## Fast Thermal Evaporation in Purification of 1,4-Di(pyren-1-ly)benzene

Chung-Yi Hsu,<sup>a</sup> Hung-Yin Lin,<sup>b</sup> Xuan-You Yan,<sup>a</sup> Tsung-Syun Huang,<sup>c</sup> Yan-Kuin Su<sup>c,d</sup> and Thou-Jen Whang<sup>a,d</sup>\* <sup>a</sup>Department of Chemistry, National Cheng Kung University, Tainan 70101, Taiwan, R.O.C. <sup>b</sup>Department of Chemical and Materials Engineering, National University of Kaohsiung, Kaohsiung 81154, Taiwan, R.O.C.

<sup>c</sup>Department of Electrical Engineering, National Cheng Kung University, Tainan 70101, Taiwan, R.O.C. <sup>d</sup>Department of Photonics, National Cheng Kung University, Tainan 70101, Taiwan, R.O.C.

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This work presents a fast purification system that involves a vertical quartz tube and vacuum sublimation for separation without dilution. This system produced re-crystallized 1,4-di(pyren-1-yl)benzene (DPB) in a few hours following the synthesis of pyrene derivatives with a mole yield ratio of 1,4-di(pyrene-1yl)benzene to 1-phenylpyrene (1PP) of 5:1. Differential scanning calorimetry (DSC) and X-ray diffractometry (XRD) were utilized to identify the products. Finally, the lifetimes of pyrene derivatives DPB and 1PP were determined by exponential regression to be  $10.0 \pm 0.14$  and  $3.3 \pm 0.15$  ns, respectively. From the lifetime measurements, the purity of the photo-responsive materials could be easily determined. To prevent red-shift of electro-luminescence (EL) spectra by the energy transfer mechanism, it has acquired to elect transfer layers for pure electroluminescence. A co-catalytic reaction reduced the amount of impurities and eliminated the need for the use of a solvent in the purification step.

Keywords: Purity; Sublimation; Co-catalytic.

## INTRODUCTION

Highly pure crystalline phases are required as new optical materials with high efficiency and electronic mobility for use in such applications as solar cells and infrared photo-detectors.<sup>1,2</sup> Frequently, an annealing process to re-crystallize zinc sulfide (ZnS) has been utilized in the manufacture of light emitting diodes (LEDs), lasers and flat panel electroluminescent (FEL) displays.<sup>3,4</sup> This annealing process has also been applied to improve the transport properties of single-wall carbon nanotubes by reducing the amount of impurities, which considerably affect the electronic structure of the organic materials.<sup>5,6</sup> Conjugation with organic optical materials, in which the p-delocalized electrons are responsible for the intra-molecular conduction, results in the formation of molecular crystals under the influence of the relatively weak van der Waals interactions among molecules. The electronic behavior is determined by the molecular packing<sup>7</sup> and the design of transfer layers.<sup>8</sup> Furthermore, the purity and crystal shape of organic optical materials greatly influence the thermal stability and photo-luminescence characteristics.<sup>9,10</sup>

Chromatographic columns have been widely employed to separate particular products from a crude mixture.<sup>11,12</sup> Recently, vacuum sublimation under temperature gradients has been adopted to separate highly pure optical materials by, for example, growing trans-stilbene and benzophenone single crystals.<sup>13,14</sup> For example, pure 2-(2-hydroxyphenyl)benzoxazolato lithium (LiPBO), pentacene and *sp*<sup>2</sup>-hybridized bistriphenylenyls are purified by physical vapor transport in a horizontal glass tube.<sup>6,7</sup> Although several apparatus have been applied, an accurate understanding of the temperature difference between compounds in the mixture is difficult to obtain, and the purified optical materials are difficult to elucidate.

A series of palladium (Pd)-catalyzed coupling procedures have been developed by Heck, Hiyama, Kumada, Negishi, Miyaura, Sonogashira, Suzuki and Stille.<sup>15-17</sup> The popular catalyzed method for forming aromatic carboncarbon (C-C, C=C, C=C) and carbon-nitrogen (C-N) or carbon-heteroatom bonds is of great importance in various chemical syntheses.<sup>18</sup> The catalyzed method can enhance the yield of synthesized derivatives.<sup>19</sup> To overcome practical limits of reactive efficiency, the Pd-catalyzed formation of bonds is gradually becoming an accepted procedure. Moreover, some related Pd-catalyzed assisted schemes have applied to inactive functional groups and ascend-up

Dedicated to the memory of Professor Yung-Son Hon (1955-2011).



<sup>\*</sup> Corresponding author. E-mail: twhang@mail.ncku.edu.tw

of the transformations.<sup>20-22</sup>

Usually palladium in Suzuki coupling can be pre-catalysts for popular reactions.<sup>23</sup> Nevertheless, specially designed catalysts are essential to the activation of steric hindrance or electronically un-activated compounds. The Suzuki reaction with a catalytic cycling-mechanism involves oxidative addition in advance, followed by a transmetallization step and then reductive elimination.<sup>24,25</sup> The catalytic quantity, reaction solvent environment, reaction temperature and nontoxic nature in environmental applications are discussed.<sup>26,18</sup> The activity of some catalysts is completely determined from their reactive efficiency or evidence of the yield of the reactions in which they participate.

Electroluminescence (EL) and quantum efficiency are affected by the design of the devices and the emission occurs through the doping effect or Förster energy transfer. Of which emitting color will be determined by the excited layer of the device. From the perspective of energy transfer, di-molecular derivatives were frequently discussed.<sup>27</sup> Especially for the di-molecular pyrene derivatives of polycyclic aromatic hydrocarbons, the capability to exhibit better photophysical properties are influenced by the purity of the derivatives.<sup>28</sup> One thing worth to be noted is if heterocyclic rings are not embedded into pyrene derivatives, such as the 1,4-di(pyrene-1-yl)benzene (DPB) while it is insoluble in most of the solvents, it is a major drawback in column chromatography. Therefore, the thermal evaporator system can be applied to cope with this dilemma.

## **RESULTS AND DISCUSSION**

TGA measurements were made by heating the material at a rate 10 °C/min in gaseous nitrogen. The thermal decomposition temperatures ( $T_d$ ) of the 1PP and DPB were 429 °C and 448 °C, respectively, indicating that they were thermally stable at the deposition temperature in the vacuum system. The first DSC trace that was obtained at a heating rate of 10 °C/min revealed that 1PP and DPB had melting transition temperatures of 291.8 °C and 295.3, respectively. In Table 2, DPB and 1PP of the melting temperatures differed by only 3.5 °C still can be completely separated in the system shown on Fig. 1.

An XRD spectrum was shown in Fig.2(a), the annealed sample was held for 2 hrs to induce re-crystallization and the original evaporated main products were compared. The heterological diffraction angles of DPB and 1PP were 16 to 25 degree, respectively. The difference between the diffraction peaks of the re-crystallized and the original



Fig. 1. (a) Setup of the thermal evaporator purification system with a vertical quartz-glass tube. The setup consists of vacuum, cooling, heating systems, the controlling parameters are tuned by vacuum degree, cooling temperature and the temperature of heating tube. The size of chamber is 38 cm in height, 25 cm in width, the radius of manual rotation plate is 8.5 cm and the diameter of heating tube is  $2\sim2.5$  cm. (b) Vacuum pressure and temperature profile during the thermal evaporation. To achieve the degree of vacuum less than  $1 \times 10^{-3}$  torr, the temperature of warm-up stage was kept for a period of time.



Fig. 2. (a) X-ray diffraction (XRD) spectrum of the deposited thin film. Note that diffracted degrees from 16 to 25 of (2) and (3) appeared distinct difference. Another, holding 250 °C in the vacuum for re-crystallization and original sample (2) of deposited 350 °C, was little variation in the crystal morphology. (b) Optical microscope images of the deposited crystallites. Note that 1 and 2 for BDP, 3 and 4 for 1PP, and before thermal annealing for 1 and 3, and after thermal annealing for 2 and 4.

material revealed the stability of the crystal lattice, without the need to hold the temperature in the quartz column at 250 °C and  $1 \times 10^{-3}$  torr for a long time. Fig. 2(a) presents the change in the pattern that was associated with annealing, and the enhanced signals were induced by the preferred orientation of the grown crystal. Figure 2(b) shows the crystal morphology of optical microscopy which can enhance the intensity of the XRD patterns in Figure 2(a).

The spectroscopic properties of DPB and 1PP in solution or as deposited films were measured and the homologous data were presented in Table 2. The maximum wavelengths of emission spectra were 459.3 nm and 455.7 nm for 1PP and DPB, respectively. Comparing the photoluminescence of the reference to the PL spectra from the dissolved solution or deposited film reveals that the spectrum of the film that was prepared by vacuum vapor deposition was red-shifted from that in the CHCl<sub>3</sub> solvent. The major reasons for that are the solvate-chromatic effect and intramolecular charge transfer.<sup>31-33</sup> Photo-electrical materials must be efficiently purified, and purities of over 99.98% by elemental analysis (EA, Heraeus, Vario EL *III*).

Accordingly, lifetime can be measured to determine the wave functions and distribution of electrons in a complex molecule. Such a measurement is useful in studying the dynamics of excited states and the mechanism of quenching collisions. To evaluate the lifetime, the lifetime decay apparatus was set at the photoluminescence wavelength  $\lambda_{max} = 455 \sim 460$  nm. Fluorescence decay curves were plotted using an oscilloscope and then exponential regression fitting of the data was performed to determine the lifetime. In Fig. 3(b), the fitted curves for the evaporated materials had the equations,  $I_{iDPB} = 4.46 exp(-t/\tau_{DPB})$  for DPB with  $1/\tau_{DPB} = 1 \times 10^8$ ,  $I_i = 28.2 \ exp(-t/\tau_{1PP})$  for 1PP with  $1/\tau_{IPP} = 3 \times 10^8$  and  $I_i = 56.4 \exp(-t/\tau_{mix})$  for the equivalent mixture of DPB and 1PP with  $1/\tau_{mix} = 3 \times 10^8$ . Therefore, the lifetime measurement method easily investigates the purity and may be applied to the area of synthesized optoelectronic materials. In Figure 3(b) the more lifetime of DPB and 1PP, the weaker intensities of the signal with moderate decreasing trend will be. However, the intensity of the mixture (1:1 mol%) has a deep decreasing tendency with the lifetime because the intensity is not an additive property. It means the purity will affect the lifetime profoundly.

The crude and purity material related to photo-physical properties, as shown in the Fig. 4(a). Therefore, the trait peak of absorption wavelength, in proper order near 350 nm of absorption wavelength with DPB and 1PP, and step up 350 nm for excited wavelength placed in the above sort. The maximum wavelength of the crude material lay between DPB and 1PP, while the full width at half maximum (FWHM) of the crude and pure materials is almost twice of DPB or 1PP. In the PL spectrum of Fig. 4(a). The maximum of the peak in 1PP is red-shifted about 5 nm compared with that of DPB. It is presumably ascribed to overture of that the content of intra-molecular charge transfer (ICT) effectively or aggregation of coplanar configuration. In the ultra-violet photoelectron spectrum (UPS) measurements, the investigation of the highest occupied molecular orbital of energy level (E<sub>HOMO</sub>) in DPB and 1PP was 5.68 eV and 5.60 eV, respectively. Accordingly, the lowest non-occupied molecular orbital of energy level (E<sub>LUMO</sub>) could be estimated from the optical band gap  $(E_g)$  of DPB (3.10 eV) and 1PP (3.02 eV) to be the same 2.58 eV same in both DPB and 1PP from the formula  $E_{LUMO} = E_g + E_{HOMO}$ 

The device was deposited in order of a patterned etching indium-tin-oxide (ITO) substrate as follows; (A) ITO /



Fig. 3. (a) Scheme of photon-path of lifetime measurement, (b) Exponential fitting curve for lifetime τ value of purified sample evaporated at 350 °C, mixing material (DPB: 1PP = 1:1) at 350 °C and the product evaporated at 330 °C were 10.0 ± 0.14, 3.3 ± 0.15 and 3.3 ± 0.08 ns, respectively.

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Fig. 4. (a) The photoluminescence (PL) which the excited wavelength is 350 nm. (b) The electrical characteristics of current–luminescence–voltage (I-L-V) curve measurements; (c) The electroluminescence of the proposed devices.

NPB (40 nm) / DPB (30 nm) / BCP (10 nm) / Alq3 (30 nm) / LiF (1 nm) / Al and (B) ITO / NPB (40 nm) / DPB (30 nm) / BCP (15 nm) / LiF (1 nm) / Al. The final deposited device on a patterned etching ITO substrate included ten equal effective blocks of 3.6 mm<sup>2</sup> each.<sup>34-36</sup> Figure 4(b) is the plot for the measurement of the I-L-V curve (current–brightness–voltage) from the electrical characteristics of the device. When investigating the devices A and B, the initial voltages of the characteristic features were 3.5 V and 4.0 V, respectively. The current density can be distinguishable after 6.0 V, and 258 mA/cm<sup>2</sup> and 500 mA/cm<sup>2</sup> of 9.0 V in device A and B, respectively. The luminance of one device is similar to that of the other, but device B was brighter in terms of luminance at a given current density than was device A.

Except a deposited layer of Alq3, several parameters of the deposited layers, including deposition rate, substrate temperature, and thickness were all the same between devices A and B. As shown in Fig. 4(c), for device A, the CIE Hsu et al.

1931 chromaticity coordinates were (x = 0.23, y = 0.27) and the maximum EL intensity was at 492 nm. For device B, the CIE 1931 chromaticity coordinates were (x = 0.17, y = 0.21) and the maximum EL intensity was at 461 nm. When addition of an Alq3 layer in device A, the electroluminescence spectrum was red-shift compared with device B.<sup>34</sup> Consequently, the electron transfer layer must be selected very carefully to avoid the overlap and Förster energy transfer by the fluorescence excited from emitter in the OLED device.

To improve the EL (electroluminescence) and quantum efficiency in the range of visible light, a single layer of PL (photoluminescence) dyes or doped emitted material was used in an electron transfer layer. It is necessary to use the doped material united with higher extinction (or absorption) coefficients and fluorescence of quantum yield to be in accordance with the Förster energy transfer of emission. Of which emitting color will be determined by the excited layer of the device. From the perspective of energy transfer, di-molecular derivatives formed from pyrene with higher energy gap can appropriately excite a material with lower energy gap to emit fluorescence. Di-molecular pyrene derivatives of polycyclic aromatic hydrocarbons, exhibit better photo-sensitivity, heat tolerance, electrical conductivity, emissive capacity, stability and character of physiology response, when purity of the derivatives has been promoted.

For the conventional chromatographic columns, silica gel has to be chosen carefully and it is tedious to optimize the ratio of solvents for elution efficiency. Even more, it may take the risk of losing product since it is likely to be adhered to silica gel. Generally, the best recycling ratio is about 50% and the most of the byproducts are not retrievable so that it is unfavorable for investigating the reaction mechanism. Therefore, it is impossible to recycle the materials because of contamination and it has huge impact on environment. Thermal evaporator purification system not only provides the solution to avoid the contamination discussed above and it is also time-saving in the process. The purification yield can be improved especially for the compounds that are not dissolvable in solvents, and the crystallization will be greatly enhanced by adopting this system for purification.

### **EXPERIMENTAL**

In this study, a vertical quartz-glass tube was used with a thermal evaporator system to increase the efficiency Fast Thermal Evaporation in Purification

#### Scheme I



Synthesis process of 1,4-di(pyrene-1-yl)benzene, including (a) bromination, (b) pyrene boronic ester and (c) suzuki coupling reaction. <sup>*a*</sup> The yield is shown on Table 1. <sup>*a*</sup> Abbreviations: NBS = n-bromo succinimide, DMF = dimethylformamide, THF = tetrahydrofuran, and *n*-BuLi = *n*-butyllithium.

of purification of pyrene derivatives. Scheme I represents a protocol for synthesizing pyrene derivates by the bromination of pyrene, which was the main reactant in the formation of N-bromosuccinimide (NBS) in N,N-dimethylformamide (DMF) solvent; the corresponding pyrene boronic acid was formed by the re-crystallization of 1-bromopyrene to substitute a halogen-lithium salt, which can react with n-BuLi at -78 °C in anhydrous tetrahydrefuran (THF). Following the reaction, B[(CH<sub>3</sub>)<sub>2</sub>CHO]<sub>3</sub> was esterified with pinacol, yielding the boronic ester after purification by acid pickling (~75% yield). Finally, the Suzuki coupling reaction between the 1,4-diiodobenzene and pyrene boronic ester yielded the target compound 1,4-di(pyrene-1yl)benzene (DPB) and the by-product 1-phenylpyrene (1PP). The yield efficiencies of the two compounds were affected by the catalytic diversity. Table 1 presents the relationship between the different catalysts and the yields. It shows that the use of co-catalyst  $(PdCl_2(PPh_3)_2 / PdCl_2(CH_3CN)_2)$  can attain to more possession with the main products. Both pyrene derivatives can be purified by thermal evaporization using a vertical quartz-glass tube. The structures of these compounds were elucidated by <sup>1</sup>H NMR, mass spectrometry and elemental analysis.

Figure 1(a) shows the setup of the thermal evaporator purification system with a vertical quartz-glass tube. The overall apparatus includes vacuum, cooling, thermal evaporator and detector systems, the control parameters were tuned by vacuum value, cooling and heating tube temperature of material sector. The vacuum system contains a rotary pump and a diffusion pump, when the vacuum value less than  $5 \times 10^{-2}$  torr was achieved by the rotary pump and the diffusion pump reached warming up to 100 °C the system keeps vacuuming until less than  $1 \times 10^{-3}$  torr. The evaporating only proceeds at this stage. The cooling water starts circulating to keep thermostated at 24 °C when the diffusion pump went into working. The control temperature of thermal evaporator system, as shown in Figure 1(b) presents the use of temperature gradient and vacuum evaporation to separate impurities from pyrene derivatives. After the synthesis process, the residue reactant and impurities

Catalyst /mol % / Derivatives yield %	Pd(PPh <sub>3</sub> ) <sub>4</sub>	PdCl <sub>2</sub> (PPh <sub>3</sub> ) <sub>2</sub>	PdCl <sub>2</sub> (CH <sub>3</sub> CN) <sub>2</sub>	PdCl <sub>2</sub> (PPh <sub>3</sub> ) <sub>2</sub> / PdCl <sub>2</sub> (CH <sub>3</sub> CN) <sub>2</sub>	
	5~10	3~5	3~5	1~2	1~2
1PP	20%	25%	10%	10%	
DPB	50%	60%	50%	85%	

Table 1. The yield of synthesized pyrene derivatives from various catalyst<sup>a</sup>

 $^{a}$  The yields of derivatives determined with quantity of decomposition temperature (T<sub>d</sub>) as an internal standard.

Table 2. Photoluminescence and thermal properties of synthesized pyrene derivatives

Evaporation	Photoluminescence (nm)			Thermal properties (°C)		
Temperature (°C)	$\omega_{\rm F}{}^a$	$\lambda_{ m f}^{\ b}$	$\lambda_{s}^{c}$	$T_d^{\ d}$	$T_m^{e}$	$T_c^{f}$
330 °C	$58.5\pm0.5$	$459.3\pm0.6$	$450.7\pm0.6$	$429.3\pm0.6$	$291.8\pm 0.3$	$166.0\pm1.0$
350 °C	$62.7\pm0.3$	$455.7\pm0.6$	$453.3\pm0.6$	$448.0\pm1.0$	$295.3\pm0.5$	$180.6\pm0.6$

<sup>*a*</sup>  $\omega_{\rm F}$ , full width at half maximum (FWHM); <sup>*b*</sup>  $\lambda_{\rm f}$ , maximum of emitted wavelength in the deposited film; <sup>*c*</sup>  $\lambda_{\rm s}$ , maximum of emitted wavelength in the solvent of CHCl<sub>3</sub>; <sup>*d*</sup> T<sub>d</sub>, decomposition temperature; <sup>*e*</sup> T<sub>m</sub>, melting temperature; <sup>*f*</sup> T<sub>c</sub>, crystallization temperature.

which has low-decomposition points were sublimed when heating up to 200 °C from room temperature. When the vacuum value was reduced obviously the temperature maintained constant until the vacuum value was reduced further for a period of time. Then the separation of compounds which has lower sublimation points was accomplished through sublimation when heating up to 300 °C. Therefore, by manipulating the vacuum value parameter through rising and maintaining temperature the following heating stages up to 300 and 350 °C can be completed. Impurities are usually separated effectively from solid crude if their vaporization pressures differ sufficiently. The pyrene derivatives were placed in a steel boat (sample container) under a vertical quartz tube, which was thoroughly cleaned with acetone, and then heated in steps in a dynamic vacuum, which was produced by a diffusion pump (ANELVA Model-300), in a vacuum chamber, to temperatures of 200, 300, 330 and 350 °C. Three heating coils around the quartz tube and under the crucible provided the temperature gradient. Impurities first sublimed on top of the substrate panel when it was heated to 300 °C, and then the 1PP (by-product) was evaporated at 330 °C. After the fore-mentioned impurities and by-products had been removed, the system was cooled to room temperature. Four heaters (three located in the quartz tube, one located in the crucible) were attached to the side of the quartz column and their temperatures were set to 350, 250, 350, and 25 °C, respectively. The DPB (main product) was introduced to the middle level of the quartz tube from the crucible at an evaporation temperature of 350 °C. The sample was re-crystallized on the surface of the quartz column by maintaining the temperatures of the second and third heater for two hours. The separated impurities, 1PP and purified DPB were collected either by rotating the Corning glass substrate on the top of the quartz column or on the inner surface of the quartz column when the sample was in contact with a colder surface.

Thermo-gravimetric analysis (TGA, Perkin-Elmer Pyris 1) and differential scanning calorimetry (DSC, Perkin-Elmer Pyris Diamond) were employed to evaluate the thermal properties of materials. According to experience value and approximate calculation, 80~100 °C below the decomposition temperature for the material is the maximum reference point for gradient temperature, and thereby characterize the product of the reaction and the compounds that were separated each other by temperature gradient vacuum evaporation.

Glass transition temperatures  $(T_g)$ , crystallization

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temperature ( $T_c$ ) and, melting temperature ( $T_m$ ) were determined with a differential scanning calorimeter (DSC, PerkinElmer DSC-7) at a heating rate of 5 °C/min. Decomposition temperature ( $T_d$ ) was obtained from TGA measurements using a PYRIS Diamond TG/DTA-10 with a heating rate of 20 °C/min. Highest occupied molecular orbital (HOMO) levels of the new compound were measured directly by using ultraviolet photoelectron spectroscopy (UPS, Riken Keiki AC-2), while the lowest unoccupied molecular orbital (LUMO) values were determined from the HOMO and the lowest energy absorption edge of the UV absorption spectra. Absorption and fluorescence spectra were recorded using a Hitachi-U3010 UV–Visible spectrophotometer and a Hitachi-F4500 Luminescence

An X-ray powder diffraction (XRD, Shimadzu XRD-700) spectrum of the deposited film on the Corning glasssubstrate was obtained at room temperature with a  $2\theta$  range from 5 to 50 with Cu K $\alpha$  radiation with a wavelength of 1.5418 Å. Each diffraction peak in the spectrum from 5° to 50° corresponds to two pyrene derivates. X-ray diffraction was used to identify the crystal lattice of the organic optical material.

The fluorescence lifetime investigation can be applied to characterize a pure material from its spectral time profile on the solution and the solid film.<sup>29</sup> According to the spontaneous emission transition probability formula (1)

$$N_i(t) = N_{i0} \exp(-A_i t),$$
 (1)

where the  $N_{i0}$  is photon counting of fluorescence at t = 0,  $N_i(t)$  is photon counting of fluorescence at t = t and  $A_i$  is the Einstein coefficient. Because the population is proportional to the fluorescence intensity, formula (1) could be changed to  $I_i(t) = I_{i0} \exp(-t/\tau)$ , where  $I_{i0}$  is the fluorescence intensity at t = 0,  $I_i(t)$  is the fluorescence intensity at t = t and  $\tau = 1/A_i$  defined as lifetime is the time which the population  $N_i$  decays to 1/e of the original value at t = 0.<sup>30</sup> In Fig. 3(a) a pulse excitation source, which is the third harmonic oscillation from an Nd-YAG laser with a wavelength of 355 nm, was used to excite the samples and to obtain the lifetime information.

The lifetime of the wavelength that was emitted by the pyrene derivatives was measured and electrical characterization was performed to ensure optical performance.

Materials of optoelectronic device were thermally deposited in a thermal evaporator (ULVAC, model TU020206). Those materials were N,N'-di(1-naphthyl)-N,N'-diphenyl-benzidine (NPB), (tris-(8-hydroxyquinoline)aluminum(III) (Alq3), 1,4-di(pyren-1-yl)benzene (DPB), 2,9dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP), and lithium fluoride (LiF). They played the roles as holes transport, electrons transport, blue emission, holes blocking, and insulating, respectively. The electrical characteristics of the device obtained using a programmable voltage-current source (Keithley 2400) and a scan spectrometer (Photo Research, PR650) to measure the I-L-V curve (current– brightness–voltage).

### CONCLUSIONS

Rapid developments in photoelectric science and technology are continually leading to the creation of new photoelectric materials. Not only must their material characteristics be optimized, but also their purification must be made increasingly efficient. This investigation enhanced the yield of the co-catalyst of the Suzuki coupling reaction using purification with a vertical quartz tube-evaporation device, which rapidly separates DPB and 1PP from their mixture. This method provides a faster, more yields, higher efficiency and less pollution than separations compared to traditional chromatographic columns. Improving the rate of synthesis and reducing the amount of solvent used in the purification are environmentally friendly. These parameters of purification process must be evaluated to provide a valuable application in high purified photoelectric materials which is needed to be a high performance photo-electrical device.

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