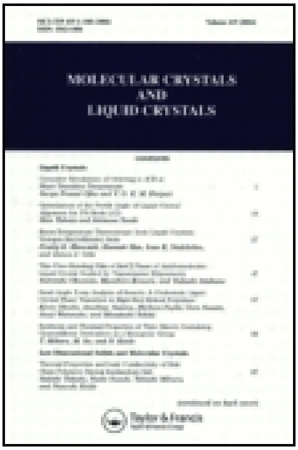

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Organic Electroluminescent Devices Fabricated with a Co-Deposited Hole Transport Layer

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ORGANIC ELECTROLUMINESCENT DEVICES FABRICATED WITH A CO-DEPOSITED HOLE TRANSPORT LAYER

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Abstract To improve the stability and durability of organic EL devices, the mixed HTL layer was fabricated by co-deposition of TPD and Alq_3 and the EL cell with the mixed HTL was compared with that with the conventional pure HTL. When more than 5 wt% of Alq_3 was added in HTL, the increase in surface roughness of ITO / TPD-Alq_3 / Alq_3 by annealing was found by AFM to be reduced dramatically. In addition, the running durability was significantly improved by addition of a small amount of Alq_3 (< 10 wt %), although the EL characteristics of the cells with mixed HTLs were slightly lowered, i.e. I-V and L-V curves shifted to the higher voltages.

INTRODUCTION

Recently, organic electroluminescent (EL) devices have been studied extensively due to the potential for commercial application such as low cost, full color, flat, and flexible display. The recent researches of EL devices which consist of organic molecular or polymeric multilayers focused on reliability and durability for practical use.[1-6] However, the limited lifetime remains still as the main obstacle for practical use. In previous papers,[7-16] we have also studied in detail the degradation mechanisms of EL devices with structure of ITO / TPD / Alq_3 / Al during their storage before EL operation and under the EL operation.

In this paper, we will report enhancement of durability of co-deposited HTLs due to thermal stability of the mixed films. We will also report the durability of the EL devices fabricated with a mixed HTL in comparison with that of conventional EL devices.

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EXPERIMENTAL

We prepared EL cells of typical two organic layers structure, i.e. ITO / HTL / ETL / Al, by using N,N'-diphenyl-N,N'-bis(3-methyl phenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD) and tris(8-hydroxyquinoline) aluminum (Alq_3) as HTL and ETL, respectively. The two-layered organic films for surface roughness measurements and for EL cells were fabricated by vapor-deposition on ITO electrodes. For mixed HTLs by co-deposition, the different deposition rates from the separate boats were monitored by a quartz crystal oscillator placed near the ITO substrate with an ULVAC CRTM 8000 controller. The thickness of all HTL, ETL, and Al was ca. 50 nm. The EL characteristics were measured by an Advantest 6145 electrometer with a Topcon BM-3 luminance meter. The surface roughness, R_a, was determined by a SEIKO SPA-300 AFM unit with a SPI-3700 probe station. Olympus Si_3N_4 triangular AFM cantilevers with a spring constant of 0.089 N m^{-1} were used.

RESULTS AND DISCUSSION

In general, instability of organic layers is one of the reason for degradation in organic EL devices. In order to confirm the instability of organic layers in the EL devices caused by Joule heating, the morphological change of organic bilayers was studied by measuring R_a, with AFM. Figure 1(a) shows the change in R_a with the annealing time at 80 °C in air on a bilayer with a pure HTL. In Fig. 1(b) are shown R_as of bilayers with mixed HTLs of various wt% of Alq_3 measured after 1 h annealing at 80°C in air. R_a of the surfaces with the pure TPD layer rapidly increased with an increase in the annealing time (Fig. 1(a)). The results are consistent with the previous observation.[11,12] In contrast, on the mixed HTLs, R_a rapidly decreased with an increase in the content of Alq_3 within 10 wt% at the same annealing conditions of 80°C in air for 1h indicating that the thermal stability of organic layers was drastically increased by adding more than 10 wt% of Alq_3 in the HTL.

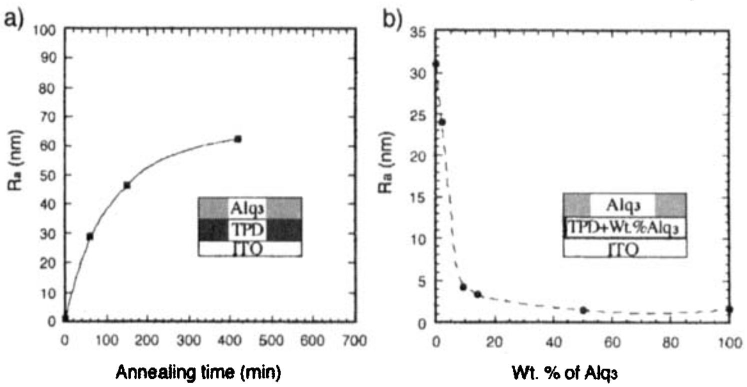

FIGURE 1 (a) Roughness (R_a) change of organic bilayers on the ITO base electrode, ITO / TPD / Alq_3, as a function of annealing time at 80 °C in air. (b) Change in roughness after 1h annealing at 80 °C in air as a function of Alq_3 concentration in mixed HTL of ITO / TPD-Alq_3 / Alq_3.

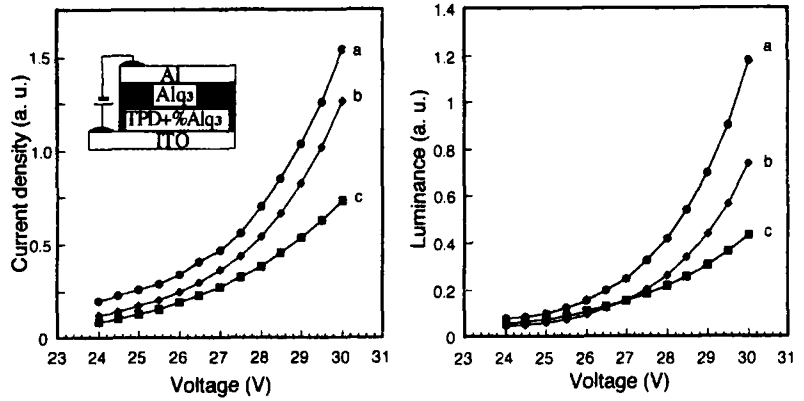

FIGURE 2 Current-voltage (I-V) and Luminance-voltage (L-V) characteristics for ITO / TPD-Alq_3/ Alq_3 / Al devices fabricated with various concentration of Alq_3 in the mixed HTL. Wt% of Alq_3 : a) 0, b) 1.6, and c) 6.2.

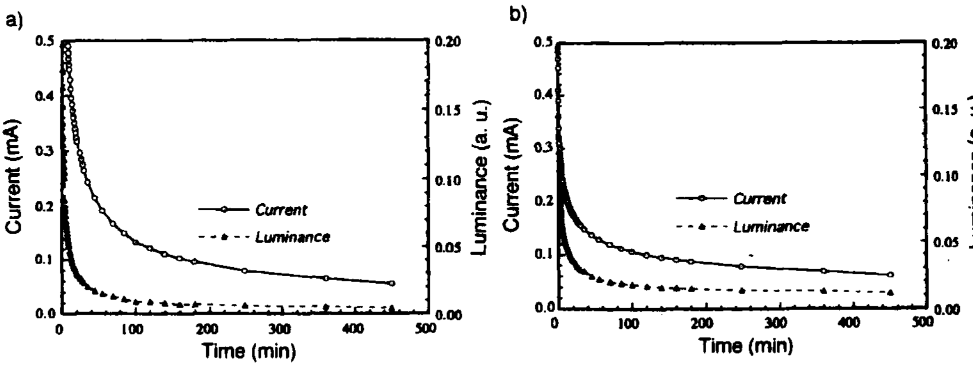

FIGURE 3 Effect of mixing of Alq_3 in the HTL on current vs. time and luminance vs time curves of the EL devices. (a) EL cell with a pure HTL, (b) EL cell with a mixed HTL containing 5 wt% of Alq_3.

To provide the effect of mixing of Alq_3 with TPD on EL characteristics, the current-voltage (I-V) and luminance-voltage (L-V) curves were measured for EL cells with a mixed HTL. Figures. 2(a) and 2(b) show I-V and L-V curves, respectively, for the cells with various concentrations of Alq_3 in the HTL. The dependence of the L-V curves on the Alq_3 concentration shown in Fig. 2(b) was similar to that of the I-V characteristics shown in Fig. 2(a). This similarity was confirmed by measuring luminance efficiency at a constant applied voltage of 20 V as a function of Alq_3 concentration in the HTL. The almost constant luminance efficiency of ca. 0.34 lm W^{-1} was observed independently of Alq_3 concentration, indicating that the EL intensity was proportional to the EL current. The the lower efficiency observed here than that reported by Tang and VanSlyke[1] may be attributable to the use of the Al cathode than the Mg-Ag cathode. As shown in Figs. 2(a) and 2(b), the I-V and the L-V curves were shifted to the higher applied voltage with an increase in the concentration of Alq_3. The shift can be explained by the increase in the barrier height for the hole injection from ITO to the HTL,[17] because ionization potential of added Alq_3 is higher than that of TPD.[18]

To compare the durability between the EL cells with the conventional pure or the mixed HTL, we investigated the current vs. time (I-t) and the luminance vs. time (L-t) curves were recorded under a constant applied voltage. Typical results observed under an applied voltage of 20 V for the EL cells with the pure TPD HTL (type A) and with the mixed HTL containing 5 wt% Alq_3 (type B) are shown in Figs. 3(a) and 3(b), respectively. It is clear from comparison between Figs. 3(a) and 3(b) that initially both the current and luminance was higher for type A than for type B, but after 20 min the luminance of type B was higher than that of type A and after 200 min even the current of type B became higher than that of type A. In other words, durability of the type B cell is higher than the type A cell. Optimization of the concentration of Alq_3 for durability is now under investigation.

CONCLUSIONS

The EL characteristics were changed by mixing Alq_3 in the TPD HTL. The EL cell with 5 wt% of Alq_3 in the mixed HTL showed longer lifetime than the conventional device.

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