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Characterizaton of tetracene-based electroluminescent devices produced by cluster beam deposition methods

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Abstract

We have fabricated and characterized heterojunction electroluminescent devices with the structure of ITO-coated glass/tetracene/(DCM-doped)Alq_3/Li:Al. The organic thin films of tetracene, Alq_3, and dye dopant DCM were deposited by applying the cluster beam deposition (CBD) methods. X-ray diffraction (XRD) and atomic force microscopy (AFM) measurements show that the flat and smooth thin tetracene films are close to a highly ordered, single crystal. Photoluminescence (PL), electroluminescence (EL) and device characteristics such as current density-voltage (*J–V*), EL intensity–voltage (*L–V*), and external quantum efficiency-current (*EQE–I*) were examined for undoped and DCM-doped devices.

Keywords: tetracene, Alq_3, DCM, cluster beam deposition, OLED

1. Introduction

Since the early EL reports based upon molecular anthracene crystals, optoelectronic devices utilizing polyaromatic hydrocarbon (PAH) molecules have long attracted attention in the development of organic light emitting diodes (OLEDs) and transistors [1-3]. The performance of some PAH thin-film transistors now stands in the state of competition with amorphous silicon transistors, which is believed to be due to the production of highly crystalline thin films. In general, PAH films with highly ordered structures have been produced by vacuum evaporation, chemical vapor deposition or pulsed laser deposition in very limited ranges of empirical deposition parameters. Another less common, but promising scheme is to apply the cluster beam deposition (CBD) method. The CBD method using weakly bound, highly directional cluster beam has been found to be efficient in depositing thin films with good surface morphology, crystallinity, and packing density at low substrate temperature [4]. Such characteristics cannot easily be attained by conventional deposition methods, and have been demonstrated in the recent investigations of pentacene-based transistors conducted by Choi and co-workers [5].

In this paper, we describe our recent fabrication and characterization studies of the double-layer-type heterojunction OLEDs using tetracene compounds. The molecular structures of the materials used and the schematic cross-sectional view of the EL device with the energy level diagram are shown in Figure 1. Tetracene, a kind of PAH species with four aligned benzene rings, has mainly been

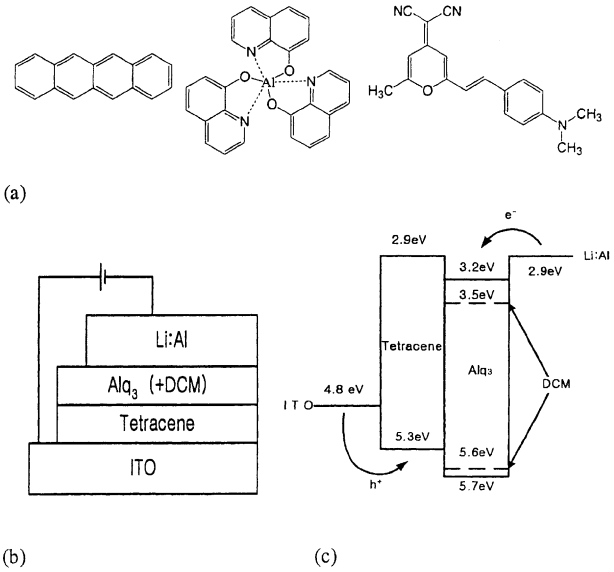

Fig. 1. Molecular structures of (a) tetracene, Alq_3, and DCM, (b) a schematic view of EL device and (c) a energy level diagram.

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used as either dopants in EL devices or conducting matrix in thin film transistors. Little information has, however, been derived on the luminescence and device performance for pure tetracene-based OLEDs. The EL devices with the structure of indium tin oxide (ITO)-coated glass/tetracene/Alq_3/Li:Al have been fabricated, in which all organic thin films were deposited by the CBD method. Tetracene and tris(8-hydroxyquinoline) aluminum (Alq_3) were employed as hole and electron transport layers, respectively. In addition, the highly fluorescent dye dopant 4-(dicyanomethylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyran (DCM) was also co-deposited in the Alq_3 layer, and the dopant effects on the device characteristics were investigated.

2. Experimental

Our home-made CBD system designed to prepare organic thin films has been described in detail elsewhere, and here only brief, relevant account is presented [4-9]. The CBD system consists of the evaporation crucible cells, the drift region, and the substrate. The chamber was pumped by a 10 in. baffled diffusion pump, and the average working pressure was maintained below $3x10^{-6}$ Torr. Organic thin films of tetracene, Alq_3, and dye dopant DCM were deposited by applying the CBD methods. The hole-transporting tetracene was first placed inside the crucible cell with a 1.0-mm diameter nozzle and was evaporated by the resistive heating at 525-545 K. Then the sublimed tetracene molecules underwent the adiabatic supersonic expansion into the drift region under high vacuum, resulting in the formation of highly directional, weakly bound cluster beam. The ensuing tetracene beam was directly deposited onto the optically transparent ITO-coated glass substrate (500 Å). For the undoped EL devices, Alq_3 as an electron transport layer was deposited on the top of tetracene layer (700 Å). For the doped EL devices, Alq_3 and dye dopant DCM loaded in separate evaporation cells were heated and co-deposited. The growth rate of the CBD film was governed by the evaporation cell temperature and was maintained about 1-2 Å/sec. The substrate was kept at room temperature throughout the deposition process. Finally, the low work-function Li:Al alloy cathode (2.9 eV) was prepared by thermal evaporation. The thickness, crystallinity, and surface morphology of the organic thin films were examined by a thickness monitor, an alpha step surface profile monitor, XRD, and AFM. The PL, EL, and device characteristics such as *J–V*, *L–V*, and *EQE–I* were recorded and investigated.

3. Results and Discussion

3.1 AFM and XRD characterization of tetracene thin films

Figure 2 shows two- and three-dimensional AFM micrographs taken for the tetracene thin films (about 5000 Å thickness) deposited on glass substrates by the CBD method. The size of grain crystallites and the root-mean-square roughness were obtained by conducting section analyses and typically measured to range from 0.45 to 0.85 μm in diameter and about 120 Å, respectively. Such quantitative values indicate that the weakly bound, neutral cluster beam experiences an effective fragmentation into mobile individual tetracene molecules resulting in the formation of smooth thin films consisting of submicrometer-sized crystallites on the glass substrate maintained at room temperature.

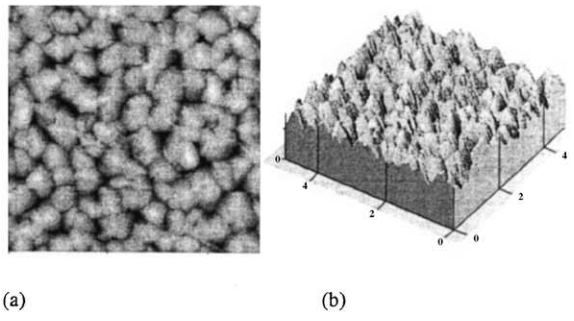

Fig. 2. (a) Two- and (b) three-dimensional AFM images for the 5 x 5 μm^2 tetracene thin films deposited on glass substrate by the CBD method.

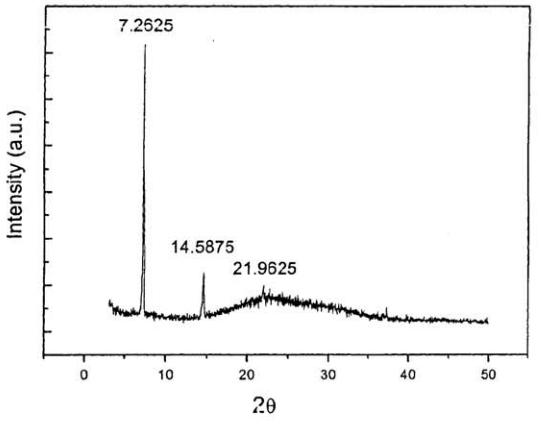

Fig. 3. A XRD pattern of the tetracene thin films (5000 Å thick) deposited on glass substrate by the CBD method. A broad maximum profile observed at 2θ of around 20–25° stems from the contribution of glass substrate.

The crystallinity of the thin CBD film is clearly demonstrated in the XRD measurements shown in Figure 3. The diffraction patterns display a highly ordered structure with the distinctive first-order peak at $2\theta \cong 7.26°$ as well as multiple higher-order peaks. The peak positions show a good agreement with the previously reported values, and the sharpness of the peaks further represents the highly ordered structure of the tetracene films prepared by the CBD methods [11].