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TECHNICAL PAPER

Optimal design of ITO/organic photonic crystals in polymer light-emitting diodes with sidewall reflectors for high efficiency

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Abstract This study aims to achieve large extraction of light emission from polymer light emitting diodes (PLEDs) via optimizing photonic crystals (PCs) and sidewall angle reflectors. Both PCs and sidewall reflectors can be resulting in increasing light emission in useful directions and reducing refection loss. 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). The optimal design of PCs and sidewall angle reflectors are presented in details. To accurately predict light extraction of the PLED, the numerical simulation tool, the FDTD method is employed. Based on the FDTD simulation, the optimal sidewall angle which can increase maximum light extraction efficiency (LEE) in our designed PLED structure is 35°. With the optical modeling of optimal sidewall angle reflectors via FDTD computation and the next step is using GA optimization to seek optimal pitch and radius of photonic crystals. According to the GA optimal result, the ratio of pitch to wavelength is 0.47 times and the ratio of radius to pitch is 0.25 times. GA is a powerful tool to cope with a complicated optimization problem with multiple variables to optimize. The PLEDs with optimized PCs and angle of sidewall reflectors would increase extraction of light emission from 20 to 26 % and the 3D FDTD calculation was conducted to explain this result.

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1 Introduction

This study is aimed to achieve high extraction of light emission from polymer light-emitting diodes with structure of photonic crystals and sidewall. For applications of large area flexible displays and illumination lights, polymer light-emitting diodes 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 (Fujita et al. 2005, 2006; Do et al. 2004; Lee et al. 2003). 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. 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 achieved if PLEDs are expected to be widely commercialized, since both low power consumption and long operational lifetime are required for a variety of displays.

Recently, in order to enhance the light extraction, several methods have been proposed to improve the light extraction efficiency, such as sidewall reflectors and photonic crystals. Kim et al. (2010) designed an advanced mesa-hole LED. Compared to a reference LED, the designed LED improved light output power by 12 %. Hui et al. (2011) presented that laser micro-machining was used to fabricate GaN LED chip with inclined sapphire sidewalls which were coated with highly reflective silver film. Hung-Pin Shiao et al. (2010) proposed to assist in forming a deep undercut sidewall and rough surface on a GaN LED.

Kuo et al. (2010) propose a simple laser scribing method to form a light guiding structure on the sapphire substrate and improve LEE of GaN-based phosphoric acid etched LEDs. Hyunsoo Kim et al. (2007) proposed using omni-directional sidewall reflectors to enhanced light output of GaN-based LEDs. Joonhee Lee et al. (2008) proposed GaN LEDs structure with two-dimensional PCs and angled sidewall deflectors. Both the PCs and angled sidewall deflectors could redirect guided photons into the surface-normal direction. Yang et al. (2009) enhance the light-extraction efficiency in nitride-based LEDs with randomly inverted pyramid sidewalls. The GaN-based LEDs with circular-gear sidewall (Wu et al. 2011) and polygonal LEDs shaped with laser micromachining (Wang et al. 2010) are investigated into increasing light extraction efficiencies. The advantages offered by PCs extraction are that, when designed properly, the emission direction can be tailored, resulting in increasing light emission in useful directions and reducing refection loss. In general, the PC pitch, PC radius and light wavelength are related to each other. The ratios of PC pitch to wavelength and PC radius to PC pitch are about 0.5 times and 0.25 or 0.3 times, respectively (Ichikawa and Babaa 2004; Lee et al. 2003).

This study presents the optimal design of PCs in PLEDs with sidewall reflectors for high efficiency. There are three 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; (4) fabricated the PC mold. Finally, Simulation results were conducted to show the increasing LEE of the proposed PLEDs structure.

2 PLED structure and light extraction efficiency

According to Hyunsoo Kim et al. (2007), they enhanced the light output of GaN-based LED by using mesa sidewall reflectors. Their designed LEDs enhance the light output by 18 %, as compared with reference LEDs. Besides, Joonhee Lee et al. (2008) fabricated a GaN LED device that contains both a 2D-PC pattern at the GaN/sapphire interface and angled sidewall deflectors at the mesa edges. Therefore, our study focuses instead on PLED. The designed PLED structure with PCs and sidewall reflectors as depicted in Fig. 1a is in a multilayer sandwich structure that comprises a glass substrate, an Indium Tin Oxide (ITO) anode, organic emitting layers, and a metal cathode.$\theta$is a sidewall profile-angle. In our designed PLED, PEDOT:PSS and $Ir(mppy)_3$ were used to be the hole transport layer and emission layer.

The extracted light energy from the PLED top surface can, based on a standard calibration setup, be further

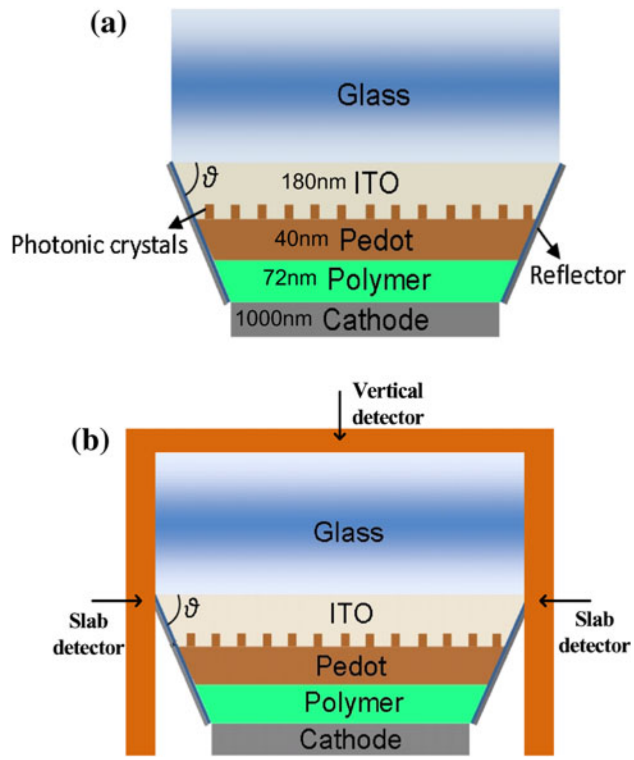

Fig. 1 **a** The PLED structure with photonic crystals and sidewall reflectors. **b** Emission efficiency detection system

categorized into those contributing to slab and vertical modes (Fujita et al. 2005); i.e.,

$$\eta_{ext} = \eta_{slab} + \eta_{vertical}, \quad (1)$$

where η_{slab} and $\eta_{vertical}$ denote emission efficiencies in slab modes and vertical modes, respectively, as shown in Fig. 1b. With proper yet easy setting modeling/size parameters to be augmented to the established FDTD model, η_{slab} and $\eta_{vertical}$ 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 $\eta_{vertical}$ is considered for later GA optimization.

3 Simulation via finite-difference time-domain

To accurately predict light extraction of the PLED, the numerical simulation tool, the finite-difference time-domain method (Noda 2000; Taflove and Hagness 2005; Choi et al. 2006; Kim et al. 2005) is employed. Yee and Chen (1997) 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. Equation (2) shows the difference form for critical terms in Maxwell's equations, as

$$\left.\frac{\partial \vec{E}}{\partial t}\right|_{t=\left(n-\frac{1}{2}\right)\Delta t} = \frac{\vec{E}^{n} - \vec{E}^{n-1}}{\Delta t} = -\frac{\sigma}{\varepsilon}\vec{E}^{n-\frac{1}{2}} + \frac{1}{\varepsilon}\nabla \times \vec{H}^{n-\frac{1}{2}}$$
$$\left.\frac{\partial \vec{H}}{\partial t}\right|_{t=n\Delta t} = \frac{\vec{H}^{n+\frac{1}{2}} - \vec{H}^{n-\frac{1}{2}}}{\Delta t} = -\frac{1}{\mu}\nabla \times \vec{E}^{n} \quad (2)$$

where E denotes the electric field, Hdoes the magnetic field, and n the time step. Also, μ is permeability; σ is conductivity and ε is dielectric constant. In the FDTD method, whereas Eq. (3) is the calculation of the electric field values and the magnetic field values is shown as Eq. (4). From Eqs. (3) and (4) under $\sigma = 0$, the equations for obtaining the x component of the electric field and the x components of the magnetic field are described as follows. The subscripts x, y and z represent the components in the directions. Figure 2 shows the flowchart of FDTD simulation. The first diagram setups the parameters of the initial electric field, magnetic field and light source at T = 0. In next step, the simulation results of electrical field and magnetic field is checked. If the results are unsatisfied for the condition T greater than T_{max}, then restart the loop until matching the condition.

$$E_x\Big|^{n+1}_{i+\frac{1}{2},j,k} = E_x\Big|^{n}_{i+\frac{1}{2},j,k} - \frac{\Delta t}{\varepsilon_{i+\frac{1}{2},j,k}\Delta z}\left[H_y\Big|^{n+\frac{1}{2}}_{i+\frac{1}{2},j,k+\frac{1}{2}} - H_y\Big|^{n+\frac{1}{2}}_{i+\frac{1}{2},j,k-\frac{1}{2}}\right] + \frac{\Delta t}{\varepsilon_{i+\frac{1}{2},j,k}\Delta y}\left[H_z\Big|^{n+\frac{1}{2}}_{i+\frac{1}{2},j+\frac{1}{2},k} - H_z\Big|^{n+\frac{1}{2}}_{i+\frac{1}{2},j-\frac{1}{2},k}\right] \quad (3)$$

$$H_x\Big|^{n+1}_{i,j+\frac{1}{2},k+\frac{1}{2}} = H_x\Big|^{n-\frac{1}{2}}_{i,j+\frac{1}{2},k+\frac{1}{2}} + \frac{\Delta t}{\mu\Delta z}\left[E_y\Big|^{n}_{i,j+\frac{1}{2},k+1} - E_y\Big|^{n}_{i,j+\frac{1}{2},k}\right] - \frac{\Delta t}{\mu\Delta y}\left[E_z\Big|^{n}_{i,j+1,k+\frac{1}{2}} - E_z\Big|^{n}_{i,j,k+\frac{1}{2}}\right] \quad (4)$$

where i, j, and k are the quantity of x, y, and z axis, respectively.

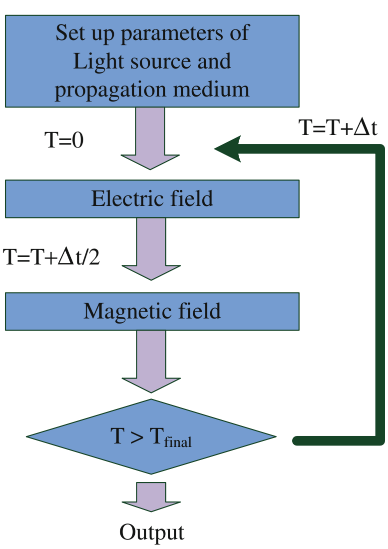

Fig. 2 FDTD process

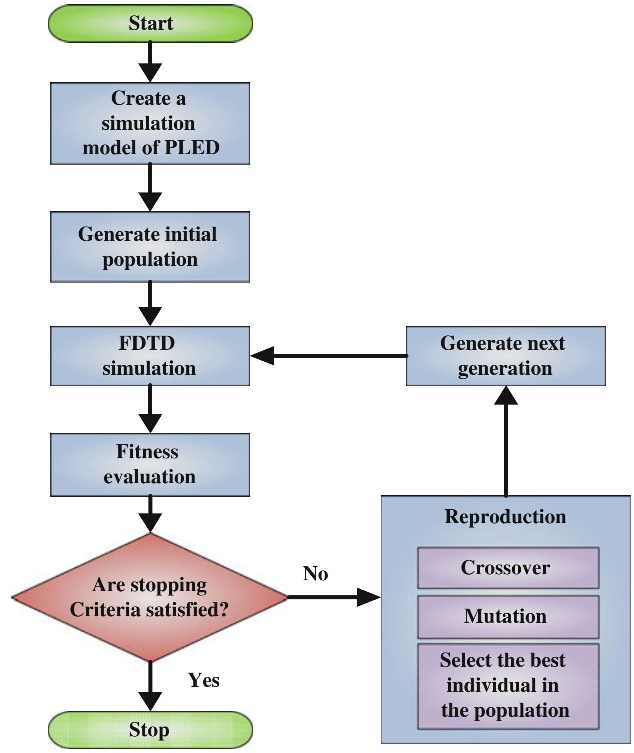

Fig. 3 A Flowchart of GA optimization on pitch and radius of the photonic crystals

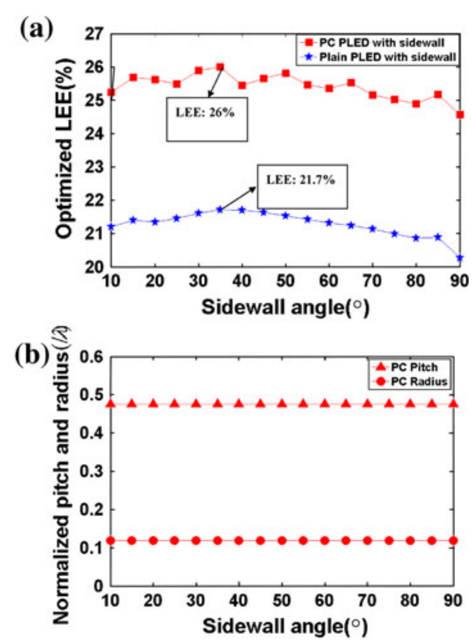

Fig. 4 **a** Light extraction efficiencies (LEEs) of a plain PLED with varied sidewall angles and GA-optimized LEEs of a PLED with photonic crystals (PC); **b** The optimal pitch and radius in terms of with wavelength (λ), which lead to maximized LEEs in subfigure (**a**)

The FDTD method has further 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. The FDTD applied herein for the PLED structure in Fig. 1a 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 wavelength 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 with 500 fs running time, 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.

4 GA optimization

4.1 Optimizing photonic crystals via genetic algorithm

The numerical optimization algorithm, genetic algorithm, is employed herein to find the optimal radius and pitch of PCs. In the GA optimization, the software Matlab with code of GA is used to link FDTD Solutions. The complete computation process for optimization is illustrated by a block flow in Fig. 3. It is seen from this figure that the first

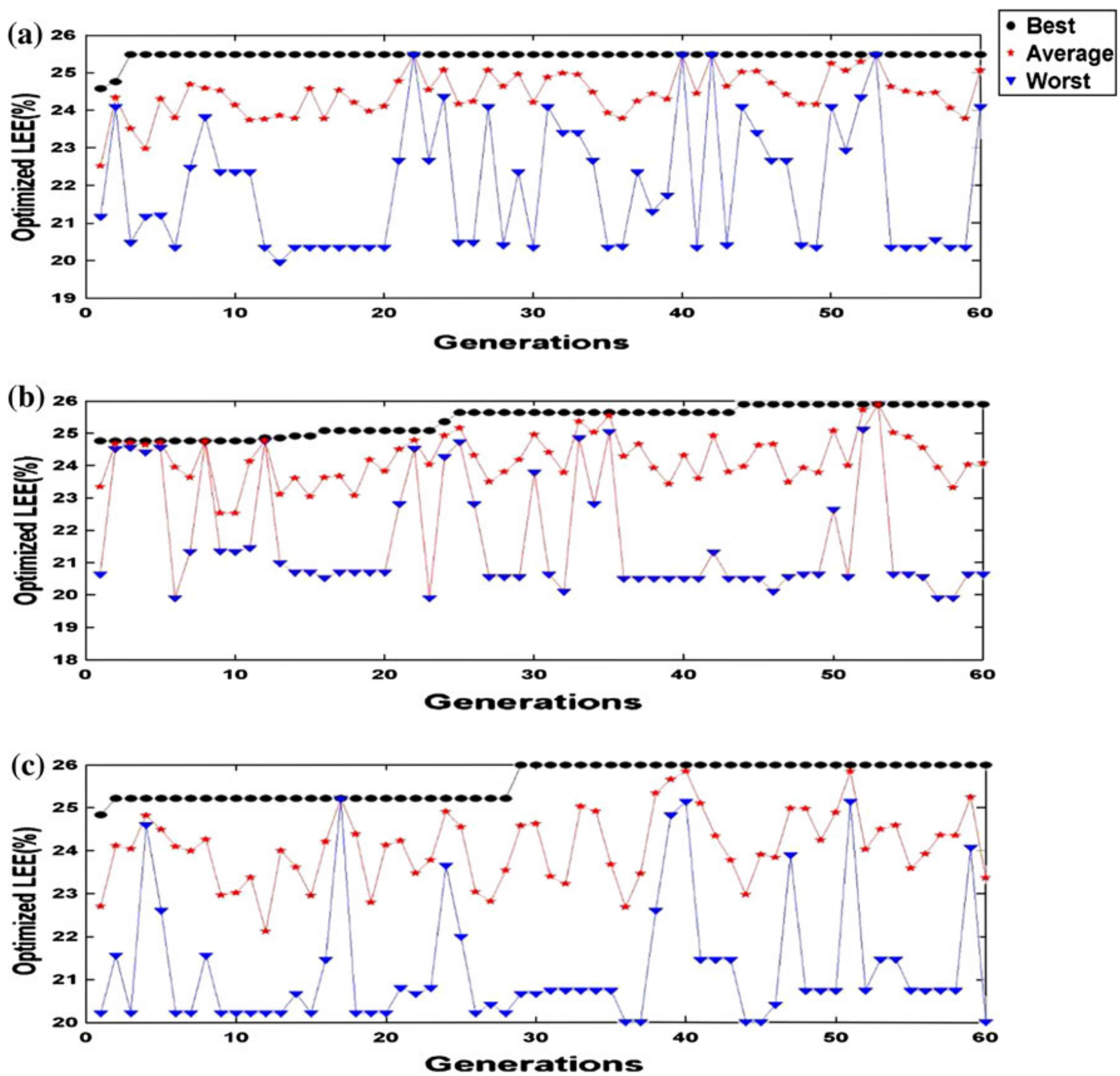

Fig. 5 Evolution of fitness function value while optimizing via GA for different sidewall angles of **a** 25°; **b** 30°; **c** 35°

step is to establish a FDTD model. Then, a population of "individual chromosome" is randomly created, which is a varied combination of design variables, PC pitch Λ and PC radius r. Note that r is in fact the radius of each hole in the PC structure in later fabricated PCs. Each individual chromosome {Λ, r} becomes a possible optimal solution to the problem, while $\eta_{vertical}$ does the equivalent light extraction efficiency to maximize. Where $\eta_{vertical}$ denote emission efficiencies in vertical modes. This light extraction efficiency is denoted as a function LEE (Λ, r), and further defined as the fitness function for GA to optimize. For the first generation, the structure of each individual is generated with a randomly chosen pitch and radius. For this given {Λ, r}, the established FDTD model is practiced to obtain LEE (Λ, r), the fitness function value. In the next step, the chromosome {Λ, r} mutates, crossover and reproduce for other possibly higher values of LEE (Λ, r) via computation based on the FDTD model. In a single generation, the optimal combination of {Λ, 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 LEE (Λ, r) reaches its stable maximum limits.

4.2 Simulation results

The software which was utilized in our simulation is FDTD Solutions. In the FDTD simulation, the PLED models with varied sidewall angles are set up firstly. Next is to calculate the light extraction efficiency of PLED with different sidewall angle and find the optimal angle which has the maximum light extraction efficiency. Sidewall reflectors could deflect the photons that reach the mesa edges along the waveguide into the surface-normal direction and help enhance the surface extraction efficiency. The LEE of different sidewall angles were calculated as shown in

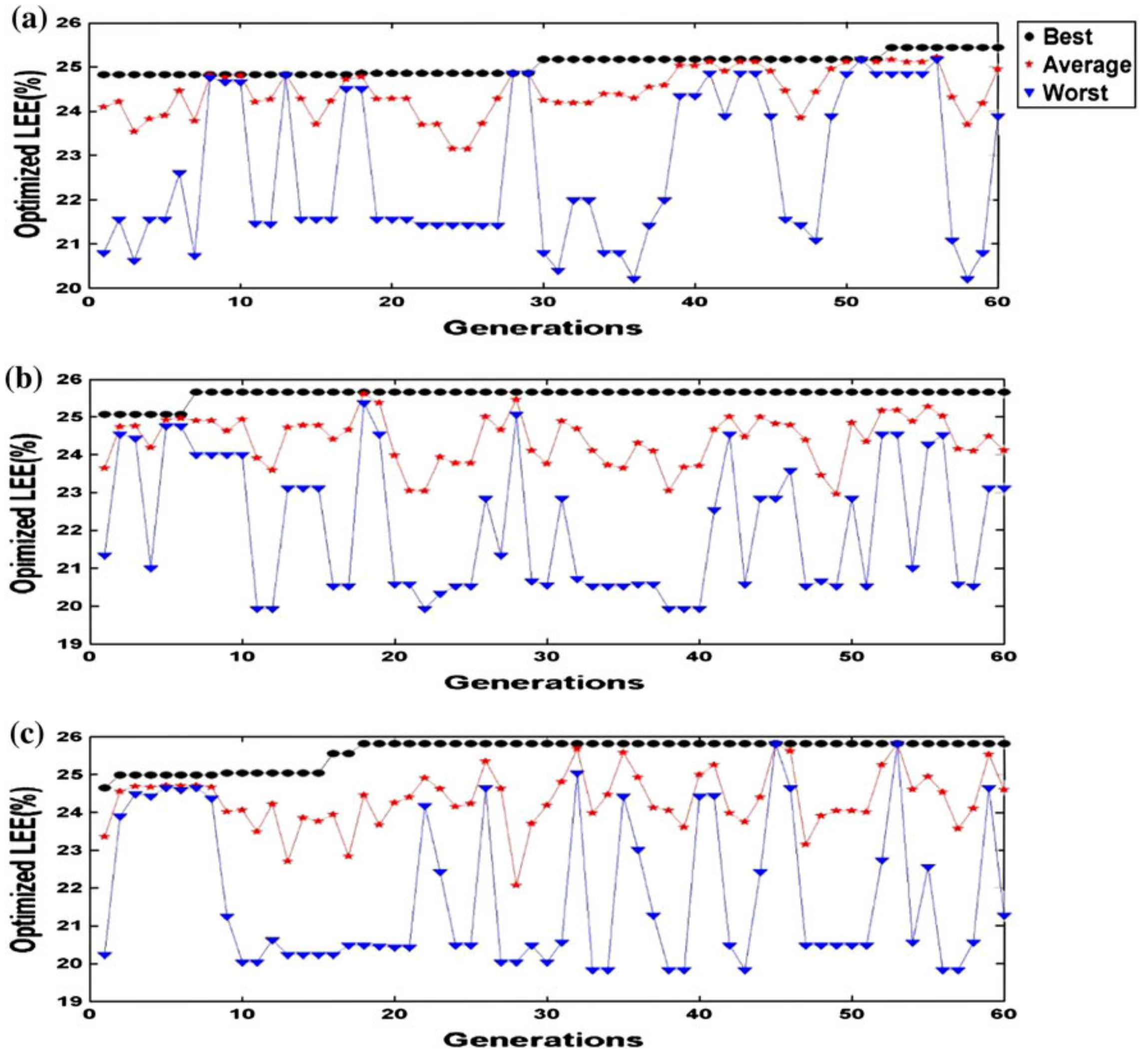

Fig. 6 Evolution of fitness function value while optimizing via GA for different sidewall angles **a** 40°; **b** 45°; **c** 50°

Fig. 4a. It is shown that the maximum light extraction efficiency with sidewall profile-angle of 35° is increased up to 21.7 %. Having calculated the LEE of different sidewall angles, find the optimal radius and pitch of PCs were fabricated in PLED structure by GA optimization. For the current study, the fitness evolutions are shown in Figs. 5, 6. To Compare all fitness evolutions, where a stable/upper limit of LEE (Λ, r) is reached at the 29th generation, giving $\eta_{vertical}$ of 26 % with {Λ, r} = {252, 63 nm} as depicted in Fig. 5(c). In our GA optimization, the results not only showed that PC pitch is 0.47 λ and PC radius is 0.25Λ in Fig. 4b but also the both of photonic crystals and sidewall angle do not affect each other for increasing LEE of PLED in our case. For comparison, a PLED without photonic crystals and sidewall reflectors are also simulated to obtain LEE (Λ, r), which is, in results, only 20 %. 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 × 2 mm^2.

5 Fabrication

The fabrication process for the photonic crystals in PLEDs is elaborated herein and shown in Fig. 7a–e. The first step is to pattern the ITO layer as shown in Fig. 7a, 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 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 thermal nano-imprint, as shown in Fig. 7b, is used to fabricate directly square lattice photonic crystal patterns. A Poly (3,4-ethylene dioxy thiophene) (PEDOT) layer with a thickness of 40 nm is next spin-coated as a hole transport layer on the PC-patterned ITO substrate (Fig. 7c). A polymer layer is next, as shown in Fig. 7d, coated on the PEDOT layer. Acetone (ACE) is again used to remove polymer in un-desired areas. Finally, a aluminum layer with 1000-nm thickness is deposited as cathode layer, as shown in Fig. 7e. In this study, thermal nano-imprint is employed to fabricate the photonic crystals in PLEDs. The technique needs a mold with nanostructures on its surface to transfer any pattern on the substrate. The fabrication process for the mold of photonic crystals is elaborated

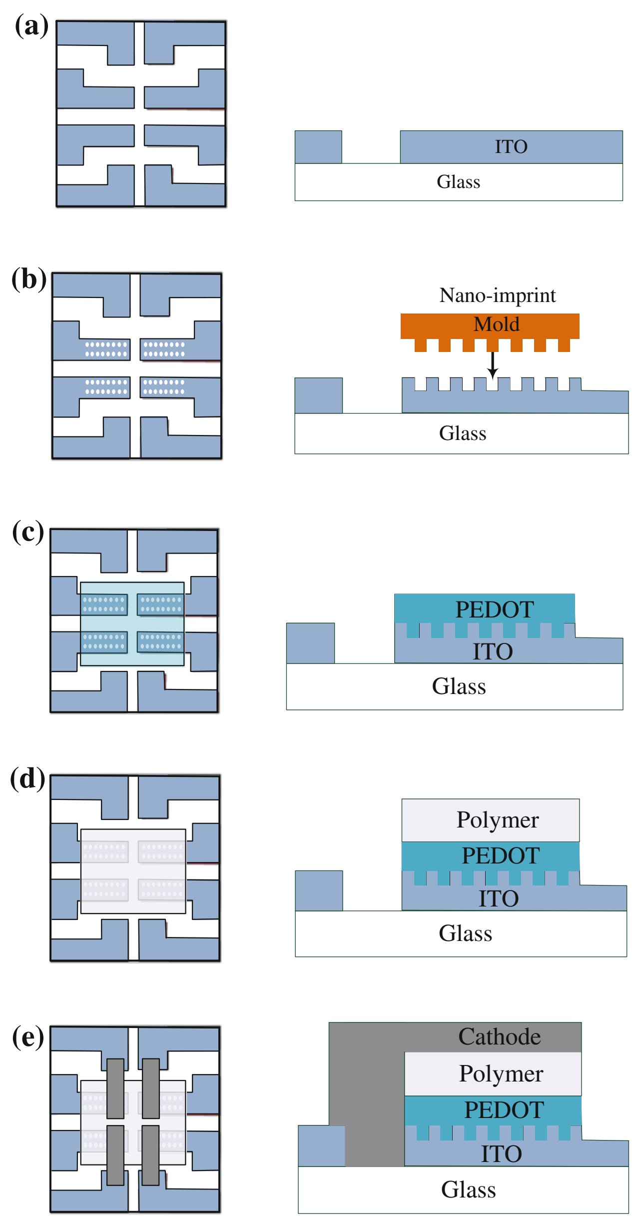

Fig. 7 **a** The pattern of ITO, **b** photonic crystals by nano-imprint, **c** PEDOT spin-coated on the ITO layer, **d** coating polymer on the PEDOT layer, **e** deposit aluminum as cathode layer

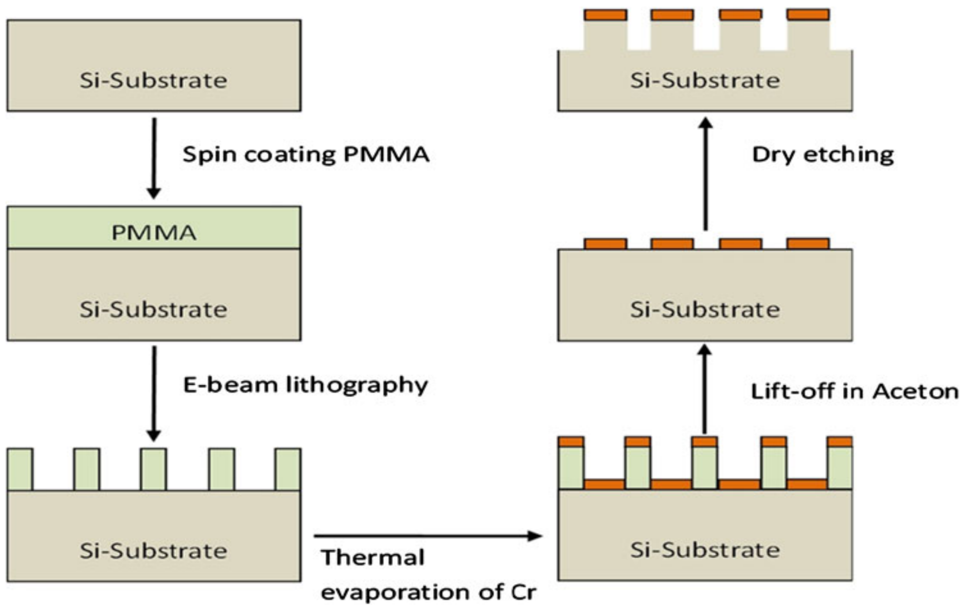

Fig. 8 The fabrication process of the PC-mold

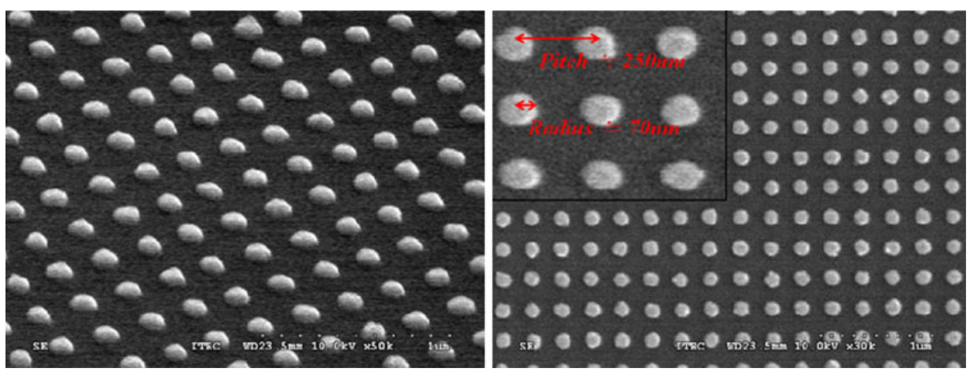

Fig. 9 The SEM Photos of the PC-mold fabricated

herein and shown in Fig. 8. In our experiments, silicon is used as the mold materials. The first step is to spin coating Polymethyl methacrylate (PMMA) on the substrate. In the second step, E-beam lithography is used to make the pattern. Chromium is next deposited by thermal evaporation and using Lift-off scheme to remove the residual PMMA. Finally, etch the Si-substrate by dry etching. A scanning electron microscopy (SEM) image on the fabricated mold is shown in Fig. 9, where the actual PC Pitch and PC radius to achieve are measured as 250 and 70 nm based on simple image processing of the photos, respectively.

6 Conclusion

A PLED with optimizing photonic crystals and sidewall reflectors is presented in this study. Enhancing the light extraction efficiency of PLEDs by optimizing the sidewall angle reflectors structure and tailoring the PC lattice parameters using genetic algorithm are demonstrated. From the FDTD simulation, the maximum light extraction efficiency with sidewall profile-angle of 35° is increased up to 21.7 %. Besides, using GA optimization to seek optimal pitch and radius of photonic crystals for different sidewall angle reflectors are completed. The ratio of pitch to wavelength is 0.47 times and the ratio of radius to pitch is 0.25 times. The LEE is enhanced to 26 %. Simulation results show excellent enhancement of the light extraction efficiency in PLEDs with photonic crystals and sidewall angle reflectors.

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