

Nano-Dots Enhanced White Organic Light-Emitting Diodes

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14. ABSTRACT Significant improvement in electroluminescent performance was resulted by incorporating nano-dots in the small-molecule host composing and solution-processing emission layers for red, green, blue and white organic light-emitting devices (WOLEDs). The proper nano-dot was cadmium selenium (CdSe) prepared in organic solvent, which was miscible with the host solutions of 4,4'-bis (carbazol-9-yl) biphenyl (CBP) doped with red dye of bis [2-(2'-benzothienyl)-pyridinato-N,C3'] (acetyl-acetonate) iridium (III), green dye of tris(2-phenylpyridine) iridium (III) and/or blue dye bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) iridium (III). The power efficiency of the red OLEDs increased from 1.7 to 2.5 lm/W on the addition of 0.006 wt% CdSe. For the green OLEDs, it increased from 7.8 to 9.9 lm/W by adding 0.006 wt% CdSe. For the blue OLEDs, it increased from 1.45 to 1.66 lm/W by adding 0.01 wt% CdSe. The incorporation effect of CdSe was the most marked on the red device and the least on the blue device; i.e. that the power efficiency increment was 47% for the red device and 14.5% for the blue device. For the WOLEDs, it changed from 3.9 to 4.5 lm/W with a 15.4% increase in power efficiency as 0.002 wt% CdSe was incorporated, accompanied by a 5.7% increase in the resulted maximum luminance.					
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Abstract

Significant improvement in electroluminescent performance was resulted by incorporating nano-dots in the small-molecule host composing and solution-processing emission layers for red, green, blue and white organic light-emitting devices (WOLEDs). The proper nano-dot was cadmium selenium (CdSe) prepared in organic solvent, which was miscible with the host solutions of 4,4'-bis (carbazol-9-yl) biphenyl (CBP) doped with red dye of bis [2-(2'-benzothienyl)-pyridinato-N,C^{3'}] (acetyl-acetonate) iridium (III), green dye of tris(2-phenylpyridine) iridium (III) and/or blue dye bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) iridium (III). The power efficiency of the red OLEDs increased from 1.7 to 2.5 lm/W on the addition of 0.006 wt% CdSe. For the green OLEDs, it increased from 7.8 to 9.9 lm/W by adding 0.006 wt% CdSe. For the blue OLEDs, it increased from 1.45 to 1.66 lm/W by adding 0.01 wt% CdSe. The incorporation effect of CdSe was the most marked on the red device and the least on the blue device; i.e. that the power efficiency increment was 47% for the red device and 14.5% for the blue device. For the WOLEDs, it changed from 3.9 to 4.5 lm/W with a 15.4% increase in power efficiency as 0.002 wt% CdSe was incorporated, accompanied by a 5.7% increase in the resulted maximum luminance.

Introduction

White organic light-emitting devices (WOLEDs) are increasingly attracting attention for their potential applications in high-quality flat-panel displays, lighting and opto-electronics.¹⁻³ WOLEDs can be fabricated by vapor deposition of electro-luminescent dyes in a single- or multi-layer structure.⁴⁻¹² However, larger area size can be made more cheaply and easily by using solution-involved printing technologies such as screen printing and ink-jet printing.¹³⁻¹⁴ Especially, better composition control over the various dyes is highly obtainable in the solution process. Some prior studies have indeed attempted to solution-mix the desired dyes into polymer hosts,¹⁵⁻²⁰ or to solution-mix the desired light-emitting polymers.²¹ Nevertheless, better performance regarding chromaticity control and stability, brightness, and efficiency is still highly desirable before the maximum potential of the applications can be truly realized.

We have proved in a prior study that high efficiency WOLEDs can be made with the key white emission layer fabricated employing a molecular host, in lieu of polymeric one, and using solution processing.²² We have also showed in another prior study that the efficiency of OLEDs can be enhanced by the incorporation of nano-dots, similar to those observed in polymer-based OLEDs (PLEDs).²³⁻²⁸

We plan in this study to develop nano-dot enhanced WOLEDs by solution-processing. To achieve, we will first investigate the

best nano-dot materials for each of the composing red, green and blue dyes. The nano-dot materials to be examined are CdSe, ZnS and CdS. The three dyes will be chosen from the ones having performed in a previous study the best reported power efficiency (5.6 lm/W at 550 cd/m²) and maximum luminance (15,000 cd/m²) in a pure WOLED (shown in the last page). The best incorporation methods and compositions of the nano-dots will be examined. The ideal nano-dot materials will then be incorporated into the luminescent-active layer of the WOLED. Processing parameters such as thickness, composition, and dispersion method and sequence will be optimized to yield better efficiency and luminance.

Technical milestones to be achieved

- Milestone I** (1st - 2nd mo) — **Nano-dots enhanced green OLEDs**
- Milestone II** (3rd mo) — **Nano-dots enhanced red OLEDs**
- Milestone III** (4th mo) — **Nano-dots enhanced blue OLEDs**
- Milestone IV** (5th – 6th mo) — **Nano-dots enhanced white OLEDs**

The significance and scientific impact of each milestone.

Milestone I — Nano-dots enhanced green OLEDs

High brightness and high efficiency are most easily fabricated in green OLEDs. The incorporation effect of nano-dots can be clearly identified in these devices. The obtained knowledge will form the important base for more advanced study for the other devices like red, blue, and white OLEDs.

Milestone II — Nano-dots enhanced red OLEDs

Chromaticity of red OLEDs is sensitive to the host polarity and the red-dye concentration. It may also be affected by the presence of the nano-dots. Investigation of the effect of nano-dots is crucial for later fabrication of white OLEDs with desired chromaticity.

Milestone III — Nano-dots enhanced blue OLEDs

Blue OLEDs typically have the poorest brightness and efficiency. Blue emission plays a key role in the entire performance of a white OLED. Any positive effect of the addition of nano-dot is highly desirable. Highest blue-emission improvement will be exploited to warrant the success of fabrication of WOLEDs with much improved performance.

Milestone IV — Nano-dots enhanced white OLEDs

Nano-dot incorporated white OLEDs will be made and

characterized. The results will be compared against with those without the nano-dots. Highest white-emission improvement will be exploited. No scientific reports have yet revealed any related studies or findings concerning the topic. WOLEDs with a solution-processing white emission layer with much improved luminescent characteristics will make large-area display and illumination possible in a roll-to-roll mass production. Important scientific breakthrough and technological impact may be expected when nano-dots enhanced white OLEDs are realized.

Technical milestones achieved

- Milestone I** (1st - 2nd mo) — **Nano-dots enhanced green OLEDs**
- Milestone II** (3rd mo) — **Nano-dots enhanced red OLEDs**
- Milestone III** (4th mo) — **Nano-dots enhanced blue OLEDs**
- Milestone IV** (5th – 6th mo) — **Nano-dots enhanced white OLEDs**

Experimental

Figure 1 shows the device structure fabricated. The fabrication process included firstly the spin-coating at 4,000 rpm of a 40 nm poly(ethylene dioxythiophene): poly (styrene sulfonic acid) (PEDOT: PSS) on the pre- cleaned indium tin oxide (ITO) glass. The PEDOT: PSS layer was employed for hole-injection purpose. The second step was the spin-coating at 2,000 rpm under nitrogen at the ambient temperature of a 20 nm emission layer composing a molecular host doped with one of the three red, green and blue dyes.

The incorporation of nano-dots into the host layer was done by mixing the solution of the nano-dot with the dye-doped host solution prior to the above spin-coating. After being dried at 100 °C under vacuum for 1 hr, a 15 nm hole-blocking layer of bis (2-methyl-8-quinolinolate)-4-(phenyl phenolato)-aluminum (BALq) and a 20 nm electron-transporting layer of tris(8-hydroxyl-quinoline)-aluminum (Alq₃) were sequentially deposited at 2×10^{-5} torr. Finally deposited were a 0.5 nm electron-injection layer of lithium-fluoride (LiF) and a 120 nm electrode of aluminum (Al).

The studied molecular host was 4,4'-bis(carbazol-9-yl) biphenyl (CBP) and dyes were red of bis[2-(2'-benzothienyl)-pyridinato-N,C^{3'}] (acetyl-acetonate) iridium (III) [Btp₂Ir(acac)], green of tris(2-phenylpyridine) iridium (III) [Ir(ppy)₃] and blue of bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) iridium (III) (Flrpic).

Two series of nano-dots were investigated, one of which was in aqueous solution and the other in organic solution. The studied nano-dots were cadmium tellurium (CdTe) and cadmium selenium (CdSe). The CdTe nano-dot that prepared in aqueous solution was found immiscible with the host solution prepared in organic solvent such as toluene, resulting in no performance improvement. However, the CdSe nano-dot prepared in organic solvent was miscible with the dye-doped host solution and the emission layer with good film integrity was obtainable. Figure 2 schematically shows the structure of the CdSe nano-dot studied.

The resultant electro-luminescent characteristics were determined by using Minolta LS110 luminance meter and Photo Research PR650 spectrophotometer. All the measurements were carried out at the ambient condition.

Results and discussion

CdSe nano-dot enhanced red OLEDs

Figure 3 shows the molecular structures of the red dye of **Btp₂Ir(acac)** and the host of CBP and the photo of the resultant red OLED at emission. The device composes structure of ITO/PEDOT: PSS/CBP: **6 wt% Btp₂Ir(acac)**: x wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

Figure 4 shows the effect of CdSe nano-dot incorporation on the resultant brightness of the red light-emitting device. The maximum brightness first increases with the increase of the incorporation concentration of the CdSe nano-dot. It then starts to decrease as the CdSe nano-dot incorporation concentration is greater than 0.004%.

Figure 5 shows its effect on the current density. The resultant current density is unanimously higher for the red light-emitting devices incorporated with the CdSe nano-dots than that without.

Both the brightness and current density curves shift toward the lower voltage side, indicating the addition of the CdSe nano-dots has enhanced the injection of carriers. Lower driving voltage is then resulted and brighter emission is observed. A maximum brightness of 6,100 cd/m² is observed as the

incorporation concentration is 0.004%. Whilst, it is 5,200 cd/m² before the addition of the nano-dots.

Figure 6 shows the effect of CdSe nano-dot incorporation on the power efficiency of the red light-emitting device. A maximum power efficiency of 2.5 lm/W is obtained as the incorporation concentration is 0.006%. Whilst, it is 1.7 lm/W without the addition of the nano-dots.

Table I summarizes the effects of the incorporation concentration of CdSe nano-dots on the red light-emitting devices. All the CdSe nano-dot incorporated red devices need a driving voltage lower than that required to emit the device without nano-dot. The driving voltage is defined herein as the voltage required to generate a brightness greater than 1 lumen (cd/m²).

CdSe nano-dot enhanced green OLED

Figure 7 shows the molecular structures of the green dye of Ir(ppy)_3 and the host of CBP and the photo of the resultant green light-emitting device at emission. The device composes structure of ITO/PEDOT: PSS/CBP: 6 wt% Ir(ppy)_3 : x wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

Figure 8 shows the effect of CdSe nano-dot incorporation on the resultant brightness of the green light-emitting device. The maximum brightness decreases with the incorporation of the CdSe nano-dot. The maximum brightness is of 30,700 cd/m^2 without the nano-dot and drops to from 25,700 to 26,700 cd/m^2 as the incorporation concentrations of the nano-dots are ranging from 0.002% to 0.006%.

Figure 9 shows its effect on the current density. The resultant current density is also unanimously lower for the green light-emitting devices incorporated with the CdSe nano-dots than that without.

Figure 10 shows the effect of CdSe nano-dot incorporation on the power efficiency of the green light-emitting device. A maximum power efficiency of 9.9 lm/W is obtained as the incorporation concentration is 0.006%. Whilst, it is 7.8 lm/W without the addition of the nano-dots.

Table II summarizes the effects of the incorporation concentration of CdSe nano-dots on the green light-emitting devices. The driving voltages are nearly all the same for all the green devices with or without CdSe nano-dot; i.e. that they are around 4 V.

CdSe nano-dot enhanced blue OLED

Figure 11 shows the molecular structures of the blue dye of Flrpic and the host of CBP and the photo of the resultant blue light-emitting device at emission. The device composes structure of ITO/PEDOT: PSS/CBP: 8 wt% Flrpic: x wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

Figure 12 shows the effect of CdSe nano-dot incorporation on the resultant brightness of the blue light-emitting device. Figure 13 shows its effect on the current density. The maximum brightness continuously increases with the increase of the incorporation concentration. The maximum brightness is the highest at 0.01% incorporation concentration, which is 7,700 cd/m². Whilst, it is 6,200 cd/m² without the nano-dot.

Figure 14 shows the effect of CdSe nano-dot incorporation on the power efficiency of the blue light-emitting device. Best enhancement is observed at 0.01% incorporation concentration, which is 1.66 lm/W. Whilst, it is 1.45 lm/W without the nano-dot.

Table III summarizes the effects of the incorporation concentration of CdSe nano-dots on the blue light-emitting devices. Similar to those observed in the green devices, the driving voltages are nearly all the same for all these blue devices with or without CdSe nano-dot; i.e. that they are around 5.5 V.

CdSe nano-dot enhanced WOLED

Figure 15 shows the molecular structures of the three dyes of red, green and blue and the host of CBP and the photo of the resultant white light-emitting device at emission. The device composes structure of ITO/PEDOT: PSS/white emission: x wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

Figure 16 shows the effect of CdSe nano-dot incorporation on the resultant brightness of the white light-emitting device. The brightness first increases as CdSe increases to 0.002wt%, and then decreases as CdSe further increases to 0.004wt% and above. The maximum brightness is the highest at 0.002% incorporation concentration, which is 9,475 cd/m². Whilst, it is 8,963 cd/m² without the nano-dot. Figure 17 shows its effect on the current density.

Figure 18 shows the effect of CdSe nano-dot incorporation on the power efficiency of the white light-emitting device. Best enhancement is observed at 0.002wt% incorporation concentration, which is 4.5 lm/W. Whilst, it is 3.9 lm/W without the nano-dot.

Table IV summarizes the effects of the incorporation concentration of CdSe nano-dots on the white light-emitting devices. The incorporation of CdSe shows little effect on the resulted emission; e.g. its original color at 100 cd/m² is nearly

pure white with CIE coordinates of (0.342, 0.386) without CdSe and remains nearly unchanged with CIE coordinates of (0.342, 0.389) with 0.002wt% CdSe.

Conclusions

CdSe nano-dots are successfully incorporated via solution-processing into the small-molecule host employed emission layers to enhance the electro-luminescent characteristics of the red, green and blue light-emitting devices as well as their composing white light-emitting devices. The enhancement is especially marked in power efficiency. The power efficiency of the red OLEDs increased from 1.7 to 2.5 lm/W on the addition of 0.006 wt% CdSe. For the green OLEDs, it increased from 7.8 to 9.9 lm/W by adding 0.006 wt% CdSe. For the blue OLEDs, it increased from 1.45 to 1.66 lm/W by adding 0.01 wt% CdSe. The incorporation effect of CdSe was the most marked on the red device and the least on the blue device; i.e. that the power efficiency increment was 47% for the red device and 14.5% for the blue device. For the WOLEDs, it changed from 3.9 to 4.5 lm/W with a 15.4% increase in power efficiency as 0.002 wt% CdSe was incorporated, accompanied by a 5.7% increase in the resulted maximum luminance.

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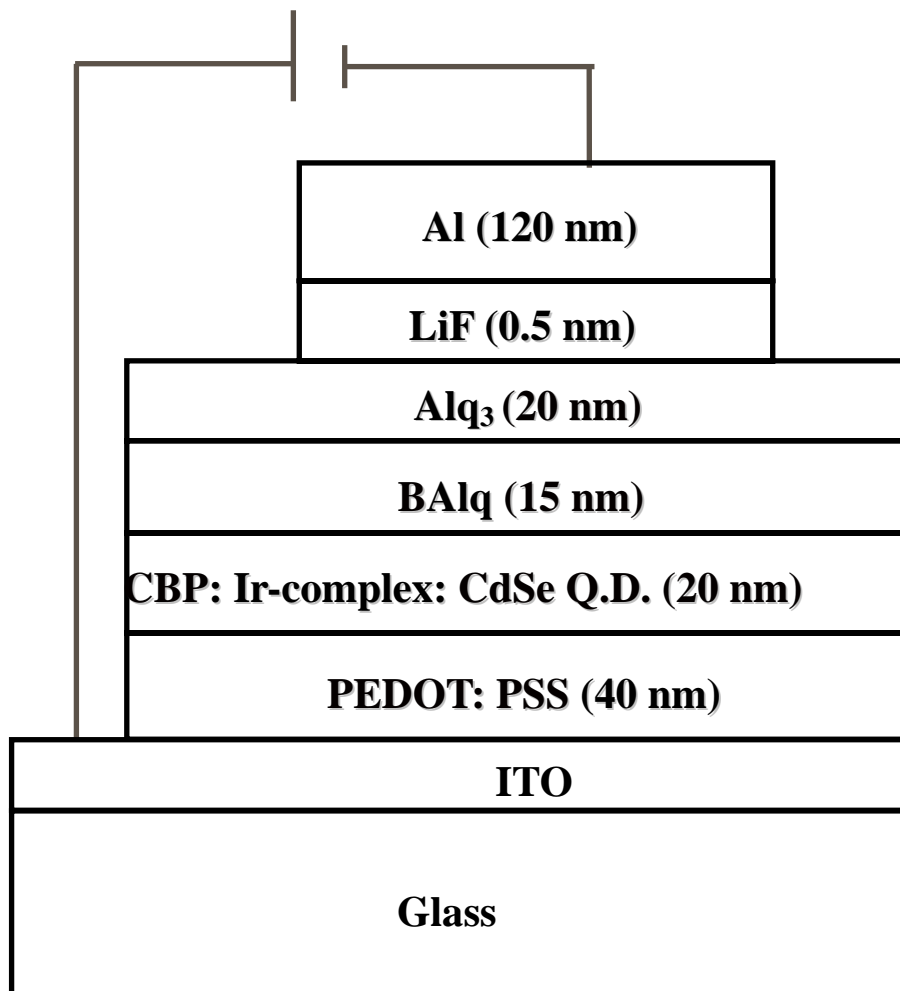


Figure 1. Device structure of the studied nano-dot enhanced OLEDs.

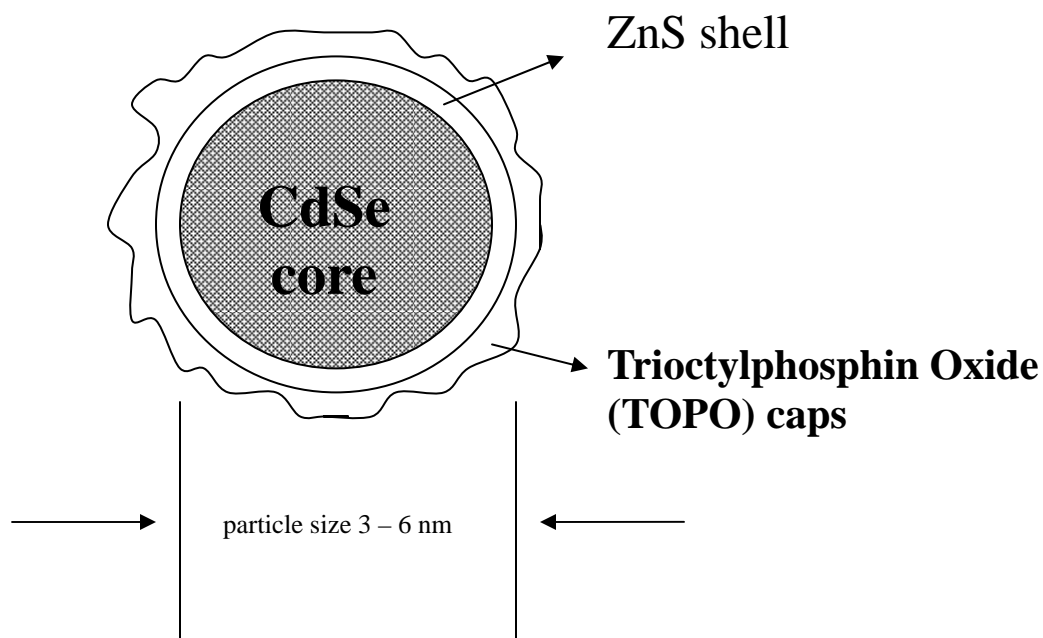
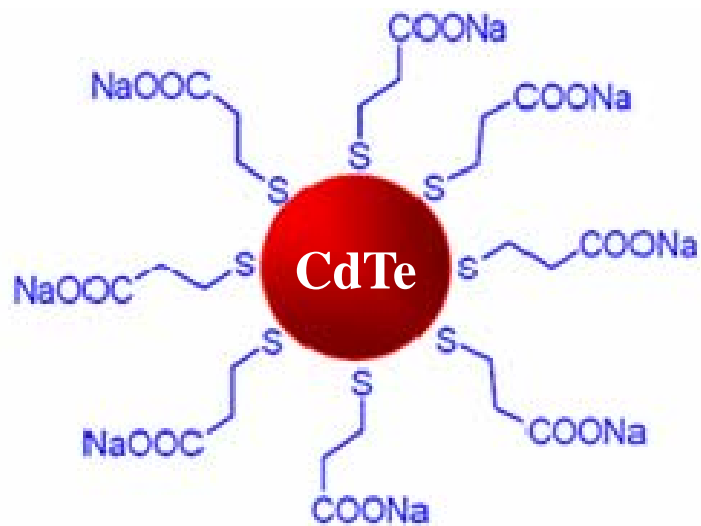
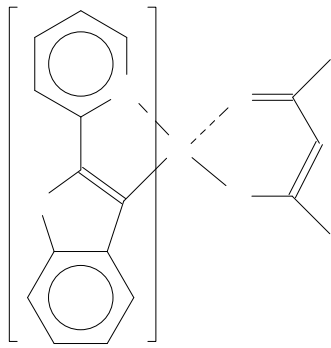
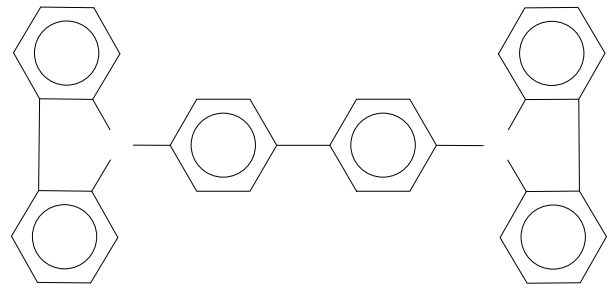


Figure 2 Schematic structures of the studied CdTe (top) and CdSe (bottom) nano-dots.



Btp₂Ir(acac)



CBP

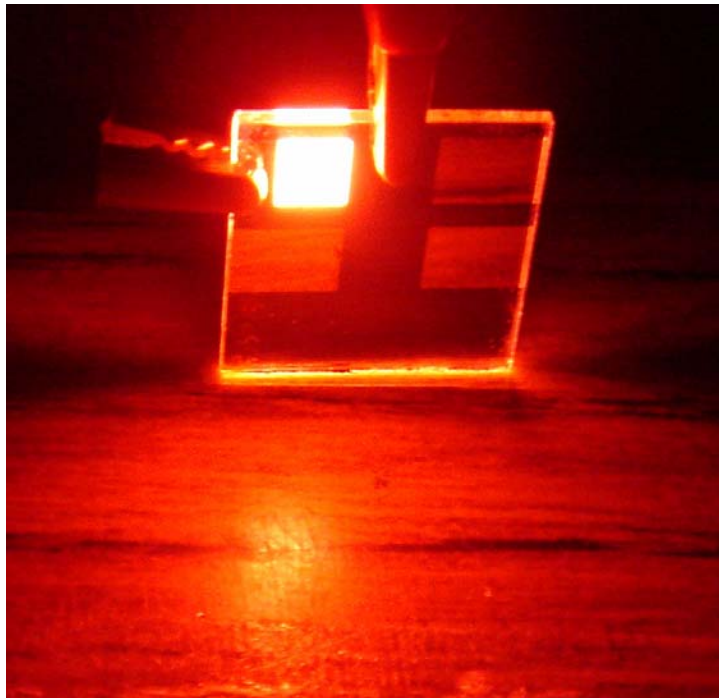


Figure 3. Molecular structures of the studied red dye Btp₂Ir(acac) and the CBP host (top) and photo of the resultant luminescence for the device composing ITO/PEDOT: PSS/CBP: 6 wt% Btp₂Ir(acac): X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al (bottom).

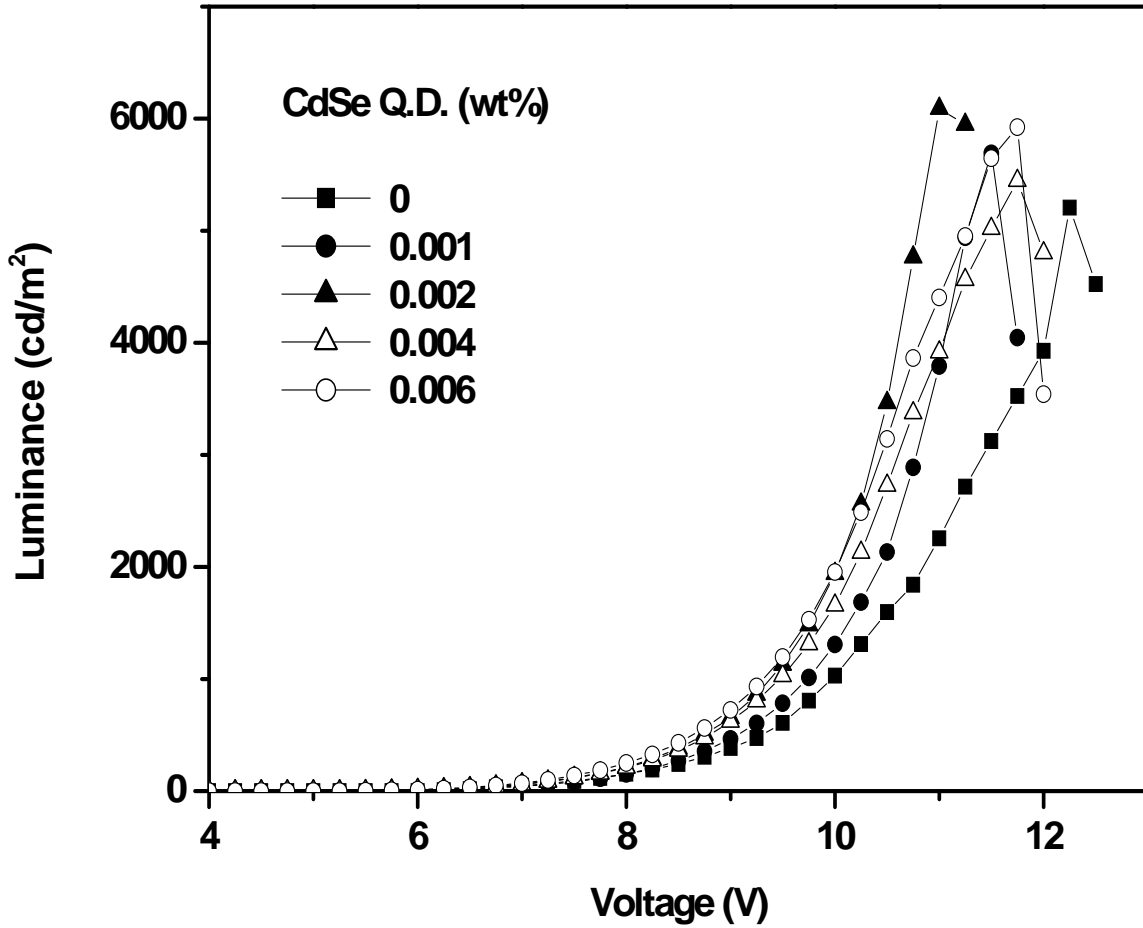


Figure 4. Doping concentration effect of CdSe nano-dot on the luminescent behaviors of the red device of ITO/PEDOT: PSS/CBP: 6 wt% Btp2Ir(acac): X wt% CdSe quantum dots/BAIq/Alq3/LiF/Al.

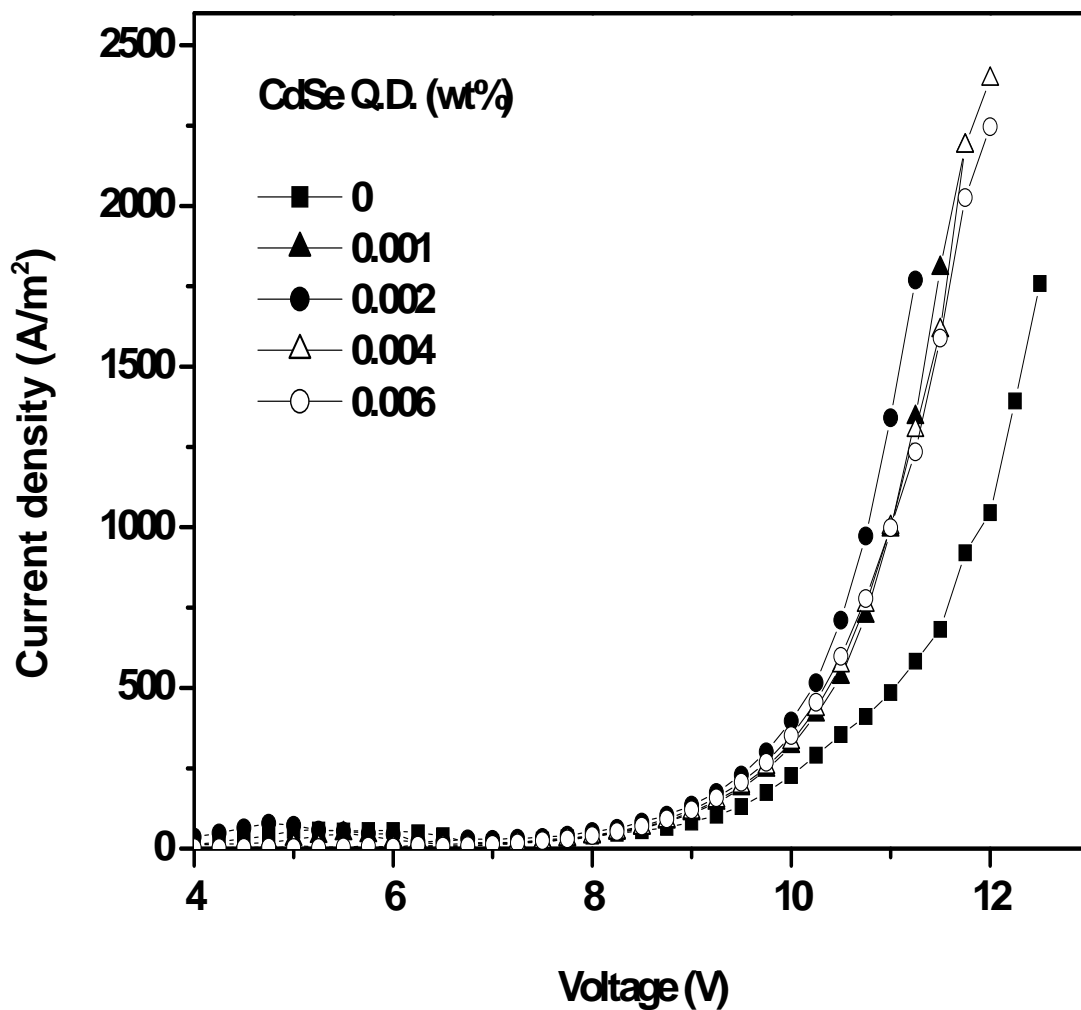


Figure 5. Doping concentration effect of CdSe nano-dot on the current density behaviors of the red device of ITO/PEDOT: PSS/CBP: 6 wt% Btp2Ir(acac): X wt% CdSe quantum dots/BAIq/Alq3/LiF/ Al.

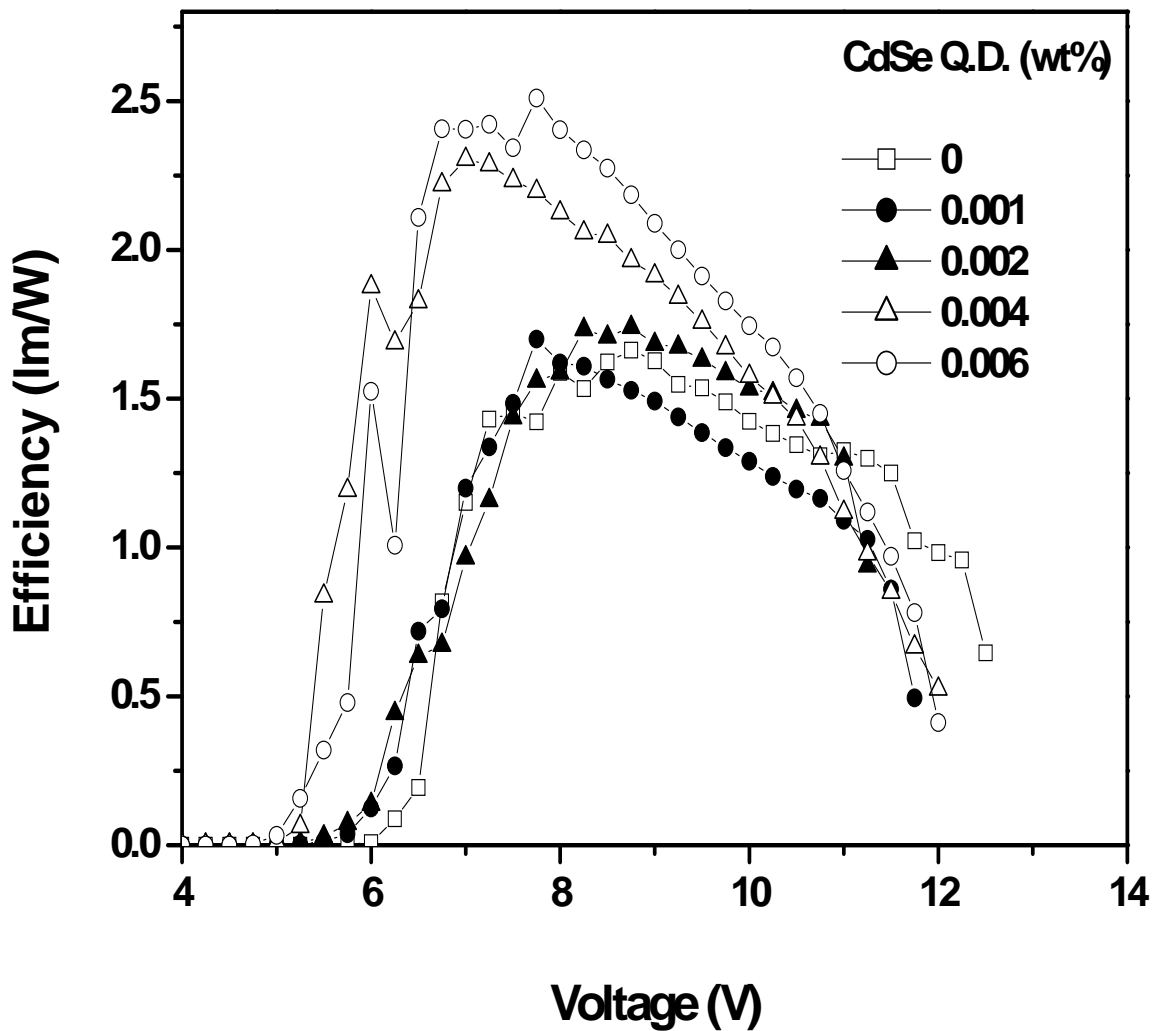
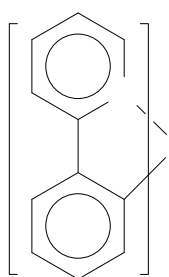


Figure 6. Doping concentration effect of CdSe nano-dot on the power efficiency of the red device of ITO/PEDOT: PSS/CBP: 6 wt% Btp2Ir(acac): X wt% CdSe quantum dots/BAIq/Alq3/LiF/ Al.

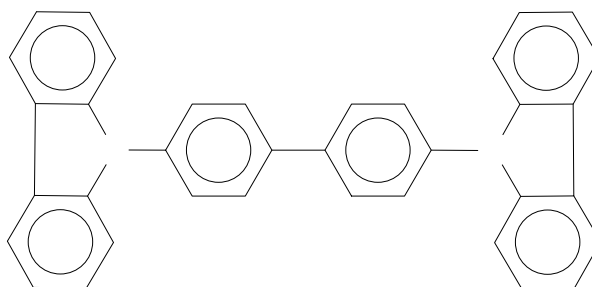
Table I. Doping concentration effect of CdSe nano-dot on the luminescent characteristics of the red light-emitting device.

CdSe Q.D. concentration (wt%)		Turn-on voltage (V)	Maximum efficiency (lm/W) [the corresponding luminance (cd/m ²)]	Maximum luminance (cd/m ²)
Red ^a	0	6	1.7 [306]	5,200
	0.001	5.25	1.7 [157]	5,500
	0.002	5.25	1.7 [562]	5,900
	0.004	5.25	2.3 [62]	6,100
	0.006	5.5	2.5 [114]	5,700

^a red dye of Btp₂Ir(acac)



Ir(ppy)₃



CBP

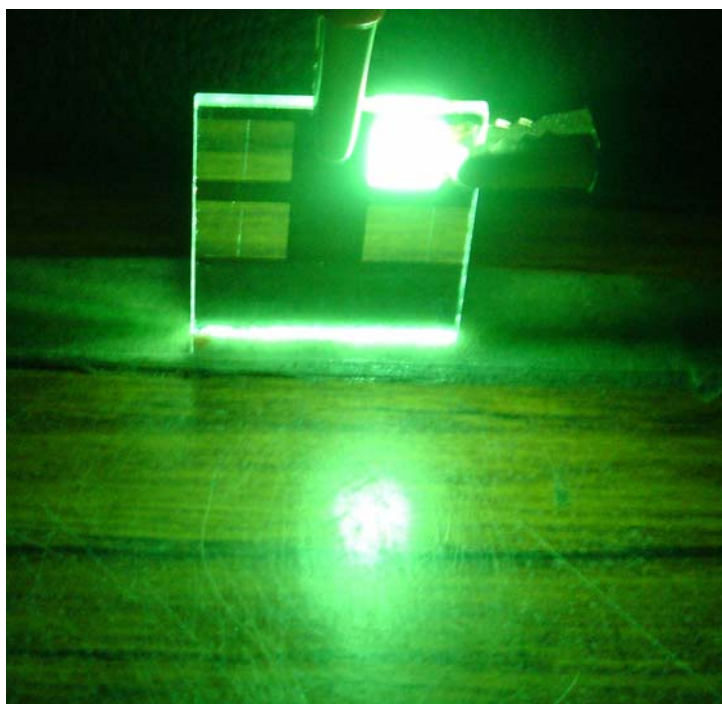


Figure 7. Molecular structures of the studied green dye Ir(ppy)₃ and the CBP host (top) and photo of the resultant luminescence for the device composing ITO/PEDOT: PSS/CBP: 6 wt% Ir(ppy)₃: X wt% CdSe quantum dots /BAIq/ Alq₃/ LiF/ Al (bottom).

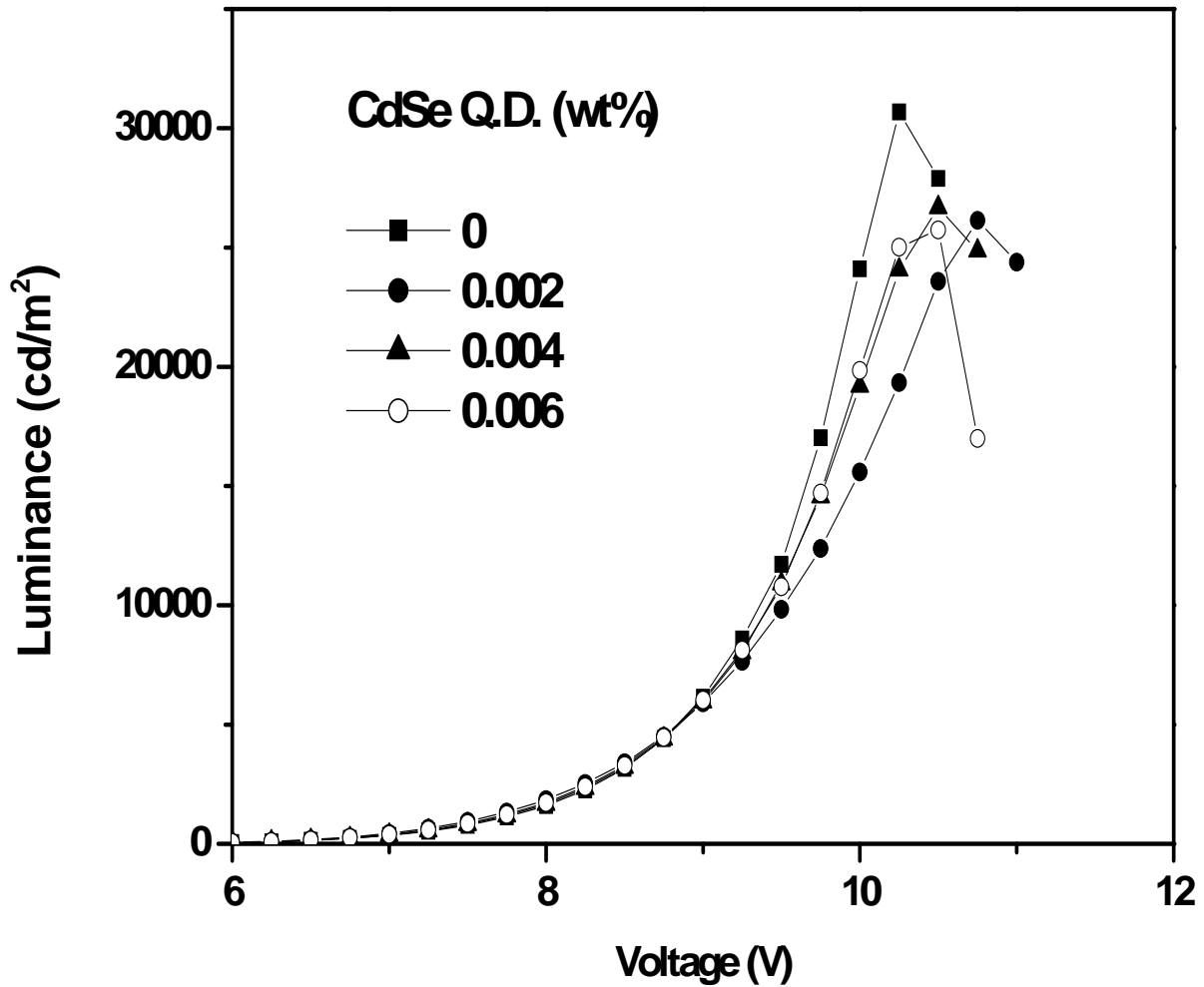


Figure 8. Doping concentration effect of CdSe nano-dot on the luminescent behaviors of the green device of ITO/PEDOT: PSS/CBP: 6 wt% Ir(ppy)₃: X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

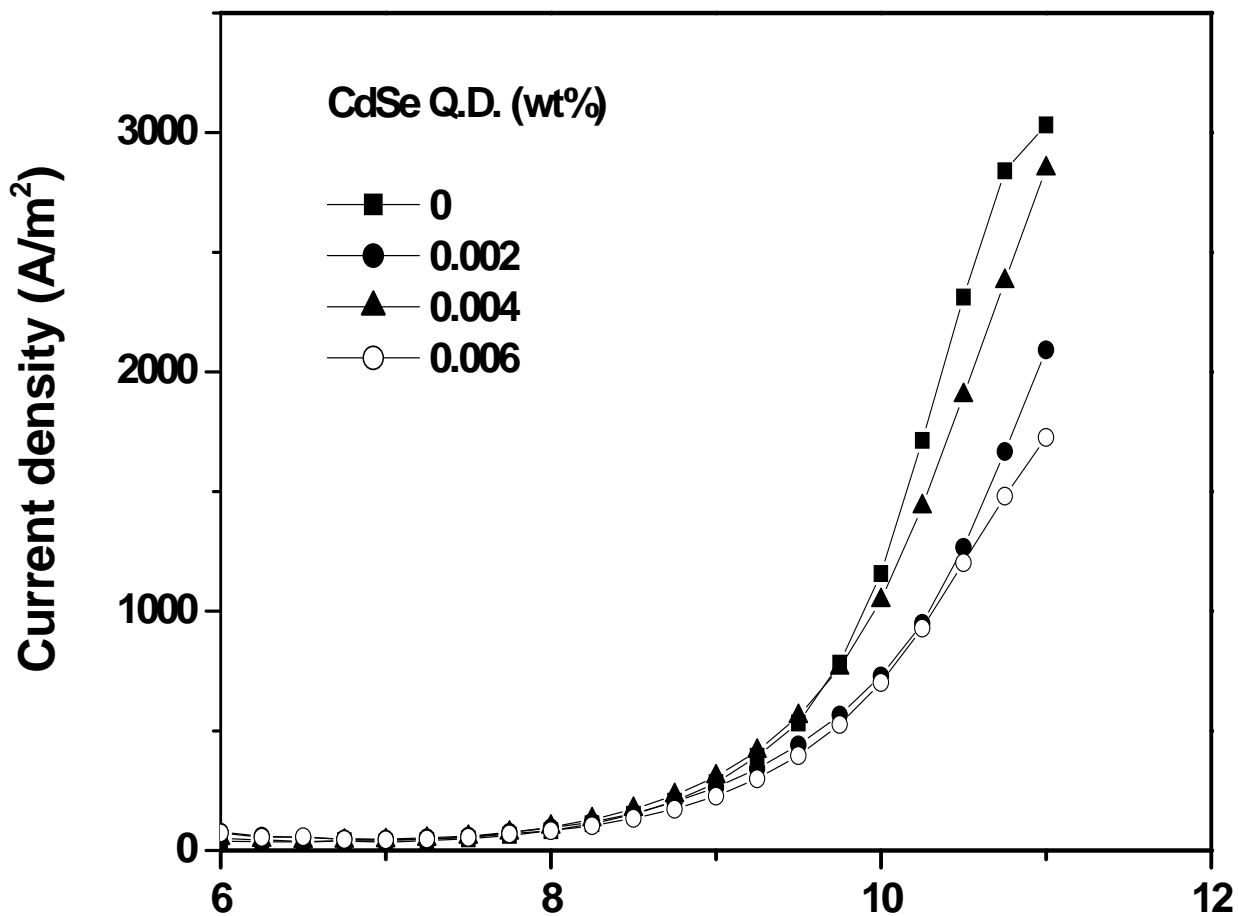


Figure 9. Doping concentration effect of CdSe nano-dot on the current density behaviors of the green device of ITO/PEDOT: PSS/CBP: 6 wt% Ir(ppy)₃: X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

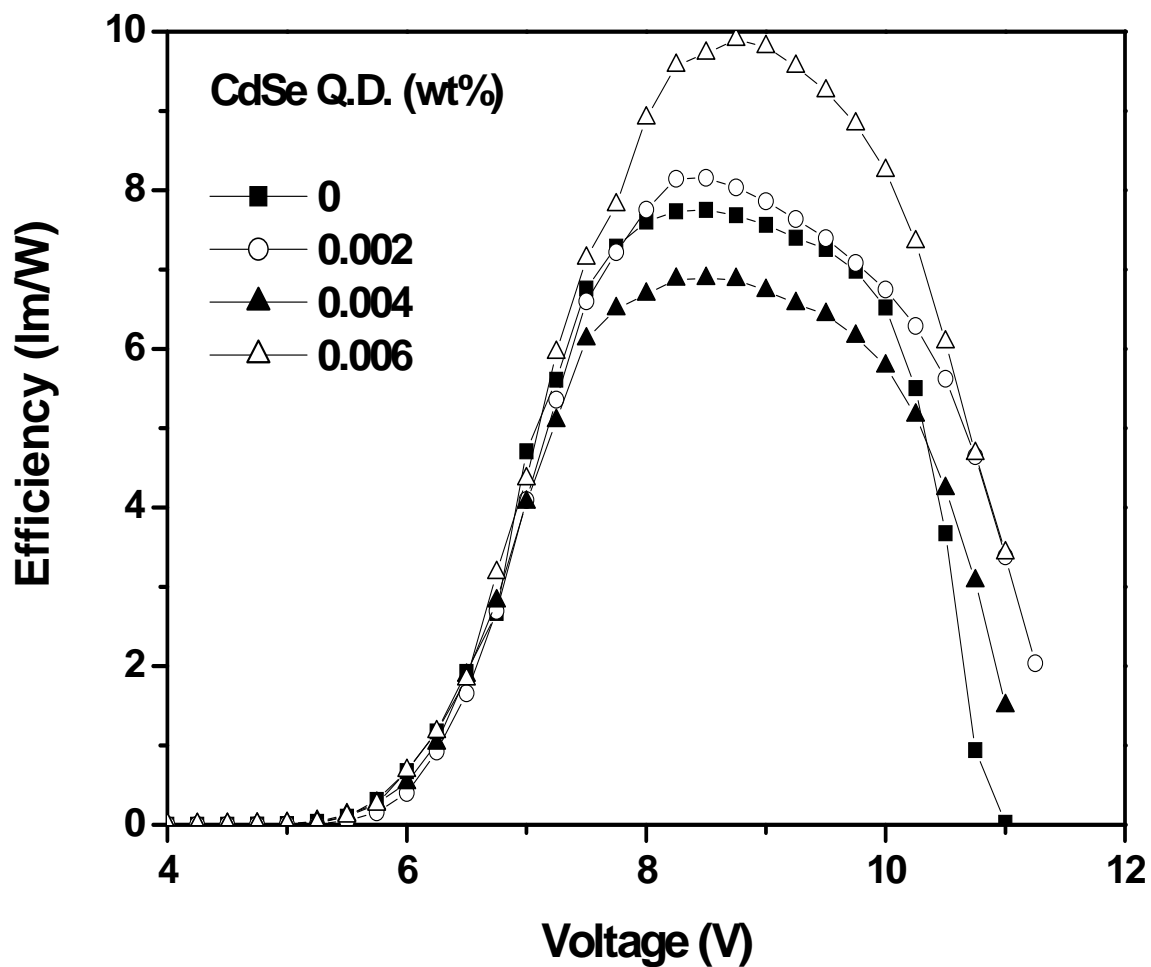
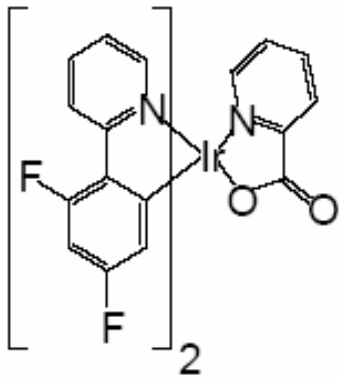


Figure 10. Doping concentration effect of CdSe nano-dot on the power efficiency of the green device of ITO/PEDOT: PSS/CBP: 6 wt% Ir(ppy)₃: X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

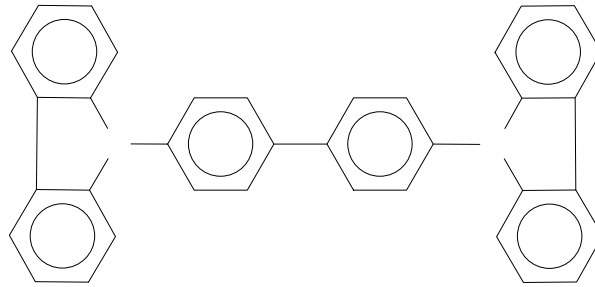
Table II. Doping concentration effect of CdSe nano-dot on the luminescent characteristics of the green light-emitting device.

CdSe Q.D. concentration (wt%)		Turn-on voltage (V)	Maximum efficiency (lm/W) [the corresponding luminance (cd/m²)]	Maximum luminance (cd/m²)
Green^a	0	4	7.8 [3163]	30,700
	0.002	4	8.2 [3395]	26,100
	0.004	3.75	6.9 [3206]	26,700
	0.006	4	9.9 [3279]	25,700

^agreen dye of Ir(ppy)₃



Firpic



CBP

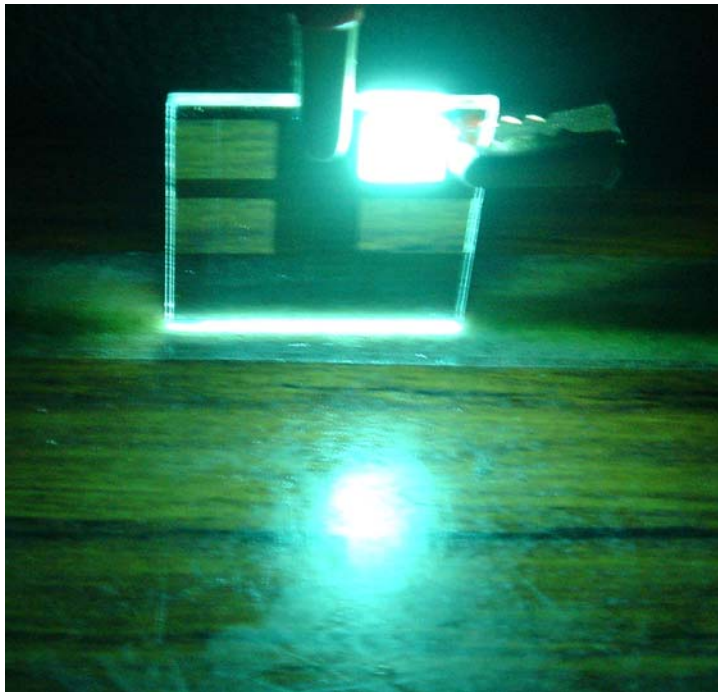


Figure11. Molecular structures of the studied blue dye Firpic and the CBP host (top) and photo of the resultant luminescence for the device composing ITO/PEDOT: PSS/CBP: 6 wt% Firpic: X wt% CdSe quantum dots /BAIq/ Alq₃/ LiF/ Al (bottom).

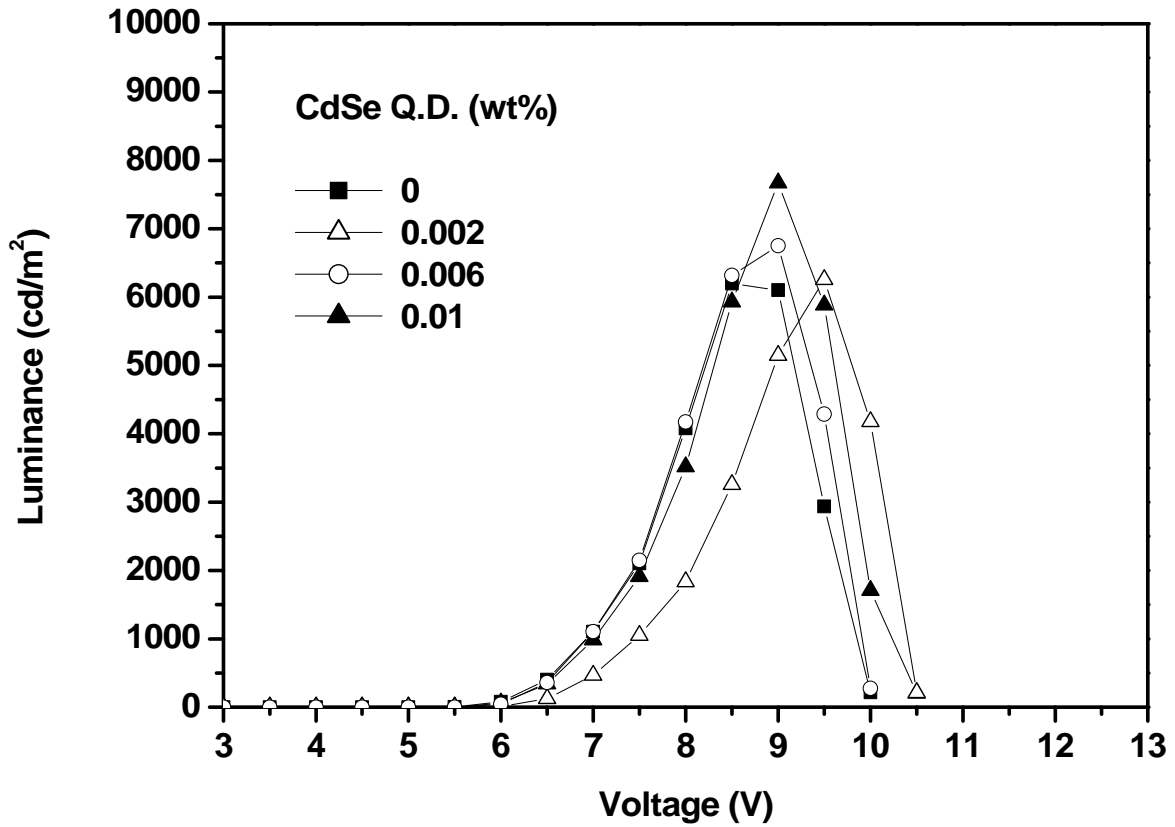


Figure 12. Doping concentration effect of CdSe nano-dot on the luminescent behaviors of the blue device of ITO/PEDOT: PSS/CBP: 8 wt% FIrpic: x wt% CdSe quantum dots/BAIq/Alq₃/LiF/Al.

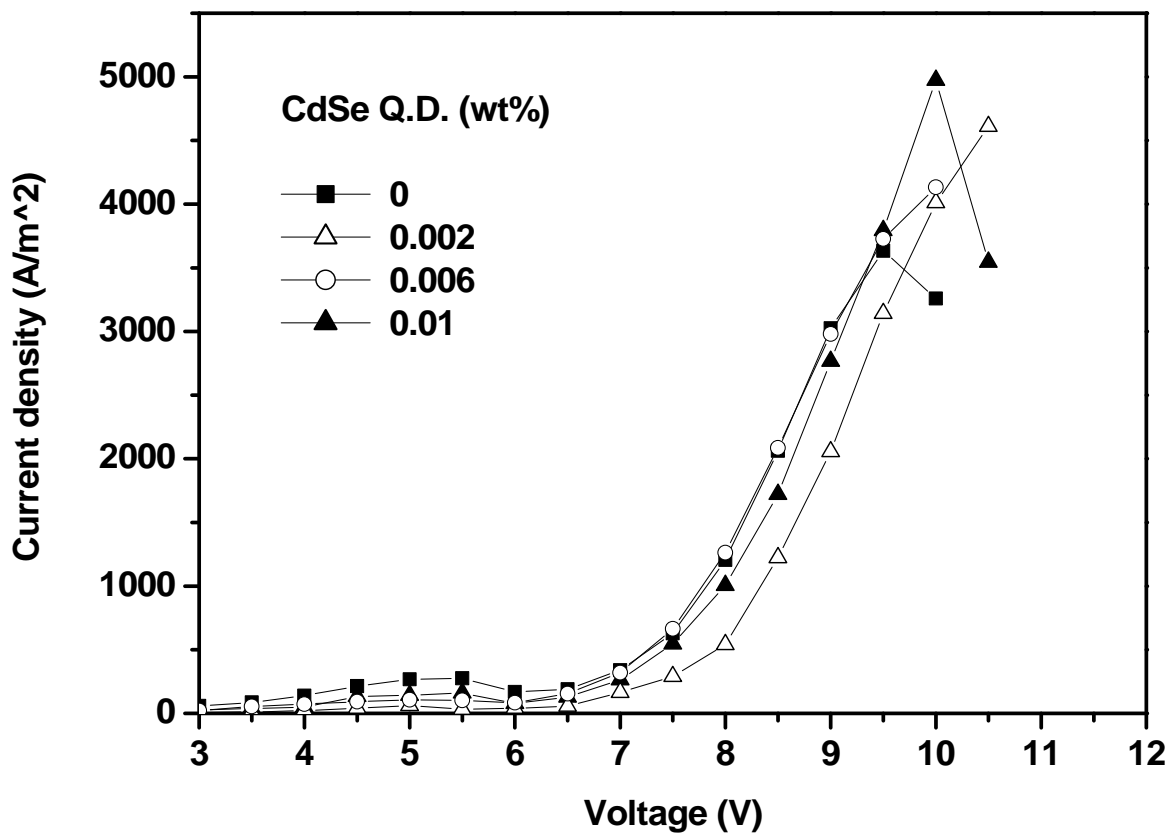


Figure 13. Doping concentration effect of CdSe nano-dot on the current density behaviors of the blue device of ITO/PEDOT: PSS/CBP: 8 wt% FIrpic: x wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

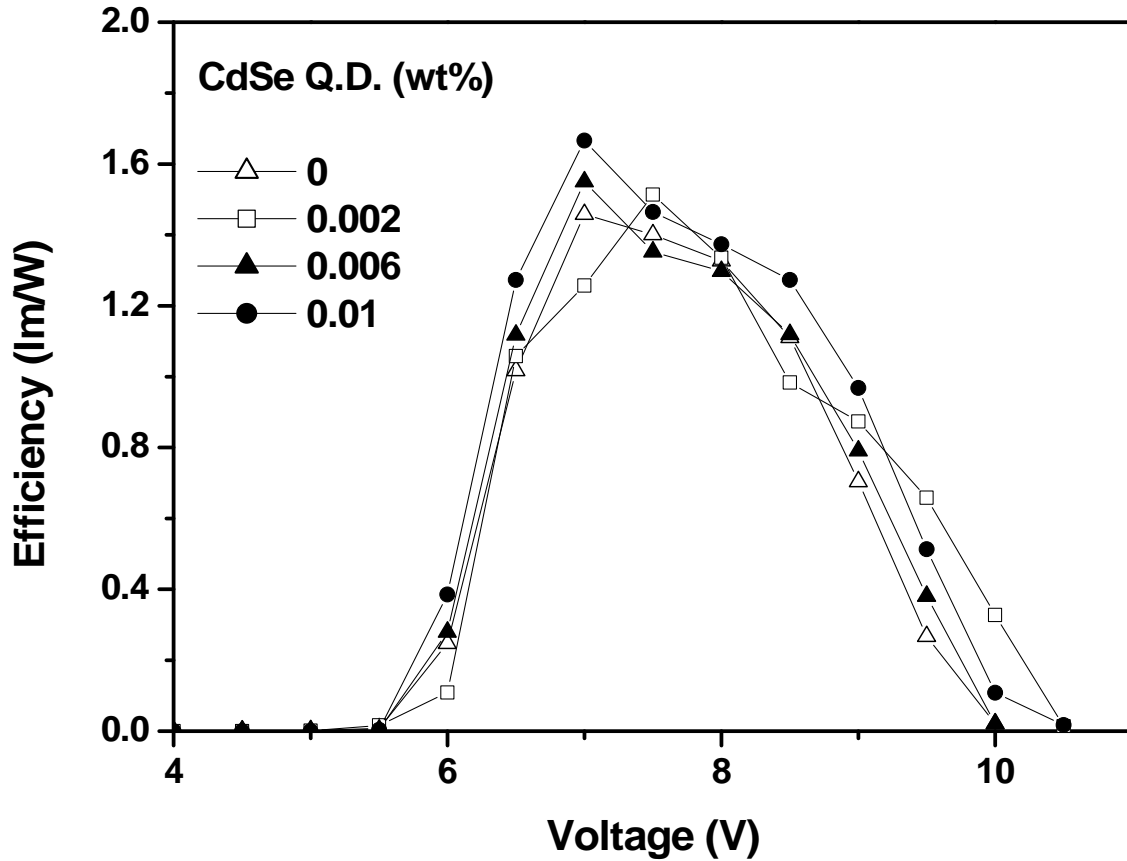
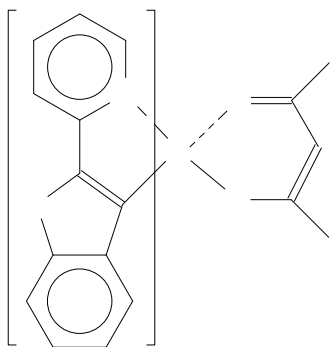


Figure 14. Doping concentration effect of CdSe nano-dot on the power efficiency of the blue device of ITO/PEDOT: PSS/CBP: 8 wt% FIrpic: X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

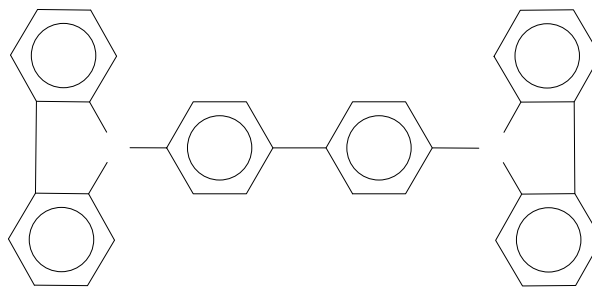
Table III. Doping concentration effect of CdSe nano-dot on the luminescent characteristics of the blue light-emitting device.

CdSe Q.D. concentration (wt%)		Turn-on voltage (V)	Maximum efficiency (lm/W) [the corresponding luminance (cd/m ²)]	Maximum luminance (cd/m ²)
Blue ^a	0	5	1.45 [1104]	6,169
	0.002	5.5	1.51 [1048]	6,256
	0.006	5.5	1.55 [1106]	6,751
	0.010	5.5	1.66 [983]	7,671

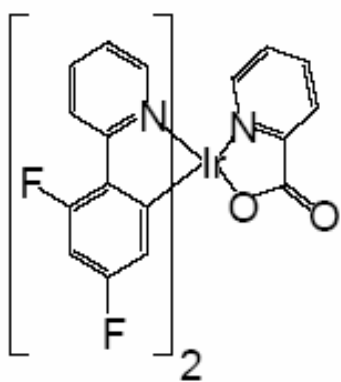
^ablue dye of Firpic



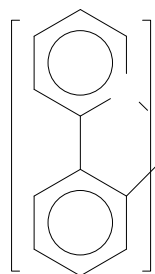
Btp₂Ir(acac)



CBP



Ir(ppy)₃



FIrpic

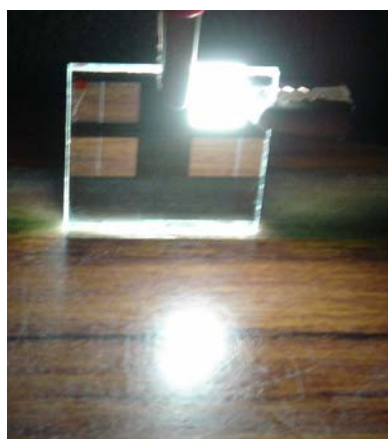


Figure 15. Molecular structures of the studied red Btp₂Ir(acac), green Ir(ppy)₃ and blue(FIrpic) dyes and the CBP host (top) and photo of the resultant luminescence for the device composing ITO/PEDOT: PSS/white emission: X wt% CdSe quantum dots /BAIq/ Alq₃/ LiF/ Al (bottom).

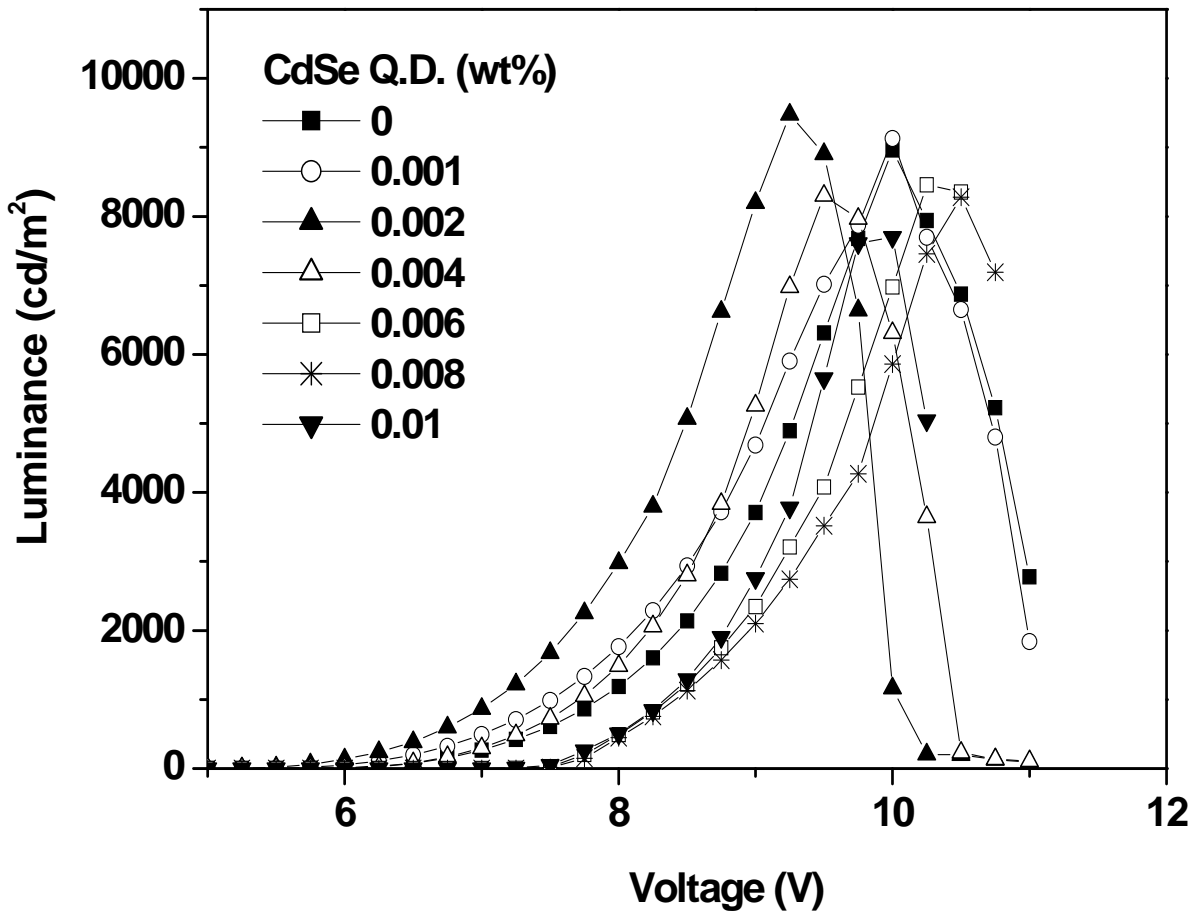


Figure 16. Doping concentration effect of CdSe nano-dot on the luminescence of the white device of ITO/PEDOT: PSS/white emission: X wt% CdSe quantum dots/BAlq/Alq₃/LiF/ Al.

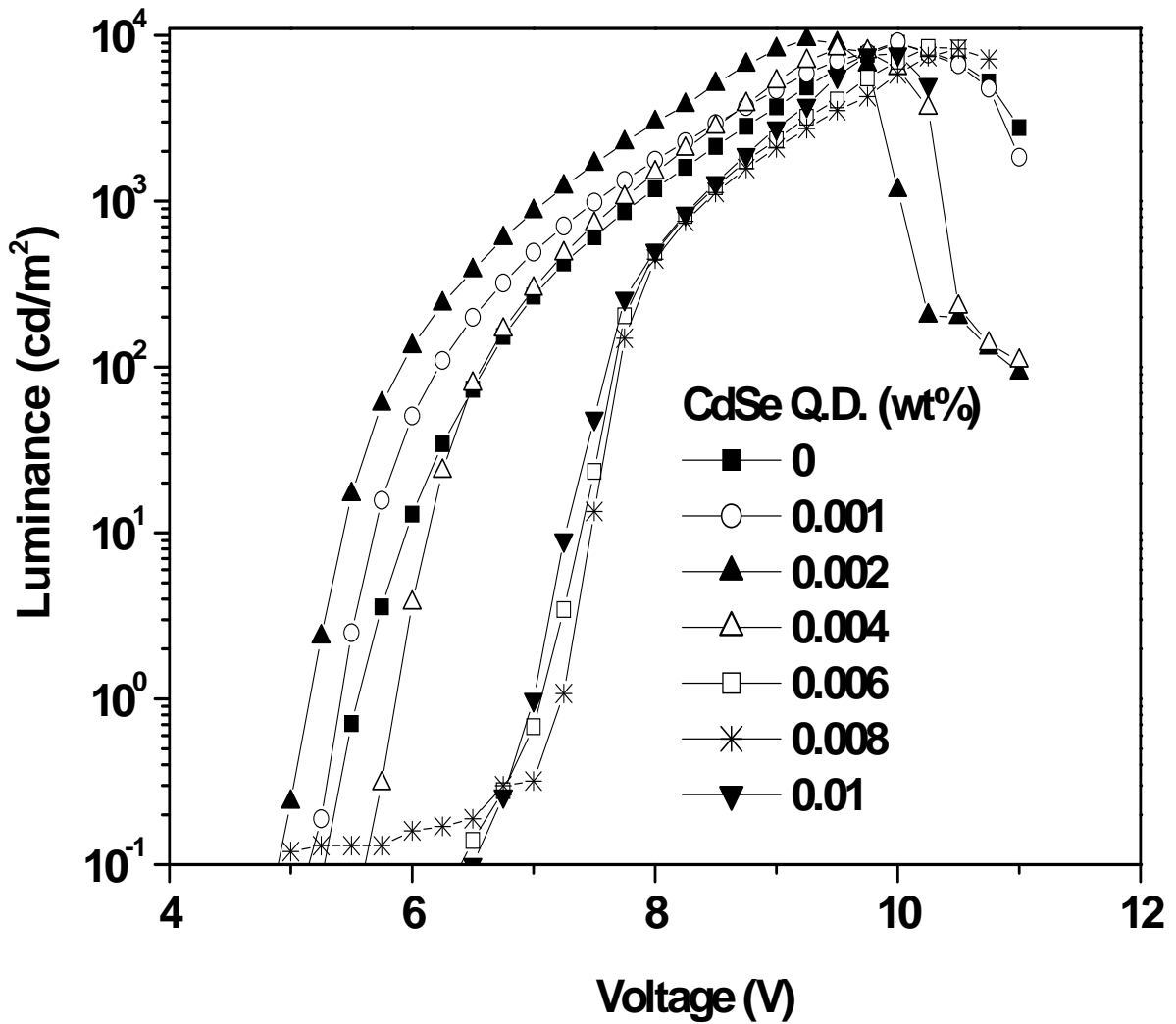


Figure 16. Doping concentration effect of CdSe nano-dot on the luminescence of the white device of ITO/PEDOT: PSS/white emission: X wt% CdSe quantum dots/BAIq/Alq₃/LiF/ Al.

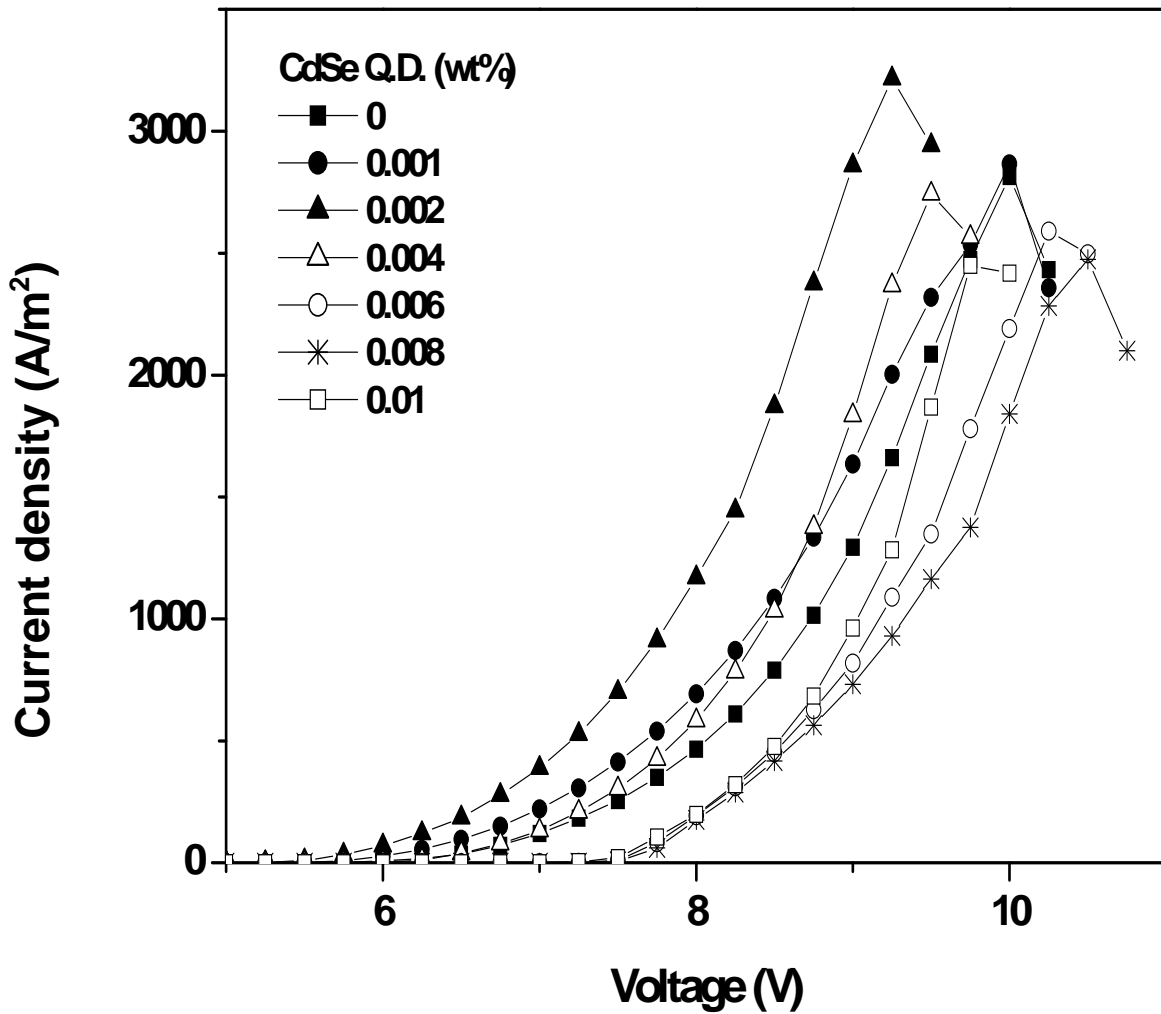


Figure 17. Doping concentration effect of CdSe nano-dot on the current density of the white device of ITO/PEDOT: PSS/white emission: X wt% CdSe quantum dots/BAIq/Alq3/LiF/ Al.

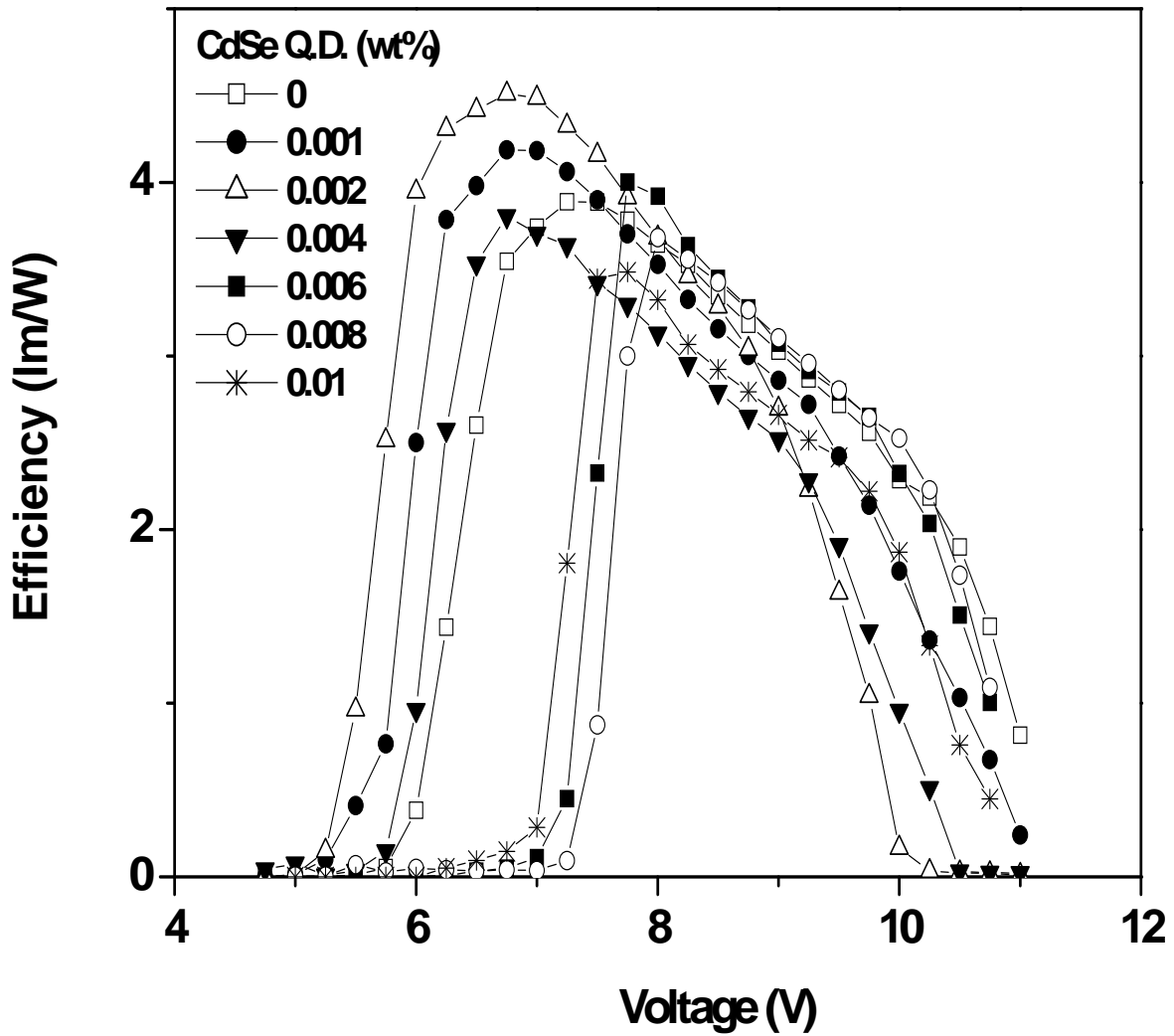


Figure 18. Doping concentration effect of CdSe nano-dot on the power efficiency of the white device of ITO/PEDOT: PSS/white emission: X wt% CdSe quantum dots/BAIq/Alq3/LiF/ Al.

Table IV. Doping concentration effect of CdSe nano-dot on the luminescent characteristics of the white light-emitting device.

CdSe Q.D. (wt%)	Max. Luminance (cd/m²)	Max. Efficiency (lm/W)	CIE (x,y) at 100 (cd/m²)	CIE (x,y) at 10000 (cd/m²)
0	8963	3.9	(0.342, 0.386)	(0.3210, 0.391)
0.001	9128	4.2	(0.347, 0.380)	(0.322, 0.387)
0.002	9475	4.5	(0.342, 0.389)	(0.323, 0.390)
0.004	8301	3.8	(0.339, 0.388)	(0.323, 0.385)
0.006	8457	4.0	(0.342, 0.377)	(0.335, 0.377)
0.008	8277	3.7	(0.348, 0.380)	(0.339, 0.380)
0.01	7700	3.5	(0.341, 0.371)	(0.331, 0.370)