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CHARACTERIZATION AND OPTIMAL DESIGNS OF ADENINE LIGHT-EMITTING DIODES

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ABSTRACT

We discuss the design of bilayer organic light emitting diodes (OLEDs) formed using adenine sandwiched between indium tin oxide (ITO) and Al, which were fabricated into four types of device structure. The electroluminescence intensities were characterized as a function of the current and voltage. The efficiency was significantly greater in bilayer devices in which adenine were used. The performance of green light emission with adenine-based devices was significantly greater than that reported to date (we find a luminance of -4227 cd/m^2 at a voltage of -8 V). The multi-layer OLED devices were formed of ITO, *N,N'*-bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)benzidine (NPB), adenine, Tris(8-hydroxyquinolinato)aluminium (Alq3) and aluminium (Al). We find that the thickness of the adenine light-emitting layer determined the brightness and efficiency, as well as the operating voltage. The thickness of the adenine layer was optimized to improve the efficiency of the OLEDs.

Key Words: BioLED, Adenine, Luminance

INTRODUCTION

Digital displays using organic light emitting diodes (OLEDs) are widely used in commercially available mobile phones, computer monitors, personal digital assistants (PDAs) and television displays (Kamtekar *et al.*, 2010). OLEDs are hetero-junction devices that are composed of electron and hole transport layers, as well as an emissive layer (Mathew *et al.*, 2001). Electrons from the cathode and holes from the anode are transported to the emissive layer, and form an excited state, which then relaxes to ground state, giving rise to electroluminescence (EL). Organic materials are usually chosen for the emissive layer that have a large energy gap, and where a low charge density exists, which leads to excitation and emission by injecting electrons and holes (Howard, 2004). In 1987, Tang and Vanslyke first reported high-performance double-layer EL devices, in which electrons and holes recombined within an organic emitting layer. With this simple OLED structure, however, disorder limited the transport of charge carriers into the emitting zone (Bernius *et al.*, 2000). For this reason, multilayered OLEDs devices have been developed, in which a well-ordered transparent conducting oxide anode, or hole transport layer (HTL), as well as an emissive layer (EML) an elec-

tron transport layer (ETL) and a metal cathode were used, each with specific functionality (Yersin, 2004). The improved efficiency offered by these multilayer structures has enabled the current technology, which has been implemented using two types of organic material: small molecules and polymers. Tang *et al.* (1987) first introduced small-molecule OLEDs, including coordination complexes such as tris(8-hydroxyquinolinato)aluminum (Alq3), which is a common compound of quinoline rings with luminescence properties (Montes *et al.*, 2006). Such compounds, including perylene, rubrene and quinacridone derivatives, exhibit emission at wavelengths in the range 400–600nm (Sato *et al.*, 1998). Triphenylamine derivatives have been used as HTLs (Bellmannet *et al.*, 1998), and Alq3 has been used not only as the emitting layer, but also as the ETL (Cheng *et al.*, 2000). The first invention of a polymer OLED used poly(p-phenylene vinylene), which emits light when connected to an external voltage (Burroughes *et al.*, 1990). Poly(p-phenylene vinylene) and polyfluorene derivatives are also widely used, and the substitution of side chains can alter the color of the emitted light (Heeger *et al.*, 1993). Those polymers are more stable and soluble than small molecules, and are easier to process (Kiebooms *et al.*, 2001). Typically, polymer OLEDs formed by spin-coating and

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inkjet printing have better workability, lower costs and are more suitable for large-area displays than small molecules, which typically require vapor deposition. Small molecules generally have longer lifetimes and better color purity; however, despite the improved color purity, many of colors cannot be achieved using existing small molecules. For this reason, a diverse range of compounds is required for using emission layer as well as electron and hole transport layers.

In this work, a new small molecule candidate is investigated for applications in OLED devices: adenine. Four different device structures featuring adenine were fabricated, with the following layer sequences: indium tin oxide (ITO)/adenine/Al, ITO/adenine/Alq3/Al, ITO/*N,N'*-Bis(naphthalen-1-yl)-*N,N'*-bis(phenyl)benzidine(NPB)/adenine/Al and ITO/NPB/adenine/Alq3/Al. The current–voltage characteristics, luminance–voltage characteristics, and the EL intensities were investigated.

MATERIALS AND METHODS

Adenine was diluted with dimethyl sulfoxide (DMSO) at a concentration of 1,000mg/ml, and 1ml of the solution was placed in a cuvette for measurements using an ultraviolet (UV) spectrophotometer at a wavelength of 365nm. The luminescence was monitored using a Minolta spectroradiometer (CS-1000, Japan), as shown in Figure 1.

To fabricate the OLED devices, transparent and conductive ITO was spin-coated on glass substrates purchased from Samsung Corning. The ITO layer had a surface resistance of $8.8 \pm 1.8 \Omega$ and a thickness of $1850 \pm 200 \text{ \AA}$. Photolithography was used to form stripe patterns. First, the ITO layer was coated with a photoresist (PR) developer using spin-coating, and baked at 100°C to remove any residual solvent and to harden the surface of the PR (Kang et al., 2015). Positive and negative PR processes were carried out to form the stripe patterns. The positive PR was removed using UV exposure, and the negative PR was non-UV. The patterned layer was hardened for 10 min at 130°C , and the ITO layer was etched for 3 min at 40°C , and placed into an asher for 10 min at 60°C in a nitrogen atmosphere. The patterned ITO layer was cleaned using ultra-sonication for 30 min in acetone, and then in MeOH and isopropyl alcohol for 5 min at 60°C . Finally, it was exposed to UV irradiation (184.9nm) for 20min, at a distance of 1°C .

Vacuum evaporation was carried out at 5.5×10^{-6} Torr in a heated chamber, and a quartz crystal microbalance (QCM) was used to measure the thickness of the coating during deposition. The rate of vacuum evaporation was $1\text{--}1.5 \text{ \AA}/\text{s}$. Four types of adenine-based multilayer OLED

structures were formed: ITO/adenine/Al, ITO/NPB/adenine/Al, ITO/adenine/Alq3/Al and ITO/NPB/adenine/Alq3/Al, as shown in Figure 2. The OLED devices were controlled using Labview (version 8.0) using a desktop personal computer and general-purpose interface bus (GPIB). The devices were placed in a darkroom and current was applied using a source–measure unit (SMU) (Keithley 236) PR (Kang et al., 2015). The emitted light was collected using a photodiode, and the resulting current was measured using a picoammeter (Keithly 487), to convert the signal into a luminance. The spectrum of the emitted light was measured using a spectroradiometer (Minolta CS-1000) to determine the red–green–blue (RGB) color coordinates.

RESULTS

Photoluminescence (PL) spectra were obtained using UV excitation at 365nm and measured over a broad range of wavelengths from 400 to 800nm, as shown in Figure 3. The optical emission was stronger in the range 380–560nm, which corresponds to green. The maximum luminance was $1.7 \text{ cd}/\text{m}^2$ with a wavelength of 417nm.

Figure 4 shows the current–voltage characteristics of the ITO/adenine/Al device with various thicknesses of the adenine layer in the range 30–100nm. With an adenine thickness of 30 nm, a current of 197mA flowed when the operating voltage was 14V. With an adenine thickness of 40 nm, a current of 97mA flowed when the voltage was 8V. The highest luminance was achieved with a thickness of 80nm, which was $1.6 \text{ cd}/\text{m}^2$ at a voltage of 23V, as shown in Figure 5. The emission spectrum was investigated in the ranges 380–400 and 780–800nm, which corresponds to color coordinates of X: 0.1526 and Y: 0.2364, respectively, as shown in Figure 6.

Figure 7 shows the current as a function of voltage with the device structure ITO/adenine/Alq3/Al with thicknesses of the adenine layers in the range 20–80nm and a 40-nm-thick Alq3 layer. For a 20-nm-thick adenine layer, a current of 10mA flowed when the operating voltage was 20V. When the thickness of the adenine layer was 20, 40 or 60nm, less light was emitted than with the ITO/adenine/Al structures. The luminance as a function of voltage is shown in Figure 8 with an adenine layer that was 20-nm-thick optical emission was observed at voltages of 10 V or greater, and the largest emission was $13 \text{ cd}/\text{m}^2$ at 17V. The device emission spectrum was examined at wavelengths around 400nm, as shown in Figure 9.

Figure 10 shows the current–voltage characteristics of devices with the ITO/NPB/adenine/Al layer sequence for thicknesses of the adenine layer in the range 40–80nm, and where the NBP film was 40nm thick. When the adenine layer was 40nm thick, a current of 24 mA flowed

when a voltage of 15V was applied. For the 60-nm-thick film, the current was 25mA when the voltage was 20V. When the adenine layer was 80nm thick, the current was 21mA when the voltage was 27V. Figure 11 shows luminance as a function of voltage. For the 40-nm-thick adenine film, optical emission was observed at voltages of 10V or greater. A maximum luminance of 1.6cd/m² was observed at 15.5V. For the 60-nm-thick film, optical emission was observed at voltages greater than 16 V, and the maximum luminance was 2.6cd/m² at a voltage of 20V. For the 80-nm-thick film, optical emission was observed at voltages of 20V or more, and the maximum luminance was 1.8cd/m² at 27V. The emission spectrum was examined in the range 450–480 nm, as shown in Figure 12.

Figure 13 shows the current as a function of voltage for the ITO/NPB/adenine/Alq3/Al devices with thicknesses of adenine layers in the range 40–60nm. The NPB and Alq3 layers were 40-nm-thick. When the adenine layer was 40nm thick, the current was 23mA when operating voltage was 15V. For the 60-nm-thick film, the current was 12mA when the voltage was 20V. Figure 14 shows the luminance as a function of voltage. With the 40-nm-thick adenine film, optical emission was observed at voltages of 8 V or greater, and a maximum luminance of 4,227cd/m² was observed at 14V. For the 60-nm-thick film, optical emission was observed at voltages of 12V or greater, and the maximum luminance was 2,603cd/m² at 16V. The emission spectrum was examined at wavelengths around 540nm, as shown in Figure 15; this emission wavelength corresponds to green.

DISCUSSION

Over the past decade, organic light-emitting devices have seen considerable development. The flexibility of these devices, coupled with the thin-film characteristics and transparency has resulted in a considerable global market. OLED technologies have made a significant contribution to the display industry. Academic and industrial research groups have developed this technology to advance the quality and workability of new materials (Borchardt, 2004). The light-emitting material is typically sandwiched between metallic materials, which are designed for efficient charge injection from the electrodes. Alq3 is widely used as electron-transport and light-emitting layers (Mathew *et al.*, 2001). There has been much interest in utilizing biological molecules, for example, cytochrome, myoglobin, hemin and chlorophyll (Tajima *et al.*, 2003; Tajima *et al.*, 2005; Ikeda *et al.*, 2005; Shimatani *et al.*, 2005; Tajima *et al.*, 2006). These materials are known to exhibit effective electron transport properties in biological systems.

We fabricated OLED devices that employ adenine as the light-emitting layer, together with ITO, NPB, Al and Alq3as charge-transport layers. Vacuum deposition was used to fabricate uniform thin films (Tang and VanSlyke, 1989; Kido and Lizumi, 1998). PL from adenine was observed at wavelengths in the range 380–560nm. Adenine is a purine derivative, and is related to cellular respiration, forming adenosine triphosphate (ATP), nicotinamide adenine dinucleotide (NAD) and flavin adenine dinucleotide (FAD), is involved in protein synthesis, and is a base of deoxyribonucleic acid (DNA) and ribonucleic acid (RNA).

OLED devices combined with adenine were investigated, in which the various thickness adenine films were formed. The simplest structure was a single-layer of adenine sandwiched between ITO and Al. When the thickness of the adenine layer was 30nm, the device exhibited a larger electric current than with a thickness of 100nm. We observed a broad optical emission spectrum. With the ITO/adenine/Alq3/Al structure, this luminance was lower when the adenine layer was thicker; however, this structure exhibited a six-fold increase in the luminance compared with the ITO/adenine/Al devices. Furthermore, characteristic adenine emission was observed at a wavelength of less than 400nm. OLED devices that incorporate DNA thin films as electron blocking layers have been reported, and significant increases in the luminance and luminous efficiency were observed (Hagen *et al.*, 2006). The ITO/NPB/adenine/Alq3/Al structure exhibited the lowest operating voltage, and also exhibited the largest EL intensity, with a peak luminance at a wavelength of 510nm.

Choi *et al.* (2006) reported that the performance and stability of OLED devices is an important factor, and that interfacial issues such as poor adhesion are significant. Adenine is a biomolecule related to electrical and optical properties in nature, and has potential applications in optoelectronic devices. This biomolecule can be modified; for example, DNA and cetyltrimethyl ammonium may be chemically coupled so that it is insoluble in water but soluble in alcohol (Hagen *et al.*, 2006). Furthermore, naturally occurring biomaterials are a renewable resource and are inherently biodegradable.

We have shown that adenine can lead to high photoemission efficiency in OLEDs. Adenine functions as both a hole-transport and light-emitting layer, and has previously been shown to function as an electron-blocking layer. This study of adenine OLEDs indicates that their favorable charge transport properties, relative low cost, minimal pre-fabrication processing, and natural origin makes them attractive for applications in natural optoelectronic devices.

CONCLUSION

The purpose of this study was to investigate the effect of adenine light emitting materials for OLED devices. These results were showed that adenine has potential light-emitting activity and its multilayered devices have effects on the electrical and luminance performances. Naturally occurring substances as adenine has advantages of renewable organic compound, environmental friendly and low cost to OLED industries for future. Base on this study will be contributed to fundamental technology of OLED application of light emitting materials.

ACKNOWLEDGEMENT

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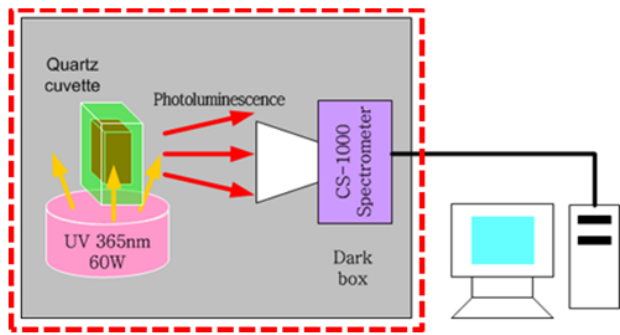


Figure 1: A schematic diagram of photoluminescence analysis.

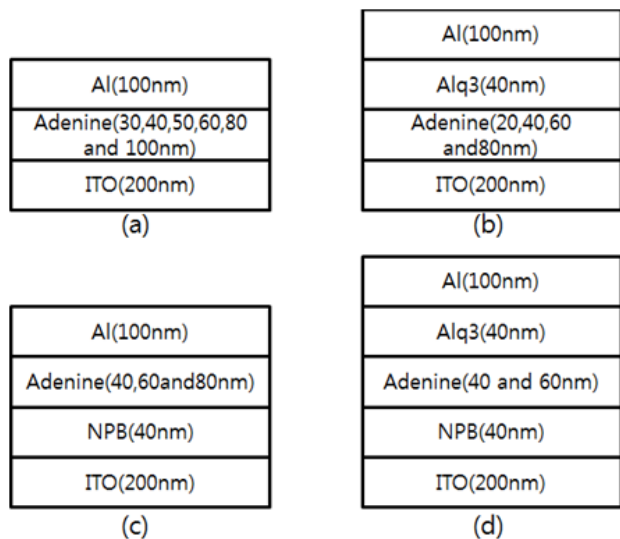


Figure 2: The OLED device structures.(a) ITO/adenine/Al, (b) ITO/adenine/Alq3/Al, (c)ITO/NPB/adenine/Al and (d)ITO/NPB/adenine/Alq3/Al.

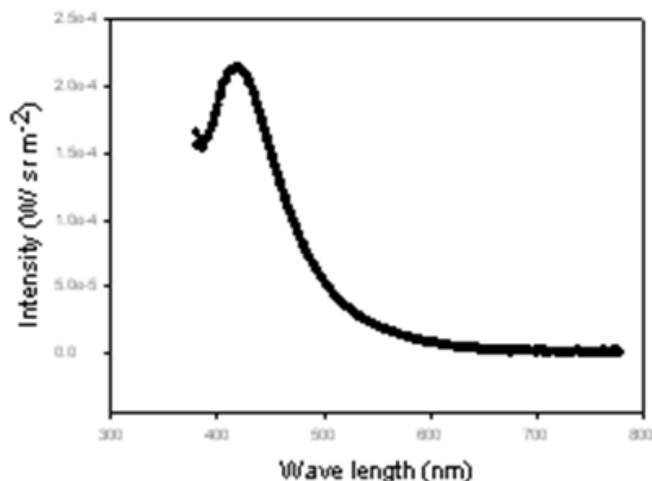


Figure 3: Photoluminescence spectra of adenine.

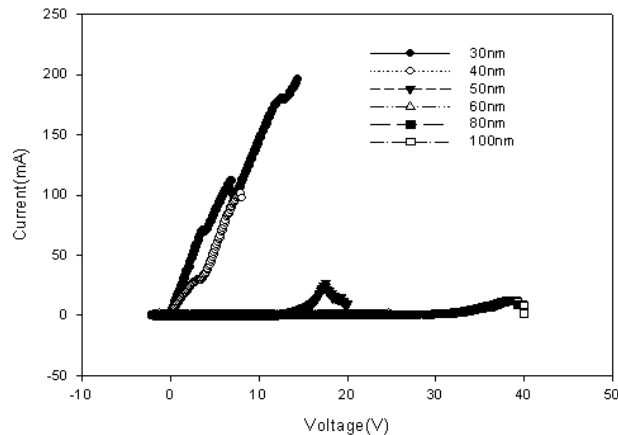


Figure 4: Current as a function of voltage for the ITO/adenine/Al device structure with adenine layers of various thicknesses.

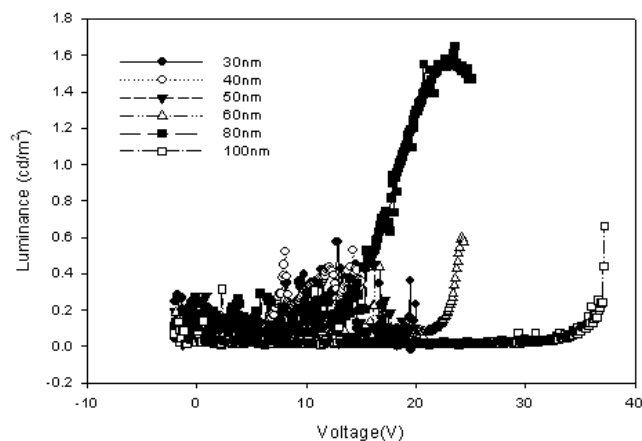


Figure 5: Luminance as a function of voltage for the ITO/adenine/Al device structure with adenine layers of various thicknesses.

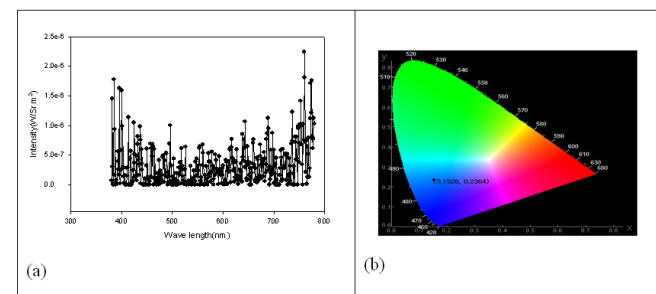
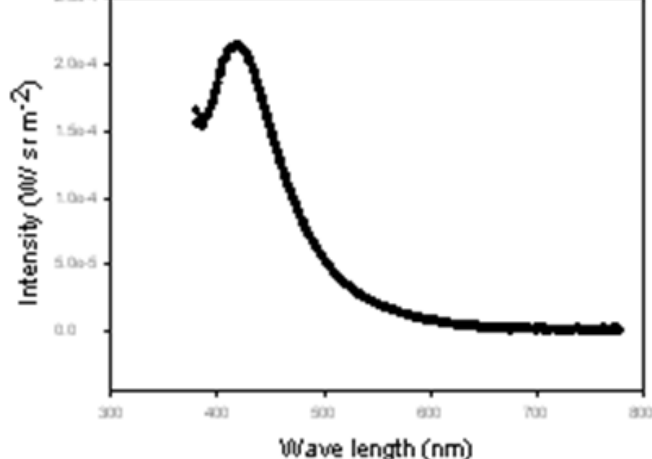


Figure 6: (a) EL intensities and (b) color coordinates of the ITO/adenine/Al devices.

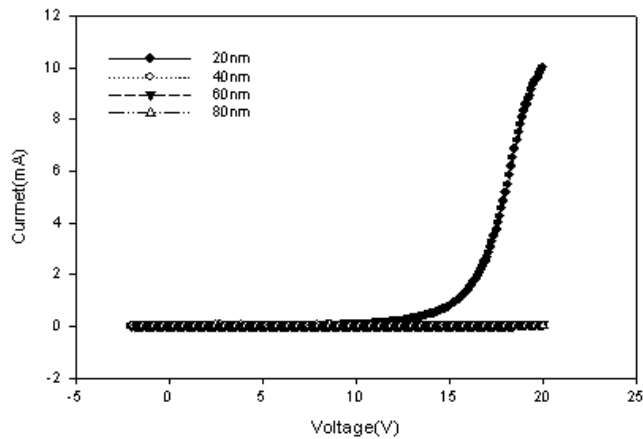


Figure 7: Current as a function of voltage for the ITO/adenine/Alq3/Al devices with adenine layers of various thicknesses.

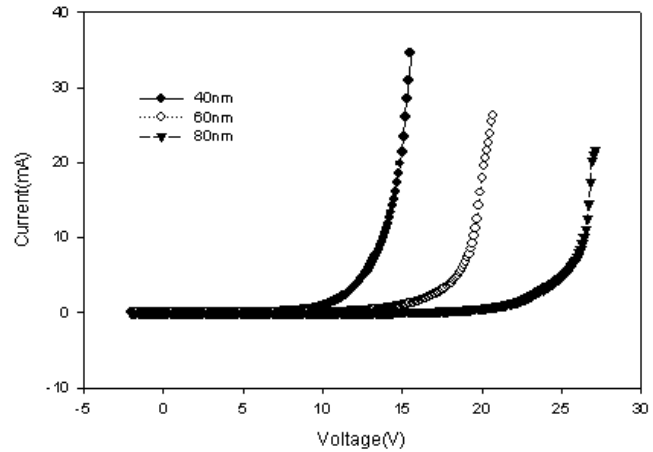


Figure 10: Current as a function of voltage for the ITO/NPB/adenine/Al devices with adenine layers of various thicknesses.

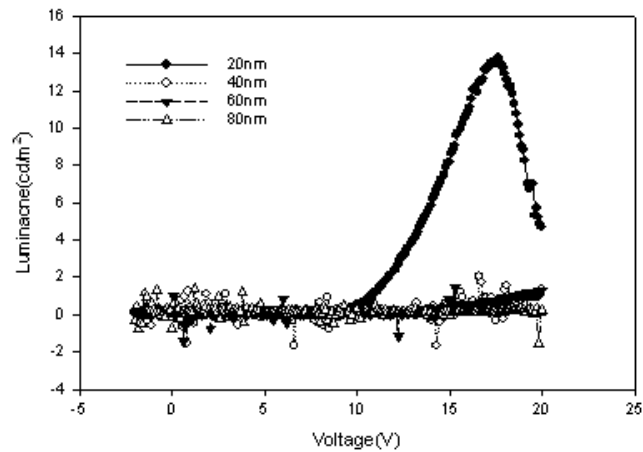


Figure 8: Luminescence as a function of voltage for the ITO/adenine/Alq3/Al devices with adenine layers of various thicknesses.

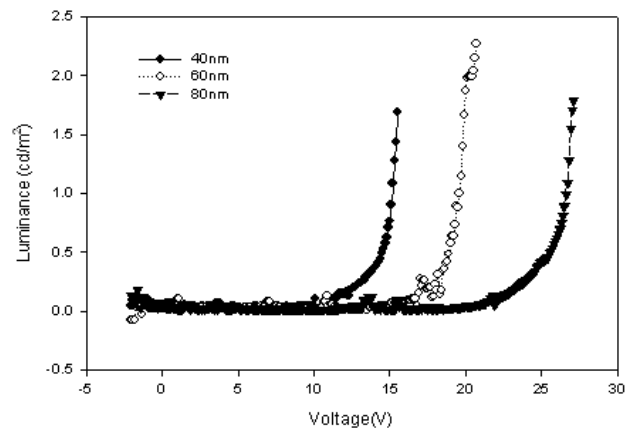


Figure 11: Luminescence as a function of voltage for the ITO/NPB/adenine/Al devices with adenine layers of various thicknesses.

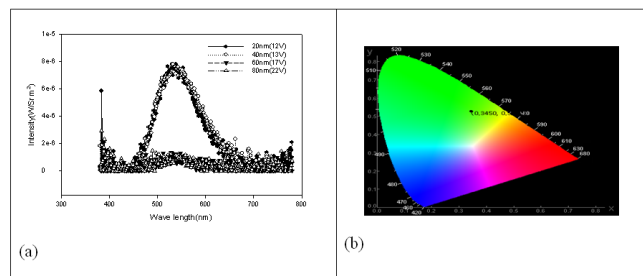


Figure 9: (a) EL intensities and (b) color coordinates of the ITO/adenine/Alq3/Al devices.

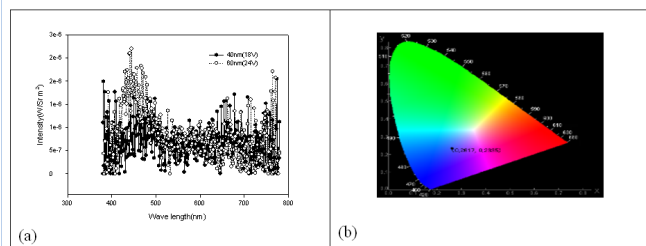


Figure 12: (a) EL intensities and (b) color coordinates for the ITO/NPB/adenine/Al devices.

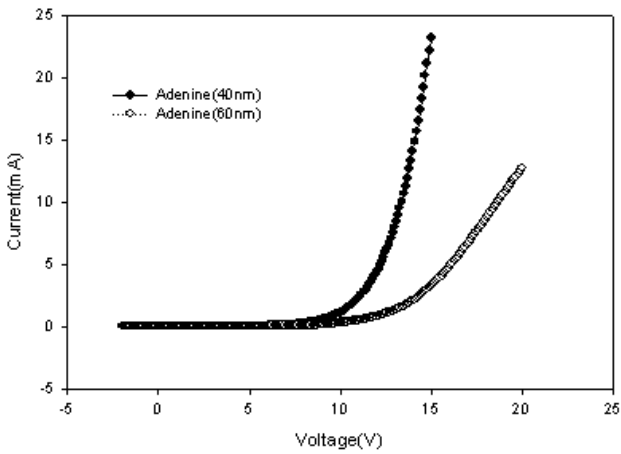


Figure 13: Current as a function of voltage for the ITO/NPB/adenine/Alq3/Al devices with adenine layers of various thicknesses.

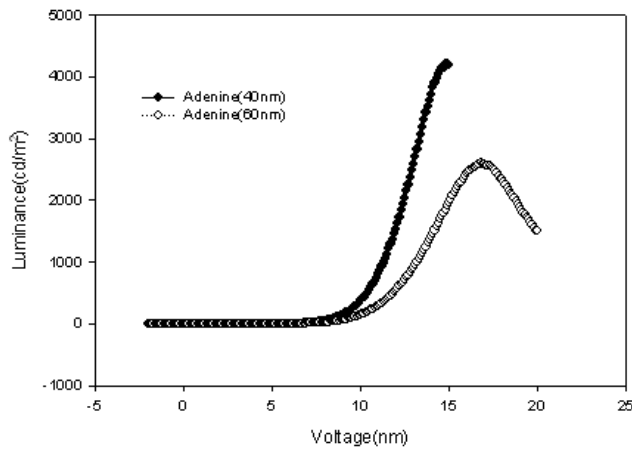


Figure 14: Luminance as a function of voltage for the ITO/NPB/adenine/Alq3/Al devices with adenine layers of various thicknesses.

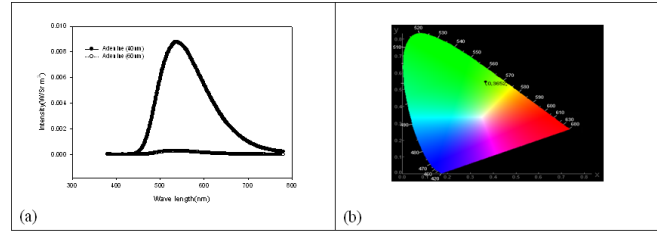


Figure 15: (a) EL intensities and (b) color coordinates for the ITO/NPB/adenine/Alq3/Al devices.