



## Optimal design and fabrication of ITO/organic photonic crystals in polymer light-emitting diodes using a focused ion beam

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### ABSTRACT

This study aims to achieve high directionality and large extraction of light emission from polymer light-emitting diodes via optimizing photonic crystals (PCs). The optimization is achieved through the optical modeling using a 3D finite-difference time-domain (FDTD) method and the intelligent numerical optimization technique, genetic algorithm (GA). In results, the PC period, depth and filling factor can be found. The FDTD method has been proven very effective for modeling complex multilayer PLED devices in which the electron-transporting layer is only a few tens of nanometers away from a metallic layer. On the other hand, GA is a powerful tool to cope with a complicated optimization problem with multiple variables to optimize. The optimized PCs are later fabricated by focused ion beams (FIB) for the PLED. With desired PCs, an increase of the extraction efficiency in excess of 46% is achieved experimentally and the 3D FDTD calculation explains this result faithfully.

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### 1. Introduction

This study is aimed to fabricate polymer light-emitting diodes with high directionality and large extraction of light emission. For applications of large-area flexible displays and illumination lights, polymer light-emitting diodes (PLEDs) serves as a very important technology due to the advantages such as low driving voltage, high response, high luminance, wide viewing angle and low cost [1–6]. This PLED is an organic electronic component made by combining organic and inorganic materials, such as organic luminescent layers, organic electron hole injection layers, a metal electrode, and a transparent oxide electrode [4]. Nonetheless, PLEDs have yet to be developed to meet the overall efficiency standards required by industry. Efforts have been devoted to improve the performance of PLEDs in functionality, operating mechanism, and fabrication methods for each of the organic and inorganic materials used for constructing the PLED. Among them, improving the luminous efficiency of PLEDs is the key issue which must be resolved if PLEDs are to be widely commercialized, since both low power consumption and long operational lifetime are required for a variety of displays.

In recent years, there has been increasing interests in photonic crystal (PC) structures for PLEDs, a typical of which is similar to our design is shown in Fig. 1a. The advantages offered by PC extraction are that, when designed properly, the emission direction can be tailored, resulting in increasing light emission in useful directions and reducing reflection loss. However, in this way, the performance improvement level by PC-LEDs is limited to factors of 2.5–3 times, despite much higher efficiency increase has been predicted theoretically. These limitations are due in part to a sub-optimized vertical design that does not achieve maximum interaction of the guided modes with the PC, and the use of non-optimal lattice size and other parameters. When the periodicity is chosen correctly, the modified in-plane wave vector falls within the air escape cone, resulting in extraction to air at an angle dependent on the specific lattice parameters within this range. This allows one to tailor the emission profile of the PLED with the PC lattice parameters. In this work, the light extraction increases higher than previously demonstrated [5], in addition to directional control of emitted light by the improved tailoring of both the vertical structure and the PC lattice.

Currently, most of interests are in 2D PC structures, since they are suitable to standard planar fabrication technologies. There have been much modelings of various PC structure types from square lattice to Penrose' type lattices [6]. Due to the limitations of current lithographic and pattern transfer processes, these structures have not been fully investigated experimentally. For example, the direct write e-beam lithography is quite flexible. It still requires pattern

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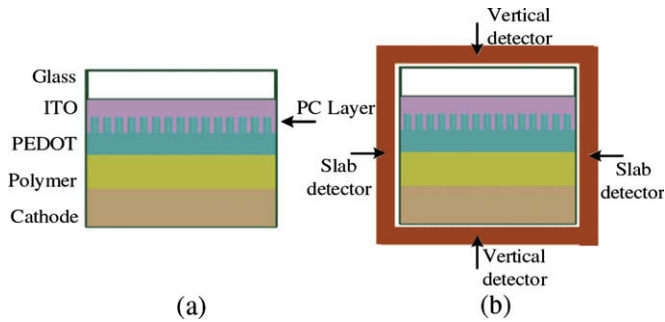


Fig. 1. (a) Schematic of PC-PLED with ITO/PEDOT photonic crystal layers. (b) Emission efficiency detection system.

transfers through multiple mask layers before applying standard etching techniques (RIE/ICP). Therefore, an alternative nanomachining technique of the focused ion beam (FIB) [7] is employed in this study, which requires no mask as the component is etched directly and thus implementing conceptual designs in relatively fast speed. For PC fabrication, it is necessary to etch accurate and repeatable structures via FIB, and also important to understand the limitations of FIB etch process and design the component with these in mind.

## 2. Materials and methods

This work uses the numerical algorithms to establish optical models for evaluating and optimizing PC performance. The algorithms include the finite-difference time-domain method (FDTD) for optical modeling and genetic algorithm (GA) for optimization. There are four steps to achieve the goals, which consist of (1) the FDTD simulation, (2) the light extraction efficiency calculation/analysis, (3) optimal design of PCs by GA, and (4) fabrication.

### 2.1. Simulation via finite-difference time-domain

The designed PLED structure as depicted in Fig. 1 is in a multilayer sandwich structure that comprises a glass substrate, an ITO anode, organic emitting layers, and a metal cathode. To accurately predict light extraction of the PLED, the numerical simulation tool, the finite-difference time-domain method (FDTD) [8–10] is employed. Yee [10] introduced the FDTD method to be used to solve Maxwell's equation. The FDTD method for solving Maxwell's equations has been useful to compute electromagnetic in the time-domain. This method has further been proven very effective for modeling complex multilayer PLED devices [11], in which the electron-transporting layer is only a few tens of nanometers away from a metallic layer.

The FDTD applied herein for the PLED structure in Fig. 1 assumes no optical losses from the organic/inorganic polymer transparent layers, and emits light reflected at the metal interface. The emitted excitons inside the PLED are modeled in FDTD as 20 fs long Gaussian oscillating dipole pulses that have a center frequency of 530 nm and a full bandwidth at half maximum of 50 nm. Since the spatial distribution of excitons in real PLEDs act in such manner that they can be treated as incoherent sources, the pulses used in the FDTD calculations would mimic realistic emission properties successfully, provided the spectral distribution is similar to that of the real source. In our FDTD calculations, sufficient dipole sources with equal numbers of mutually orthogonal  $x$ -,  $y$ -,  $z$ -polarizations are assumed distributed randomly throughout the active area. With light sources assumed, the terms in Maxwell's equations are next represented in difference forms, including those pertaining to magnetic and electric fields [9]. The ensuing calculations follow the proce-

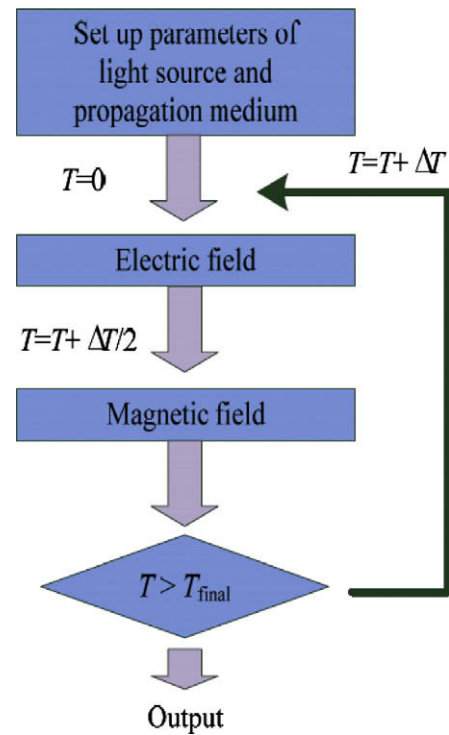


Fig. 2. FDTD process.

cedure illustrated in Fig. 2. The parameters and light source are first set up, which is followed by simulation on electrical field at a given time  $T = 0$  and magnetic fields at the time in a lag of half computation time interval,  $\Delta T$ . Simulation results of electrical and magnetic fields are checked if the results are unsatisfied for that  $T$  greater than  $T_{final}$ , for the re-start of simulation in the next round. This calculation is repeated until  $T$  reaches the final time  $T_{final}$ .

### 2.2. Light extraction efficiency

With the optical modeling via FDTD completed and then successfully predicting light emission profile on the top surface of the PLED, the next step is to evaluate light extraction efficiency via computing emission efficiency within 30 cone degree of the top emission profile. The computed efficiencies would be used by later GA optimization to seek optimal pitch and size of photonic crystals. In this way, favorable emission directionality and extraction efficiency can be achieved.

The total external emission efficiency  $\eta_{ex}$  is commonly defined as product of the internal quantum efficiency  $\eta_{in}$  and light extraction efficiency  $\eta_{ext}$  [1]; thus,

$$\eta_{ex} = \eta_{in}\eta_{ext}. \quad (1)$$

Note that the internal quantum efficiency  $\eta_{in}$  is mainly decided by the physical properties of the thin polymer layer (as shown in Fig. 1) and taking consideration the losses due to side reflection and absorption; thus,  $\eta_{in}$  is no way improved by varied designs of photonic crystals. Therefore, the current study focuses on elevating extraction efficiency  $\eta_{ext}$ , possibly by better design pattern and size of photonic crystals. In this way,  $\eta_{ex}$  can finally be increased.  $\eta_{ext}$  can be computed by the FDTD technique, stated in the previous subsection. The computed  $\eta_{ext}$  is ready to be optimized by GA. Note that Eq. (1) reflects that only a small fraction of the total photons generated inside the PC-PLED films could escape from the internal structure, due to total internal reflection and wave-guiding effects of high-index layers. In fact, the light extraction efficiency of PLEDs

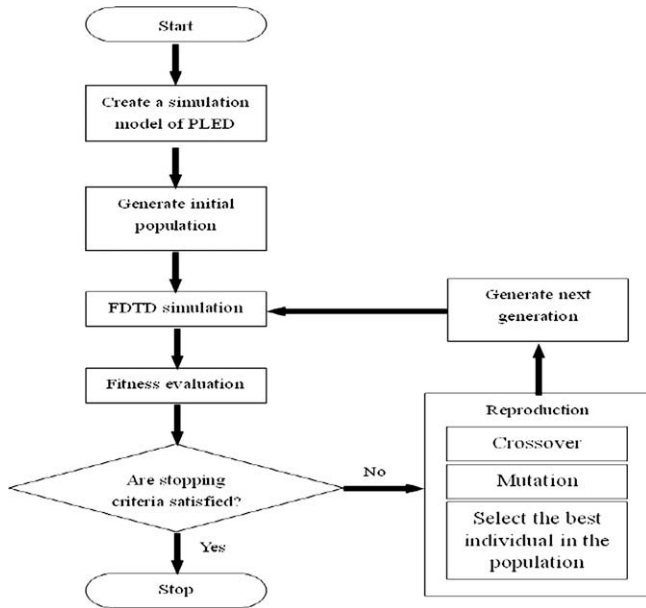


Fig. 3. GA optimal flowchart.

constructed to date is typically only 20% of the internal efficiency. Therefore, in order to further improve the external quantum efficiency, one has to find ways to increase the light extraction efficiency. When full-color-mobile PLED displays become commercially viable, the problem of low extraction efficiency must be overcome. Several out-coupling schemes have been implemented in the hope of improving emission direction and light extraction from PLEDs.

The extracted light energy from the PLED top surface can, based on a standard calibration setup, be further categorized into those contributing to slab and vertical-modes [4]; i.e.,

$$\eta_{\text{ext}} = \eta_{\text{slab}} + \eta_{\text{vertical}} \quad (2)$$

where  $\eta_{\text{slab}}$  and  $\eta_{\text{vertical}}$  denote emission efficiencies in slab modes and vertical-modes, respectively, as shown in Fig. 1b. With properly yet easily set modeling/size parameters to be augmented to the established FDTD model,  $\eta_{\text{slab}}$  and  $\eta_{\text{vertical}}$  can be calculated individually without difficulties. For most practical usages of a PLED, only the part of extraction light in vertical-mode is accounted for emission performance. Thus, only vertical-mode emission efficiency  $\eta_{\text{vertical}}$  within 30 cone degree of the top emission profile is considered for later GA optimization.

### 2.3. Optimizing photonic crystals via genetic algorithm

The numerical optimization algorithm, genetic algorithm (GA), is employed herein to find the optimal size and pitch of photonic crystals (PCs), with aim to maximizing the vertical efficiency  $\eta_{\text{vertical}}$  defined in Eq. (2). The complete computation process for optimization is illustrated by a block flow in Fig. 3. It is seen from this figure that the first step is to establish a FDTD model by the techniques given in Section 2.1. Then, a population of “individual chromosome” is randomly created, which is a varied combination of design variables, PC pitch  $\lambda$  and PC size  $r$ . Note that  $r$  is in fact the radius of each hole in the PC structure in later fabricated PCs. Each individual chromosome  $\{\lambda, r\}$  becomes a possible optimal solution to the problem, while  $\eta_{\text{vertical}}$  does the equivalent light extraction efficiency (LEE) to maximize. This light extraction efficiency is denoted as a function  $E(\lambda, r)$ , and further defined as the fitness function for GA to optimize.

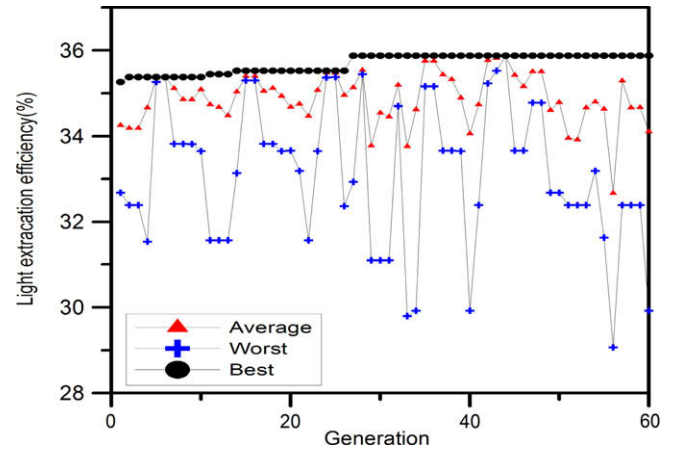


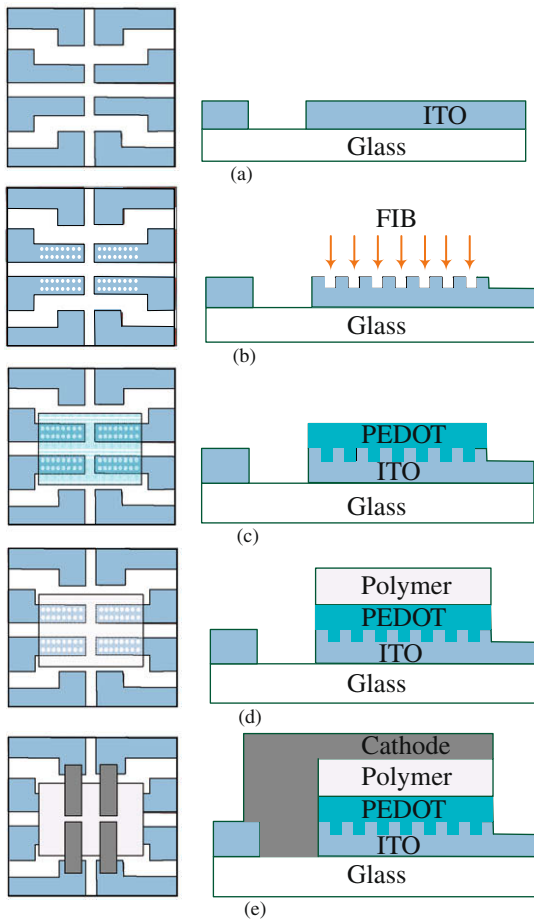
Fig. 4. Evolution of fitness function value.

For the first generation, the structure of each individual is generated with a randomly chosen pitch and radius. For this given  $\{\lambda, r\}$ , the established FDTD model is practiced to obtain  $E(\lambda, r)$ , the fitness function value. In the next step, the chromosome  $\{\lambda, r\}$  mutates, crossover and reproduce for other possibly higher values of  $E(\lambda, r)$  via computation based on the FDTD model. In a single generation, the optimal combination of  $\{\lambda, r\}$  can then be determined based on some mutations and reproduction. This optimization can be continued for a number of generations until the evolution of the optimized  $E(\lambda, r)$  reaches its stable maximum limits. For the current study, the fitness evolution is shown in Fig. 4 for generations, where a stable/upper limit of  $E(\lambda, r)$  is reached at the 28th generation, giving  $\eta_{\text{vertical}}$  of 36% with  $\{\lambda, r\} = \{350 \text{ nm}, 98 \text{ nm}\}$ . For comparison, a PLED without photonic crystals are also simulated to obtain  $E(\lambda, r)$ , which is, in results, only 28%. Thus, the extraction efficiency can be improved by optimizing PC structural parameters.

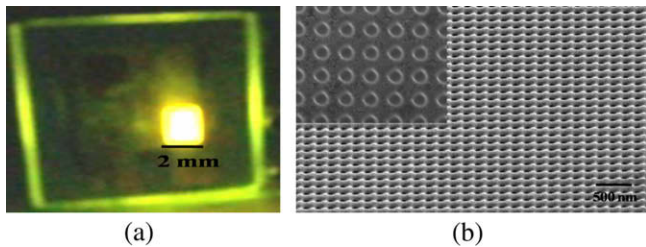
Note that in the computation each time for GA, the light-emitting dipole pulses last only 20 fs, and the total photon number available from the pulses is finite and fixed. It is also interesting to observe that the time-integrated photon energy extracted into the air increases asymptotically with time. The photons in the ultrashort pulses leak out of the waveguide as the guided photons propagate along the high-index layer. However, one cannot wait indefinitely to collect all the photons. Whereas in real PLED displays, the finite pixel size of the device places an upper limit on the travel time of photon pulse after which all the photons are effectively outside a given pixel. One simple way to take this into consideration is to use the values of the integration at the time when the photon generated at the center of a pixel arrives at the boundary of the pixel. To compare the extraction efficiency of PLEDs, a standard light propagation time of 500 fs calculated as the averaged light propagation time is needed to reach the boundary of a pixel of size  $2 \times 2 \text{ mm}^2$ .

### 2.4. Fabrication

The fabrication process for the photonic crystals in PLEDs is elaborated herein and shown in Fig. 5a–e. The first step is to pattern the indium tin oxide (ITO) layer as shown in Fig. 5a, after ITO standard cleaning process. The ITO layer is then coated photoresist. The photoresist in exposed areas is lithographically patterned by exposing UV-light through the mask that enables the removal of the photoresist in un-desired areas. HCl is next utilized to etch the unwanted ITO layer, and then acetone (ACE) for cleaning the remained photoresist on the patterned ITO. In the second step, the FIB, as shown in Fig. 5b, is used to fabricate the pre-opti-



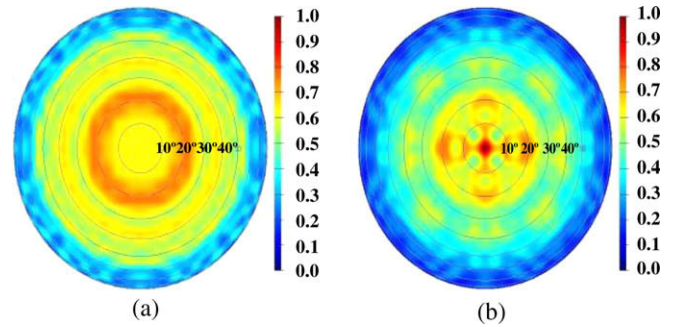
**Fig. 5.** (a) The pattern of ITO, (b) photonic crystal etching by using FIB, (c) PEDOT spin-coated on the ITO layer, (d) coating polymer on the PEDOT layer, (e) deposit aluminum as cathode layer.



**Fig. 6.** Optical image of a PC-LED, (b) scanning electron micrographs of optimizing photonic crystal patterns.

mized square lattice photonic crystal patterns. A PEDOT (poly(3,4-ethylene dioxy thiophene)) layer with a thickness of 40 nm is next spin-coated as a hole transport layer on the PC-patterned ITO substrate (Fig. 5c). A polymer layer is next, as shown in Fig. 5d, coated on the PEDOT layer. Finally, an aluminum layer with 100 nm thickness is deposited as cathode layer, as shown in Fig. 5e.

The crucial step of the stated process is the application of FIB for desired photonic crystals. The FIB system utilized has a model no. as FEI Nova 200 FIB, which is in laboratory mounted to the specimen chamber of a scanning electron microscope (SEM) for observing PC structure and control FIB quality. The FIB produced a tightly focused beam of  $\text{Ga}^+$  ions (rated at less than 10 nm minimum spot size) with an energy of 30 KeV. The beam is deflected electronically over a field size of  $\approx 110 \mu\text{m}$  by an external input in the form of X/Y analog signals that are digitally generated with a step size of



**Fig. 7.** FDTD calculation results for far-field profiles of (a) non-PC-LED and (b) PC-LED.

1.7 nm. A custom software is developed to generate desired PC pattern and then translate design information composed of basic geometrical objects into waveforms which scan the beam over the sample to fabricate the desired pattern.

The FIB for producing larger-area nanostructures could be accomplished by the vector scanning or raster. The scanning guidelines can be distilled based on the following considerations. Firstly it is observed that the outcome of FIB depends strongly upon the precise beam path and its timing. However, the FIB scanning rate should sometimes be slowed down to dissipate the heat generated by FIB fabrication. Secondly, it is anticipated that less-rough surfaces could be obtained, when the sidewalls are fabricated using a trajectory parallel to the sidewall. Note also that to fabricate an arbitrary hole-shape, a sequence of beam paths that were locally parallel and separated by an appropriate pitch (e.g. 50 nm) should be designed. It is expected that these filled the holes with the outermost path coinciding with the user-specified shape. The scanning speed should be almost constant and consistent along these paths. Finally, the FIB machine used can offer, based on experimental trails, a focused Ga beam with estimated diameter of 10 nm. The smallest feature can be fabricated is around 30 nm with the design parameters employed in this work.

### 3. Results and discussion

PC slabs in a square area with 20,410 holes are successfully fabricated by FIB. Each hole is produced one at a time to form a large lattice in a width of  $50 \mu\text{m}$ . For validating the performance of the optimized design, a sample of period for  $\lambda = 350 \text{ nm}$  is prepared. The radius of the sample is 98 nm. The experimental optical emission profile is shown in Fig. 6a. Fig. 6b shows a micrograph of the fabricated square lattice in the photonic crystal pattern by using FIB. For comparison, calculated normalized far-field profiles are shown in Fig. 7a and b. Fig. 7a shows the profile with a non-PC-LED and Fig. 7b does the optimized PC-LED. Through FDTD calculation, the PC-LED of  $\lambda = 350 \text{ nm}$  registers an enhancement amounting to 46% relative to that of the conventional LED without a PC pattern. A maximum increase in vertical output power of 3.5 times is experimentally achieved for a lattice period of 350 nm, when the viewing cone contains an angle less than  $30^\circ$ . This large increase is obtained by controlling both the vertical-mode profile and the lattice constant to extract the modes. This increase in directional emission exemplifies the specific advantages of PC, when optimal lattice constants are employed. Therefore, strong enhancements in light extraction efficiency and directional emission can be achieved by properly-designing PC lattice, based on the measured emission profile of the PLED.

#### 4. Conclusions

In this study, high directionality and extraction of light emission from polymer light-emitting diodes via optimizing photonic crystals is proposed. Enhancing the light extraction and directionality of PC-PLEDs by optimizing the vertical structure and tailoring the PC lattice parameters using genetic algorithm is demonstrated. A maximum increase in vertical output power by 3.5 times is experimentally achieved for a lattice period of 350 nm when the viewing cone contains an angle less than 30°. From the PC-PLEDs fabricated by the optimized photonic crystal pattern using FIB, an increase of the extraction efficiency in excess of 46% is achieved experimentally and the FDTD calculation explains this result faithfully. In conclusion, the performance of the fabricated large-area PCs by FIB reaches a level of satisfaction in terms of extraction efficiency and directionality for a number of applications, including the high-performance active-matrix PLED and flexible displays.

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