Journal of the Korean Vacuum Science & Technology; JKVST
Vol.6, No.1, 2002, pp.51~54

The Stability and Indium Diffusion from ITO to PPV Layer of Polymer Light Emitting Devices with/without PI Blocking Layer

Seongjin Cho, Dongkyu Park, Taewoo Kwon, Dongsun Yoo*, Ilgon Kim*

Department of Natural Science, Kyungsung University, Pusan 608-736, Korea
**Department of Physics, Changwon national University, Changwon, 641-773, Korea*
(Received)

Abstract

Polymer EL devices of glass/ITO/PI/MEH-PPV/Al structure were fabricated using spin coating and the Ionized Cluster Beam deposition technique. PMDA-ODA type thin polyimide films which can be used as a impurity blocking layer of EL device were deposited by ICB. According to our previous results, the packing densities of polyimide films were subject to change and depend on their deposition condition. By inserting a PI layer with various thickness and packing density, I-V characteristics and life time of the devices were investigated to determine the role of a interlayer. The blocking of impurity diffusion from ITO to luminescent layer were confirmed by XPS.

1. Introduction

Since the organic and polymeric electroluminescence devices solve the major drawbacks of current displays such as LCDs, PDPs and CRTs, the study on the EL devices has been considered as a hot research topic in recent years [1,2]. One of the major obstacles for the device is due to a poor device stability. Some important reasons that cause the inferiority of the device stability are known to be poor interface structure, unbalanced carrier injection and impurities diffused from both electrodes to the light emitting polymer layer [3,4]. Recently, it has been reported that by inserting an insulating layer between the anode and the light emitting layer, the stability and electroluminescence efficiency was significantly enhanced [5,6]. The enhancements are attributed to an improved balance between hole and electron injections and blocking of the diffusion of impurities such as In and Sb from ITO [4]. But the exact contribution of the PI buffer layer was not clearly undersyood. In this paper, we will evaluate the relationship between diffused impurities and device stability by direct measuring half luminosity life time of the devices with or withour PI buffer layers. As a impurity blocking layer of polymer electroluminescence device, PMDA-ODA type polyimide(PI) having various thickness and various packing densities were inserted between the ITO and MEH-PPV polymer light emitting layer using ICB deposition method. ICB deposition method is a very unique technique that uses accelerated ionized clusters for thin film formation enabling flexible control of the film properties. Accelerated energetic clusters collide with the substrate surface and break into small fragments, allowing the molecules to migrate to the surface of the substrate. A smooth interface, enhancements of nucleation, and shallow implantation effects can also be expected [7,8]. Due to these effects, films fabricated by ICB deposition have a high packing density, good adhesion, and low surface roughness. In this paper, we focused on

investigating the relationship between the impurity diffusion from ITO to light emitting layer and the device stability. The improved performance such as reducing the operating voltage, enhancing the thermal stability and quantum efficiency in the device were observed due to the existence of PI interlayer.

2. Experimental

PI films were formed on ITO substrate by ICB under 10^{-6} torr. The detailed description of the ICB system has been published elsewhere [9]. To retain stoichiometry, the crucible temperatures for forming PMDA and ODA clusters were held at 210 and 180℃, respectively. The as-deposited film on the substrate at 70℃ was a polyamic acid (PAA) film. Since the PAA is the precursor of PI, the PAA film being cured at 250℃ for 1 hour in vacuum transformed to PI. The operation pressure of the ICB system was maintained in the low 10^{-6} torr range during the film deposition. Bilayer EL devices were fabricated to investigate the performance of the ICB deposited PI insulating layer as shown in Fig.1. A precleaned 2.5 X 2.5 cm^2 ITO substrate with 20 Ω/sq was used for the device. A strip about 5mm wide of SiO_2 (150 nm) was deposited on the ITO to avoid short circuiting of the electrodes during device testing. MEH-PPV was spin-coated on the top of ICB deposited PI layer followed by deposition of Al cathode. The I-V-L characteristics of devices with different PI interlayer thickness were measured and compared to the devices without having a PI layer.

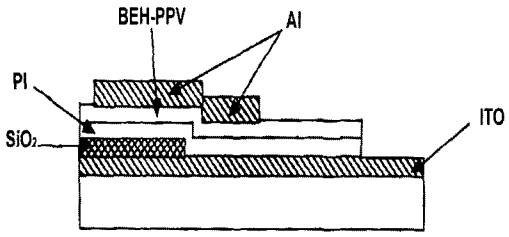

Fig. 1 Device structure with PI interlayer

And the stability of the devices were also examined by measuring half-luminosity time, i.e. the period of light output becomes to half. In order to measure the depth profile of the diffused impurity concentration in PPV layer, Ar sputtering ion gun was used and the X-ray photoelectron spectroscopy(XPS) were taken continuously during sputtering.

3. Results and Discussion

The chemical properties of cured films (PI at 250℃ for 1 hour in vacuum) and as-deposited films (PAA) were measured by FT-IR and XPS respectively. The details were reported elsewhere[10]. During curing process, anhydride carbonyl and amide peaks were considerably decreased, while the imide peaks increased remarkably, which indicates the progress of subsequent reaction and imidization. The I-V-L results for the devices are shown in Fig. 2. The results show that the efficiency and device performances vary with different PI interlayer thickness, but there is no considerable evidences on relationship between PI interlayer thickness and efficiency. According to our previous results on metal diffusion in ICB deposited PI which were

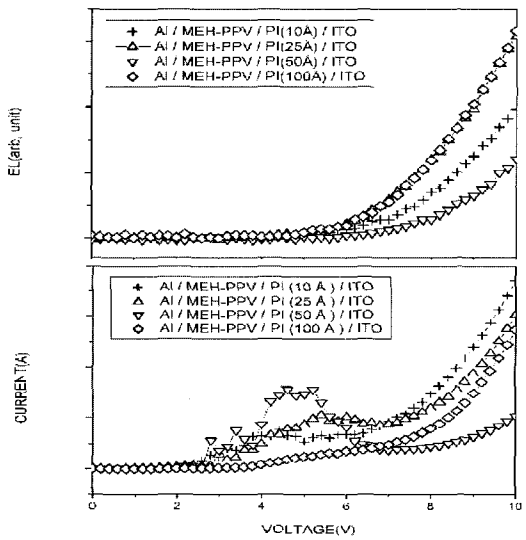

Fig. 2. I-V-L characteristics of the devices

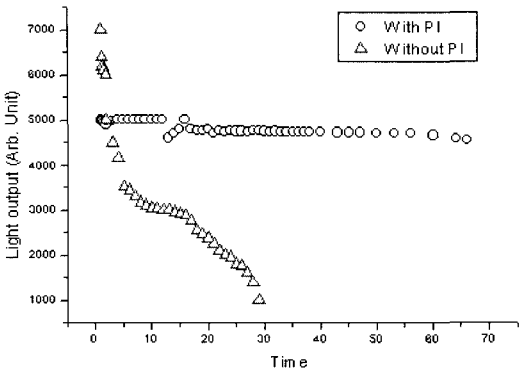

Fig. 3. Life Time of the device

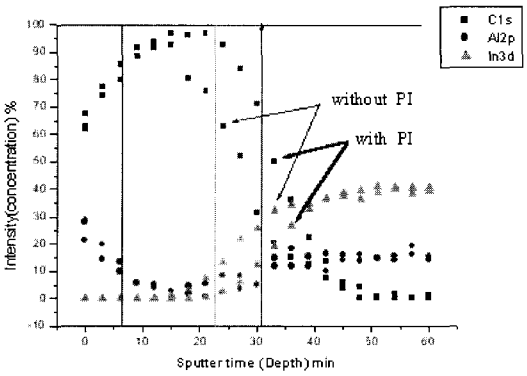

Fig. 4. Depth profile of the devices (XPS results).

investigated by XPS and Rutherford backscattering spectroscopy (RBS), the diffusivity of metal atoms into PI films strongly depended on the deposition conditions [8]. Fig. 3 shows the life time of the devices with or without PI buffer layer. The half-luminosity time for the device with no PI buffer layer drops very rapidly but that of the devices with buffer layer stands relatively long time. Even the half-life time were measured for various devices with PI buffer layer, the thickness and packing density variations does not seems to affect on life-time. But they affect device performances such as current density and device temperature during operation etc.. As the thickness of the PI interlayer increased, the resulting turn on voltage was increased due to enlargement of the potential drop within interlayer. The injected carrier density was decreased with the increasing interlayer thickness which resulted more balanced charge injection and provided more stable device performance within certain range of thickness. According to the XPS results as shown in Fig.4, the indium diffusion is strongly controlled by the presence of PI buffer layer and the indium diffusion stopped in PI buffer layer. This means that the ICB deposited PI film blocks interdiffusion of the quenching centers effectively. Therefore stability of the devices with ICB deposited PI interlayer were remarkably enhanced.

In conclusion, the device stability and efficiency were enhanced by inserting ICB deposited PI which acted as a hole blocking layer to balance charge injection and also acted as a buffer layer for choking diffusion of impurities into light emitting layer.

Acknowledgment

This work was supported by grant from the Basic Research Program of the Korea Science & Engineering Foundation (2000-2-11400-009-3) and in part by Research Grant(2000) of Kyungsung University.

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