Fabrication of natural DNA-containing organic light emitting diodes

Eliot F. Gomez[a], Hans D. Spaeth[a], Andrew J. Steckl[a] and James G. Grote[b]
[a] Nanoelectronics Laboratory, University of Cincinnati, Cincinnati, OH USA 45221-0030
[b]US Air Force Research Laboratory, Wright-Patterson Air Force Base, Ohio 45433-7707 USA

ABSTRACT

The process of creating natural DNA-containing bio-organic light emitting diodes is a fascinating journey from salmon fish to the highly-efficient BiOLED. DNA from salmon sperm is used as a high-performance electron blocking layer, to enhance the efficiency of the BiOLED over its conventional OLED counterpart. An overview of the BiOLED fabrication process and its key steps are presented in this paper.

Keywords: deoxyribonucleic acid, DNA, CTMA, BiOLED, OLED, device fabrication, thin films

1. INTRODUCTION

Organic light emitting diodes are being developed for many applications including solid state lighting and a variety of displays ranging from smartphones to TVs. The incorporation of natural DNA in OLED structures has resulted [1] in significant improvements in both light output (brightness) and emission efficiency [2]. In this paper, we review the processes used for BiOLED fabrication with particular attention to the transformation of raw natural DNA into thin films which can be incorporated into BiOLEDs [3]. A typical DNA-containing BiOLED structure is shown schematically in Fig. 1. The structure is built on a glass substrate which is coated with a transparent electrode of indium-tin oxide (ITO). A conductive organic polymer (PEDOT:PSS) layer is deposited by spin-coating in order to facilitate injection of holes from the anode (ITO). A DNA (modified with a surfactant) thin film is deposited next, also by spin-coating. The device is then introduced into an ultra-high vacuum system where the remaining organic layers and the Al cathode are deposited by thermal evaporation.

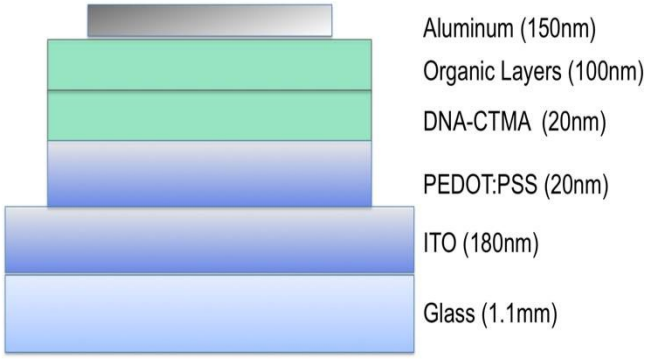

Figure 1. Schematic diagram of typical BiOLED structure, including layer thicknesses.

2. DNA PROCESSING

The DNA to be used in BiOLEDs begins as waste salmon sperm, milt, and roe sacs. These materials are available in large quantities and at relatively low cost, as they are undesirable for human consumption, unlike the salmon meat and eggs. As shown in Fig. 2a, the processing begins with DNA extraction, followed by the removal of protein from the material. Once the DNA is in this purified form, it is freeze-dried (Fig. 2b). The freeze-dried DNA powder is the starting material for BiOLED fabrication. At this stage, the DNA strands are in the molecular weight range of millions of

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Daltons. In order to arrive at a desired lower molecular weight (which results in increased conductivity), DNA dissolved in water is sonicated using a Misonix S-4000 sonicator (see Fig. 3) at 90% amplitude (18000J per 10 second cycle) for 10 cycles to achieve a 200kDa molecular weight.

In this form, the DNA remains water-soluble and insoluble in organic solvents, such as alcohols. For use in conventional thin film fabrication methods, aqueous solubility is undesirable since it leads to poor thin film formation by spin coating. To convert the DNA to organic solubility, the DNA is complexed with the surfactant cetyltrimethylammonium (CTMA) chloride (CTAC), via an ion-exchange reaction. This reaction substitutes CTMA for Na ions associated with the DNA base pairs, resulting in DNA-CTMA and NaCl in aqueous solution (Fig. 2c). The resulting DNA-CTMA precipitate is captured by filtering followed by thoroughly rinsing with water to remove the NaCl and any excess CTAC. The DNA-CTMA is then dried under vacuum and low heat, yielding the dry white granules shown in Fig. 2d. Next, in preparation for thin film formation, the DNA-CTMA is dissolved in an organic solvent. The chosen organic solvent for this process is butanol, due to its low vapor pressure and a viscosity suitable for spin-coating. The DNA-CTMA granules are mixed in butanol at a 0.25wt% ratio for 12 hours.

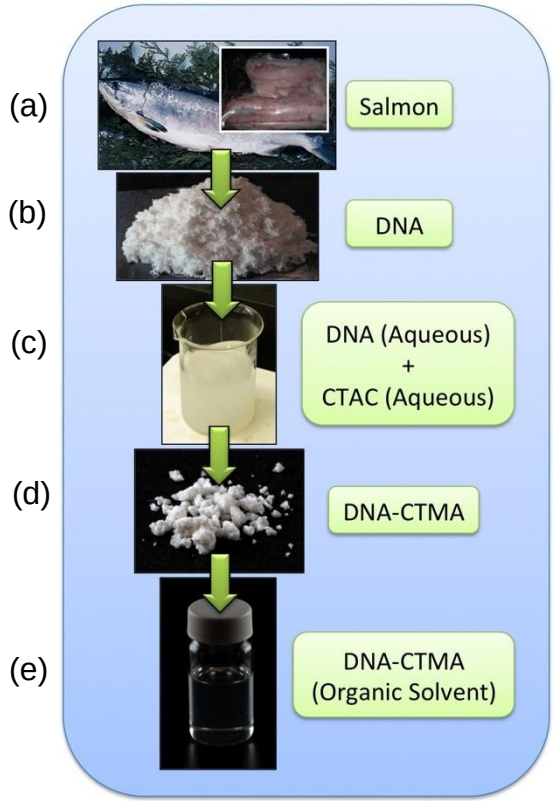

Figure 2. Natural purified DNA is extracted from salmon, purified, freeze-dried, and sonicated. The surfactant CTAC is added to produce the DNA-CTMA complex which not soluble in water but is soluble in organic solvent. At this point the DNA is ready for thin film fabrication [2].

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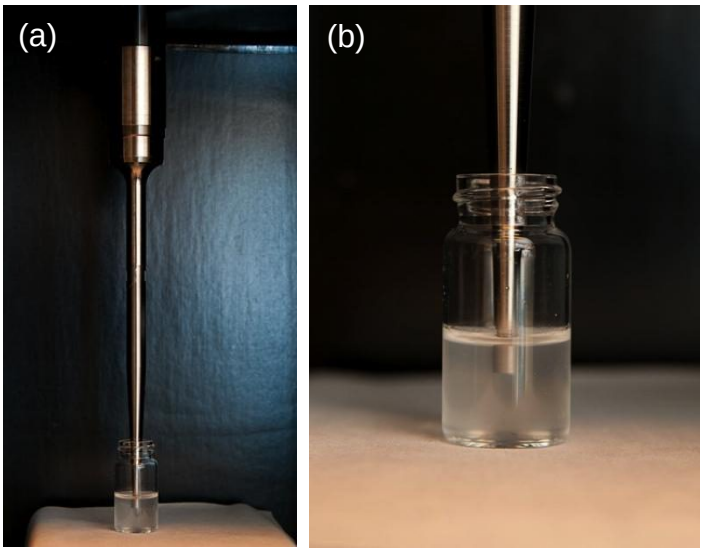

Figure 3. Sonication process used to reduce the DNA molecular weight: (a) Misonix S-4000 sonicator; (b) close-up of sonic horn tip.

3. FABRICATION

Each of the layers in this structure has been selected to perform specific roles in the OLED and has been optimized in terms of thickness. The ITO layer serves as a transparent electrode, patterned to provide one portion of the geometry of the devices. Since ITO is a transparent material light emitted in the device can be outcoupled through the glass substrate. The ITO is sputtered onto a clean 2-inch glass wafer and annealed to enhance both optical transparency and conductivity of the layer. PEDOT:PSS (Heraeus Materials Technology LLC) is the next layer in the device stack and serves as a hole injection layer. PEDOT is filtered and spin-coated on top of the ITO wafer using a Laurell Technologies (WS-400B-6NPP/Lite) Spin Coater at 500rpm for 8 seconds and 2000rpm for 20 seconds as shown in Fig. 4. The PEDOT:PSS is subsequently baked at 125°C for 15 minutes. The substrate is allowed to cool and then the DNA-CTMA solution is spin-coated on top of the baked PEDOT:PSS. The DNA-CTMA in butanol is spread at 500rpm for 8 seconds and then reduced to the desired thickness by spinning at 6000rpm for 20 seconds. The film is allowed to air dry for 10 minutes. The edge of the substrate is cleaned with a cotton swab to remove the unwanted conductive PEDOT:PSS between the electrodes.

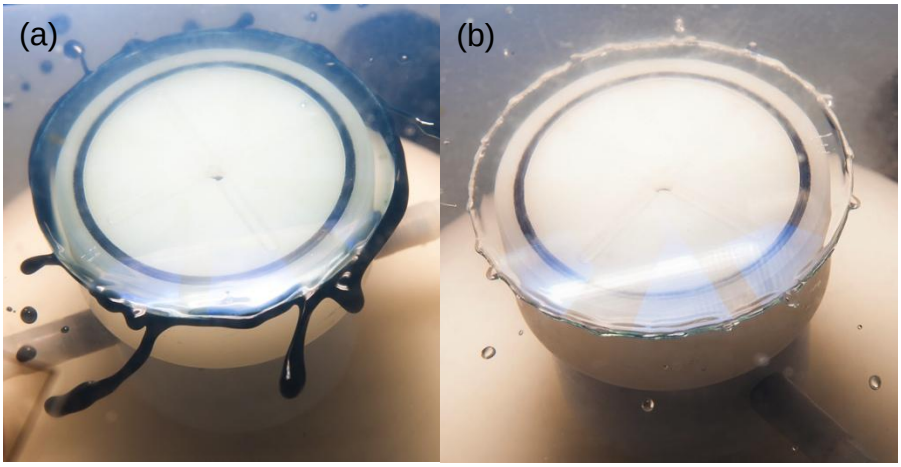

Figure 4. Spin-coating process of layers in BiOLED structure: (a) PEDOT:PSS; (b) DNA-CTMA.

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Fig. 5 shows the major steps for device fabrication, including spin-coating and molecular beam epitaxy (MBE) deposition. Representative photographs after each major step are included in Fig. 5. After spin-coating, the next segment of device fabrication is a stack of small molecule organic layers deposited by MBE through a shadow mask (also shown in Fig. 5). An MBE system, shown in Fig. 6 is an ultra high vacuum system capable of achieving pressures of $\leq 10^{-9}$ Torr. The substrates with the wafers are loaded using a mechanical arm from an easily accessed load lock chamber annexed to the main chamber. Once the substrates are positioned in the main chamber, the substrate rotates to ensure uniform deposition. Effusion cells are shown protruding from the bottom of the main chamber. The effusion cells contain crucibles loaded with the organic materials that evaporate or sublime when heated to a particular temperature. The evaporated or sublimed material plumes out of the crucible and is deposited onto the substrate. The primary advantages of the MBE system are extremely pure material deposition, excellent layer quality, and precise thickness control.

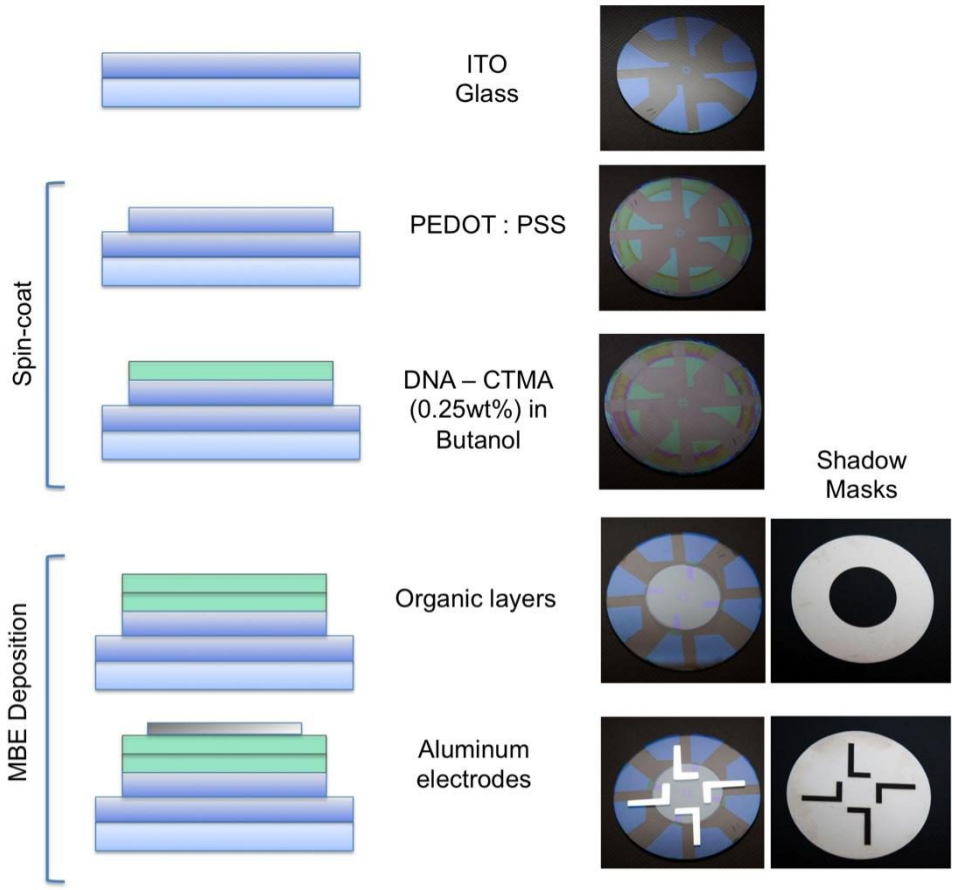

Figure 5. The process of the fabrication of OLED includes the first section of spin-coating PEDOT:PSS and DNA-CTMA; the final steps deposit the remaining organic layers and the aluminum electrodes via thermal evaporation.

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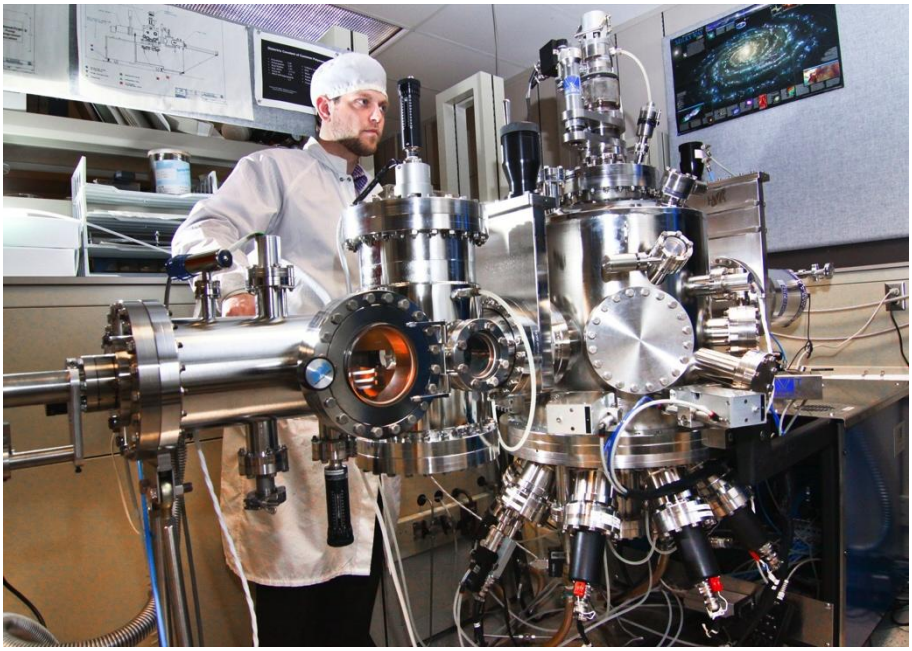

Figure 6. MBE vacuum deposition system to deposit highly purified organic layers and electrodes. The load lock is located on the left half of the system and the main chamber is on the right. The main chamber contains 7 effusion cells located on the bottom flange of the chamber.

The type and the emission color of the OLED determine the order and the materials that are deposited. The materials that are deposited for a green fluorescent device are NPB, Alq_3, BCP, Alq_3, and LiF/Al. The NPB layer serves as a hole transport layer, the first Alq_3 layer as an emitting layer, the BCP as a hole blocking layer, and the second Alq_3 layer as an electron transport layer, and the LiF serves as a transition to the aluminum electrode. The thickness of each layer is an important parameter that can be precisely controlled by deposition temperature, time, and verified by a Inficon XTC/2 crystal monitor. The aluminum electrode layer is MBE deposited through another mask (see Fig. 5), defining the remainder of the device geometry.

4. BIOLED OPERATION

The electrical role of each layer is governed by the HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) levels of the material. These levels are loosely analogous to the concept of band-gap in inorganic semiconductor materials. The HOMO and LUMO levels of the BiOLED, designated in the diagram of Fig. 7, serve to funnel the electrons and holes into the layer selected as the emitting material. The electrons and holes may recombine in the emitting layer to release a photon of the color characteristic of that material.

In conventional OLED structures, a hole blocking layer (BCP) is introduced to enhance the electron-hole interaction in the emitting layer. In the BiOLED structure, we have introduced a DNA film as an electron blocking layer placed on the other side of the emitting layer resulting in further enhancement of radiative recombination. In addition to being an excellent electron blocking layer (due to its high LUMO level), the DNA-CTMA layer has a HOMO level which allows unimpeded hole transport into the emitting layer. Figure 7 also includes photographs of operating BiOLEDs emitting green and blue light.

5. CONCLUSIONS

In this paper we have described the fabrication process which enables the incorporation of natural DNA polymers into organic light emitting diodes. We have shown that this process is compatible with conventional OLED fabrication

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techniques and that the resulting device structure is a very efficient light emitter. In conclusion, we believe that DNA-containing BiOLEDs have a "bright future".

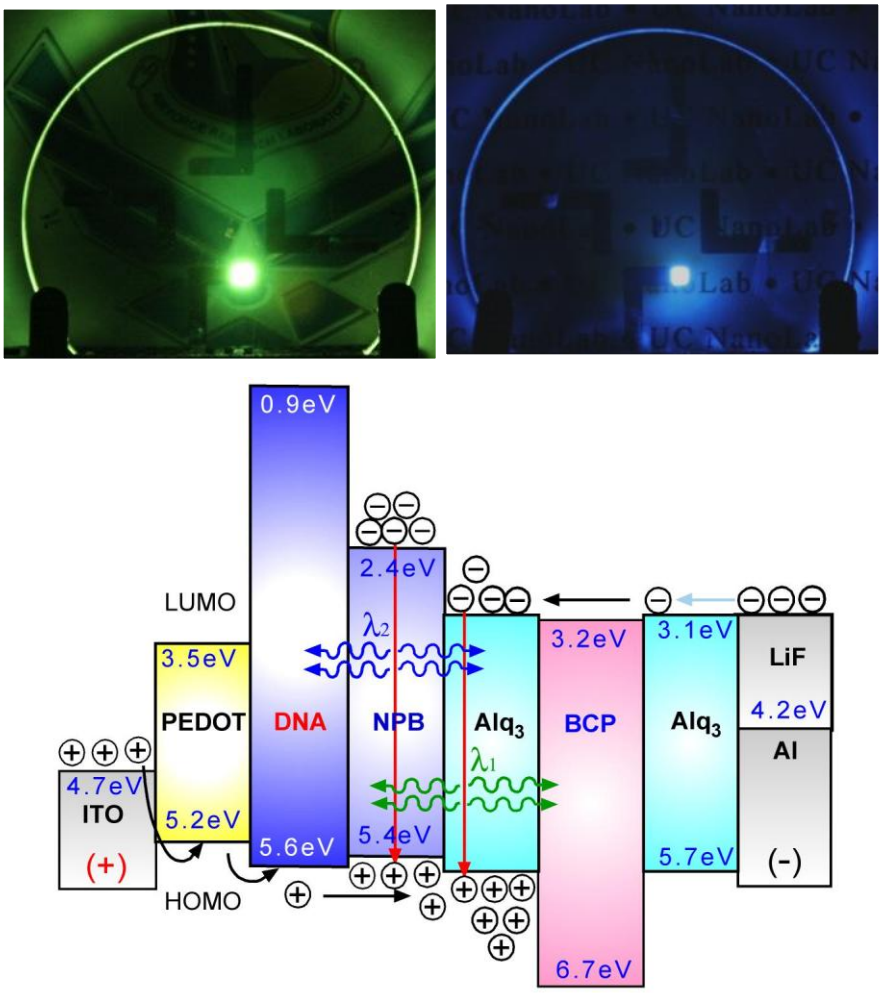

Figure 7. The completed DNA OLED in operation (top) and HOMO-LUMO levels showing the electron blocking properties of the DNA layer [4].

6. ACKNOWLEDGEMENTS

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