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Dielectric nanoparticles for the enhancement of OLED light extraction efficiency

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ABSTRACT

This work reports the use of dielectric nanoparticles placed at glass substrate in the improvement of light extraction efficiency of organic light emitting diode (OLED). The nanoparticles will act as scattering medium for the light trapped in the waveguiding modes of the device. The scattering efficiency of dielectric nanoparticles has been calculated by Mie Theory. The finite difference time domain (FDTD) analysis and simulation estimate the effect of dielectric nanoparticles on the light extraction efficiency of OLED. The efficiency depends upon the diameter, interparticle separation and refractive index of dielectric nanoparticles. It is shown that the dielectric nanoparticles layer can enhance the light extraction efficiency by a factor of 1.7.

1. Introduction

Organic light emitting diodes (OLEDs) are flat large area light sources with medium luminance in nature. OLEDs have attracted a great deal of attention due to their advantages of high internal quantum efficiency, low power consumption, low cost, high brightness and high contrast than those of semiconductor based LEDs. Organic materials are easier to fabricate on flexible substrate because of their intrinsic property. In OLED, the emission wavelength can be tuned easily by the use of appropriate dopants in organic emissive layer [1,2]. OLEDs have been of keen interest due to their potential applications in illumination, flat panel displays, and solid state lightning etc.

Radiative decay of molecular excited states is the underlying principle of light generation mechanism in organic light emitting diodes. The internal quantum efficiency (IQE) defined as the ratio of total number of photons generated in the emissive layer to the number of ejected electrons is only 25% if only radiative decay of singlet excitons takes place for light generation. In order to achieve high IQE, OLEDs must be able to harvest the radiative decay of both singlet and triplet excitons that form within them. Despite achieving almost 100% IQE [3], OLED's external quantum efficiency (EQE) defined as the ratio of generated photons coming out of the device to the photons generated inside the device is limited to 20% in conventional cases [4]. A large fraction of light (~80%) is lost due to the different mechanisms takes place inside the device as shown in Fig. 1. Approximately 30% of generated light is lost due to surface plasmons modes formed at the metal/organic interface [5]. 20% loss of generated light is related to waveguide mode formed at organic/glass interface. Approximately 30% of light is lost due to substrate mode which comes from the high refractive index of the substrate.

Various approaches have been reported in the literature to extract power from these modes and thereby to increase the extraction efficiency of OLED. Use of textured substrate, surface roughening and micro lens arrays has been suggested to extract the power from OLED [6–9]. Use of high index substrate has been reported to avoid the formation of waveguide modes [10]. However, high index contrast leads to trapping of light in the substrate. Power coupling to substrate modes can be reduced by inserting silica aerogel layer between glass and ITO in planar OLED [11]. Use of photonic crystals, microstructures and nanostructures has been reported to extract the power from waveguide and surface plasmons modes [12–16]. Use of scattering structures is another approach to improve the extraction efficiency of OLED [17]. Siang et al. demonstrated the enhancement of out coupling efficiency of OLED due to the effect of volumetric light scattering [18]. Incorporation of ordered monolayer silica microspheres, which act as scattering medium enhanced the light extraction of planar OLED [19]. External quantum efficiency could be enhanced by 1.5 times by incorporation of polymer layer with dispersed scattering nanoparticles [20].

In this paper, we propose to incorporate a layer of dielectric nanoparticles at the glass substrate to break the waveguiding caused by ITO and glass index contrast. The scattering efficiency of dielectric nanoparticles has been calculated by Mie theory [21] and the proposed structure has been simulated and analyzed by FDTD method, using commercially available software (Lumerical FDTD solutions) [22]. The parameters such as refractive index, diameter and interparticle separa-

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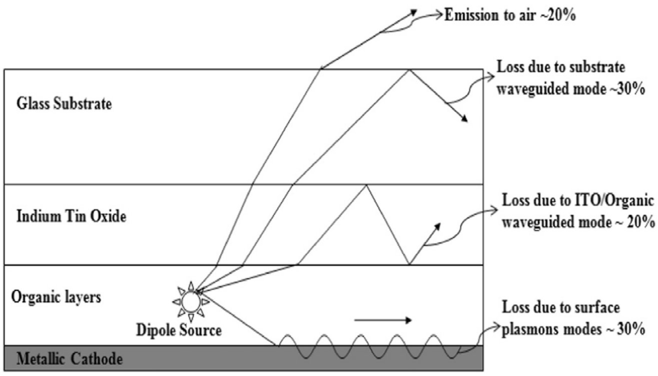

Fig. 1. A simplified OLED showing various trapped modes.

tion of dielectric nanoparticles are optimized so that more power can be extracted into the glass substrate from organic layer. Almost 1.7 times enhancement in the light extraction efficiency has been achieved by using layer of dielectric nanoparticles at glass substrate.

2. Proposed structure and method of analysis

The conventional OLED device set as reference is shown in Fig. 2(a). The structure consists of a glass substrate, indium tin oxide (ITO) as anode, N,N-Di(naphthalene-1-yl)-N,N′-diphenyl-benzidine (NPD) acting as hole transport layer (HTL), tris(8-quinolinolato) aluminium (Alq_3) as emissive layer (EL), Lithium Fluoride (LiF) as electron transport layer (ETL), aluminium (Al) acting as cathode. The proposed structure shown in Fig. 2(b) consists of a single layer of dielectric nanoparticles at glass substrate followed by ITO/NPD/Alq_3/LiF/Al. There will not be any problem of absorption by nanoparticles as dielectric material is optically lossless. Nanoparticles are assumed to be spherical in shape. Mie theory and finite difference time domain (FDTD) method have been used to analyze the effect of nanoparticles.

For spherical particles, Mie theory is an exact solution to Maxwell's equations to calculate the optical properties of non-absorbing (dielectrics) and absorbing (metal) sphere embedded in a homogenous medium [21]. According to Mie theory, the scattering, extinction and absorption efficiencies are given by [21]

$$Q_{sca}=\frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2+|b_n|^2)\,Q_{ext}=\frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)Re(a_n+b_n)$$

$$Q_{abs}=Q_{ext}-Q_{sca}$$

where a_n *and* b_n are the Mie coefficients [21], n is an index running from 1 to ∞ and $x=\boldsymbol{k}.\boldsymbol{r}$ is the size parameter. $\boldsymbol{k}$is the wavenumber in ambient medium and $\boldsymbol{r}$ is the radius of the sphere. For infinite series, n is truncated to n_{max}, i.e.

$$n_{max}=x+4x^{1/3}+2$$

As we know that dielectric nanoparticles have no dissipation in the visible range,$Q_{abs}=0$ which implies

$$Q_{sca}=Q_{ext}$$

To find the enhancement in light extraction efficiency of proposed structure we have used FDTD method.

FDTD is a powerful way of solving Maxwell's equations at very small scales [23,24] and is also very applicable for the study of OLED devices [25]. We have numerically analyzed the proposed structure using the simulation software Lumerical FDTD solutions [22]. Though 3D simulation provides more reliable results but to save simulation time, we did 2D FDTD simulation with transverse electric polarizations as FDTD simulations are computationally very heavy. Park et al. showed that the relative enhancement ratio for 2D and 3D simulations are same [26]. They also found that external quantum efficiency ratio observed in the experiment was close to that obtained from the FDTD simulation. The following parameters were chosen for the numerical simulation. Thicknesses of layers were set to be as: ITO (100 nm), NPD (65 nm), Alq_3 (60 nm), LiF (1 nm), Al (40 nm). It may be noted that 100 nm thickness of ITO layer is above the dielectric nanoparticle layer. Diameter (d) and interparticle separation (s) of nanoparticles were optimized for maximum extraction of light from the device. Refractive index and extinction coefficient of ITO, NPD, Alq_3 and Al were taken from literature [27–29] whereas refractive index of glass substrate and LiF were set to be 1.5 and 1.39 respectively. Refractive index (n_{np}) of dielectric nanoparticles was varied from 1.1 to 2.1.

Generation of light takes place in the emissive layer as the injected electrons and holes recombine to generate photons. The photons are generated by spontaneous emission process and each photon has random direction, phase and polarization. The exact treatment of this process can be described quantum mechanically in terms of photons but we have considered the generated light classically by using electromagnetic point dipole sources. A single dipole was chosen for finite computation and placed in the centre of active layer. Since FDTD simulations directly calculate the various field components i.e. it is vectorial in nature; we can't expect an incoherent addition of fields by placing two sources together in the same simulation. So, in order to have a point dipole source we need to make use of transverse electric(TE) polarized dipoles with orthogonal orientations in two separate simulations and then add the two responses incoherently. In order to estimate the total power generated in active layer, the dipole source was surrounded by a monitor. To avoid the unnecessary reflection of light, perfectly matched layer (PML) boundary conditions had been placed at all boundaries except at cathode side [30]. We used the metallic boundary at cathode side. The FDTD mesh size is also a very crucial aspect as this decides the convergence of calculated fields and the results as well. The span of the simulation region needs to be large enough such that there is enough propagation distance for the light to be fully extracted from the emission layer. The enhancement in the light extraction efficiency was determined by calculating the far field distribution of emissive dipole source. The far field intensity distribution was detected by monitor placed in the air above the glass substrate.

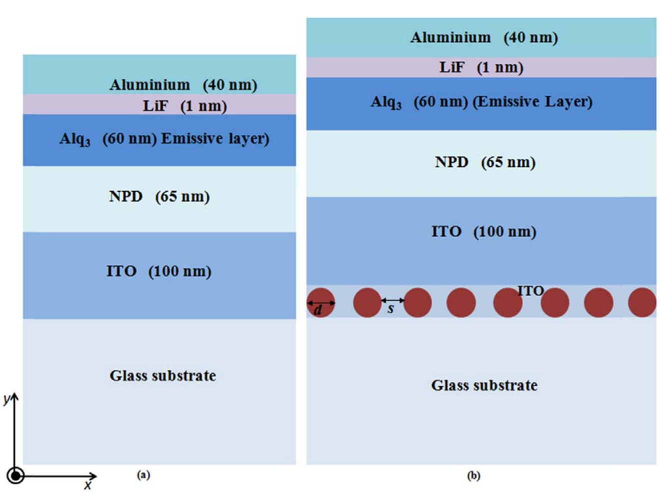

Fig. 2. Schematic design of (a) conventional OLED (b) proposed OLED with nanoparticles.

3. Numerical results and discussion

When calculating the light extraction efficiency, we calculate the fraction of useful power emitted from the OLED device relative to the total power emitted from the active layer. In FDTD simulation, the monitor placed in air receive the light extraction efficiency of OLED with nanoparticles and conventional OLED for various parameters. The

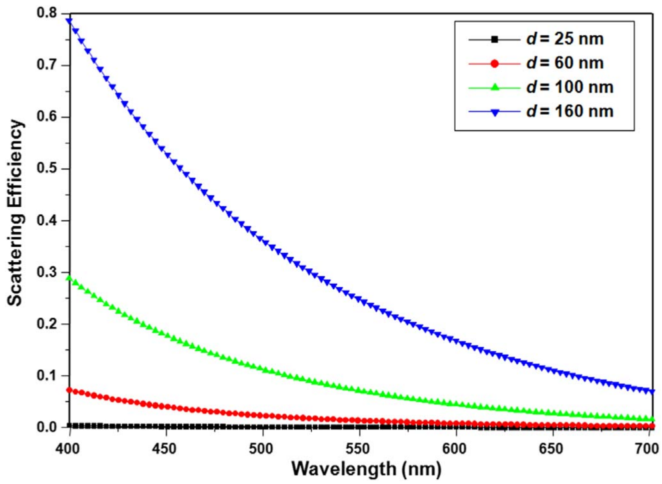

Fig. 3. Scattering efficiency as a function of wavelength for different diameters of nanoparticles.

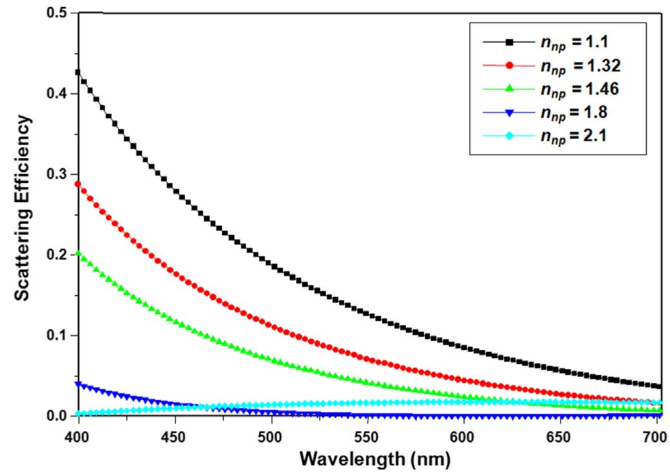

Fig. 4. Scattering efficiency as a function of wavelength for different refractive indices of nanoparticles.

enhancement was calculated by ratio of light extraction efficiencies of OLED with nanoparticles to the conventional OLED. Even with the use of dielectric nanoparticles, the light could not be completely extracted because the organics and metals absorb the light before it is coupled out of the device. To achieve the maximum enhancement in light extraction efficiency, several parameters like diameter (d), interparticle separation (s) and refractive index (n_{np}) of nanoparticles were optimized.

We have studied the optical scattering properties of dielectric nanoparticles in order to examine their suitability for enhanced performance of OLED. All these dielectric nanoparticles do not have dissipative properties in the spectral range of operation of OLED and hence they have an added advantage of not absorbing a part of incident energy and losing it in the form of joule heating. The dielectric nanoparticles do not exhibit the dipolar plasmonic resonances as they have positive dielectric constant over visible range so. Fig. 3 shows the calculated scattering efficiencies of isolated dielectric nanoparticle (n_{np}=1.32) with diameter in range from 25to 160 nm in ambient medium of ITO. From the curve, we can see that the scattering efficiency increases from d=25 nm to d=160 nm. This means larger nanoparticles are generally more desirable for extracting light from the device due to their greater ability to scatter more light. Large size nanoparticles tend to induce significant electrical effects [31]. However, in the proposed structure the nanoparticles being between glass and anode layer do not fall in the path of current and so are not expected to alter the electrical properties. Fig. 4 shows the scattering efficiencies of dielectric nanoparticle (diameter=100 nm) with n_{np} varied from 1.1 to 2.1. Nanoparticle with n_{np}=1.1 shows the maximum scattering efficiency and with n_{np}=1.8 shows the minimum scattering efficiency. The ITO medium refractive index is not fixed to 1.8 but it is varying with wavelength [27]. That's why there will be scattering by nanoparticles of n_{np}=1.8. This shows that the scattering of light by nanoparticles strongly depends on the index contrast.

Fig. 5(a) and (b) shows the far field intensity distribution patterns proportional to $|E|^2$, where E is the electric field, for conventional OLED and OLED with nanoparticles. From the patterns we can see that by adding the dielectric nanoparticles more radiation is directed outside from the emission layer of OLED. Addition of nanoparticles in the OLED design results in scattering and hence coupling out the emitted light from waveguide modes. Diameter of nanoparticles plays an important role in scattering out the light from the device. To find the effect of diameters in light extraction, the enhancement in light extraction efficiency was calculated as a function of wavelength for different diameters of nanoparticles as shown in Fig. 6 while all other parameters like refractive index and interparticle separation were kept fixed to 1.1 and 0 nm (i.e. no spacing). The diameter was varied from 25 nm to 160 nm. As we can see from the plot that enhancement in extraction increases for d increasing from 25 nm to 100 nm but it decreases at d=160 nm. This shows that for the larger diameter, the light extraction efficiency was not increasing because the number of scattering sources was decreasing in the simulation region of analysis. The enhancement can be seen in the wavelength range from approximately 440 nm to 600 nm. Therefore, one can achieve extraction efficiencies higher than that of planar OLED by setting the operating wavelength of OLED at any of these wavelengths. Light has to be extracted out from waveguided modes of ITO/organic interface. Because of the property of waveguide, we get resonance for particular wavelength at which scattering by nanoparticles will be maximum. So, in our structure approximately 1.7 times enhancement can be seen with d=100 nm at around 500 nm wavelength.

Any change in the interparticle separation of nanoparticles also affects the scattering, which results in a change in extraction efficiency. To show this we have investigated the effect of interparticle separation of dielectric nanoparticles on the enhancement in light extraction efficiency with wavelength and the results are shown in Fig. 7. We have fixed the diameter and refractive index to 100 nm and 1.1. Interparticle separation was varied from 100 nm to 200 nm. As seen from the figure, enhancement in extraction efficiency decreases from 1.7 times to 1.58 times as the interparticle separation increases from 0 nm to 100 nm. Also, there is resonance wavelength shift of approximately 10 nm as the interparticle separation is raised from 0 nm to 100 nm. The peak experiences a blue shift and reduced strength with increasing interparticle separation indicating the diminishing of coupling effect between two spherical nanoparticles. These reductions in peak strength with interparticle spacing reflect the decay of field distribution between the particles. From the figure, we note that maximum enhancement (almost 1.7 times) is achieved with no interparticle spacing (i.e. s=0 nm).

Next, we have studied the effect of refractive index of nanoparticles on enhancement in light extraction efficiency. To illustrate this, in Fig. 8 we have plotted the enhancement in light extraction efficiency curve with wavelength corresponding to different refractive index of nanoparticles. Diameter of nanoparticles was fixed to 100 nm with no interparticle separation (s=0 nm). The curve shows the maximum enhancement of almost 1.7 times with refractive index 1.1. The scattering of light by the nanoparticles strongly depends on index contrast. Here the index contrast is in between refractive index of dielectric nanoparticles and ITO (n=1.8). We varied the refractive index of nanoparticles from 1.1 to 2.1. From the figure, we can infer that enhancement in light extraction efficiency decreases from 1.7 times to 1.4 times as n_{np} increases from 1.1 to 1.46 whereas it

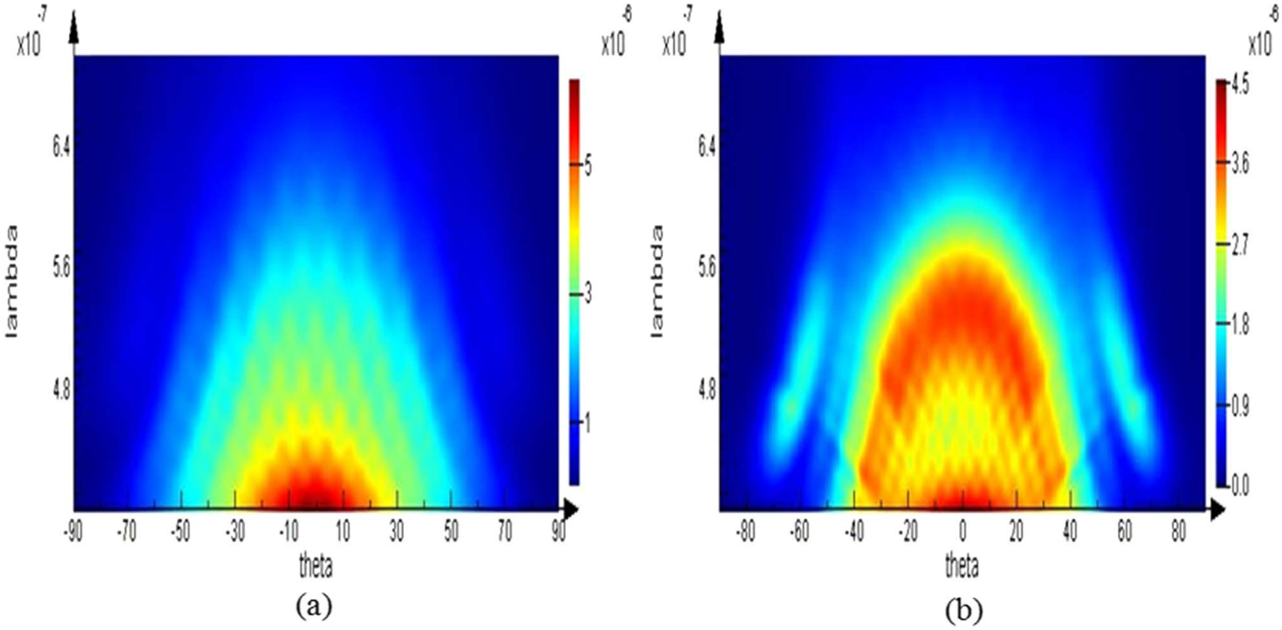

Fig. 5. Far field intensity distribution pattern for (a) conventional OLED (b) proposed OLED with nanoparticles.

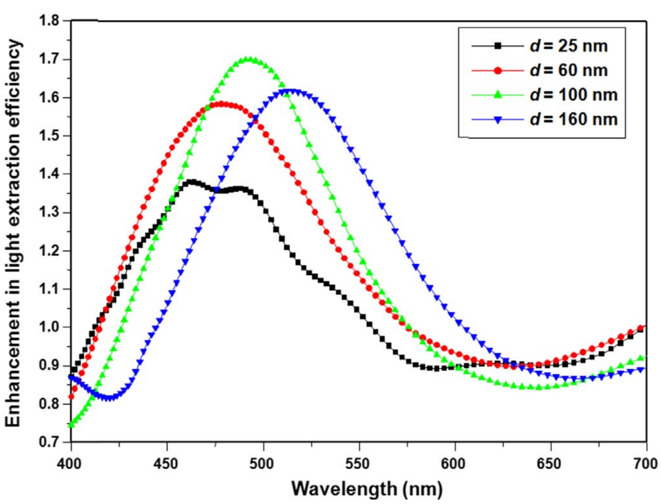

Fig. 6. Enhancement in light extraction efficiency as a function of wavelength for different diameters of nanoparticles.

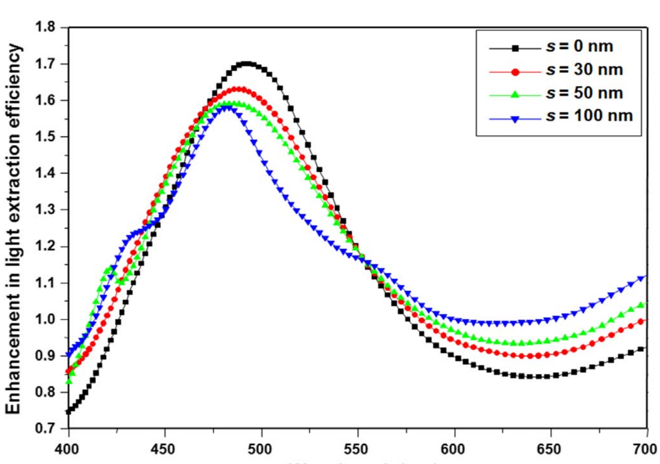

Fig. 7. Enhancement in light extraction efficiency as a function of wavelength for various interparticle separations of nanoparticles.

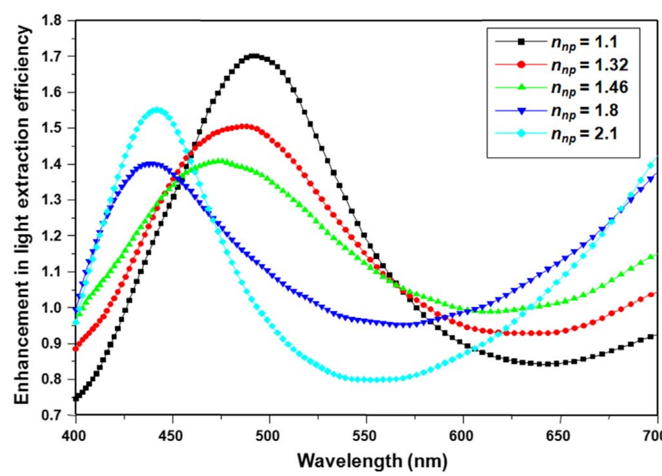

Fig. 8. Enhancement in light extraction efficiency as a function of wavelength for different refractive indices of nanoparticles.

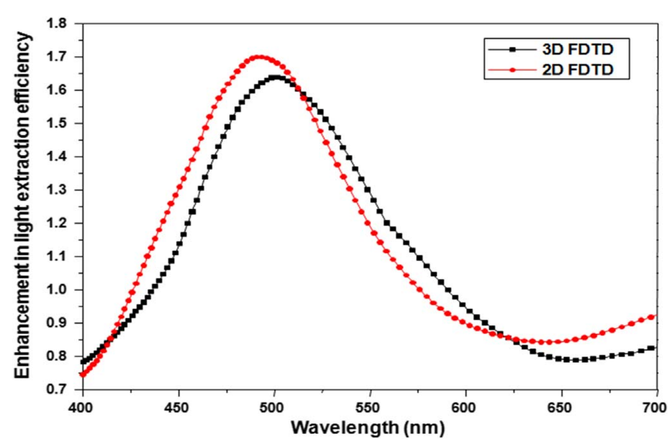

Fig. 9. Comparison of enhancement in light extraction efficiency for 2D and 3D FDTD simulation.

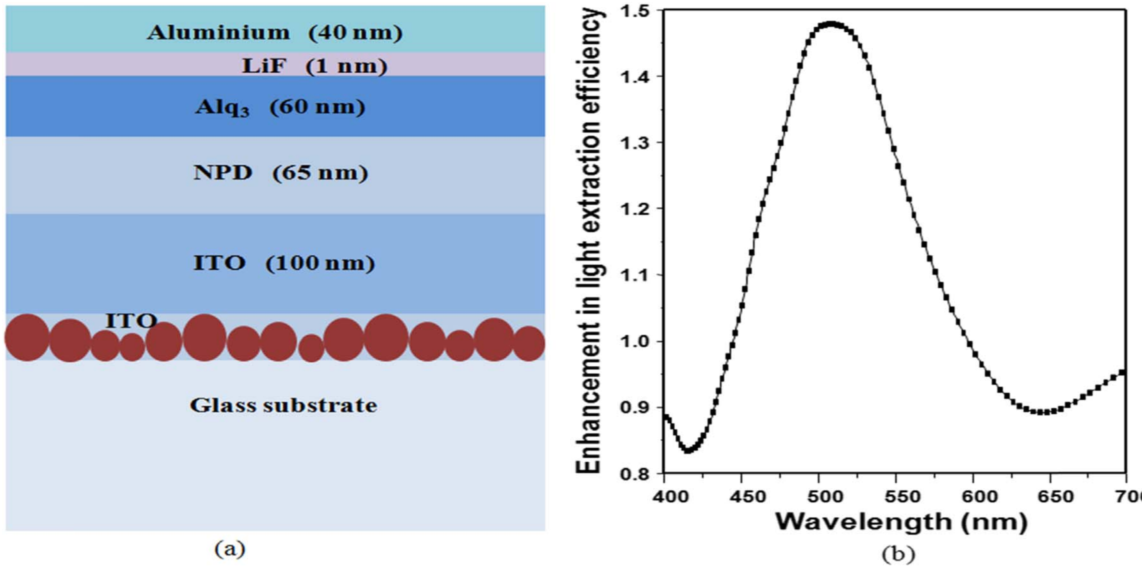

Fig. 10. (a) Proposed structure with assorted diameter nanoparticles. (b) Enhancement in light extraction efficiency as a function of wavelength for assorted diameter nanoparticles.

increases from 1.4 times at n_{np}=1.8 to 1.55 times at n_{np}=2.1. Due to selective reflection of the nanoparticles, light scattering is highly dependent on wavelength of light. Low refractive index (n_{np}=1.1 and n_{np} =1.32) dielectric nanoparticles show the enhancement in light extraction efficiency in the wavelength range 430–570 nm whereas high refractive index (n_{np}=1.8 and n_{np}=2.1) nanoparticles show the enhancement in wavelength range 400–500 nm and 630–700 nm. Enhancement can be seen in the whole visible spectrum with n_{np}=1.46. So, one can use refractive index of dielectric nanoparticles according to the choice of operating wavelength. Hence, mesoporous silica (n_{np}≈1.1), teflon (n_{np}≈1.3), silica (n_{np}≈1.46), aluminium oxide (n_{np}≈1.8) and zirconia (n_{np}≈2.1) dielectric materials can be used for nanoparticles.

For more realistic results one should in principle perform 3D simulation. However, 3D simulations are highly computationally extensive and time consuming. Also, the relative enhancements from both 2D and 3D simulations are expected to be similar [26]. To verify this, we have also carried out 3D FDTD simulation of the proposed structure for d=100 nm, s=0 nm, n_{np}=1.1 and compared our results with 2D simulation. Fig. 9 shows the plot of enhancement in light extraction efficiency versus wavelength for 2D and 3D FDTD simulation. From the curve, we can see that the enhancement ratios of light extraction efficiency for both 2D and 3D simulations are not significantly different. This confirmed that 2D simulation can be used to examine the trend of the light extraction efficiency. So, to save computational efforts and time, we have limited ourselves to 2D simulations.

We have also investigated the effect of assorted diameter dielectric nanoparticles layer (deposited on glass substrate) on the enhancement of light extraction efficiency. Fig. 10(a) shows the structure containing assorted diameter dielectric nanoparticles and Fig. 10(b) shows the plot of corresponding enhancement in light extraction efficiency with wavelength. We have varied the diameter from 80 nm to 120 nm in a random manner while keeping the refractive index of nanoparticles as 1.1 with no interparticle spacing (i.e. s=0 nm). The plot shows that enhancement of 1.5 times in light extraction efficiency is achieved with assorted diameter nanoparticle.

The results clearly show that by using dielectric nanoparticles which act as scattering medium the waveguiding light can be coupled out from the device.

4. Conclusion

In order to increase the light extraction efficiency of the OLED, the dielectric nanoparticles layer was inserted at glass substrate to extract the trapped light inside ITO-organic layer. Effects of refractive index, diameter and interparticle separation of dielectric nanoparticles were investigated to obtain the maximum enhancement in light extraction efficiency. As a result, it was estimated that the light extraction efficiency of the dielectric nanoparticles OLED could be enhanced up to 1.7 times than that of conventional device for the nanoparticles refractive index 1.1, the diameter of 100 nm and no interparticle separation.

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