

Organic electroluminescent devices with Langmuir–Blodgett films of an amphiphilic complex with an 8-hydroxyquinoline as an emitter

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Electroluminescent (EL) devices with a single layer structure [indium-tin-oxide/emitting layer (Langmuir–Blodgett films)/Al] are synthesised using an intensely fluorescent amphiphilic complex, bis(*N*-hexadecyl-8-hydroxyquinoline-2-carboxamide)lanthanum [La(HQ)₂(H₂O)₄Cl] as the emitter material; green–yellow electroluminescence is observed with a low drive voltage (9 V).

Organic electroluminescent (EL) devices have recently received much attention because they can potentially produce emissions of all colours from blue to red in accordance with the wide selection of organic emitting materials, and because of their possible applications as large-area light-emitting displays,^{1–7} which are difficult to achieve using inorganic light-emitting cells.

Thin aromatic hydrocarbon films that fluoresce could have practical significance as large area electroluminescent panels if ultra-thin high quality aromatic hydrocarbons films can be constructed.^{8,9} Tang *et al.*^{1,2} found that the 8-hydroxyquinoline-aluminium (Alq₃) complex can be used as an emitting element in EL devices. As an emitting material, Alq₃ has excellent properties such as highly stable film formation, high carrier transport and good heat resistance. In the EL diode Tang *et al.* designed, high external quantum efficiency, luminous efficiency and brightness are achievable at a driving voltage below 10 V. Hamada *et al.*⁶ have developed other organic EL devices with 8-hydroxyquinoline–metal complexes, such as Znq₂, Beq₂, Mgq₂, Zn(mq)₂, Be(mq)₂ and Al(prq)₃ (mq and prq are 2-methyl-8-hydroxyquinoline and 7-propyl-8-hydroxyquinoline, respectively), as the emitter. The emissions of these devices are green or blue and they have a luminance of more than 3000 cd m⁻². For example, the EL device with Znq₂ has a luminance of 16 200 cd m⁻², higher than the luminance of Alq₃ (15 800 cd m⁻²). There is no report of Langmuir–Blodgett (LB) films of 8-hydroxyquinoline complexes being used as an emitting layer. LB film techniques make it possible to prepare organic functional ultra-thin films with a controlled thickness at a molecular size and well-defined molecular orientation,^{10,11} and the long chain 8-hydroxyquinoline complexes have strong fluorescence at room temperature.^{12,13} Therefore, if these amphiphilic 8-hydroxyquinoline derivatives complexes can be incorporated in LB films, these films will be of higher quality than the general vacuum deposited films and may have potential applications in molecular electronic devices.¹²

The synthesis of the amphiphilic ligand, *N*-hexadecyl-8-hydroxyquinoline-2-carboxamide (HHQ) and its lanthanum complex, La(HQ)₂(H₂O)₄Cl, which has high fluorescence efficiency,¹² were carried out by a previously reported method.^{12,14} The molecular structure of La(HQ)₂(H₂O)₄Cl is shown in Fig. 1. The EL device has a single layer structure: ITO/emitting layer (9- or 19-layer LB films)/Al, as shown in Fig. 1. The substrate is an indium-tin-oxide (ITO) coated glass with a sheet resistance of about 20 Ω/□, and it was cleaned by the known standard procedure.¹⁵ The LB films of the La(HQ)₂(H₂O)₄Cl amphiphilic complex (the emitting layer) were

prepared by a conventional LB technique. The amphiphilic complex was spread from a chloroform solution on a subphase of Millipore-filtered water. The transfer was performed at 0.025 N m⁻¹ with a dipping speed of 3 mm min⁻¹. After the various LB films were made aluminium to a thickness of 300 nm was deposited at 10⁻⁵ Torr onto the LB film surface as the top electrode with a deposition rate of 1.0 nm s⁻¹. The emitting area was 2 × 3 mm². The luminance of the EL devices was measured with a luminance meter. All measurements were carried out at room temperature in air under DC bias conditions.

Fig. 2 shows the relationship of current density and voltage of the EL devices with 9- and 19-layer LB films. The shapes of the curves are strongly dependent on the thickness of the LB films of La(HQ)₂(H₂O)₄Cl (luminance layer). The driving voltage increases with increasing LB film layer. The driving voltage for

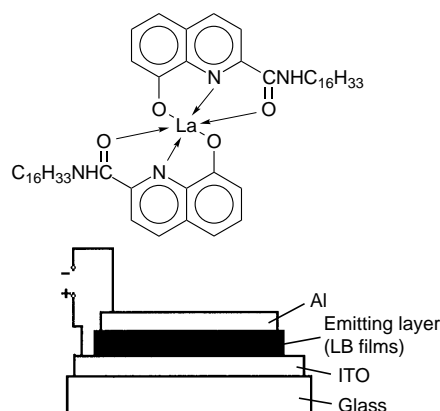


Fig. 1 Configuration of the EL cells and the molecular structure of the material used in this study

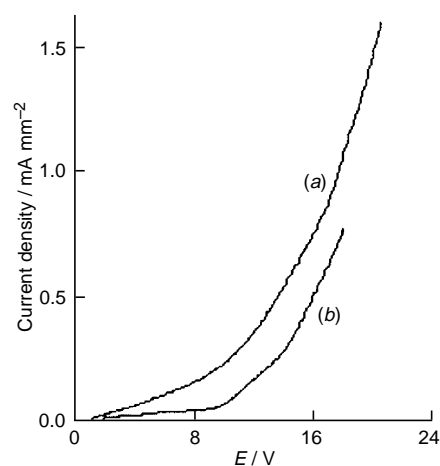


Fig. 2 The relationship of current density and voltage of the EL cell with the emitter of the La(HQ)₂(H₂O)₄Cl LB films: (a) 9-layer, (b) 19-layer

the cell with 19-layer LB films was 9 V. At a given voltage, the current density for the EL cell with a thinner LB film emitter is higher than that for the thicker one. The curves can be fitted to an injection-limited model where the electron current is limited by electron emission from the cathode into the $\text{La}(\text{HQ})_2(\text{H}_2\text{O})_4\text{Cl}$ LB film layer.

The luminance-current density characteristics of the EL cell with the emitter of the 19-layer $\text{La}(\text{HQ})_2(\text{H}_2\text{O})_4\text{Cl}$ LB films are shown in Fig. 3. Green-yellow emission with a luminance of about 330 pw was achieved which was enhanced with the injection current elevation in the region of 0.1–1.00 mA mm^{-2} .

Comparison of the EL emission spectrum of the cell with the PL spectrum of 19-layer $\text{La}(\text{HQ})_2(\text{H}_2\text{O})_4\text{Cl}$ LB films¹² showed

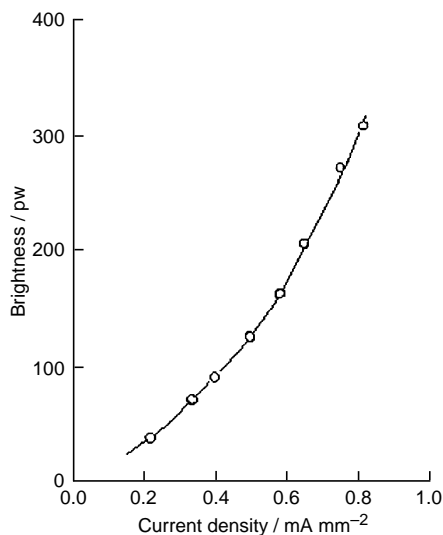


Fig. 3 The relationship of brightness and current density of the EL cell with the emitter of the 19-layer $\text{La}(\text{HQ})_2(\text{H}_2\text{O})_4\text{Cl}$ LB film

that the spectrum does not change greatly. The PL and EL peak wavelengths and the half values of these two spectra are 520, 515 and 170, 200 nm, respectively. The EL emission spectrum is independent of the driving voltage and current. This result indicated that the radiative recombination of injected electrons and holes takes place in the $\text{La}(\text{HQ})_2(\text{H}_2\text{O})_4\text{Cl}$ LB films.¹

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