Hole Injection Layer by Ion Beam Assisted Deposition for Organic Electroluminescence Devices

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Abstract

The ultra thin hole injection layer (HIL) was deposited on an indium-tin-oxide (ITO) anode by using an ion beam assisted deposition (IBAD) for the fabrication of an polymeric electroluminescence device for the first time. The device with the HIL deposited by IBAD has higher external quantum efficiency than the device with the HIL by conventional thermal evaporation. It is found that the deposited HIL by IBAD has high surface coverage on ITO anode in a few nm regions because the HIL prepared has high adatom mobility by ion beam energy.

1. Objectives and Background

During the last years organic light emitting diodes (OLEDs) have become the focus of intense research due to their possible applications in flat panel displays. The device efficiency of OLEDs have been improved by the modification their structure and the development of new materials.[1)] Many reports have shown to improve the device efficiency of OLEDs by using low work function metals for cathode, or proper hole (or electron) injection configurations.[2-5)]

For effective hole injection, indium-tin oxide (ITO) is used as anode due to its good transparency, low resistivity, and relatively high work function. In contrast, the surface properties of the ITO directly affect the characteristics of the device such as shorting and low device efficiency. Recently, it has been shown that the insertion of a ultra thin insulating interlayer of SiO_2,[6)] CuO_x[7)], and LiF layers[8)] between the ITO electrode and the hole transport layer (HTL) OLED structures lowers the turn-on voltage and increases the efficiency compared to typical devices without the insulating layer.

The hole injection layer (HIL) has been prepared by conventional thermal evaporation. The resulting layer has the surface morphology of island shape because of the low deposition energy of the evaporated elements.[9)] Therefore, the surface of the HIL is discontinuous below a few nanometers in thickness and it has low effective contact area as it is covered by the organic HTL. The hole injection mechanism of the insulating layer is explained by tunneling effect in general but also the surface dipole effect must be considered as far as the insulating layer fully covers on an indium-tin-oxide (ITO) anode,[10)] because the layer of a few angstrom thick could exactly not be explained by tunneling theory. The ion beam assisted deposition (IBAD) processes can control the film growth formation by various energetic ion of deposited material. The mechanism of IBAD has been reviewed in the literature.[11)] The device with continuous HIL layer prepared by IBAD has higher current density and better device efficiency than the layer deposited by thermal evaporation from above reasons.

2. Results and discussion

Figure 1 and 2 show the relation between current density vs. voltage (*I-V*) and brightness vs. voltage (*L-V*), respectively. The current density of the device not having the LiF hole injection layer was about 47.7 mA/cm^2 and its brightness was about 6000 cd/m^2 at 8 V. In contrast, at the same voltage, the current density and brightness of the LiF-deposited device obtained from thermal evaporation were about 58.6 mA/cm^2 and 7620 cd/m^2, respectively. And those of the corresponding device prepared by the IBAD technique were 71.7 mA/m^2 and 10908 cd/m^2, respectively. It was found that *I-V* and *L-V* curves are shifted to the lower level of operating voltage by an inserted LiF layer. The device including the LiF hole injection layer by IBAD showed a high current density and a high brightness at the given voltage.

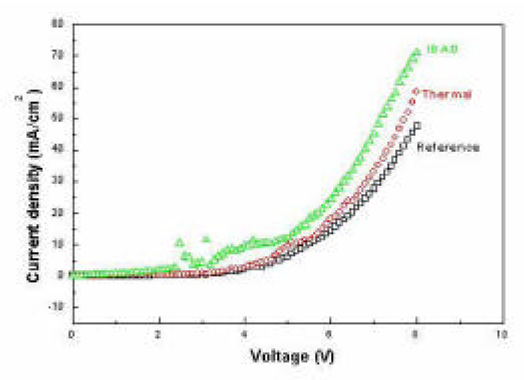

Figure 1. The current density and voltage characteristics of the devices without LiF layer, with LiF layer by thermal evaporation and IBAD.

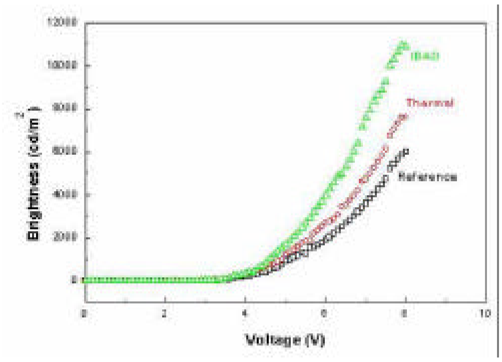

Figure 2. The Brightness and voltage characteristics of the devices without LiF layer, with LiF layer by thermal evaporation and IBAD.

Fig. 3 is a graph to compare the external quantum efficiency of various devices (EQE) prepared by different processes. The EQE of the devices not having a LiF layer and including the LiF layer prepared by thermal evaporation and IBAD were 1.1, 1.2, and 1.4 %, respectively under a given current density of 30 mA/cm^2. It was found that the EQE of the device with LiF by IBAD also has higher than that of the case of the device with LiF by conventional thermal evaporation.

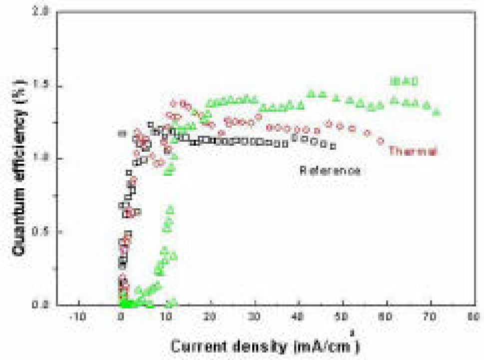

Figure 3. External quantum efficiency versus voltage curves of the devices without LiF layer, with LiF layer by thermal evaporation and IBAD.

It has been known that the presence of an insulating LiF layer between ITO and HTL enhanced hole injection by tunneling effect. Zhu et al$^{8)}$ reported that LiF deposited on an ITO surface by thermal evaporation is most likely formed by coalescence of islands from nucleation site and the thickness of LiF layer entirely covered on ITO is a few nanometers. In this region, the island formed LiF layer on ITO surface diminishes the effective contact area for carrier injection. In contrast, the ultra thin LiF layer by IBAD on the ITO substrate has more uniform surface than the layer by thermal evaporation because the deposition element of adatom mobility is enhanced by energetic ion during IBAD process. Therefore, the device with LiF by IBAD has higher quantum efficiency than the device with LiF by conventional thermal evaporation. In order to better understand the role of by IBAD, we investigated the work functions of various LiF thicknesses on an ITO surface and the surface coverage of LiF layer by each deposition method.

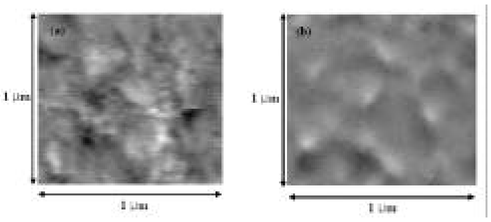

Figure 4. The AFM images of LiF (1.5 nm) on ITO by thermal evaporation and IBAD

Table 1 shows the relation between LiF layer thicknesses versus effective work function. This work was done to investigate the electrical characteristics of the LiF layer prepared by thermal evaporation and IBAD. The values of work function were measured by using a surface analyzer AC-2 manufactured by RIKEN KEIKI Co., Ltd. The measurement of effective work function by this technique has been proved to be adequate for the present ITO substrate. The LiF thicknesses adopted for the present test is 0, 1.0, 1.5, 3.0, and 5.0 nm, respectively. The work function of the bare ITO is about 4.8 eV, which is close to that given in the literature. In the case of thermal evaporation, the work function increases as a LiF thickness increases from 0 nm to 5.0 nm and then, saturates. The work function of the LiF layer deposited by IBAD sharply increase until 1.5 nm and then, the value is almost saturated after 1.5 nm thickness. Therefore, in the case of IBAD, the unsaturated value such as under 1.5 nm thick region maybe due to the partially covered LiF on the substrate, while the saturated value such as upper 1.5 nm region results from the full coverage of LiF on it. Fig. 4 (a) and (b) show the atomic force microscopy (AFM) images obtained over an area 1 µm × 1 µm for the surface of 1.5 nm thick LiF layer on ITO by thermal and IBAD, respectively. The deposited LiF layer by thermal shows the partially covered ITO surface. In contrast, the LiF by IBAD has a fully covered ITO surface and smooth surface morphology at 1.5 nm thickness region. It is indicated that the deposition element of adatom mobility is enhanced by energetic ion during IBAD process, moreover, the homogeneously deposited hole injection layer by IBAD has a uniform electrical field distribution between inorganic ITO anode and organic HTL. The insulating LiF layer was also introduced for charge injection by tunneling effect moreover. Here, note that tunneling probability is related to insulating layer thickness. The device efficiency is affected by the thickness of insulating layer. That is, increase in layer thickness reduces tunneling probability above complete coverage region, while hole injection is influenced by dipole moment below incomplete coverage region because the insulating LiF layer has a high dipole moment. The work function of the ITO anode increases with increasing coverage of LiF in its submono layer region because the charge of LiF can be dipole-oriented towards the surface to increase work function. Moreover, the existence of the interfacial dipole layer allows energy level alignment various with the modification of the interface. However, as the surface is entirely covered by the insulating layer, the effect of the surface dipole diminishes and its tunneling effect become dominate for hole injection since the surface of ITO is entirely covered with insulating layer. Upper this region, it is important that the tunneling is affected the hole injection. From these reasons, the LiF hole injection layer by IBAD has high spontaneous dipole and high tunneling probability because the continuous insulating layer by high adatom mobility has thinner thickness than that of LiF by conventional thermal evaporation.

3. Conclusion

In summary, the ultra thin LiF layer was deposited by the IBAD technique to make a hole injection layer between HTL and ITO. Such as continuous hole injection layer had high current density and brightness at a given thickness. The device efficiency of the corresponding LiF layer was higher than that of the LiF layer by conventional thermal evaporation.

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