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RESEARCH ARTICLE

# Fabrication of Organic Light Emitting Diodes (OLEDs) using the Lamination method in a Vacuum-Free Environment

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**Abstract**

Organic Light Emitting Diodes (OLEDs) have recently become one of the fastest-growing technologies in the world. The challenge in OLED fabrication, especially larger-area OLEDs, is its relatively high costs and complexity. The lamination method in a vacuum-free environment is an approach to simplify and reduce the cost of fabrication. This paper reports our latest progress on OLEDs fabricated using the said method and condition. The processing parameters were explored and optimized. Spin coating the emissive Layer (PFO) at 1300 rpm and the anode (TC-07-S) at 3000 rpm yield the best results in terms of current conduction and success rate. Laminating the OLEDs at 160 °C, with 245 N of force, and for 30 seconds, gave the best results in terms of previously stated parameters. Furthermore, the constituting materials of the OLEDs were explored. It was found that TC-07-S as an anode, PFO as the light-emitting material, a 30-micrometer thick aluminum foil as the cathode, and Kapton as the dielectric and adhesive material yielded the best results. These results may pave the way for other innovative methods to fabricate OLEDs with simple and affordable processes.

**Keywords:** Lamination method, Organic Light Emitting Diodes (OLEDs), PFO, Spin coating, Vacuum-free Environment

## 1. Introduction

Lighting is the deliberate use of artificial or natural light sources for our benefit. It is an integral part of our lives. Conventional artificial light sources such as incandescent and

fluorescent lighting are still the most dominantly used lighting sources, which tend to have a low power-to-light conversion ratio, low colour performance, low uniformity, and are environmentally unfriendly [1]. Through a novel working principle based on the Electroluminescence (EL) phenomenon, Organic Light Emitting Diodes (OLEDs) offer to solve these setbacks [2].

Since first demonstrated by Tang and van Slyke in 1987 [3], there have been many breakthroughs in this technology, particularly in parameters where OLEDs had lacked beforehand, such as the lifetime, fabrication cost, colour accuracy performance, and colour tunability [4]. Advancements on this front have made OLEDs commercially very successful, with market sizes expected to be 18.6 billion US$ for OLED displays and 65 million US$ for OLED lighting in 2026 globally [5]. OLEDs also offer new possibilities relating to their applications, as OLEDs can be fabricated on thin and flexible substrates. This possibility lets OLEDs pave the way to the development of a new generation of devices, such as bendable electronic wearables [6].

Citing their power-to-light conversion ratio and flexibility, OLEDs are also expected to replace conventional lighting. Therefore, OLEDs need to be fabricated with larger areas. Developing large-area OLED technology has been a priority of research. However, flexible and large-area devices have had cost and fabrication complexity problems, stemming from the conventional fabrication methods of OLEDs. Conventional fabrication of OLEDs often involves vacuum deposition to deposit layers of the consisting materials [7]. These methods are unsuitable for flexible, large-area devices as the cost of fabrication increases significantly. Thus, other methods must be utilized.

The spin coating deposition method is one way to deposit large areas of OLEDs. Spin coating involves the deposition of OLED layers in the form of a solution, spread over the entire substrate by applying centrifugal force. The spin coating process can fabricate OLEDs at a fraction of the cost of vacuum deposition methods [8].

However, traditional spin coating methods are incompatible with OLEDs involving more than one spin coated material (multi-layers). Spin coating a material layer on top of the preceding solution is impossible as the solvents used for the next layer will dissolve the preceding layer. The lamination method is an alternative approach to fabricating OLEDs in two parts. The first part contains the transparent, solution-based anode and the first substrate. The second part contains the solution-based emissive layer coated over the second substrate, which also works as the cathode. These two parts are bound together to form an OLED using an adhesive material, moderate heat and pressure, hence the term "lamination method" [9].

These advancements have allowed the development of flexible thin film OLEDs with a relatively large-emitting area and a relatively satisfactory performance, low fabrication cost and low fabrication complexity [10]. Despite this, previous works still required some materials to be deposited under vacuum conditions, thereby making fabrication cost and complexity remain as issues. There is an opportunity to explore a fabrication scheme based on the lamination method entirely done in a vacuum-free environment.

This article outlines our progress focusing on OLED fabricated using the lamination method in a vacuum-free environment. At the beginning of this article explains

the general working principle and how light is generated through electroluminescence in OLEDs. Each composition of OLEDs is then explained and the process of optimizing the materials are detailed. The performance parameters will be discussed, such as current-voltage (I-V) characteristics, threshold voltages, luminance output, and success rates. Finally, a reliable fabrication method on a vacuum-free environment is proposed and a comment on the future research direction on lamination-based, vacuum-free-fabricated OLEDs will be provided.

## 2. Working Principles

### *2.1 Organic Materials in OLEDs*

The term "organic" in OLEDs refers to the organic materials that form an OLED. These materials are compounds that consist of carbon chains in the form of small molecules or polymers with semiconductor properties. Organic semiconductors are similar to inorganic semiconductors but differ in their conductivity mechanism due to Coulomb effects. This difference is caused by intra-molecular Van der Waals forces in organic semiconductors, which are often weaker than in inorganic semiconductors.

Hence, their intrinsic conductivity and light absorption/emission are commonly less than for inorganic semiconductors. However, these properties of organic semiconductors can be amply harnessed to create traditional semiconductor devices (Field-effect transistors, LEDs, photovoltaics). On the other hand, organic semiconductors have high tunability (stemming from its variety), which means that their electrical and absorptive/emissive properties can easily be modified through chemical processes, unlike inorganic semiconductors.

While the bandgaps of inorganic semiconductors are most often between 0 to 2 eV, the bandgaps of organic semiconductors are usually larger. Thus, the generation of charge carriers in organic semiconductors is often achieved by (i) the use of electrodes, (ii) optical excitations, and (iii) doping [11]. OLEDs are designed with number 1 in mind. In OLEDs, the organic materials are sandwiched between two metallic electrodes.

### *2.2 Carrier Movements in OLEDs*

HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) are energy levels that denote the excitability of electrons of a particular organic semiconductor. HOMO is the outermost orbital that contains electrons, while LUMO is the innermost orbital that can receive electrons. The energy gap between these two orbitals is considered the bandgap of the organic semiconductor. Every organic semiconductor has its own LUMO and HOMO levels, depending on their geometries. Due to an electric field application, the LUMO interacts with the electrons injected by the cathode in an OLED. The HOMO interacts with holes injected by the anode. These charges will then migrate into the organic emissive material. This concept is illustrated in Figure 1.

### *2.3 Light generation in OLEDs*

Both inorganic and organic semiconductors generate light by a mechanism called recombination. Recombination is when an electron fills a hole to combine and

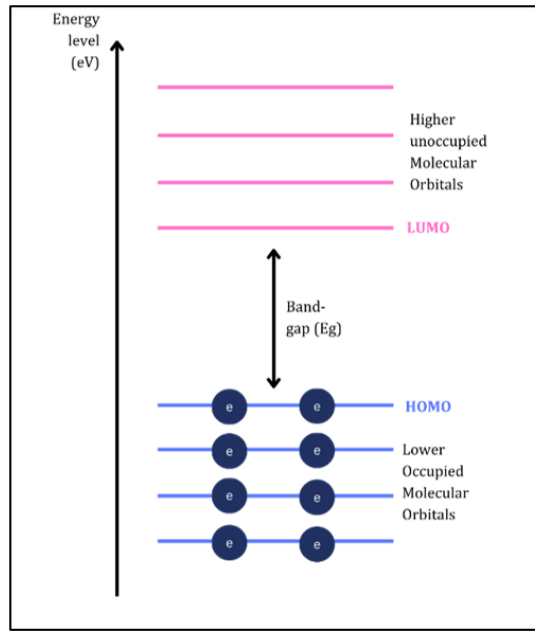


**Figure 1.** Illustration of HOMO and LUMO. The y-axis is the energy level. In organic semiconductors, there are many levels of molecular orbitals outside the band gap, in which the two closest to the band gap are the HOMO and the LUMO.

cancel each other out (Figure 2(a)). It is a process in which an electron goes from a higher energy state to a lower one, thereby depositing energy in a material. The recombination movement is illustrated in Figure 2(b). The areas where recombination happen are called recombination channels, and channels that can generate light are called radiative channels. Radiative channels are only available in materials with the ability to produce photons during the recombination process, i.e., direct bandgap semiconductors, which have the low point of the LUMO (conduction) band directly over the peak of the HOMO (valence) band.

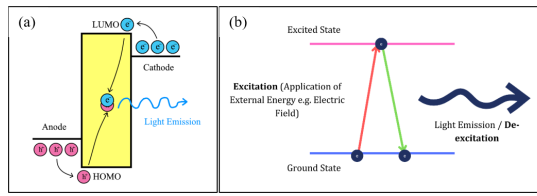


**Figure 2.** (a) The cathode, which has a lower work function of will inject electrons to the LUMO, while the anode, which has a higher work function of will inject holes to the HOMO. With the application of an electric field, the electrons and holes will drift across the barriers and enter the emissive layer's LUMO and HOMO respectively. (b) With recombination mechanisms, such as de-excitation, the electrons will recombine with the holes generating photons, i.e., emitting light.

Due to organic semiconductors having a weaker Van der Waals force, a lower dielectric constant, and a larger band gap, the recombination occurring in organic semiconductors occurs via excitons (electron-hole pairs bound to each other) instead of by band-to-band recombination (Figure 3) [12]. This recombination happens by applying an electric field on the electrodes, making major electron-hole movements possible. Thus, the process is denoted as "electroluminescence". Electrons move from the LUMO, and holes move from the HOMO of the emissive material, recombining in the form of excitons. Next, the charges inside the excitons convene, and light emission happens. The wavelength/colour of the light depends on the organic semiconductor chosen as the emissive layer. The gap between the energy levels of LUMO and HOMO of the emissive layer in question will determine the colour of the light.

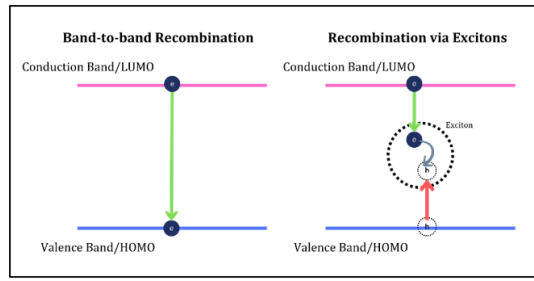


**Figure 3.** Band-to-band recombination and recombination via Excitons. Recombination via excitons, a more complex recombination process, is more occurrent in organic semiconductors due to their dissimilar properties.

### *2.4 General Structure of OLEDs*

OLEDs comprise layers in which organic semiconductor(s) are sandwiched between two electrodes. The number of layers depends on the design of the OLED, but at least one layer, the emissive layer (EML), situated between two electrodes, is needed for device operation. Additional layers for substrate, passivation, carrier transport, blocking, and injection can be added to complement the EML to enhance performance. Common layers are explained in Table 1. Figure 4(a) shows common OLED structures, and Figures 4(b) and 4(c) illustrate carrier movements in simple, EML-only and multi-layer OLEDs.

## 3. Methods

### *3.1 Fabrication Process*

The fabrication begins in the preparation process. The materials are prepared accordingly. The substrates underwent ultrasonic cleaning to remove dust and debris from the substrates. The cleaning agents, acetone and ethanol, were dried out under a heating treatment. Once preparation is complete, the spin coating process is done on a Yowa Riken K-359SD-1 spin coater.

After the solutions (anode and EML) are spin coated, the solutions underwent heat treatment to evaporate the solvents. It was found that heating the spin coated

**Table 1.** Common layers utilized in OLEDs, their intention, their prerequisite properties, and common examples.

| Material | Specification | | |
|---|---|---|---|
| | Intended to | Must Be | Common Examples |
| Substrate | Be deposited for all constituting materials | Transparent, insulating, and flexible, high chemical stability | Plastic, Glass, Transparent foils |
| Anode | Be given a positive voltage to send holes to the EML | High work function (~5 eV), transparent | ITO, PEDOT: PSS, TC-07-S |
| Hole Injection Layer (HIL) | Ease the hole movements/migration | Organic semiconductor, transparent, have a HOMO between the work function of Anode and HOMO of the HTL, high hole mobility | m-MTDATA, copper phthalocyanine [13] |
| Hole Transport Layer (HTL) | Same as HIL, but can block electrons as well | Organic semiconductor, transparent, have a HOMO between the HIL/work function of the anode and HOMO of the EML, High LUMO, high hole mobility | NPB, TPD, TAPC [14] |
| Emissive Layer (EML) | Be the main Emitting layer | Direct band-gap organic semiconductor | Alq3, MEH-PPV, PFO |
| Electron Transport Layer (ETL) | Ease the hole movements/migration and block holes | Organic semiconductor, have a LUMO between the work function of Cathode and the LUMO of the EML low HOMO, high electron mobility | PBD, BND [15] |
| Cathode | Be given a negative voltage to send electrons to the EML | Low work function (~4 eV) | LiF-Al and Magnesium-silver |
| Passivation | To encapsulate/protect the materials inside the OLED | Low oxygen and water vapor transmission rates, high chemical stability | Epoxy Resin, Plastic. Glass |

solutions at relatively lower temperatures (30–50 °C) for 5 minutes yielded the best results. These formations can then be assembled and laminated. We determined that the lamination process done at a temperature of 165 °C, a force of 245 N, and a time of 30 seconds produced the best results.

Through these experiments, a final OLED structure and fabrication scheme was proposed (Figure 5). With these process parameters, we were able to fabricate a relatively large-area OLED device that emits light. The total fabrication time is about two to three hours and was done entirely in a regular environment. Further optimizations are also done regarding the materials that are utilized to build the OLEDs, with each of the layers explained in the following.

### *3.2 I-V and Luminance Measurement Method*

The I-V characteristics of the OLEDs were measured using a Hewlett-Packard 4145B Semiconductor Parameter Analyzer. The current is measured with a voltage sweep between 0 and 10 volts, with a 250-mV interval. The data were then compiled and graphed using the Origin software. Three main parameters were analyzed, namely threshold voltage, curve shape, and maximum current. Luminance testing is done with an application of voltage to the OLED using a DC power supply and a camera with controlled exposure settings.

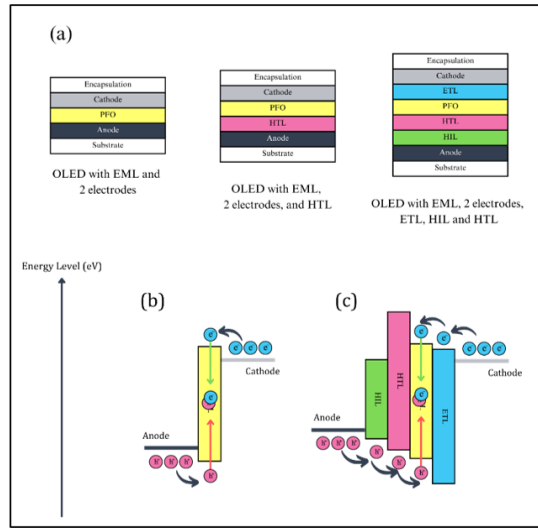


**Figure 4.** (a) Common OLED structures, from the simplest to the most complex. The simplest one contains two electrodes and one active layer, the emissive layer, with no enhancing layers. A moderately complex structure often has one or two enhancing layers, which is often the HTL. More complex structures usually have 3 or even more active enhancing layers. (b) and (c) Differences of electron-hole movements in a simple and more complex OLED structures. With the addition of more layers, the barrier that the electrons and holes must leap is reduced, which, if designed and implemented properly, can reduce turn-on voltages and increase efficiency.

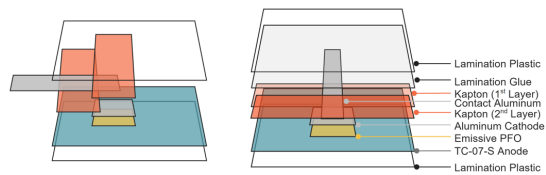


**Figure 5.** The optimized structure of the OLEDs. The purpose of each layer will be explained in section 3.3 through 3.8.

## 4. Results

### *4.1 Substrate and Passivation*

Substrates in OLEDs are employed as the underlying layer that holds materials above them. Thus, it is an insulator and is not a part of the actual OLED. It must be transparent to let light through. Some candidates for substrates are Glass, PET, and Plastic. In the result, the plastic substrate yielded the best results. The plastic substrate yields the lowest threshold voltage and highest success rate, emits the brightest luminance, and keeps a flexible structure. For passivation/encapsulation, different materials can be used, such as plastic and epoxy resin. Plastic was chosen as it is cheaper and does not require additional fabrication steps.

### *4.2 Anode*

Anodes in OLEDs are employed to inject holes into the OLED system. They have a low work function, commonly in the 4 to 5 eV range. They must at least be semi-transparent to let light through the device and to the outside. Some candidates for Anodes are ITO ($\varphi = 4.5eV$), FTO ($\varphi = 4.4eV$), PEDOT: PSS ($\varphi = 5eV$), and TC-07-S ($\varphi$ $5eV$) [10]. ITO is deposited in a PET substrate, while FTO is deposited in a glass substrate. PEDOT: PSS and TC-07-S are solutions that must be deposited/grown over a substrate. This deposition is done at a speed of 3000 rpm, growing an anode layer about 3 μm thick. TC-07-S is a modified PEDOT: PSS manufactured by Denshi Kako Co. Ltd.

Based on our findings, the spin-coated anodes performed significantly better than the rest. Performance metrics include threshold voltage, luminance, and success rate. This improvement might be caused by the Lamination method, which is more suited for spin-coating. The diethylene glycol modification on TC-07-S from PEDOT: PSS improved performance, with TC-07-S anode OLEDs emitting light at lower voltages (<9V) than the regular PEDOT: PSS anode OLEDs (>15V).

### *4.3 HTL*

Experiments have been conducted to find a suitable HTL for our OLED structure, but more options have yet to be available. The limitation is that the HTL must be coated on top of ITO/FTO anodes, which based on our findings, are better anodes for our OLEDs. If the HTL is coated on top of other solution-based anodes such as PEDOT: PSS and TC-07-S, the solvent of the HTL will dissolve those previous layers. However, an experiment employed PEDOT: PSS as HTL, directly spin coated above ITO. The HTL-employed PEDOT: PSS was able to reduce the OLED threshold voltage, but it is unclear whether this structure can match the performance of the OLED with a TC-07-S anode. Its optical performance (luminance), however, was not satisfactory.

### *4.4 Emissive Layer*

PFO, or Poly(9,9-di-n-octylfluorenyl-2,7-diyl) (also often abbreviated as F8) is an emissive material for OLED with a polymer structure. It is an organic semiconductor based on fluorene. This material must be dissolved in Toluene with a weight ratio of 1% before spin coating and growing a layer of it. This material has been demonstrated to generate blue light with wavelength peaks of 426 nm (primary) and 490 nm (secondary), as shown in Figure 6 for reference.

Spin coating of PFO is found to be the best at a speed of 1300 rpm, growing a thin layer about 15 μm thick. Only one coating is needed to yield the best result.

Other attempts to use other emissive materials were also made, particularly on Tris(8-hydroxyquinoline) aluminum or more commonly known as Alq3. Alq3, dissolved in chloroform with spin coating, was demonstrated to be used for the emissive materials of OLED by Sarjidan et al. [13]. Using ITO as an anode, OLEDs using spin coated Alq3+chloroform were fabricated and successfully showed diode current-voltage characteristics. However, a magenta-violet coloured luminance was only seen instantaneously.

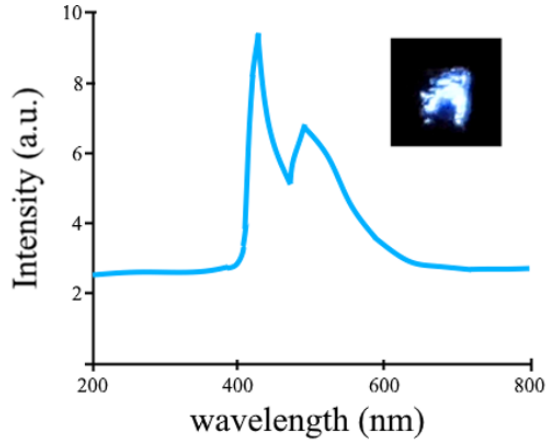


**Figure 6.** Wavelength spectrum of PFO (smoothed), inset a photograph of the light emission. The primary peak can be seen around 426 nm and the secondary peak can be seen around 490 nm. The resulting light emission is a blue light with a slight tint of violet which agrees with the calculated mean wavelength of 464 nm.

### *4.5 Cathode*

Experiments were made to compare different cathodes to be employed with the OLED. Sheets of copper and aluminum were used as cathodes, with aluminum showing the best luminance and current-voltage characteristics. This result stems from the unsuitable work function of copper, despite it being an excellent electrical conductor. It is also noted that pure aluminum was used instead of the more common aluminum with lithium fluoride (LiF) added. This decision was made to avoid growing LiF in a vacuum environment.

The thickness of the aluminum cathode was also explored. Aluminum with a thickness of 30 μm was found to yield the best luminance and current-voltage characteristics. The thickness of the cathode influences the resistance, making the rate of electron injection to the OLEDs different. However, physical properties also matter, and the thinner the cathode, the more flexible the OLED will be, and it is assumed that the physical property influences fabrication difficulty as well.

### *4.6 Dielectric*

The dielectric material is not part of the OLED but is still integral to the OLED design. The dielectric material separates the two electrodes, avoiding a short circuit. Dielectric materials used must have an adhesive component for ease of fabrication. Lamination glue and Kapton® tape films were used as dielectric materials, with Kapton yielding better luminance, lower threshold voltages, and higher success rates. This improvement comes from Kapton's resilience to heat and pressure.

With Kapton's adhesive and insulating properties, the OLEDs can be further optimized by employing a second Kapton layer to ease the lamination process. The second Kapton increased the fabrication success rate (from 57% to 84%) and increased

the OLED emitting area, as shown in Figures 7(a) and 7(b).

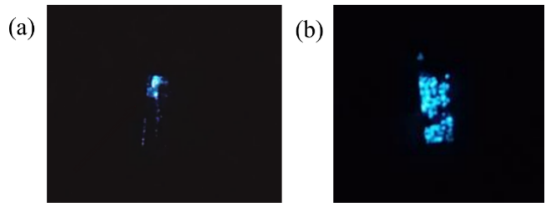


**Figure 7.** (a) Light emission for Kapton as dielectric material and (b) Light emission for Kapton as dielectric and adhesive material.

## 5. Conclusion

We have devised an OLED structure and fabrication method based on the Lamination method in a vacuum-free environment. Using the most suitable anode, cathode, dielectric, and optimization of the deposition techniques, we have demonstrated OLEDs with a diode current-voltage characteristic and a blue light emission. Our OLED was able to be activated at lower voltages than 9V (from the previous 15V) and increased the success rate from 56% to 84%. The outlook of the forward direction is to explore new methods, adding additional performance-enhancing layers such as ETL and HTL. Their addition is yet to be realized as the lamination method is currently only possible for up to two spin-coated layers, but with more adaptive methods such as transfer-printing, novel OLED fabrication methods on a vacuum-free environment can be explored.

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