Extended Abstracts of the 2001 International Conference on Solid State Devices and Materials, Tokyo, 2001, pp. 636–637

F-8-2

Electroluminescent Properties of OLEDs with a Rubrene Sub-Monolayer Inserted between Electron-and Hole-Transport Layers

Michio Matsumura and Takumi Furukawa

Research Center for Solar Energy Chemistry, Osaka University
1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan
Phone:+81-6-6850-6695 Fax:+81-6-6850-6699 E-mail: matsu@chem.es.osaka-u.ac.jp

1. Introduction

Coumarin, dimethylquinacridone, rubrene, etc. have been utilized as the dopants of organic EL devices. When these dopants are added to host materials at about a few percent, they emit light efficiently. Coumarin, quinacridone, and the derivatives are considered to accept the excitation energy form the host materials. On the other hand, rubrene is considered to directly recombine electrons and holes on the molecule [1]. Because of this property of rubrene as the dopant, it shows unique electroluminescent properties [1, 2].

Here, we report that (1) rubrene shows efficient electroluminescence, when its sub-monolayer is inserted between Al-hydroxyquinoline complex (ALQ) and diamine layers, (2) the current-voltage curve of this device shifts toward lower voltage than that of a device without the rubrene layer, and (3) the efficiency of this device is high even at low current densities, in contrast to the device without the rubrene layer.

2. Experimental

Devices were fabricated using ALQ and N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine (TPD), which were purchased from Dojindo Laboratories and Japan Joryu Co., respectively. Rubrene and quinacridone were obtained from Sigma-Aldrich Fine Chemicals and Ciba Specialty Chemicals, respectively. ITO-coated glass plates were purchased from Japan Sheet Glass Co., and were washed with acetone, followed by treatment with oxygen plasma. Typical EL devices without the rubrene or quinacridone layer had the structure of glass/ITO/TPD(60 nm)/ALQ(60 nm)/cathode(200 nm). We used a codeposited Mg:Ag (10:1) layer as the cathode. All the organic layers were deposited successively in the same chamber without breaking the vacuum.

3. Results and Discussion

Figure 1 shows typical EL spectra obtained from the devices which have a 0.05-nm-thick rubrene layer placed in different regions. The devices have the rubrene layer at the ALQ/TPD interface (Type A), in the ALQ layer at 5 nm from the interface (Type B), and in the TPD layer at 5 nm from the interface (Type C). The spectra of the Type A and Type B devices are mostly due to the fluorescence of rubrene, while

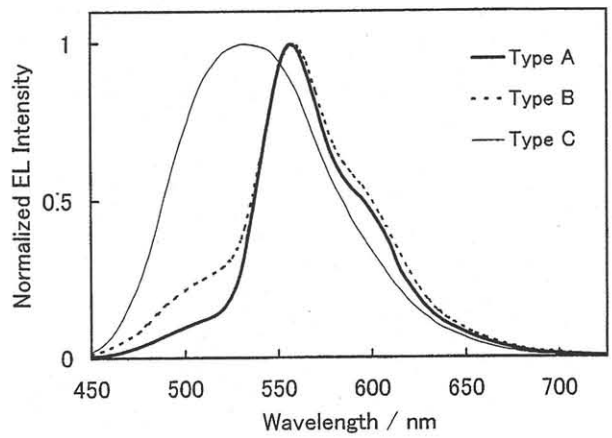

Fig. 1 EL spectra from devices with a 0.05 nm-thick rubrene layer placed at different positions.

the spectrum of the Type C device is due to the fluorescence of ALQ. The absence of the emission from rubrene in the Type C device is understood by considering the electron-blocking property of the ALQ/TPD interface. The EL efficiency is the highest for the Type A device, and the efficiency reaches its maximum when the average thickness of the rubrene layer is about 0.05 nm. The efficiency of this device reaches as high as 5.8 cd A^{-1}, which is higher than that of the device without the rubrene layer (3.2 cd A^{-1}). The

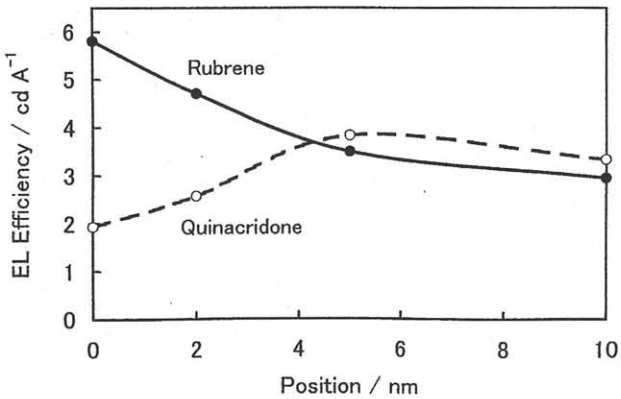

Fig. 2 EL efficiency vs position of rubrene (solid line) and quinacridone (broken line) layers inserted in the ALQ layer.

efficiency of the device is high even if compared with the efficiency of a device with a rubrene-doped TPD layer, i.e., ITO/TPD(40 nm)/rubrene(5 wt%)-doped TPD(20 m)/ALQ(60 nm)/cathode, which has an efficiency of 4.5 cd A^{-1}.

Figure 2 shows the dependence of the EL efficiency (cd/A) on the position of the 0.05-nm-thick rubrene layer inserted in the ALQ layer. As the rubrene layer moves into the bulk of the ALQ layer, the efficiency decreases. The lowering of the efficiency is accompanied by a decrease of emission from rubrene and an increase of emission from ALQ. When the rubrene layer was placed in the TPD layer at a distance greater than 5 nm from the ALQ/TPD interface, the emission was due to ALQ, as shown in Fig. 1, and the EL efficiency was almost the same as that of the devices without a rubrene layer, i.e., 3.2 cd A^{-1}.

The broken line in Figure 2 shows the EL efficiency for devices with a 0.05 nm-thick quinacridone layer inserted in the ALQ layer. Quinacridone accepts excitation energy from ALQ by the Förster mechanism, and emits strong EL and PL when doped in the ALQ layer [3]. However, when a 0.05-nm thick quinacridone layer was placed at the ALQ/TPD interface, the emission was from both quinacridone and ALQ, and the efficiency was low. As the position of the quinacridone layer moved into the bulk of the ALQ layer to about 5 nm from the interface, we observed strong emission from quinacridone and the efficiency increased. The different tendencies observed between the devices with a rubrene layer and those with a quinacridone layer are attributable to the different functions of these dopants, as discussed in the Introduction section.

The current-voltage curve of the device with a 0.05 nm-thick rubrene layer at the ALQ/TPD interface shows the onset of the current lower than that of the device without the rubrene layer by about 2 V. This is probably due to the effective hole trapping by the rubrene molecules. The holes accumulated at the interface enhances the electric field over the ALQ layer, which assists the electron injection process from the cathode [4].

The EL efficiency (cd/A) for the device with a rubrene layer is high even at low current densities, as shown by the solid line in Fig. 3. This is in contrast to the property of the device without the rubrene layer (see the broken line). In the case of the device without a rubrene layer, holes are injected into the ALQ layer *via* the TPD layer, and the holes recombine in the ALQ layer. At low voltages the recombination region extends deep into the ALQ layer [5]. If the recombination takes place in the deep region of the ALQ layer, the efficiency is low, because the thickness of the ALQ layer (about 6o nm) is designed so that the efficiency is high for the recombination near the ALQ/TPD interface. On the other hand, in the case of the device with a rubrene layer placed at the ALQ/TPD interface, rubrene molecules rap the

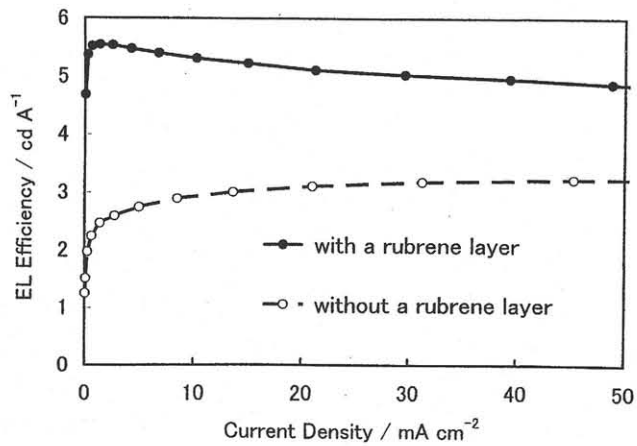

Fig. 3 EL efficiency vs current density for devices with and without a 0.05 nm-thick rubrene layer at the ALQ/TPD interface.

holes, and the recombination occurs on the rubrene molecules. In other words, the recombination takes place at the position as designed, and the efficiency is high even at low voltages or at low current densities.

4. Conclusion

All the properties observed for the device with a rubrene sub-monolayer are favorable for fabrication of efficient OLEDs.

References

[1] H. Murata, C. D. Merritt and Z. H. Kafafi: IEEE J. Sel. Top. Quantum Electron., **4**, 199 (1998).

[2] M. Matsumura and T. Furukawa, Jpn. J. Appl. Phys., to be published.

[3] S. Kundu, K. Fujihara, T. Okada and M. Matsumura: Jpn. J. Appl. Phys., **39**, 5297 (2000).

[4] M. Matsumura, A. Ito and Y. Miyamae: Appl. Phys. Lett., **75**, 1042 (1999).

[5] M. Matsumura and Y. Jinde, IEEE Trans. Elec. Dev., **44**, 1229 (1997).