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Metal chelates as emitting materials for organic electroluminescence

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Contents

Abstract

Recent developments in the design and use of vapor-deposited thin-film metal chelates as organic electroluminescent materials for display applications are presented. The material issues pertaining to color, emission efficiencies, and operational stability are described. © 1998 Elsevier Science S.A.

Keywords: Organic electroluminescence; OLED; Materials; Devices; Aluminium Chelate; Display

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1. Introduction

Organic electroluminescent (EL) devices are of growing interest in various display applications because of their high luminous efficiency and capability of emitting

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many colors throughout the visible spectrum. Much progress has been made since the discovery of a *tris*(8-hydroxyquinolinato)aluminum (AlQ_3)-based multilayer thin-film device by Tang and co-workers in 1987 [1,2]. This phenomenon is attributed largely to the continuing discovery of new and improved EL materials. From small fluorescent molecules (for a recent review see ref. [3]) to conjugated polymers [4,5], intense research effort in both industrial and academic laboratories has yielded organic EL devices with remarkable efficiencies [6] and operational stability [7–9]. These advances have in turn stimulated considerable commercial interest in developing this new display technology that offers significant advantages over those of the current flat panel displays [10].

This article is written primarily from a chemist's perspective. It is intended to focus on the most significant developments in AlQ_3 chelate derivatives that have been designed and synthesized specifically for organic EL application. A more extensive review on organic electroluminescent materials and devices can be found in a newly published book [11]. The device principle, applications requirements and reliability issues of organic electroluminescent devices have also been discussed recently [12].

2. Device structure and fabrication

A basic multilayer structure of a thin-film organic EL device is sketched in Fig. 1. The electron-transport layer is AlQ_3, the emitting layer is AlQ_3 doped with a

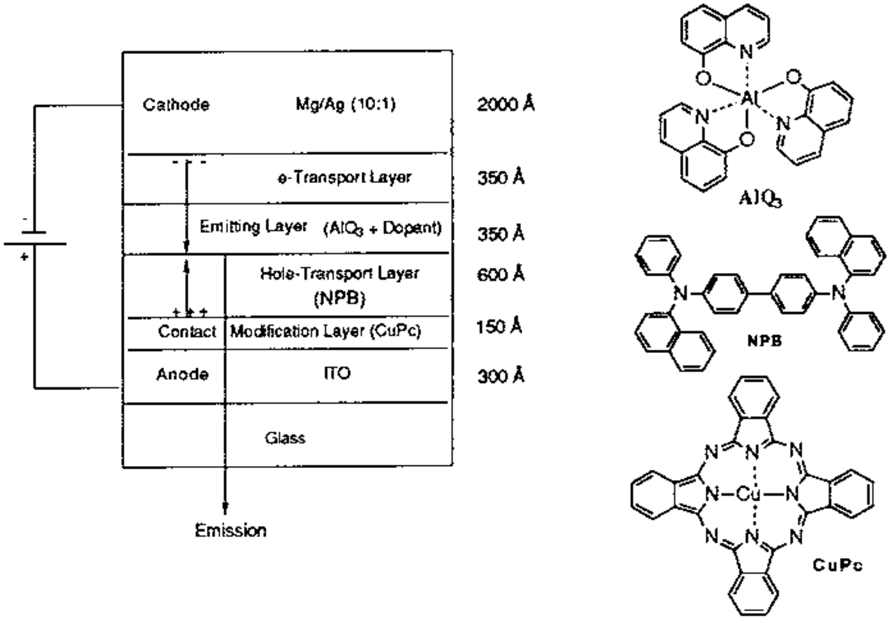


Fig. 1. Multilayer structure of a thin-film EL device

fluorescent dye, and the hole-transport layer is α-naphthylphenylbiphenyl amine (NPB). An additional layer, copper phthalocyanine (CuPc), was inserted between the NPB layer and the indium-tin-oxide (ITO) electrode, which is typically 300 Å thick with a sheet resistance of about 100 ohms/square.

All organic layers were prepared in a vacuum chamber (about 1×10^{-5} Torr) by vapor deposition using resistively heated tantalum boats. Typically, the deposition rate was 4 Å/s. After the deposition of the organic layers and without a vacuum break, the Mg:Ag (10:1) electrode was deposited on top of the organic layers using separately controlled sources. The deposition rate for Mg and Ag was typically 10 and 1 Å/s, respectively. The active area of the EL device, defined by the overlap of the ITO and the Mg:Ag electrodes, was 0.1 cm². The EL device was completed with encapsulation in a dry argon glove box.

The device is current driven and the light emission whose wavelength usually corresponds to the photoluminescence of the emitting material is measured from the transparent glass side, as shown in Fig. 1. A typical standard EL cell based on AlQ_3 without any dopant has an initial luminance of about 510 cd/m² driven at a constant current density of 20 mA/m². The initial luminance–current–voltage (L–J–V) and the corresponding L–J characteristics of a standard EL device are shown in Fig. 2. The luminous power efficiency has a maximum value of 0.73 lm/W at about 9.5 V and the emission spectrum peaks at around 528 nm with a FWHM about 100 nm, which is independent of current levels [7].

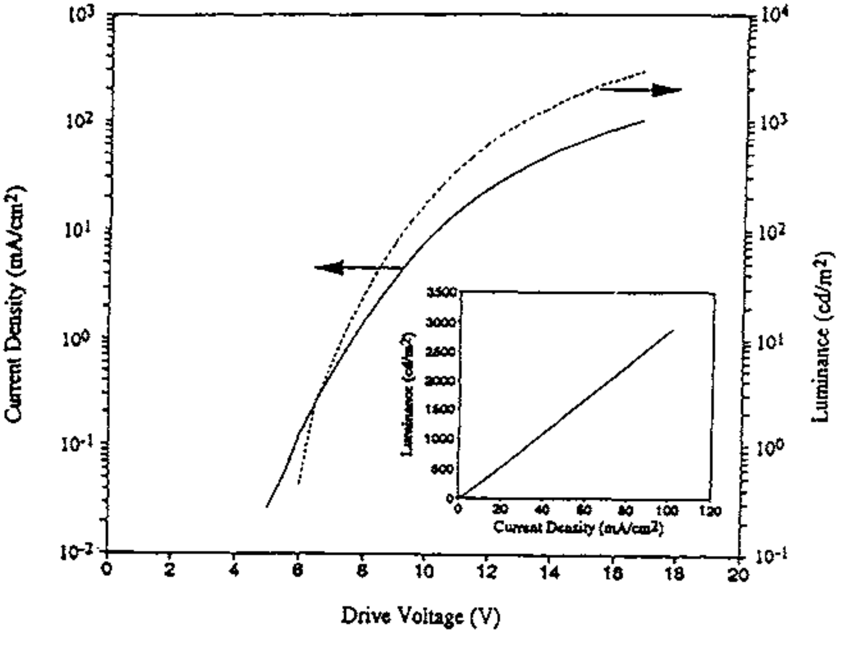


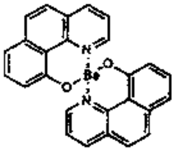
Fig. 2. Luminance–current–voltage characteristics of the undoped AlQ_3 EL device. (The inset shows the luminance vs. forward bias current.)

Using this multilayer structure and several other variations, we have shown that the electron–hole pair recombination can be largely confined to the interface between the hole-transport (NPB) and electron-transport emitting (AlQ_3) layers. By spacing this interface at a sufficient distance from the contacts, the probability of quenching near the metallic surfaces is greatly reduced. As a result, the EL emission is very efficient.

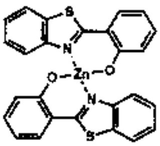
The AlQ_3 layer in the EL structure described above is responsible for the broadband green EL emission. Other EL colors can, in principle, be obtained by choosing different organic fluorescent materials as emitters. Indeed, numerous fluorescent molecules have been found to produce EL emissions of other hues. However, the preferred method for tuning the EL colors is by doping a guest molecule into a host emitting material, such as AlQ_3 [13]. By direct excitation or via energy transfer from the host, the guest molecule emits with its characteristic luminescence. In addition to tuning the EL colors, this scheme also provides a means of enhancing the overall EL efficiency by fully capitalizing on the strong fluorescence of the guest. In the doped AlQ_3 system, a greater than three-fold increase in efficiency has been realized over the undoped system [14]. As will be discussed later, the AlQ_3/dopant emitter also greatly enhances the operational stability of the organic EL device.

3. Material considerations

To be useful as emitting materials for vapor-deposited organic light-emitting devices (OLED), metal chelates must be isolable, thermally stable, highly fluorescent in the solid state, thin-film forming upon vacuum deposition, and also capable of transporting electrons. Many metal chelates are highly luminescent in solution, but only a handful of chelates can satisfy all of the above requirements for EL applications. The difference lies in the fact that in solution, most metal complexes can exist with open coordination sites that are stabilized by additional solvating molecules, such as water. This solvation is no longer possible for metal chelates that are isolated as solids. Therefore, many fluorescent metal chelates with open coordination sites are not thermally stable unless they are filled by additional ligands.



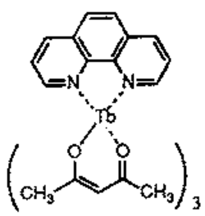
$BeBq_2$



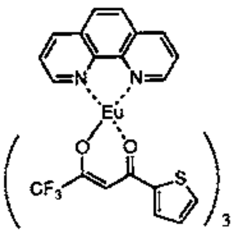
$Zn(BTZ)_2$

Most of the luminescent EL metal chelates known to date contain metal ions that belong to Group II or III of the periodic chart. Within Group III metals, Al^{3+}

(with coordination number 6) is the most important, while in Group II, Be^{2+} and Zn^{2+} (with coordination number 4) are the most prominent electron-transport emitting materials for electroluminescence. Besides AlQ_3, the most noteworthy beryllium chelate is the green fluorescent *bis*(10-hydroxybenzo[h]quinolinato)beryllium known as $BeBq_2$ [15]. EL cells fabricated using rubrene doped (emission $\lambda_{max} = 562$ nm) in $BeBq_2$ have recorded a half-decay time of over 15 000 h [16] at an initial luminance of 100 cd/m^2. A recent example of zinc chelate is the electron-transporting *bis*[2-(2-hydroxyphenyl)benzothiazolato)zinc known as $Zn(BTZ)_2$, which showed greenish white emission with a very broad EL spectrum (FWHM = 157 nm, emission $\lambda_{max} = 486$, 524 nm). It exhibited a high luminance of 10 190 cd/m^2 at an applied voltage of 8 V, making it a good candidate for a rare single-component white emitter [17].



Tb(AcAc)$_3$Phen



Eu(TTFA)$_3$Phen

There are also a number of lanthanide complexes, such as *tris*(2,4-pentanediono)-1,10-phenanthroline terbium(III) [18] and *tris*[4,4,4-trifluoro-1-(2-thienyl)-1,3-butanediono]-1,10-phenanthroline europium(III) [19], that have been used as emitting materials to provide nearly monochromatic green ($\lambda_{max} = 543$ nm) and red emissions ($\lambda_{max} = 613$ nm) in organic electroluminescence. Both Tb^{3+} and Eu^{3+} ions have nine coordination sites which must be completely filled with additional ligands, such as 1,10-phenanthroline, in order to produce vacuum sublimable chelates. Besides their inherent extremely sharp emission bands, one of the motivations to use lanthanide complexes as emitters in EL devices is their potentially high internal quantum efficiency. In general, organic molecular compounds produce emission due to their π–π^* transitions resulting in luminescence half-bandwidths of about 80 to 100 nm. Spin statistic estimations lead to an internal quantum efficiency of dye-based EL devices limited to only about 25%. In contrast, the fluorescence of lanthanide chelates is based on an intramolecular energy transfer from the triplet state of the organic ligand to the 4f energy states of the metal ion. Therefore, the theoretical internal quantum efficiency is, in principle, not limited.

In the following sections, we will discuss several important classes of light-emitting aluminum chelates used in organic electroluminescent devices according to their ligand structures.

3.1. 8-Hydroxyquinoline

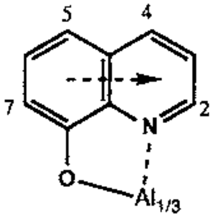
Without doubt, the most important milestone in the material research which led to the development of a stable organic EL device was the discovery of *tris*(8-hydroxyquinolinato)aluminum (AlQ_3), which is used as an electron-transport emitting layer. AlQ_3 belongs to a class of metal chelates known as the "inner complex salt" [20] in which the central metal ion, Al^{3+}, is surrounded by three bidentate 8-hydroxyquinoline anions. The chelate net charge is, therefore, zero and its coordination sites are completely filled. As a result, AlQ_3 is a remarkably stable material which can be sublimed without decomposition at a temperature of about 350 °C. Its DMF solution photoluminescence quantum efficiency is about 11% [21] and its thin-film room-temperature PL quantum efficiency is reported to be around 32% independent of film thickness from 100 Å to 1.35 μm [22]. Electron mobility of AlQ_3 was estimated to be around 10^{-5} $cm^2/V\,s$ [23] with its hole mobility only about 1/100th as much [24].

In practice, aluminum chelates have been found to provide the best combination of stability and efficiency. The *tris*-chelate AlQ_3 has been studied most extensively and, as mentioned before, is used mostly as the host material for the green and red emitting device configurations. The nature of the metal ion has been shown to influence the emission color, efficiency, stability and evaporability of the metal complex. A few general rules which govern the fluorescence of metal chelates of 8-hydroxyquinoline have been formulated [25,26]. (1) Chelates with metal ions that are paramagnetic are essentially non-fluorescent due to a high rate of intersystem crossing from the excited singlet to triplet state (e.g. Cr, Ni). (2) Fluorescence is reduced with increasing atomic number of the metal ion, also caused by an increase in the rate of intersystem crossing known as the heavy atom effect. For example, InQ_3 is less fluorescent compared to GaQ_3 which, in turn, is less fluorescent than AlQ_3. (3) As the covalent nature of the metal–ligand bonding (primarily metal–nitrogen) is increased, the emission shifts to longer wavelength. For example, the chelates formed with Al, Ga, and In emit at progressively longer wavelengths of 532, 545, and 558 nm, respectively. Conversely, more ionic metal–ligand bonding results in a blue shift. For example, MgQ_2 emits at a shorter wavelength (500 nm) compared to the analogous ZnQ_2, which emits at 557 nm.

One of the best attributes of AlQ_3 is its propensity to form a good thin-film upon vacuum deposition, and it has a relatively high T_g (175 °C) as well. Vapor-deposited AlQ_3 films consist of a solid solution of two geometric isomers (called meridianal and facial) that may hinder the recrystallization of AlQ_3 films and explain their long-term stability in a glassy form. From a dye chemist's point of view, AlQ_3 can also be considered as a metallized pigment, which is invariably more stable than a "dye". In addition, it also possesses organic ligands that may provide the best compromise for mediating phase incompatibility between a metallic phase (Mg:Ag) cathode and a pure organic phase (hole-transport material). The molecular geometry of AlQ_3 that was determined by single-crystal X-ray crystallography is roughly shaped as a ball [27]. It is believed to have little, or no, propensity to form exciplex with an electron-rich hole-transporting molecule at the interface.

3.2. 5-Substituted 8-hydroxyquinolines

Many derivatives of AlQ_3 have been designed and synthesized for use as organic light-emitting materials, among which the C-5 substituted ones are the most prevalent. It is known that the electronic π–π^* transitions in AlQ_3 are localized on the quinolate ligands. The filled π orbitals (HOMOs) are located on the phenoxide side of the quinolate ligand, and the unfilled π^* orbitals (LUMOs) are on the pyridyl side. According to a prediction made by molecular modeling [28], substitution of an electron-withdrawing substituent at C-5 or C-7 position of the phenoxide side of the quinolate ligand will cause a blue-shift of the absorption spectrum of AlQ_3, while an electron-withdrawing substituent at C-4 or C-2 position of the pyridyl side will cause a red-shift. The direction of spectral shift will be reversed if the substituents are electron-donating instead at the respective positions. In the following table, the shifts of photoluminescence of several substituted AlQ_3 are compared with that of the parent AlQ_3 in solution. From this data, it is apparent that the blue-shift caused by substituting an acceptor group at C-5 (e.g. CN) [29] is small compared to the donating effect at C-4 (e.g. Me). This result is also consistent with the ZINDO semi-empirical MO calculation [30].

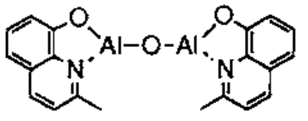


Group	4-Me	5-Me	5-F[28]	5-Cl[22]	5-CN	7-*n*-Pr[13]
$\Delta\lambda_{max}$	-10 nm	+31 nm	+15 nm	+10 nm	-3 nm	+35 nm

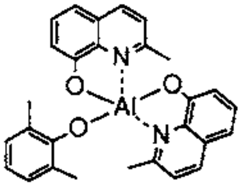
3.3. 2-Methyl 8-hydroxyquinoline

Many efforts have been spent to shift the emission of AlQ_3 to the blue. A blue electroluminescent emitter is essential for the development of a full-color display based either on the "color changing medium" technology [31] or the RGB filtered white emission. For the above mentioned *tris*(4-methyl-8-hydroxyquinolinato)aluminum(III), although blue shifted from AlQ_3, its emission at around 515 nm is still not suitable for use as the blue emitter for display purposes.

Interestingly, methyl substitution at C-2 position prevents the formation of stable *tris*-chelate with aluminum. Attempts to isolate a *tris*-chelate instead results in the formation of an oxo-bridged complex, $(QAl)_2O$, which emits at a peak wavelength of 490 nm [32]. However, because of the unfilled coordination sites remaining in



$(QAl)_2O$

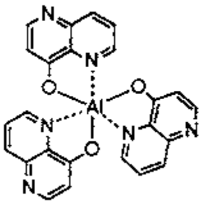


$Q_2Al\text{-}OAr$

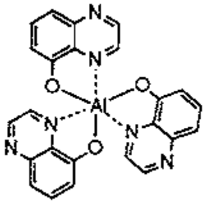
the Al^{3+}, its stability is far from satisfactory. A more stable blue emission was achieved by using the modified chelate $Q_2Al\text{–}L$, where Q is "8-hydroxy-2-methylquinoline" and L is a phenolic or an aryl carboxylate derivative [33]. A representative example is shown as structure $Q_2Al\text{–}OAr$. A bulky substituent, such as 2,5-dimethylphenol which effectively shields the Al^{3+} from nucleophilic attack, was found to improve stability.

3.4. 4-Hydroxy-1,5-naphthyridine and 5-hydroxyquinoxaline

From the molecular design point of view, the best way to alter the $\pi\text{–}\pi^*$ transition without distorting the molecular shape of AlQ_3 is by incorporation of a "heteroatom" into the 8-hydroxyquinoline ring system. The first example of such a ligand modification synthesized for OLED application was 5-hydroxyquinoxaline whose aluminum chelate is shown as AlX_3 [28]. As predicted by the ZINDO calculation, its PL emission at 580 nm is red-shifted 60 nm relative to AlQ_3. This large bathochromic shift can be attributed to the intrinsic electron-withdrawing nature of nitrogen, which replaces the "CH" at the 4-position of the quinolate ligand.



$Al(NQ)_3$



AlX_3

Recently, we have applied this methodology to effect a hypsochromic shift of AlQ_3 by replacing the "CH" at the 5-position of the quinolate ligand with nitrogen. Indeed, the new $Al(NQ)_3$ can be readily prepared from the modified ligand, 4-hydroxy-1,5-naphthyridine, by following a literature procedure [34]. Its photoluminescence at 440 nm is blue-shifted about 90 nm from that of AlQ_3.

Fig. 3 compares the PL of new $Al(NQ)_3$ with those of AlQ_3 and the blue aluminum

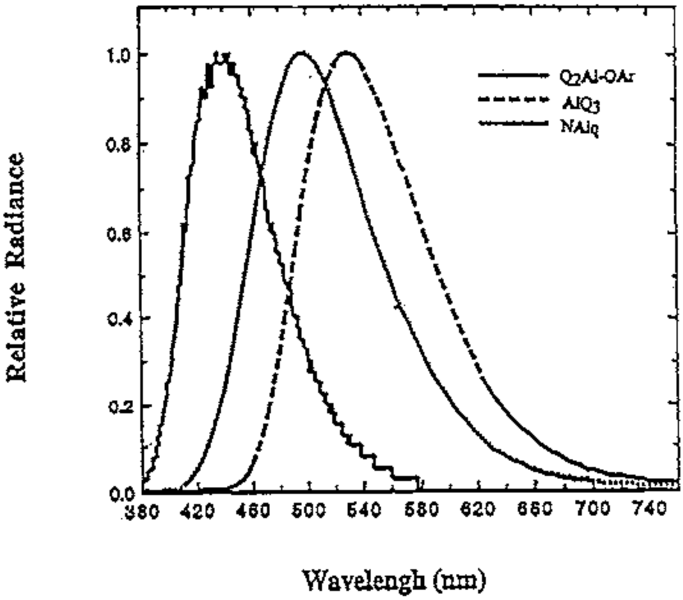


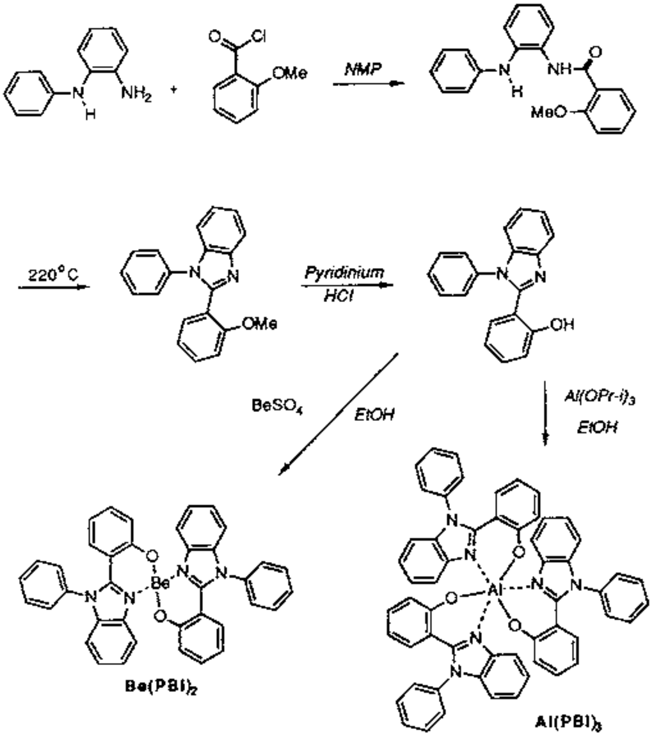
Fig. 3. Photoluminescence spectra of $Al(NQ)_3$, AlX_3, and AlQ_3.

chelate, $Q_2Al–OAr$. Single-crystal X-ray analyses of both the *trans*(1,2,6)-form of AlQ_3 and $Al(NQ)_3$ confirmed that they have an essentially identical molecular shape with both Al–N and Al–O bond lengths equal to (2.04 ± 0.02 Å) and (1.84 ± 0.02 Å) [35], respectively.

3.5. *o-(N-phenyl-2-benzimidazolyl)phenol*

We have recently discovered another bidentate ligand, *o*-(*N*-phenyl-2-benzimidazolyl)phenol, which complexes with Al^{3+} and Be^{2+} to form stable, thermally evaporable, and blue-emitting metal chelates abbreviated as $Al(PBI)_3$ and $Be(PBI)_2$, respectively [36].

The synthesis is outlined in Scheme 1. Thus, to a solution of *N*-phenyl-1,2-phenylenediamine in *N*-methyl pyrrolidinone (NMP) was reacted with an equal molar of *o*-anisoyl chloride at room temperature under nitrogen to give a 96% yield of *N*-phenyl-2-(*o*-anisoyl)benzamide. The latter was cyclized by heating at 220 °C under 0.3 atm of positive nitrogen pressure to the corresponding benzimidazole. Demethylation was conveniently accomplished by heating *N*-phenyl-2-(*o*-anisyl)-benzimidazole with excess pyridinium hydrochloride to reflux at around 200 °C to give, after passing through a short silica gel column, the bidentate ligand, *o*-(*N*-phenyl-2-benzimidazolyl)phenol in 80% yield. The *tris*-chelate was synthesized by refluxing 3 equiv. *o*-(*N*-phenyl-2-benzimidazolyl)phenol with aluminum *i*-propoxide in dry ethanol under nitrogen to give 91% yield of $Al(PBI)_3$. This material was purified further by sublimation at 345 °C under a constant 2 Torr argon pressure before subjecting to device fabrication. Similarly, the beryllium chelate, $Be(PBI)_2$,



Scheme 1. Synthesis of Al(BPI)$_3$ and Be(PBI)$_2$.

can be prepared in 71% yield by heating the bidentate ligand, *o*-(*N*-phenyl-2-benzimidazolyl)phenol with beryllium sulfate tetrahydrate in ethanol in the presence of NaOH.

The EL device was fabricated under 1×10^{-5} Torr by sequentially depositing onto a clean ITO-coated glass substrate as follows: [ITO/150 Å CuPc/600 Å NPB/400 Å Al(PBI)$_3$/2000 Å Mg:Ag (10:1)]. The light output from this EL device was 216 cd/m^2 when it was driven by a current of 20 mA/m^2 and at a bias voltage of 11 V. The EL color is blue with 1931 CIE color coordinates of $x = 0.153$; $y = 0.125$. From the device configuration, it is concluded that the blue-emitting Al(PBI)$_3$ is also electron transporting.

In a similar device, using Be(PBI)$_2$ in place of Al(PBI)$_3$, the light output was 504 cd/m^2 when it was driven by a current source of 20 mA/m^2 and at a bias voltage of 8.7 V. The blue EL color according to 1931 CIE color coordinates is $x = 0.154$; $y = 0.151$.

4. Stability

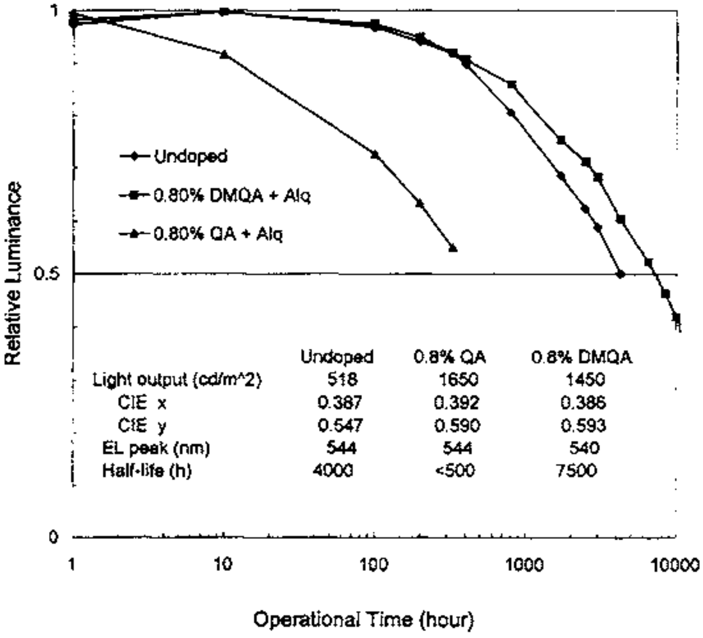
Practical applications using organic EL devices are only feasible if the devices are sufficiently long-lived under normal operating conditions. However, most organic

EL devices reported so far have a rather short operational lifetime (with some reports which did not even mention stability at all), ranging from a few hours to several hundred hours. This instability has been attributed to the deterioration of the organic [37] as well as the electrode layers [38] and is highly dependent on the device configuration [39].

For "undoped" cells, highly stable organic electroluminescence devices based on vapor-deposited AlQ_3 thin films have been reported recently [7]. The improvement in stability is derived from several factors including: (1) a multilayer thin-film structure with a CuPc stabilized hole-injection contact; (2) a NPB hole-transport layer; and (3) an a.c. drive waveform. These emissive devices have shown an operational half-life of about 4000 h from an initial luminance of about 510 cd/m^2.

This stability can be improved further with the judicious selection of dopants in the host AlQ_3 emitting layer. Fig. 4 compares the stability and EL performance of three green AlQ_3 devices driven by a constant current of 20 mA/cm^2, using an undoped and two doped emitters. For the quinacridone (QA)-doped AlQ_3, the initial luminance (1600 cd/m^2) is almost tripled compared to that of the undoped device because of the enhanced luminance efficiency. The stability is, however, rather poor, providing a $T_{1/2}$ of only about 300–400 h. With *N*,*N*-dimethylquinacridone (DMQA) as dopant, an EL device with superior stability and high luminance has been achieved. The $T_{1/2}$ is more than 7000 h with an initial luminance of 1400 cd/m^2.

These examples show that the EL stability can be profoundly affected by the particular choice of dopant, among other factors. The $T_{1/2}$ value is dependent on



	Undoped	0.8% QA	0.8% DMQA
Light output (cd/m^2)	518	1650	1450
CIE x	0.387	0.392	0.386
CIE y	0.547	0.590	0.593
EL peak (nm)	544	544	540
Half-life (h)	4000	<500	7500

Fig. 4. Stability and EL performance of doped AlQ_3 EL devices.

the luminance level produced by the EL device, and a longer $T_{1/2}$ is obtainable for operation at a lower luminance. Thus, a half-life in excess of 50 000 h can be reasonably projected for the DMQA-doped AlQ_3 device operating at an initial luminance of about 100 cd/m^2 [10].

With these lifetimes, applications ranging from directly modulated fast-switching light sources for optical imaging to large-area, full-color, flat panel displays can be envisioned. Whereas much more work remains to be done, particularly in further improving the EL stability, we are confident that organic electroluminescence will prove to be an important technology with a "bright" future.

5. Outlook

Tremendous progress has been made in organic electroluminescence, particularly in the last few years. We have witnessed an explosion of R&D efforts coming from both industrial and academic laboratories. As this review has shown, prototypical materials such as the ubiquitous AlQ_3/dopant have achieved luminous efficiency well in excess of several lm/W, and EL operation at 100 cd/m^2, with half-life greater than 10 000 h has been demonstrated.

Further improvement in the organic EL device performance can be anticipated as more materials are being designed and synthesized by chemists specifically for EL applications. The molecular design criteria for high fluorescence efficiency and glass transition temperatures are well understood. The classic rules of rigid and sterically hindered molecular structures are generally applicable to the design of organic EL materials. Less clear, however, is the dependence of the carrier transport characteristics on the molecular structure. As the carrier transport is related to the magnitude of the injection current through the EL device (the light output), it is important to be able to design EL materials with superior hole or electron mobilities. This task is challenging, considering that there are additional constraints that other EL material parameters, such as fluorescence efficiency, cannot be degraded significantly in the process.

Although organic EL devices have achieved long operational stability, the material issues underlying the EL degradation are still poorly understood. Probable causes for degradation such as interfacial interactions between layers, electrochemical and excited-state reactions, and thin-film phase changes have been suggested. However, none have been conclusively proven to be the key degradation mechanism. More analytical effort would be needed in sorting out the degradation pathways. Particularly important is the identification of chemical species responsible for the device instability, as such analysis could provide insight into the design of new and improved EL materials.

Commercial interest in organic EL technology has been the main driving force in fueling the recent research and development activities. Anticipated applications are mainly in flat panel displays, including backlights for LCD, alphanumeric displays, and dot-matrix panels for both low and high information content. Already, high-resolution monochrome green dot-matrix displays with 256 × 64 pixels by Pioneer

Electronic Corp. [40] and an RGB [75 × (7 × 3)] multicolor prototype EL display by Idemitsu Kosan Co. have been demonstrated [41]. [The fabrication of a QVGA (320 × 240) monocolor display has just been announced by Idemitsu Kosan Co., at the recent SID 97 meeting in Boston, MA, 15 May 1997.] Also, TDK Corp. has recently announced plans to develop an advanced active matrix display.

In comparison with other more mature display technologies such as LCD, organic EL is still in its adolescent stage. However, its promise of low power consumption and excellent emissive quality with a wide viewing angle is unique among display technologies. These advantages will no doubt be successfully exploited and commercial use for organic EL devices will be realized in the near future.

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