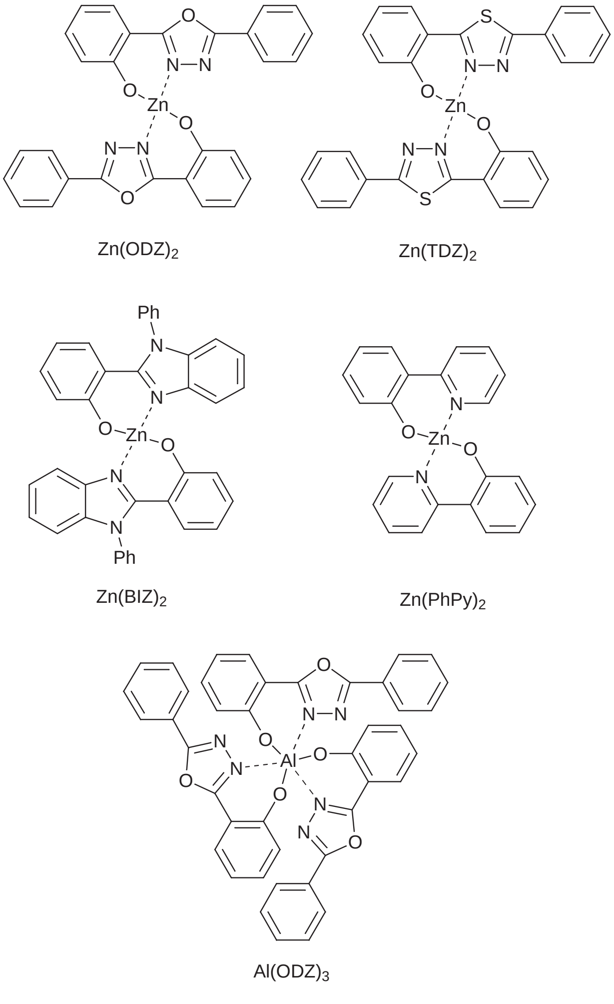

Zn(ODZ)$_2$ Zn(TDZ)$_2$
Zn(BIZ)$_2$ Zn(PhPy)$_2$
Al(ODZ)$_3$

Fig. 1 Structures of the complex molecules with polycyclic ligands

Each synthesized ligand molecule has an =N(*o*-phenol) type coordination site. ODZ and TDZ have one donating group (O, S) and one electron withdrawing group in the imino ring. BIZ, BTZ and BOX has one electron donating group (N, S, O) in the coordinated imino ring while PhPy contains no hetero group.

ODZ and TDZ were synthesized by cyclization of the corresponding 1,2-diacylhydrazine. Cyclization with thionyl chloride yielded ODZ, and cyclization with phosphorus pentasulfide yielded TDZ. BIZ was obtained by condensation of *o*-anisic acid and *N*-phenyl-1,2-phenylenediamine. The methoxy group of BIZ intermediate was demethylated in the course of the condensation reaction. PhPy was synthesized by Grignard

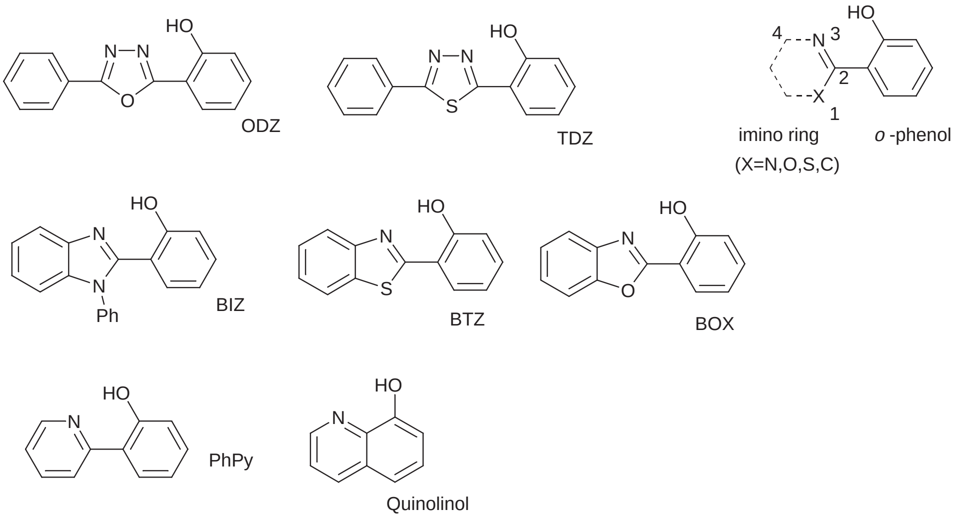
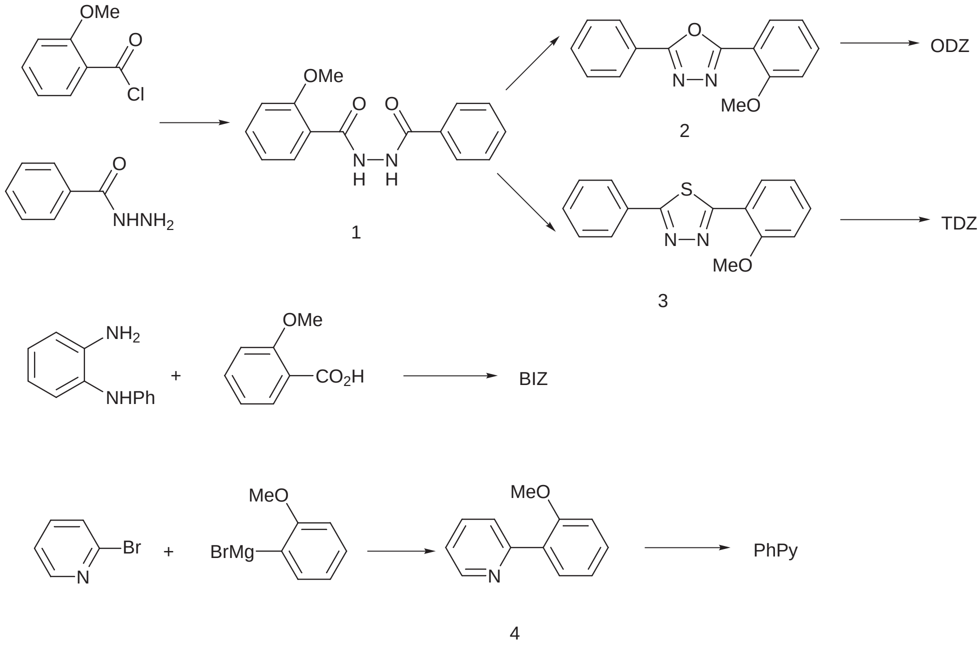


Fig. 2 Structures of the polycyclic ligand molecules



Scheme 1 Synthetic schemes for the target ligands

coupling of *o*-methoxybromobenzene with nickel phosphine catalyst[6] followed by demethylation with BBr_3.

EL devices were fabricated by vacuum deposition of the organic materials and must be stable to heating during the deposition process, hence thermal durability is important in these materials. The complexes synthesized were thermally stable up to 300 °C. No phase transitions of the complexes were observed by DSC measurements between room temperature and 300 °C.

All of the complexes exhibited strong fluorescence. PL spectra of the metal complexes are summarized in Table 1. These emitting materials showed blue or green fluorescence. Both $Zn(PhPy)_2$ and $Zn(quin)_2$[7] coordinate to zinc *via* imino nitrogen and hydoxyl oxygen. $Zn(PhPy)_2$ consists of a polycyclic ligand while $Zn(quin)_2$ consists of a fused ring ligand. The PL peaks of $Zn(PhPy)_2$ and $Zn(quin)_2$ occur at 455 and 542 nm, respectively.[7] The PL peak of the polycyclic ligand complex was at a shorter wavelength than that of the fused ring ligand complex.

PL spectra of $Zn(BIZ)_2$, $Zn(BOX)_2$[4] and $Zn(BTZ)_2$[5], which contain an electron dontating group in each fused imino ring were compared. $Zn(BIZ)_2$ exhibited a sharp PL spectrum. The spectral halfwidth ($\Delta\lambda$) of $Zn(BIZ)_2$ was less than those of the $Zn(BOX)_2$ and $Zn(BTZ)_2$. Broadening of the PL spectrum appears to be suppressed when the nitrogen group is introduced as a donating group into the ring compared to oxygen and sulfur. The PL peak red-shifted as the electron-donating character of the heteroatoms increased [ionization potential of heteroatom/eV:[8] 14.53 for N, 13.62 for O and 10.36 for S, PL peak wavelength/nm: 440 for $Zn(BIZ)_2$, 478 for $Zn(BOX)_2$[4] and 487 for $Zn(BTZ)_2$[5] with large spectral expansion towards longer wavelengths].

A similar relationship between the PL peak wavelength and electron-donating character was found in $Zn(ODZ)_2$ and $Zn(TDZ)_2$. These complexes contain 3,4-diazole moieties with one donating group (O or S) at the 1 position. The PL peak of $Zn(TDZ)_2$ is more red shifted than that of $Zn(ODZ)_2$ by *ca.* 50 nm as the electron-donating character of the heteroatom increased. In both cases the PL spectrum was affected strongly by the introduction of a sulfur atom with a broadening of the spectrum for $Zn(BTZ)_2$ and a large red shift for $Zn(BTZ)_2$.

The nature of the metal center (Zn or Al) did not affect the PL spectrum with the spectra of $Zn(ODZ)_2$ and $Al(ODZ)_3$ being the same. On the other hand, in chelete complexes

Table 1 Photoluminescence (PL) and electroluminescence (EL) results for the complexes

		PL		EL[a]			
emitting material	PL, EL color	λ_{max}/nm	$\Delta\lambda$/nm	λ_{max}/nm	$\Delta\lambda$/nm	max. luminance/cd m^{-2} (applied voltage/V)	luminous efficiency[b]/lm W^{-1}
$Zn(ODZ)_2$	blue	457	73	460	66	3000 (17.6)	0.45
$Zn(TDZ)_2$	green	505	73	505	100	1000 (18)	0.13
$Zn(PhPy)_2$	blue	455	56	483	91	5300 (14)	0.45
$Zn(BIZ)_2$	blue	438	51	450	73	800 (19)	0.15
$Al(ODZ)_3$	blue	455	73	460	100	400 (22)	0.08
$Zn(BOX)_2$[c]	blue	478	112				
$Zn(BTZ)_2$[d]	whitish green	487	157				

[a] EL device structure: [ITO/TPD (70 nm/emitting layer (70 nm)/Mg:Ag (180 nm)]. [b] Value at luminance 100 cd m^{-2}. [c] Ref. 4. [d] Ref. 5.

having fused ring ligands (8-quinolinolate),[7] the PL peak wavelength is affected by the coordinated metal [542 nm for $Zn(quin)_2$ and 519 nm for $Al(quin)_3$)].

The synthesized metal complexes were used as emitting materials of EL devices after conventional vacuum-vapor deposition. The device structures were [ITO/hole transport layer (70 nm)/emitting layer(70 nm)/Mg:Ag(10:1, 180 nm)]. The hole transport layer was composed of TPD (*N*,*N′*-dimethyl-*N*,*N′*-*m*-ditolylbenzidine),[9] and the emitting layer was composed of the synthesized metal complexes. These complexes formed uniform, smooth and clear films. No crystallization was found in the films over several months. The EL peak wavelengths λ_{max} and half-widths ($\Delta\lambda$) of the complexes are listed in Table 1. The PL and EL spectra were almost identical, which implies that the EL emission originated from the metal complex layer in the EL device, and that no exiplex is formed between the hole transport layer and the emitting layer to reduce EL efficiency. The EL color of the devices was blue for $Zn(ODZ)_2$, $Zn(PhPy)_2$, $Zn(BIZ)_2$ and $Al(ODZ)_3$ and green for $Zn(TDZ)_2$. A blue color was characteristic for complexes based on polycyclic ligand systems while complexes based on fused ring systems emit green light in most cases.[7,10] The maximum luminance and luminous efficiency of these devices are given in Table 1. The EL devices fabricated with $Zn(ODZ)_2$, $Zn(TDZ)_2$ and $Zn(PhPy)_2$ exhibited a high luminance >1000 cd m^{-2}. $Zn(PhPy)_2$ showed the highest luminance value of 5300 cd m^{-2}. In luminance–current density measurements of the EL devices, the luminance was found to be proportional to the injection current. The high luminance and charge injection efficiency of the EL devices imply electron transporting ability of the complexes.[7]

In summary, we have synthesized metal–chelate complexes based on polycyclic aromatic novel ligands containing =N(*o*-phenol) moieties as coordination sites. The EL and PL wavelength of the polycyclic system was shorter than those of corresponding fused ring systems. The peak wavelengths of the EL and PL spectra were red shifted by the introduction of electron donating atoms into the imino ring of the ligands. Especially, sulfur was found to induce a large red shift. The nature of the coordinated metal in the complexes based on polycyclic systems did not affect the PL spectrum. In EL devices these materials exhibited an electron transporting ability and emitted blue or green light.

Experimental

All metal–chelate materials were purified by the train sublimation method reported by Wanger *et al.*[11] Yields of 70–80% were obtained for the metal–chelate complexes.

A Perkin-Elmer DSC7 instrument was used for the DSC measurements and ^1H NMR spectra were recorded at 90 MHz on a JEOL FX90Q spectrometer. Chemical shifts refer to Me_4Si as internal standard. IR spectra were recorded on a JASCO FT/IR-5M spectrophotometer. EL and PL spectra were measured with a multichannel analyzer (Hamamatsu photonics PMA-11) upon irradiating with a UV lamp ($\lambda=350$ nm). The EL devices were fabricated by the vacuum deposition method. A 70 nm layer of TPD was deposited on a substrate consisting of glass slides precoated with indium tin oxide (with sheet resistance of 15 $\Omega/\square^{-1}$), by thermal evaporation under a vacuum of 10^{-4} Pa. A 70 nm layer of emitting material was then deposited on the TPD layer. A top electrode was subsequently deposited by coevaporation of Mg and Ag (10:1 atomic ratio) under a vacuum of 10^{-5} Pa. The luminance of the device was measured with a Minolta photometer (nt-1/3° P). EL measurements were carried out under a nitrogen atmosphere.

Syntheses

1-Benzoyl-2-(2-methoxybenzoyl)hydrazine 1. To an ice-cooled suspension of 15.34 g (0.11 mol) of benzoylhydrazide and 11.96 g (0.11 mol) of sodium carbonate in 184 g of dioxane was added 19.16 g (0.11 mol) of *o*-methoxybenzoyl chloride dropwise over 10 min with stirring under a nitrogen atmosphere. The reaction mixture was stirred for 1 h at room temperature (r.t.) then for 1 h at 90 °C. After cooling to r.t., to the mixture was added 400 ml of water. The reaction mixture was then filtered and washed with water. The white solid was dried *in vacuo* to give 21.0 g (71%) of **1**: mp 134 °C. ^1H NMR (90 MHz,$CDCl_3$) δ 4.10 (s,3H), 7.18–6.96 (m,2H), 7.62–7.40 (m,4H), 7.98–7.80 (m,2H), 8.19 (d,1H,$J=8.4$), 9.79 (br,2H). IR(KBr) 3260, 3050, 1630, 1475, 1290, 1240, 1100, 755, 705, 595 cm^{-1}. MS for $C_{15}H_{14}N_2O_3$: calc. 270(M), found 270.

2-(2-Methoxyphenyl)-5-phenyl-1,3,4-oxadiazole 2. A mixture of 5.5 g (20 mmol) of **1**, 20 g of thionyl chloride and 0.3 g of pyridine was stirred at 80 °C under a nitrogen atmosphere. After 4 h, the reaction mixture was cooled and poured onto 200 g of crushed ice. The reaction mixture was then filtered and washed with water. The white solid was dried *in vacuo* to give 5.2 g (quantitative) of **2**: mp 91 °C. ^1H NMR (90 MHz,$CDCl_3$) δ 3.99 (s,3H), 7.00–7.18 (m,2H), 7.40–7.63 (m,4H), 7.94–8.21 (m,3H). IR(KBr) 3050, 3000, 2950, 2850, 1605, 1535, 1475, 1270, 1020, 745, 710, 685 cm^{-1}. MS for $C_{15}H_{12}N_2O_2$: calc. 252(M), found 252.

2-(2-Hydroxyphenyl)-5-phenyl-1,3,4-oxadiazole (ODZ). To a solution of 5.13 g (20 mmol) of **2** in 50 ml of CH_2Cl_2 at -78 °C was added 3 ml (32 mmol) of BBr_3 in 20 ml of CH_2Cl_2 dropwise over 10 min. The reaction mixture was allowed to warm slowly to room temperature. Water (50 ml) was added to the reaction mixture carefully, then the reaction mixture was extracted twice with 30 ml of CH_2Cl_2. The organic layer was dried over Na_2SO_4. The solvent was removed by evaporation to yield a white solid. Recrystallization of the residue from 200 ml of ethanol gave ODZ as a white crystalline solid: 3.89 g (82%): mp 165 °C. ^1H NMR (90 MHz,$CDCl_3$) δ 6.95–7.23 (m,2H), 7.35–7.78 (m,4H), 7.87 (d,1H,$J=8$), 8.16 (m,2H), 10.19 (s,1H). IR(KBr) 3200, 3160, 3060, 1625, 1590, 1540, 1490, 1410, 1240, 1070, 750, 710, 690 cm^{-1}. MS for $C_{14}H_{10}N_2O_2$: calc. 238(M), found 238.

2-(2-Methoxyphenyl)-5-phenyl-1,3,4-oxathiazole 3. A mixture of 5.5 g (20 mmol) of **1**, 5.5 g (12 mmol) of P_4S_{10} and 50 ml of xylene was stirred at 140 °C for 4 h. After cooling, 140 ml of water was added to the reaction mixture. This was heated at 80 °C for 1 h. Sodium hydrogen carbonate was added to the water layer until a pH of 6 was obtained. The xylene layer was washed with water and dried over Na_2SO_4. The solvent was removed by evaporation to yield a white solid. The residue was dried *in vacuo*. The crude product was chromatographed (SiO_2, $CHCl_3$) to yield **3** as a pale yellow solid, 3.5 g (65%): mp 137 °C. ^{1}H NMR (90 MHz,$CDCl_3$) δ 4.02 (s,3H), 6.98–7.22 (m,2H), 7.35–7.59 (m,4H), 7.92–8.20 (m,2H), 8.52 (d,1H,$J=8$Hz). IR(KBr)3050, 3000, 2940, 2830, 2700, 2530, 2560, 1595, 1450, 1410, 1300, 1260, 1015, 725, 680 cm^{-1}. MS for $C_{15}H_{12}N_2OS$: calc. 268(M), found 268.

2-(2-Hydroxyphenyl)-5-phenyl-1,3,4-oxathiazole (TDZ). To a solution of 3.5 g (13 mmol) of **3** in 40 ml of CH_2Cl_2 at -78 °C was added dropwise 17.5 ml of a solution of 1.0 M BBr_3 in CH_2Cl_2. The reaction mixture was allowed to warm slowly to room temperature overnight. Water (50 ml) was added to the reaction mixture carefully, then the reaction mixture was extracted twice with 30 ml of CH_2Cl_2. The organic layer was dried over Na_2SO_4. The solvent was removed by evaporation to yield TDZ as a white solid: 1.75 g (53%): mp 128 °C. ^{1}H NMR (90 MHz,$CDCl_3$) δ 6.85–7.43 (m,3H), 7.43–7.68 (m,4H), 7.91–8.08 (m,2H), 11.48 (s,1H). IR(KBr) 3060, 3030, 2920, 2730, 2630, 2570, 1600, 1455, 1430, 1310, 1260, 1100, 1000, 750, 680, 600 cm^{-1}. MS for $C_{14}H_{10}N_2OS$: calc. 254(M), found 254.

2-(2-Methoxyphenyl)pyridine 4. To a mixture of 1.28 g (53 mmol) of Mg and 1 ml of THF was added a solution of 10 g (53 mmol) of *o*-methoxybromobenzene in 15 ml of dry THF. The reaction mixture was refluxed for 30 min. This Grignard solution was added dropwise to a mixture of 7.5 g (47 mmol) of 2-bromopyridine, 0.25 g of 1,3-bis(diphenylphosphinopropane)nickel(II) chloride and 15 ml of dry THF over 10 min at 0 °C. The reaction mixture was allowed to warm to room temperature and stirred overnight. To the reaction mixture was added 30 ml of water and the solvent was then evaporated. The residue was partitioned between 70 ml of water and 50 ml of $CHCl_3$. The organic layer was dried over Na_2SO_4 and evaporated to yield **4** as a light brown oil: 8.84 g (quantative). ^{1}H NMR (90 MHz,$CDCl_3$) δ 3.82 (s,3H), 6.91–7.50 (m,4H), 7.55–7.90 (m,3H), 8.7 (d,1H,$J=6$Hz). IR(KBr) 3060, 3000, 2950, 2840, 1600, 1585, 1500, 1460, 1425, 1260, 1240, 1025, 755 cm^{-1}. MS for $C_{12}H_{11}NO$: calc. 185(M), found 185.

2-(2-Hydroxyphenyl)pyridine (PhPy). To a solution of 8.84 g (48 mmol) of **4** in 50 ml of CH_2Cl_2 at -78 °C was added dropwise 49.5 ml of a solution of 1.0 M BBr_3 in CH_2Cl_2. The reaction mixture was allowed to warm slowly to room temperature overnight. Water (60 ml) was added to the reaction mixture carefully, then the reaction mixture was extracted twice with 100 ml of CH_2Cl_2. The organic layer was dried over Na_2SO_4. The solvent was removed by evaporation to yield PhPy as a light brown oil: 3.35g (41%). ^{1}H NMR (90 MHz,$CDCl_3$) δ 6.79–7.35 (m,4H), 7.65–7.95 (m,3H), 8.52 (d,1H,$J=6$), 14.35 (br,1H). IR(KBr) 3400, 3060, 2950, 2840, 1600, 1505, 1495, 1260, 1025, 830, 760 cm^{-1}. MS for $C_{11}H_9NO$: calc. 171(M), found 171.

2-(2-Hydroxyphenyl)-1-phenylbenzimidazole (BIZ). A mixture of 2.0 g (13 mmol) of *o*-anisic acid, 2.42 g (13 mmol) of *N*-phenyl-1,2-phenylenediamine and 5 ml of *o*-dichlorobenzene was heated at 170 °C for 10 h, then at 220 °C for 30 h. The crude product was chromatographed (SiO_2, 50% $CHCl_3$–hexane) to yield BIZ as needles, 0.37g (10%): mp 119 °C. ^{1}H NMR (90 MHz,$CDCl_3$) δ 6.55 (t,1H,$J=8$), 6.89 (d,1H,$J=8$), 7.01–7.50 (m,7H), 7.52–7.73 (m,3H), 7.75–7.91 (m,1H), 13.53 (br,1H). IR(KBr) 3050, 2600, 1580, 1480, 1380, 1280, 1255, 810, 740, 700 cm^{-1}. MS for $C_{19}H_{14}N_2O$: calc. 286(M), found 286.

Bis[2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato]zinc $Zn(ODZ)_2$. To a suspension of 500 mg (2.1 mmol) of ODZ and 5 g of methanol was added chloroform until the suspension was clear. To this solution was added a solution of 230 mg (1.0 mmol) of zinc acetate in 1.55 g of methanol and the mixture was heated at 70 °C for 1.5 h. The solvent was removed by evaporation. The residue was washed with water and dried *in vacuo* to give a pale yellow solid. The crude materials were purified by the train sublimation method to give $Zn(ODZ)_2$. Anal. Calc. for $C_{28}H_{18}N_4O_4Zn$: C,62.30; H,3.36; N,10.38. Found: C,62.04; H,3.33; N,10.14%.

Bis[2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxathiazolato]zinc $Zn(TDZ)_2$. To a suspension of 597 mg (2.3 mmol) of TDZ, 204 mg (2.3 mmol) of morpholine and 5 g of methanol was added a solution of 258 mg (1.2 mmol) of zinc acetate and 1.7 g of methanol with stirring. The mixture was heated at 70 °C for 1 h and the solvent was removed. The residue was washed with water and dried *in vacuo*. The crude materials were purified by the train sublimation method to give $Zn(TDZ)_2$. Anal. Calc. for $C_{28}H_{18}N_4O_2S_2Zn$: C,58.80; H,3.17; N,9.80. Found: C,58.63; H,3.13; N,9.56%.

Bis[2-(2-hydroxyphenyl)pyridinato]zinc $Zn(PhPy)_2$. To a solution of 816 mg (4.8 mmol) of PhPy, 419 mg (4.9 mmol) of piperidine, 3 g of methanol and 10 g of chloroform was added a solution of 523 mg (2.4 mmol) of zinc acetate in 3.5 g of methanol. The solvent was evaporated slowly for 5 days. The residue was washed with water and dried *in vacuo* to give a pale yellow solid. The crude materials were purified by the train sublimation method to give $Zn(PhPy)_2$. Anal. Calc. for $C_{22}H_{16}N_2O_2Zn$: C,65.12; H,3.97; N,6.90. Found: C,64.70; H,3.97; N,6.70%.

Bis[2-(2-hydroxyphenyl)-1-phenylbenzimidazolato]zinc $Zn(BIZ)_2$. To a solution of 365 mg (1.3 mmol) of BIZ, 114 mg (1.3 mmol) of piperidine and 2 g of methanol was added a solution of 140 mg (0.64 mmol) of zinc acetate in 0.92 g of methanol. The reaction mixture was stirred for 6 h. The precipitates were filtered off and washed with water and dried *in vacuo*. The crude materials were purified by the train sublimation method to give $Zn(BIZ)_2$. Anal. Calc. for $C_{38}H_{26}N_4O_2Zn$: C,71.76; H,4.12; N,8.81. Found: C,71.39; H,4.13; N,8.66%.

Tris[2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato]-aluminium $Al(ODZ)_3$. To a solution of 1.00 g (4.2 mmol) of ODZ and 0.75 g (8.6 mmol) of morpholine in 190 g of ethanol was added a solution of 0.187 g (1.4 mmol) of aluminium chloride in 3.15 g of ethanol. The reaction mixture was heated at 80 °C for 4 h. The precipitates were filtered off and washed with water and dried *in vacuo*. The crude materials were purified by the train sublimation method to give $Al(ODZ)_3$. Anal. Calc. for $C_{42}H_{27}N_6O_6Al$: C,68.29; H,3.68; N,11.38. Found: C,68.32; H,4.08; N,10.96%.

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