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Research Article

Production of organic light-emitting diode with fluorescence featured quinoline derivative

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ABSTRACT

High-priced coating devices limit producing electronic devices and circuit applications widely in laboratories. Simply In this study spin coating technique was used to create surface thin films. Also with this method, an OLED (Organic Light Emitting Diode) device was practically produced. OLED device includes mainly HTL (hole transfer layer), fluorescent layer (light-emitting layer), and an ETL (electron transfer layer). Light-emitting layers in OLED experimental studies are frequently done with commercially produced expensive fluorescence polymers. As an example, MEH-PPV (Poly[2-methoxy-5-(2'-ethyl-hexoxy)-1,4-phenylenevinylene]), Alq3 (Tris-(8-hydroxyquinolinato) aluminum) are mostly known and used fluorescent semiconductor polymers. Alternative to these fluorescent polymers, three different produced quinoline ligand products has fluorescent feature were evaluated. After comparing the fluorescence yields of the produced three complexes, it was seen that 5,7-dibromo-8-hydroxyquinoline has the highest fluorescent response from the others. OLED device production was done with a commercial MEH-PPV (commercial) fluorescent product, and produced (5,7-dibromo-8-hydroxyquinoline). Designed OLED device illumination spectrum was found in the UV (ultraviolet) region. It was concluded that this quoniline product can use as a fluorescent material to produce an OLED device.

Keywords: thin film, OLED, fluorescence, UV, 8-hydroxyquinoline.

Floresans özelliklere sahip kinolin türevi kullanılarak organik ışık yayan diyot üretimi

ÖZ

Bu çalışmada ışık yayan diyot üretimi için spin kaplama tekniği kullanılarak yüzeyde ince filmler oluşturulmuştur. Bu yöntemle pratik olarak bir OLED (Organik Işık Yayan Diyot) cihazı üretilmiştir. OLED cihazı temel olarak HTL (delik transfer katmanı), floresan katmanı (ışık yayan katman) ve bir ETL (elektron transfer katmanı) içerir. OLED deneysel çalışmalarında ışık yayan tabakalar genellikle ticari olarak üretilen fiyatları yüksek olan, floresans özelliklere sahip polimerler kullanılarak yapılır. Bu floresan özelliklere sahip yarıiletken polimerlere alternatif olarak üretilen ve floresan özelliğine sahip kinolin ligand ürünü incelenmiştir. Üretilen üç kinolin kompleksinin floresan verimleri karşılaştırıldıktan sonra 5,7-dibromo-8-hidroksikinolin ürününün diğer iki sentezlenen üründen daha yüksek floresan tepkisine sahip olduğu görülmüştür. OLED cihaz üretimi için sentezlenen bu floresan özellikli kinolin türevi (5,7-dibromo-8-hidroksikinolin) ve MEH-PPV (ticari) floresans özellikli ürün kullanılmıştır. Tasarlanan OLED cihazının aydınlatma spektrumu UV (ultraviyole) bölgesinde olduğu görülmüştür. Bu çalışma sonucunda ince film kaplama ile OLED üretilebileceği görülmüştür. Ayrıca üretilmiş kinolin türevinin bir OLED cihazı üretmek için floresan ara katman malzemesi olarak da kullanılabileceği sonucuna varılmıştır.

Anahtar Kelimeler: ince film, OLED, floresans, UV, 8-hidroksikinolin.

1. INTRODUCTION

The development of semiconductor polymer-based products has significant application potential in the scientific and industrial fields. This technology finds application areas from the production of optoelectronics based organic light-emitting diodes, mobile phone screens, new-generation high-resolution television screens to the photovoltaic cells that generate electricity with solar energy. After the 2000 years of discovering and developing the conductive polymers,[1] innovative studies were increased on organic electronics particularly the semiconducting polymers that were used in optoelectronic devices; such as organic light-emitting

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diodes (OLEDs) and organic photovoltaic cells (OPVs). A luminescent layer between the anode ITO (indium tin oxide) coated glass) and the cathode (aluminum, Ga-In-Eu alloys) forms an OLED device. Applying a voltage (3V to 15V) between anode and cathode would give a characteristic light. Light spectrum isdirectly with luminescent material's bandgap. A band model can be used to describe the OLEDs working principle. When a potential difference is applied between the anode and the cathode; molecules would electrically be transferred into an excited state. In OLED device this excited state would return to the ground state by illumination. The color of the emitted light is determined by the scale of energy gap (Eg) between the HOMO (highest occupied molecule orbital) and LUMO (lowest unoccupied molecule orbital) levels. Photochemical basics of luminescence were explained in the literatüre.[2,3] For the emission of light electric potential must drift charges through the coated polymer layers.

When the band structure is investigated; firstly electrons are injected into the LUMO of molecules close to the cathode and holes are injected into the HOMO of molecules close to the anode. For this process, it is crucial, that the Fermi levels of the electrodes fit well to the energy levels of the polymer layer. Also, intimate contact between all layers is necessary for charge transport. Injected charges drift through the polymer layer from molecule to molecule in opposite directions. These hopping processes are necessary because there are energetic barriers between the molecules, which the electrons have to overcome for an efficient current flow.[4] For the emission of light, a thin layer of the electroluminescent polymer has to be placed in a diode between a transparent ITO anode (indium tin oxide) and a low work function metal cathode. Polymer-based new generation semiconductors can be used instead of high-priced commercially used electro-optic materials such as gallium arsenide (GaAs), cadmium telluride (CdTe), and CdSe (Cadmium Selenide).[4] The development of new organic-based materials will also contribute greatly to the development of electro-optical device technology and sensor technology.[5] For this purpose in this study hydroxyquinoline derivative materials were produced. Synthesized material was used to make an OLED device.

When 8-hydroxyquinoline (8-HQ) complex evaluated it would be seen that, it has got large application fields from medical usage to chemical process applications.[6] 8-Hydroxyquinoline (8-HQ), an important quinoline derivative known as oxine, has been used as a fungicide in agriculture and a preservative in the textile, wood, and paper industries.[7] 8-HQ possesses potent coordinating ability and good metal recognition properties, which means it is widely used for analytical and separation purposes as well as for metal chelation.[8] Its aluminium complex is a common component of organic light-emitting diodes (OLEDs). Variations in the substituents on the quinoline rings effect its luminescence properties.[9] In the photochemically induced excited-state ionic isomers are formed in which the hydrogen atom is transferred from oxygen to nitrogen.[10] The complexes as well as the heterocycle itself exhibit antiseptic, disinfectant, and pesticide properties[11] functioning as a transcription inhibitor. On the other hand, 5,7-dibromo-8-hydroxyquinoline has effective antibacterial, antiprotozoal, antiamoebic, antifungal, bacteriostatic and fungistatic activities, particularly used in treating the intestinal amebiasis.[12] As well as its pharmaceutical importance, 5,7-dibromo-8-hydroxyquinoline is also used in spectrophotometric study and solvent extraction proces.[13]

By this article a preliminary study was described which was performed using organic fluorescence coated surfaces. Synthesized 5,7-dibromo-8-hydroxyquinoline, has shown good fluorescence properties. It was seen that this quoniline product can be used successfully in semiconductor diode device production, after coated as a thin film.

2. MATERIALS AND METHODS

High luminescence efficiency fluorescent products such as MEH-PPV (Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene]), Alq3 (tris-(8-hydroxyquinoline)aluminum)) materials are widely used as an efficient luminescent layer material. In this comparison, OLED device with MEH-PPV product over the PEDOT-PSS ((Poly (3, 4-ethylenedioxythiophene): polystyrene sulfonate) hole transfer layer was also produced. Basically, the OLED circuit is produced over a glass substrate, the ITO (indium tin oxide) layer serves as an anode and there is a luminescent fluorescence coated material over it it, at last layer lithium florür (LiF) the electron transfer layer coated to increase the device luminescence efficiency. The lithium fluoride layer is an electron transfer layer. This thin layer was coated by dropping the LiF–water (0.7 mg/100 mL) solution over the surface and evaporating the water ingredient from the surface in a 70 degree heated incubator for one hour. Finally negative electrical connection was attached over the electron transfer layer. Gallium Indium Eutectic, known as wood metal (at liquid form), is used as the cathode connection contact, which facilitates to use over glass surface layer (Figure 1c).

2.1 Chemicals and reagents

MEH-PPV, "Poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene" was purchased from Sigma-Aldrich, MEH-PPV which has got 2.3 eV band gap, average molecular weight is between 40,000-70,000. Fluorescence wavelength is between 493 nm and 554 nm. Orbital energy HOMO is-5.3 eV and LUMO is -3 eV. Electron transfer layer LiF with 99.995% purity was purchased from Sigma-Aldrich. Poly(3,4-

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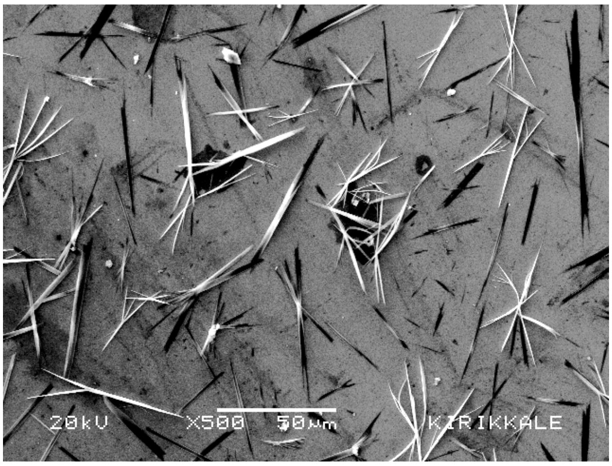
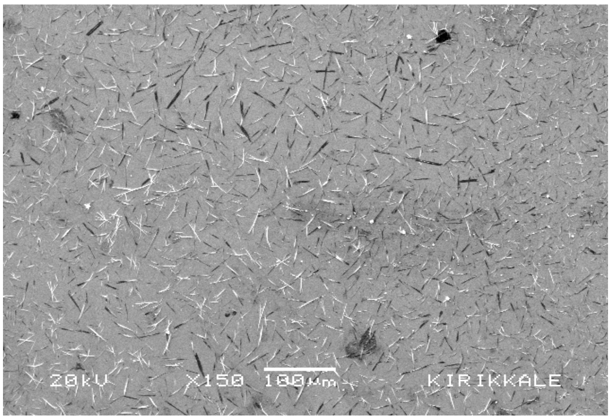
ethylenedioxythiophene) polystyrene sulfonate (PEDOT-PSS) is a widely used (HTL: Hole Transfer Layer) conductive polymer (3-4)% in water. It is widely used in sensor and OLED production stages. This product was also purchased from Sigma-Aldrich. ITO (indium tin oxide) coated glasses with 10 ohm /cm surface resistivity was also used for anode main substrate part, which purchased from Sigma-Aldrich. Liquid metal (Gallium-Indium eutectic, Sigma-Aldrich) was also used for cathode electrical contact connections to work OLED device.



(a)



(b)

Figure 1. Pictures of the spin coated 5,7 dibromo-8-hydroxyquinoline over PEDOT-PSS, ITO glass taken under 500 a) and 150 b) magnifications are shown.

2.2 Preperation of quinoline derivatives

1,2,3,4-tetrahydroquinoline-6,8-dicarbonitrile was synthesized via nucleophilic substitution reactions reported by previous related publications.[14, 15] The 5,7 dibromo-8-hydroxyquinoline was prepared according to literature reports isolated compounds were fully characterized with melting point, elemental analysis, FT-IR, ^{1}H, ^{13}C, HMBC and HETCOR spectroscopy in these paper.[15,16] Also, 8-nitroquinoline was purchased commercially from Sigma Aldrich.

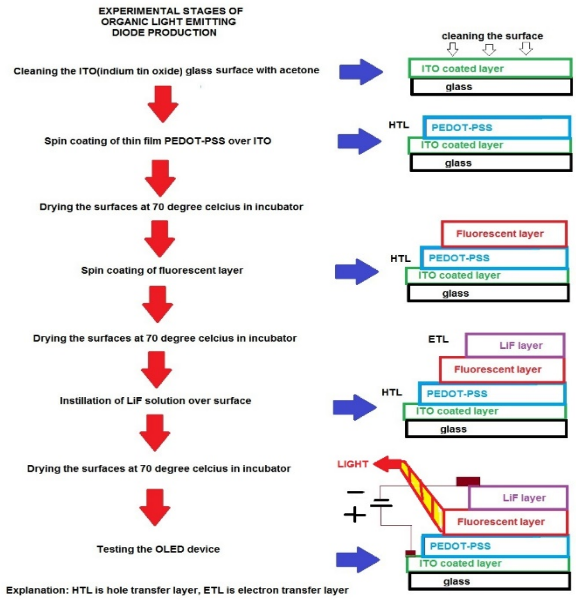
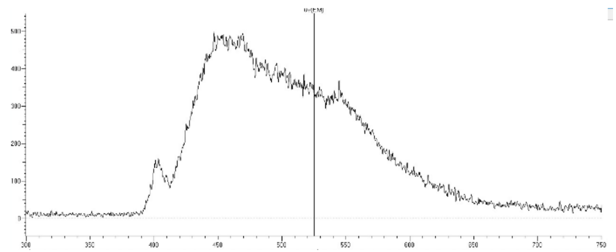


Figure 2. Experimental stages of OLED device production and explanations, ladder type coating prevents short circuit.

(a) (b)



(c)

Figure 3. a) OLED device produced by MEH-PPV with luminescence light b) OLED diode circuit made with synthesized 5,7 dibromo-8-hydroxyquinoline and its operation c) 5,7-dibromo-8-hydroxyquinoline OLED device illumination spectrum.

3. RESULTS AND DISCUSSION

3.1 Chemistry

Quinoline derivatives, determined of their photophysical properties were prepared in recent papers. In brief, 5,7-

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dibromo-8-hydroxyquinoline (c) Figure 4 was obtained by treatment of 8-hydroxyquinoline and molecular bromine in ionic reaction conditions.[6] As seen in Figure 4; 6,8-dicyano-1,2,3,4-tetrahydroquinoline (a) was synthesized by S_NAr reactions in reflux temperature.

3.2 OLED device production

Organic light emitting device experimental sequences were explained in Figure 2. At first stage all surfaces must be cleaned; surfaces were cleaned with acetone and dried in incubator. Coatings were done using a ladder type passing and coating to prevent short circuit between all previous and subsequent layers. Spin coating device was designed with an 12 volt working fan. At second experimental stage PEDOT-PSS polymer layer was spin coated and dried in incubator at 70 degree celcius. At third stage fluorescent layer was coated with spin coating. After drying surfaces at 70 degree celcius LiF layer was coated. LiF is soluble in water this solution was dropped over the fluorescent layer and dried in incubator at 70 degree celcius. After all coatings completed, OLED device was tested and photographed (Figure 3).

When the surface resistance is measured with a multimeter device, it is seen that the bare resistance of the surface with just ITO (indium tin oxide) coated glass is 36 ohm/cm. An home made spin coating device was used for thin film coating over ITO glass surfaces. After the spin-coating of organic quinoline material; surface coating resistance was measured and found as 40 ohm/cm.With the spin coating system application, the homogenous surface coating was achieved. The new coating system was provided a 46 ohm/cm surface resistance on the glass. The surface coating was also examined with the SEM (JEOL-3010) electron microscope system. (Figure 1a, b) Unlike commercial OLED polymers, the newly synthesized 5,7-dibromo-8-hydroxyquinoline (c) was prepared by reaction of 8-hydroxyquinoline with 2 equivalent moles of Br_2 under mild conditions.[6] Also final product 5,7-dibromo-8-hydroxyquinoline solution was prepared with chloroform solvent to coat surfaces. As an OLED substrate; ITO (indium tin oxide) coated glasses were used. PEDOT-PSS second coated layer was used as a hole transfer layer. As the third layer 5,7-dibromo-8-hydroxyquinoline layer was coated over the PEDOT-PSS layer. When the device was completed as explained; for as a test contact to which the negative voltage (cathode) will be given, the circuit was completed and operated with a smooth surfaced aluminum metal surface on the 5,7-dibromo-8-hydroxyquinoline fluorescent layer material. In subsequent tests, it was noticed that the surface should not be scratched. Instead of the aluminum cathode; Ga-In-Eu (wood metal - this is in liquid form) is used to prevent deterioration of the coating. Since this material, which is a liquid metal like mercury, cannot be held by a hand, a pit is formed over the circuit boards in the form of a cavity, and wood metal was placed in it. With this setup on copper, the negative voltage was given over from Ga-In-Eu metal and positive voltage was given over the anode ITO layer. Three different compounds were produced for OLED purposes. These compounds are 6,8-dicyano-1,2,3,4-tetrahydroquinoline (Figure 4a), 8-nitroquinoline (Figure 4b) and another is 5,7-dibromo-8-hydroxyquinoline (Figure 4c).

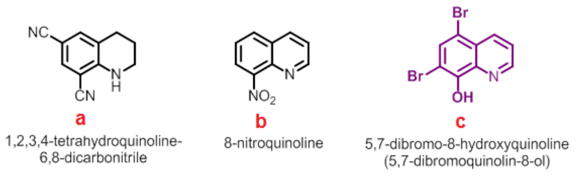

Figure 4. Main structures of substituted quinoline derivatives. Flouresans yield of synthesized three heterocyclic organic ligand substances was compared for production of OLED.

The fluorescence properties of these produced products were examined with a Hitachi-7000 spectro fluorometer device. The experimental parameters used for the measurement are High voltage 900V, scan speed 60 nm / min, Ex-slit 5 nm and Em-slit 5 nm. Fluorescence measurement were found between the 575 nm to 593 nm wavelengths in all three samples. The phosphorescence material named 1,2,3,4-tetrahydroquinoline-6,8-dicarbonitrile (a), the second sample, showed slightly more phosphorus property than the 8-nitroquinoline (b). The first sample had a peak at height of 6300 (arbitrary units) while the second sample showed a phosphorus signal at 7300 (arbitrary units) level. The third sample phosphorus signal peak height exceeded the 10000 limit level in the same measurement parameters, and the Ex-slit and Em-slit setting were reduced from 5 nm to 2.5 nm. In this way, the phosphorus radiation completely measured with the photomultiplier tube of the Hitachi 7000 system. Signal numerical value was reduced and the measurement signal at 1700 was found. The third example 5,7-dibromo-8-hydroxyquinoline (c) with the highest luminescence property of these produced samples was used to made an OLED device.

The phosphorescence peaks of all three produced products are shown in Figure 5 above. The experimental parameters used are High voltage: 700V, Ex -slit = 5 nm, Em-slit = 5 nm, scan speed 60 nm / min. It showed fluorescence in all three materials produced. In 5,7 dibromo-8-hydroxyquinolin sample, the fluorescence spectrum taken between 580 nm and 620 nm was at the highest level (Figure 4). By creating an OLED circuit with this material, it was desired to see the glow of the luminescence led circuit. Prior to this study, the OLED circuit was created using the material Poly [2-methoxy-5- (2-ethylhexyloxy) -1,4-phenylenevinylene], which was commercially available from Sigma-Aldrich, and operated by oled.

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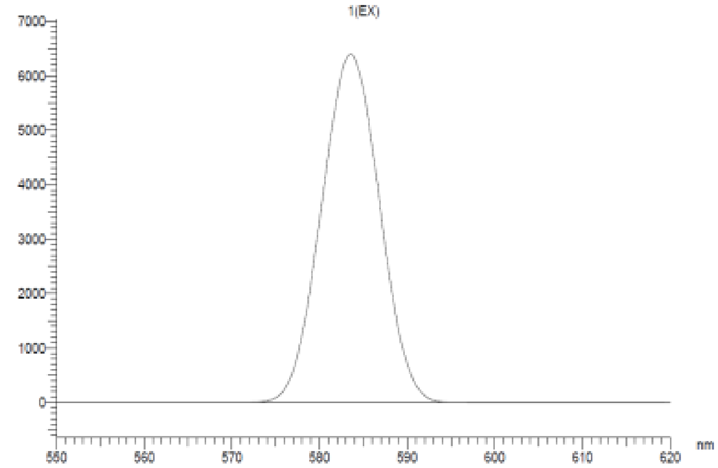
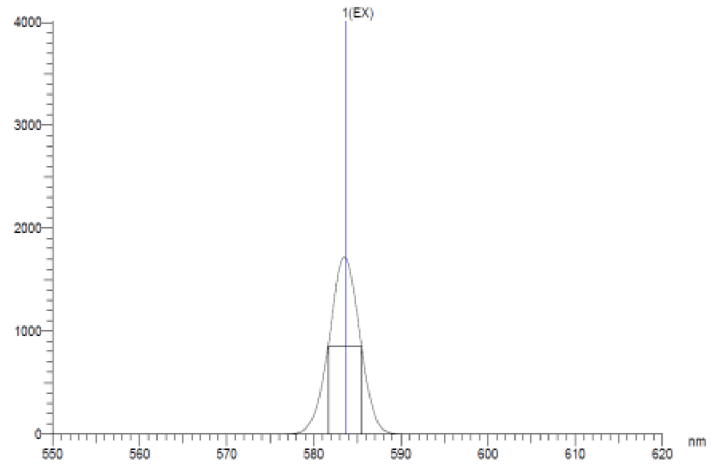
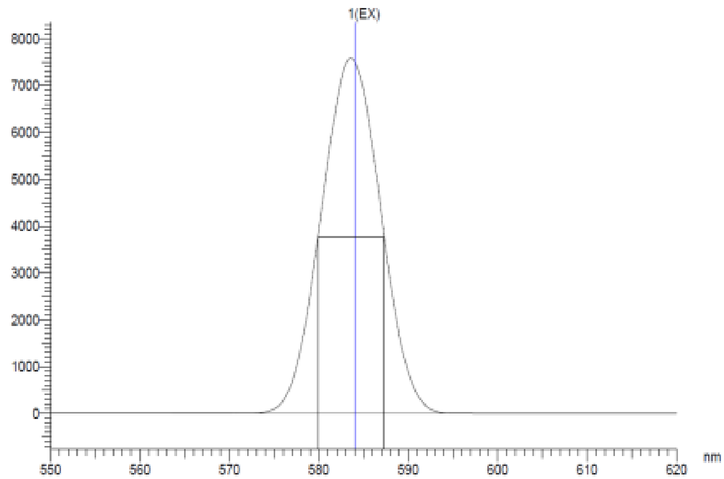
<table>
<tr><th>Fluorescence İntensity (Y axis) vs Wavelength (X axis)</th><th>Material</th><th>Fluorescence intensity (A.U.)</th></tr>
<tr><td>
</td><td>8-nitroquinoline</td><td>6300

Slit 5nm</td></tr>
<tr><td>
</td><td>6,8-dicyano-1,2,3,4-tetrahydroquinoline</td><td>1700

Slit 5nm</td></tr>
<tr><td>
</td><td>5,7-dibromo-8-hydroxyquinoline

This quinoline ligand has the maximum fluorescence response from the others,
This ligand was used for OLED device production</td><td>7300

Slit2.5nm</td></tr>
</table>

Figure 5. Fluorescence measurement peaks of polymer materials produced a) 8-nitroquinoline (b), b) 1,2,3,4-tetrahydroquinoline-6,8-dicarbonitrile (a), c) 5,7-dibromo-8-hydroxyquinoline (c).

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Luminescent light was taken with MEH-PPV product in Figure 4a. This luminescence light worked optimally at 12-13Volt values. When the value of 15Volt is reached, the OLED circuit is broken. Bare ITO surface resistance value is around 10 ohm / cm. After spin coating process over the surface coating was controlled relating the electrical resistance. The electrical conductivity of the 5,7-dibromo-8-hydroxyquinoline coated surface was measured with a multimeter. Surface conductivity was changed between 46 ohm / cm and 40 ohm / cm values over the surfaces. 5,7-dibromo-8-hydroxyquinoline OLED device has opened and experimental picture was showed in Figure 4b. OLED diode light response was defined in the UV (ultraviolet) region and recorded with HITACHI 7000 spectrofluorometer. (Figure 4c). Nowadays similar UV light sources production are necessary to simulate degradation of materials after sun exposure.[17]

4. CONCLUSION

The OLED circuit has also been successfully run. It was seen that surfaces of coated layers are very sensitive to deterioration. Scratching the surface in OLED circuits can cause damaging of the thin coated surfaces and the circuit would not to work. Therefore, taking electrical contact with liquid metals such as Gallium or Gallium Indium Eutectic will also ensure that the circuit is tested without deterioration. Crystal and polymer applications for new generation OLED coating would give the way for applications to be made with these devices as sensors. This preliminary study has shown that the OLED device can produce with limited laboratory equipments. The production of electronic components with high purity semiconductors using p and n type silicon wafers, thin-film coating systems and lithography-based manufacturing techniques require laboratories and production facilities with very high costs today. With polymer coating and using spin coating systems; device production would be easier. High school and university students would have an opportunity to produce electronic devices with the way explained, also using the practical coating techniques explained in this study. Also known or synthesized luminescent materials can be investigated by these methods even within limited laboratory equipments or devices.

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Conflict of interests

There is no conflict of interest with any person, instituon or company, etc.

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